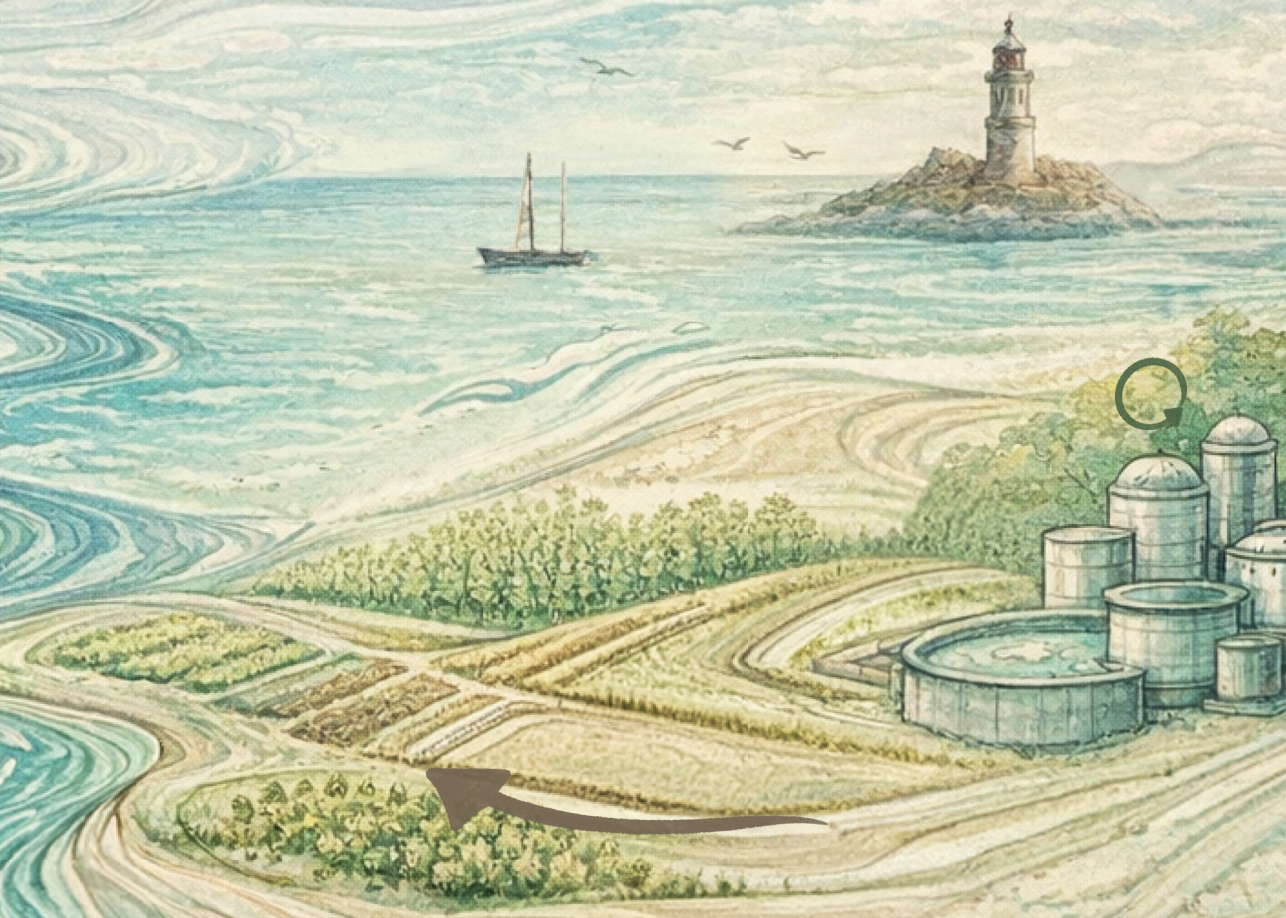
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Integrated modeling of nutrient emissions to air and water:

Synergetic solutions for the European future



Aslıhan Ural-Janssen

Propositions

1. Regional variability in human activities offers opportunities for reducing nutrient emissions to the air and surface waters simultaneously.
   (this thesis)

2. Bio-based fertilizers are by themselves not enough to reduce future nutrient emissions to the air and surface waters synergistically.
   (this thesis)

3. The plethora of scientific terms on a particular issue delays or impedes decision-making and acting.

4. While the use of AI saves time in scientific research and writing, it eliminates the originality and creativity of mankind.

5. In academia, sustaining motivation and passion outweigh research and technical skills.

6. Working with colleagues from diverse disciplinary and national backgrounds widens the horizon of PhDs beyond research.

7. The perception of maternity leave in academic careers reinforces gender disparities in science.

Propositions belonging to the thesis, entitled

Integrated modeling of nutrient emissions to air and water:
Synergetic solutions for the European future

Aslıhan Ural-Janssen,
Wageningen, 1 June 2026

Integrated modeling of nutrient emissions to air and water:

Synergetic solutions for the European future

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Integrated modeling of nutrient emissions to air and water:
Synergetic solutions for the European future

Aslıhan Ural-Janssen

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This thesis is dedicated to my parents.

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Chapters 2, 3 and 4 have been published as peer-reviewed scientific articles. The text, figures, and tables of the published articles have been adjusted to the PhD thesis format. Editorial changes were made for reasons of uniformity of presentation in this thesis. References should be made to the published articles.

1

Chapter 1

Introduction

1.1 Background

Motivation: My interest in environmental protection began in secondary school with a book called "Environmental Sciences". I was curious about how human activities contribute to environmental pollution and how to reduce this. Coming from a country surrounded by large coasts and having a strong liking for swimming, I wanted to learn the causes of coastal water pollution and ways to protect it. With a BSc in Environmental Engineering, I improved my knowledge of environmental compartments (air, water, soil) and gained an interest in air pollution control. In my MSc, I focused on modeling and coastal water pollution with nutrients in Europe. Later at the Ministry of Agriculture and Forestry in Türkiye, I learned about the European Union (EU) frameworks (e.g., Nitrates Directive), nitrate vulnerable zones, and sensitive areas. All these made me realize the complexity of controlling nutrient pollution as it affects both air and water, for instance, through pollution swapping. This scientific curiosity brought me to my PhD research.

Background: Human activities have largely influenced natural nutrient cycling. Air quality often suffers from nitrogen (N) emissions that can be deposited on land and then reach rivers and coastal waters. Besides, rivers receive not only N but also phosphorus (P) from human activities on the land, causing eutrophication issues. However, integrated assessments of both air and water pollution associated with nutrients are, so far, limited, hindering a better understanding of future trends in this pollution. Although many EU policies have been introduced, they focus either on air or water. Thus, nutrient management strategies to reduce both air and water pollution are needed, but their effects have not been explored enough for the EU areas and future years. This PhD research, therefore, focuses on exploring synergetic strategies to mitigate future nutrient-driven air and surface water pollution simultaneously. In particular, I improve our understanding of the effects of bio-based fertilizers (BBFs, see Section 1.1.7 for definition) on reducing future nutrient emissions to the air and surface waters (rivers and coastal waters) across Europe. My assessments timely contribute to decision making for these promising renewable alternatives to synthetic fertilizers while exploring the combined effects of BBFs with other nutrient management strategies. This thesis touches on the two scientific aspects: (1) the role of agriculture in nutrient-driven air, river, and coastal water pollution across the EU and United Kingdom (UK), and (2) the mitigation potential of BBFs and other nutrient management strategies in the future under global change. To address these aspects, I have developed a new version of the MARINA-Nutrients model for Europe.

The following sections introduce nutrient cycles (Section 1.1.1), sources and impacts of air (Section 1.1.2) and surface water pollution (Section 1.1.3), modeling approaches (Section 1.1.4), effects of global change on nutrient pollution (Section 1.1.5), EU policies for nutrient management (Section 1.1.6), and strategies to reduce nutrient losses in Europe (Section

1.1.7). Based on them, the knowledge gaps and research objectives are summarized in Section 1.2, followed by the thesis methodology and outline (Section 1.3).

1.1.1 Nutrient cycles

Nutrients, N and P, are essential for food production and to support all life. These nutrients cycle in the environment. Some are lost, and some are retained in the systems. Below, I briefly describe the cycling of N and P in terrestrial, atmospheric, and aquatic systems (Figure 1.1).

N cycle: N circulates across land, air, and water systems through biological and physicochemical processes that release various N forms under aerobic and anaerobic conditions (Bouwman et al., 2009) (Figure 1.1). As a unique trait, certain bacteria extract inert dinitrogen gas (N_2) from the atmosphere and convert it into biologically reactive forms, including ammonia (NH_3) and ammonium (NH_4^+), to fulfill their nutritional needs, called biological N_2 fixation (Marcarelli et al., 2022). In nitrification, bacteria converts NH_4^+ eventually into nitrate (NO_3^-), another bioavailable compound (Butterbach-Bahl et al., 2013) that can leach into groundwater and through runoff into aquatic systems (Liu et al., 2024). During mineralization and ammonification, various microorganisms decompose soil organic N compounds into NH_3, which then becomes NH_4^+ in the soil solution (de Vries et al., 2024). Denitrification is the reduction of biologically available NO_3^- to ultimately atmospheric N_2 by denitrifying bacteria under anaerobic conditions (Butterbach-Bahl et al., 2013). If the denitrification process is incomplete, intermediate products such as nitrous oxide (N_2O) can be released (Butterbach-Bahl et al., 2013). NH_3 volatilization is the conversion of NH_4^+ to NH_3, which is then lost to the atmosphere. Thereby, denitrification and NH_3 volatilization are the main pathways of N emissions to the air (Bouwman et al., 2009).

P cycle: P mainly cycles across land and water systems. The processes of P cycling include weathering, mineralization of organic P, adsorption, and desorption (Bouwman et al., 2009; Schoumans, 2015) (Figure 1.1). Chemical weathering releases phosphate (PO_4^{3-}) from rocks and minerals to the soil and water systems (Panagos et al., 2022). This inorganic P form is absorbed by plants and soil microbes, where they return organic matter by death and decay (Bot & Benites, 2005). In mineralization, certain bacteria mineralize this organic P back to bioavailable forms (Bot & Benites, 2005). In the availability of calcium, iron, and aluminum oxides, these chemicals bind P and form insoluble compounds, leading to P fixation in the soil, called adsorption (Schoumans, 2015). During desorption, the bounded P is released into the soil solution (Schoumans, 2015). P can leach into groundwater and through erosion and runoff into aquatic systems (Bouwman et al., 2009; Schoumans, 2015). In aquatic systems such as rivers and coastal waters, P can be used by organisms and settle into sediments. So-called legacy accumulation in soils and sediments becomes a long-term source of P pollution (Sharpley et al., 2013).

Nutrient cycling and human activities: Historically, the cycling of N and P has often been disrupted by human activities. As an important example, the invention and industrialization of the Haber-Bosch process (Haber, 1920) increased agricultural productivity massively with a breakthrough in fertilizer production (Erisman et al., 2008). The process enabled the synthesis of NH_4^+ from atmospheric N_2 and hydrogen, providing an almost unlimited source to produce N fertilizers (Haber, 1920). In parallel, the mining of phosphate rocks intensified to meet the growing demand for P fertilizer production (Cordell & White, 2014). While these advancements underpinned the synthetic fertilizers used in agriculture today, they came with the cost of environmental degradation. The planetary boundaries framework is developed for estimating to which extent anthropogenic pressures influence Earth system processes (Rockström et al., 2009). The estimations of boundaries and of how much they are crossed have been constantly updated (Rockström et al., 2009; Steffen et al., 2015; Richardson et al., 2023). The recent estimate indicates that six of nine boundaries have already crossed the safe operating space for humanity, including those for biogeochemical flows of N and P mainly due to agricultural use (Richardson et al., 2023). On the other hand, livestock production further escalated environmental degradation through manure application (Grizzetti et al., 2023) and through excess nutrients in livestock-dense regions (de Vries et al., 2021). Especially in the EU, tipping points (critical thresholds above which environmental damage occurs) for N associated with air and water pollution are exceeded (Schulte-Uebbing et al., 2022). Similarly, the current P flows in the EU are not within the safe operating space as a result of human activities (Grizzetti et al., 2023). All these have also impacts on human health and the environment (Table S1.1).

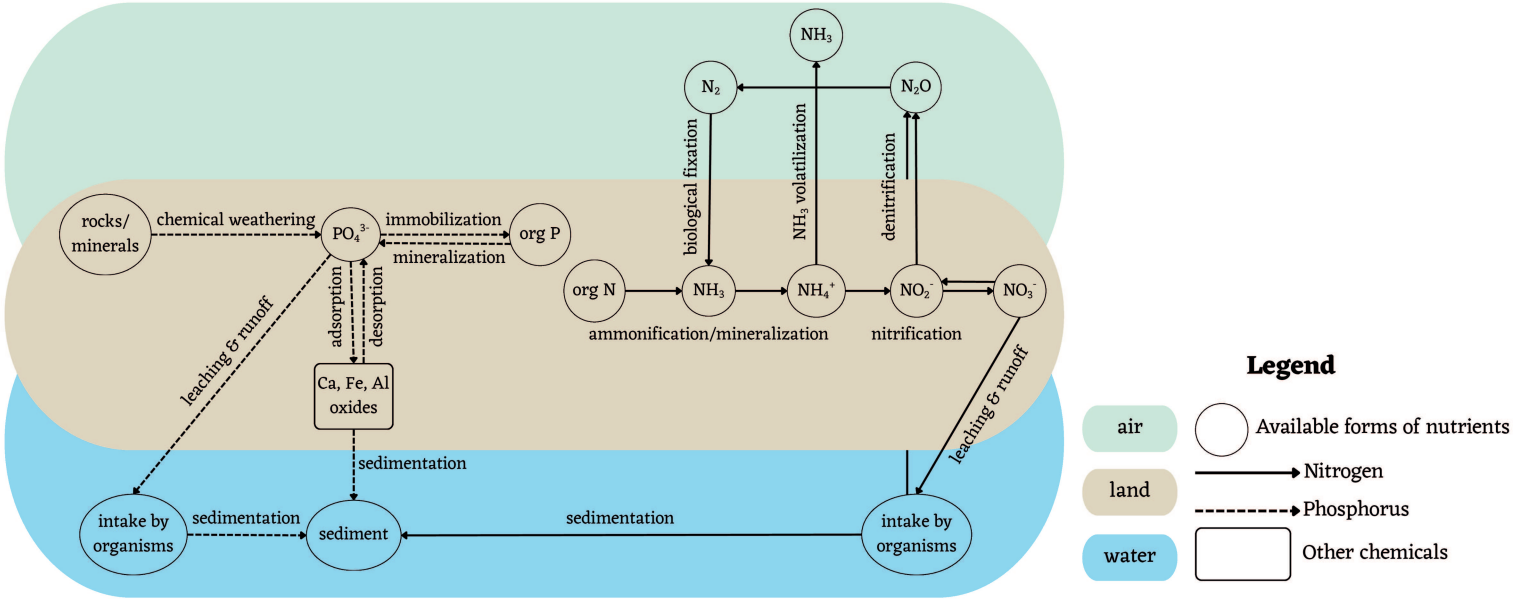


Figure 1.1 Summarized nutrient cycling of nitrogen (N) and phosphorus (P) within land, air, and surface waters (rivers and coastal waters). NH_3 is ammonia, N_2 is atmospheric nitrogen, N_2O is nitrous oxide, org N is organic N, NH_4^+ is ammonium, NO_2^- is nitrite, NO_3^- is nitrate, org P is organic P, PO_4^{3-} is phosphate, Ca is calcium, Fe is iron, and Al is aluminum.

1.1.2 Nitrogen emissions to the air: sources and impacts

Sources: Reactive N stands for oxidized and reduced N forms, including NH_3, N_2O, and nitrogen oxides (NO_x). Reactive N emissions are released to the air from sources including agriculture, energy, industry, transportation, and households (EEA, 2024b). Agriculture is often a significant contributor to these emissions, especially NH_3. Agricultural practices account for 93% of NH_3 emissions by the EU in 2022, primarily by animal production and manure handling (EEA, 2024b). Animal housing and manure storage systems are the main stages of NH_3 emissions, while the emissions from crop production are dominated by the application of synthetic fertilizer and manure (Grizzetti et al., 2023). Moreover, N_2O emissions from agricultural soils account for 30% of the total agricultural greenhouse gas (GHG) emissions by the EU in 2023 (EEA, 2025c). The N_2O is directly and indirectly released from agricultural soils (de Vries et al., 2024). Direct N_2O emissions to the air mainly originate from the application of synthetic fertilizer and animal manure (de Vries et al., 2024), urine and manure deposited on land by grazing animals, crop residues, N mineralization, and drainage of organic soils (Velthof & Rietra, 2018). Indirect N_2O emissions are released from terrestrial and aquatic systems as a result of NH_3 volatilization and/or NO_3^- leaching from surrounding agricultural areas, where N_2O is produced during nitrification-denitrification processes (Velthof & Rietra, 2018; de Vries et al., 2024). Furthermore, agricultural practices contribute to NO_x emissions relatively smaller extent (de Vries et al., 2024), as NO_x mainly originates from road transport and energy supply sectors in the EU (EEA, 2024b).

Impacts: Despite the progress that has been made, air pollution is still a significant threat to human health (Khomenko et al., 2021) and ecosystem quality in the EU (EC, 2025b; EEA, 2025e) (Table S1.1). For example, the emissions of NH_3 and NO_x contribute to aerosol/particulate matter formation (Wyer et al., 2022) through reacting with acid gases, thereby influencing the radiation balance and cooling the atmosphere (de Vries et al., 2024). The deposition of NH_3 and NO_x negatively affects forest ecosystems (e.g., decrease in biodiversity) by the acidification of forest soils (de Vries & Du, 2024). As a greenhouse gas (GHG), N_2O exacerbates climate change with a global warming potential 273 times greater than carbon dioxide (CO_2) (Forster et al., 2021). N_2O emissions contribute to the depletion of stratospheric ozone (Portmann et al., 2012), which increases ultraviolet light exposure for humans and plants (Barnes et al., 2019). This can, sometimes, result in skin cancer in humans and damage of plants (Barnes et al., 2019). Moreover, N_2O may cause respiratory diseases (de Vries et al., 2024; EEA, 2024d). On the other hand, NO_x emissions often contribute to tropospheric ozone formation, which in turn may lead to a warming effect and reductions in crop yields and grassland productivity (de Vries et al., 2024).

1.1.3 Nutrient emissions to surface waters: sources and impacts

Sources: Human activities alter nutrient cycles mainly by loading nutrient inputs to rivers. Nutrients enter rivers from many sources on land, such as agricultural runoff, aquaculture, domestic and industrial wastewater effluents (Beusen et al., 2022). Agriculture and urban-

ization (urban sewage treatment systems) are often the most important sources of nutrient pollution in the EU basins (Grizzetti et al., 2023; EEA, 2024c). Nutrients can reach rivers from diffuse and point sources. Diffuse sources often include application of synthetic fertilizers and animal manure on agricultural land, atmospheric N deposition, biological N_2 fixation, human wastewater that stays on land (not collected by sewage systems) (Vigiak et al., 2023). Point sources often discharge nutrients into rivers at a specific location, and include industrial and domestic sewage treatment systems (Vigiak et al., 2023). Nutrients in rivers from diffuse and point sources can be lost and/or retained in the river network. For example, dams and reservoirs enhance denitrification and trap P in sediments, reducing downstream transport and interrupting ecological functions (Maavara et al., 2020). Moreover, water withdrawals reduce river flow rate, altering hydraulic retention time and nutrient processing (Maavara et al., 2020; Wang et al., 2022b). All these factors alter river exports of nutrients to coastal waters.

Impacts: Despite the significant progress that has been made, river and coastal water pollution with nutrients is still a significant issue in the EU (Wolfram et al., 2021; Haase et al., 2023; EEA, 2025d). In the latest report on the implementation of the Nitrates Directive, at least one third of surface water bodies, including rivers (36%) and coastal waters (31%), are reported eutrophic due to excess nutrient inputs (EC, 2021b). In parallel with rivers and coastal waters, most of the marine waters (81%) are reported eutrophic, mainly due to the river exports of nutrients (EC, 2021b). Aquatic eutrophication leads to algal blooms (Glibert et al., 2018a), changes in aquatic vegetation, depletion of light, and an increase in sedimentation (Njock et al., 2023). As a result, oxygen depletion (Glibert et al., 2018b) and fish kills occur (Kudela et al., 2018) (Table S1.1).

1.1.4 Integrated air and water pollution modeling

Background: Understanding both the sources and state of air and water pollution is important to identify effective strategies to prevent and reduce them. Models are useful tools to quantify fluxes of pollutants, predicting the effects of changes (e.g., climate) as well as exploring the effects of mitigation strategies. Models exist for simulating nutrient flows and their sources (see below). However, integrated assessments of nutrient emissions to both air and surface waters (rivers and coastal waters) are, so far, limited. Hardly any models focus simultaneously on nutrient emissions to the air, inputs to rivers, and river exports to coastal waters. Below, the illustrative examples of models that include the agricultural sector and have been widely used for Europe are summarized (Table 1.1) followed by the reasons for developing an integrated model for nutrient emissions to the air and surface waters (rivers and coastal waters) for the EU basins (Figure 1.2 and Table 1.2).

Models for nitrogen emissions to the air: Examples of the existing models include GAINS (Amann et al., 2011), Daisy (Abrahamsen & Hansen, 2000), and STICS (Brisson et al., 2003) (Table 1.1). GAINS (Greenhouse gas - Air pollution Interactions and Synergies)

analyzes the pathways of atmospheric pollution from its anthropogenic origin to the environmental impacts, and is often used to provide model input for air pollution (JRC, 2024). The process-based model uses external activity data for the macroeconomic and sectoral (e.g., agriculture, energy) developments, and estimates emissions, mitigation potentials and costs for the major air pollutants (e.g., NH_3, NO_x) and GHGs (JRC, 2024). Smaller scale models, Daisy (Soil-plant-atmosphere system model, Rashid et al. (2022)) and STICS (Soil-crop model, Yin et al. (2020)) operate at scales such as field or farm while partially covering N losses from agricultural soils.

Models for nutrient emissions to surface waters: Examples of the existing models include GREEN (Grizzetti et al., 2008) and Riverstrahler (Billen et al., 1994) (Table 1.1). GREEN (Geospatial Regression Equation for European Nutrient losses) only focuses on losses of N and P to waters (rivers and seas) by European basins, mainly from agriculture and sewage systems (Vigiak et al., 2023). Riverstrahler is a water quality model that accounts for nutrient cycling and ecological functioning in water bodies at the scale of an entire drainage network, considering the hydro-climatic conditions, land use, and inputs from the watershed (Billen & Garnier, 2022).

Models for emissions to both air and waters: Examples of the existing models include CAPRI (Britz & Witzke, 2014), GRAFS (Billen et al., 2014), and INTEGRATOR (Reinds et al., 2011) (Table 1.1). CAPRI (Common Agricultural Policy Regional Impact Analysis) is a global agro-economic model focusing on the impacts of policies on agriculture in terms of the emissions of GHGs and NH_3, and nutrient balances, including N leaching (JRC, 2025). This partial-equilibrium model operates at the NUTS2 level for the EU (JRC, 2025). GRAFS (Generalized Representation of Agro-Food Systems) is a modeling approach that accounts for nutrient fluxes within agro-food systems, considering cropland, permanent grassland, livestock systems, and population (Billen et al., 2014). The approach is used at different scales across Europe and the world (Garnier et al., 2023). INTEGRATOR (Integrated Nitrogen Tool across Europe for Greenhouse gases and Ammonia Targeted to Operational Responses) is a process-based model that calculates GHG emissions and N fluxes from agriculture and forestry in the EU on a high spatial resolution (ca. 40,000 Nitrogen Calculation Units) (de Vries et al., 2021).

Reasoning: I identified three reasons for developing an integrated model for nutrient emissions to both air and surface waters (rivers and coastal waters) for the EU basins. The first reason is to better understand the contribution of agriculture to air and water pollution associated with nutrient emissions across the EU. There is a lack of a comprehensive assessment of nutrient emissions to the air, rivers and coastal waters (Knowledge gap 1a). *This thesis fills this gap by developing an integrated model to quantify N emissions to the air, and inputs of N and P to rivers and their exports by rivers to coastal waters of Europe.*

The second reason is to better assess integrated management measures for mitigating nutrient emissions to both air and waters simultaneously. While the EU air policies are in general at administrative scales, such as the National Emission reduction Commitments Directive (EU, 2016), water policies involve basin scales such as River Basin Management Plans (EC, 2000). The Water Framework Directive urges a holistic assessment of nutrients from sources to losses to seas through rivers (EC, 2000). There is a lack of a common scale assessment of air and surface water pollution associated with nutrient emissions to explore effective strategies to reduce this pollution across the EU (Knowledge gap 1b). *This thesis fills this gap by providing an integrated model to quantify nutrient emissions to the air and surface waters for the EU and the UK basins.*

The third, overarching, reason is to develop a spatially explicit model to better understand the effects of BBFs on reducing nutrient emissions to the air and surface waters. Most existing studies regarding the environmental impacts of BBFs are derived from laboratory (Wester-Larsen et al., 2022) or field experiments (Zilio et al., 2022). The implementation of BBFs on a larger scale has not been studied much yet (Rashid et al., 2025). To my knowledge, none of these studies consider the effects of BBFs on reducing nutrient emissions to the air, rivers and coastal waters simultaneously in the future across the EU (Knowledge gap 1c). *This thesis fills this gap by quantifying the impacts of implementing BBFs on reducing N emissions to the air, and inputs of N and P to rivers and their exports by rivers to coastal waters of Europe in the future under global change.*

To this end, a new version of the MARINA-Nutrients model for Europe is developed by coupling the MITERRA-Europe model (Velthof et al., 2009) to the existing modeling approach for nutrients and by adding new modeling elements for emissions to the air (Figure 1.2). MITERRA-Europe is short for Integrated Modeling of Nutrient Flows at Regional Level. It is a deterministic and static nutrient cycling model that quantifies the emissions of GHG, NH_3, and NO_x to the atmosphere, and flows of N and P to ground- and surface waters from crop and livestock production at NUTS2 level, where NUTS stands for Nomenclature of Territorial Units for Statistics (Eurostat, 2022; Duan et al., 2025). The model uses emission factors and fractions for leaching and runoff to simulate agricultural N and P flows based on a mass balance approach (Duan et al., 2025). On the other hand, MARINA is short for Model to Assess River Inputs of pollutaNts to seAs. The family of MARINA models include several versions for nutrients (Strokal et al., 2016; Chen et al., 2019b; Wang et al., 2020a; Wang et al., 2024). The existing MARINA-Nutrients models always addressed river and coastal water pollution (Table 1.2). The first version of MARINA-Nutrients was developed for China (Strokal et al., 2016), then this version was updated (Wang et al., 2020a). After that, MARINA-Nutrients was applied globally for N to more than 10,000 rivers (Wang et al., 2024). However, the existing MARINA-Nutrients models have never been applied to the EU rivers separately and do not include N emissions to the air (Table 1.2).

Table 1.1 Examples of existing models for nitrogen (N) and phosphorus (P) emissions from land to the air and/or water systems in Europe. GHG is greenhouse gas, NH_3 is ammonia, NO_x is nitrogen oxides, BC is black carbon, OC is organic carbon, $PM_{2.5}$ is particulate matter that is 2.5 microns or less in diameter and PM_{10} is those with a diameter of 10 microns or less, SO_2 is sulfur dioxide, NMVOCs is non-methane volatile organic compounds, N_2O is nitrous oxide, NO_3^- is nitrate, EU is European Union, and UK is United Kingdom, NUTS2 stands for Nomenclature of Territorial Units for Statistics 2, and NCUs is Nitrogen Calculation Units.

Model name	GAINS	GREEN	Riverstrahler	CAPRI	GRAFS	INTEGRATOR
Main approach	Process-based, steady state	Statistical, steady state	Process-based, steady state	Partial equilibrium, steady state	Mass balance, steady state	Process-based, steady state
Sources of nutrients	Industry, residential, transport, and agriculture sectors	Crop production, wastewater that stays on land and is not collected by sewage systems, domestic and industrial waste that goes to sewage systems	Crop and livestock production, and urban sewage systems	Crop and livestock production	Crop and livestock production	Crop and livestock production
Emissions to the air	GHG, NH_3, NO_x, BC, OC, $PM_{2.5}$, PM_{10}, SO_2, and NMVOCs	-	-	GHG, NH_3 and NO_x	NH_3 and N_2O	GHG, NH_3 and NO_x
Emissions to water systems	-	TN and TP loads to seas	Inputs of N and P to rivers from soil leaching	NO_3^- leaching to groundwater and run-off to surface water	NO_3^- leaching to ground- and surface water	NO_3^- leaching to groundwater and run-off to surface water
Spatial extent	Global, EU	All river basins draining into the European marine regions	Global, Europe	Global, EU	Global, Europe	EU and the UK
Spatial resolution	10 km – 50 km grids for the world, 1 km – 10 km grids for the EU	Hydrological sub-catchments with an average area of 7 km^2	Watersheds with the whole drainage network	NUTS2 for the EU	NUTS2 for Europe	NCUs (clusters of 1 km x 1 km grids)
Temporal extent	1990-2050, to 2070 for the EU	1990-2050	2016-2020	Up to 2030 and 2050	2014-2019	2010
Temporal resolution	5-year intervals	Yearly	Yearly	Yearly	Yearly	Yearly
Reference	(Amann et al., 2011; JRC, 2024)	(Grizzetti et al., 2008; Grizzetti et al., 2025)	(Billen et al., 1994; Billen & Garnier, 2022)	(Britz & Witzke, 2014; JRC, 2025)	(Billen et al., 2014; Garnier et al., 2023)	(Reinds et al., 2011; de Vries et al., 2021)

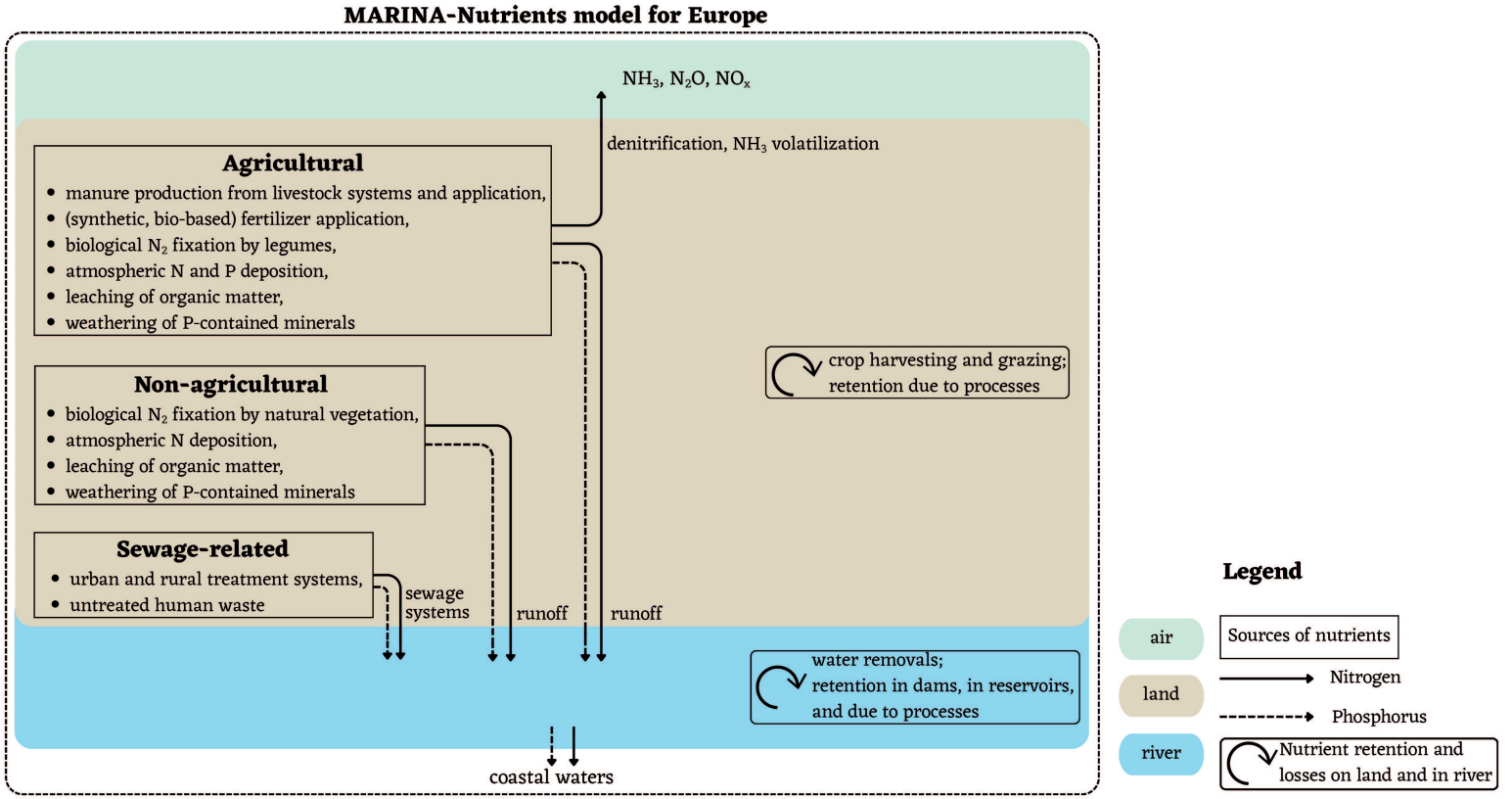


Figure 1.2 Simplified framework of the MARINA-Nutrients model for Europe in this PhD thesis. The diagram illustrates sources of nitrogen (N) and phosphorus (P), their pathways (e.g., denitrification, runoff, sewage systems), and associated processes (retentions and losses) on land and in rivers. The model quantifies the emissions of ammonia (NH_3), nitrous oxide (N_2O), and nitrogen oxides (NO_x) to the air from agricultural sources; the inputs of dissolved inorganic and organic N (DIN, DON) and P (DIP, DOP) to rivers and their exports by rivers to coastal waters from agricultural, non-agricultural, and sewage-related sources. The framework presents the model development (Chapter 2).

Table 1.2 Overview of the MARINA-Nutrients model family, along with my contribution (highlighted in bold). The first two versions of the MARINA-Nutrients model (China 1.0 and China 2.0) form the basis for the model development in this PhD research. N is nitrogen, P is phosphorus, DIN is dissolved inorganic N, DON is dissolved organic N, DIP is dissolved inorganic P, DOP is dissolved organic P, NH_3 is ammonia, N_2O is nitrous oxide, NO_x is nitrogen oxides, EU is European Union, and UK is United Kingdom. MEA is the Millennium Ecosystem Assessment scenarios. SSPs-RCPs are the combinations of Shared Socioeconomic Pathways (SSPs) and Representative Concentrative Pathways (RCPs).

Model name	MARINA-Nutrients (China 1.0)	MARINA-Nutrients (China 2.0)	MARINA-Nutrients (China 3.0)	MARINA-Nutrients (Global 1.0)	**MARINA-Nutrients (Europe 1.0)**
Model output	Inputs to rivers and river exports to coastal waters (load)	Inputs to rivers and river exports to coastal waters (load)	Inputs to rivers (load), and in-stream surface water quality (loads and concentrations)	Inputs to rivers and river exports to coastal waters (load)	**Emissions to the air, inputs to rivers, and river exports to coastal waters**
Sources	Diffuse (e.g., runoff) and point (sewage and manure discharges)	Diffuse (e.g., runoff) and point (sewage and manure discharges)	Diffuse (e.g., runoff) and point (sewage and manure discharges)	Diffuse (e.g., runoff) and point (sewage)	**For emissions: agriculture, For inputs to rivers and exports to coastal waters: diffuse (e.g., runoff) and point (sewage)**
Air emission parameters	None	None	None	None	**NH_3, N_2O and NO_x**
Water quality parameters	DIN, DON, DIP and DOP	DIN, DON, DIP and DOP	DIN, DON, DIP and DOP	DIN and DON	**DIN, DON, DIP and DOP**
Spatial extent	China	China	China	World	**EU and the UK**
Spatial resolution	Sub-basin	Sub-basin	Grid of 0.5°, polygon, county, and sub-basin	Sub-basin	**Basin and sub-basin**
Temporal extent	Annual	Annual	Annual	Annual	**Annual**
Temporal resolution	1970, 2000, and 2050	2012 and 2050	2012	2010 and 2050	**2017-2020 and 2050**
Future scenarios	MEA	SSPs-RCPs	None	SSPs-RCPs	**SSPs-RCPs**
Reference	(Strokal et al., 2016)	(Wang et al., 2020a)	(Chen et al., 2019b; Chen et al., 2022)	(Wang et al., 2024)	**This PhD thesis (Chapters 2-4)**

1.1.5 Effects of global change on nutrient pollution

Global change effects: Global change is the interplay of socioeconomic developments and climate change. These drivers might considerably contribute to nutrient-driven air and water pollution in the future. Socioeconomic development regarding population growth, changes in gross domestic product (GDP), and in dietary habits alter land use (Popp et al., 2017) and climate (Riahi et al., 2017; van Vuuren et al., 2025). Agriculture is expected to intensify (Popp et al., 2017), unless significant transitions are made (Billen et al., 2021; Kozicka et al., 2023; Simon et al., 2024). Europe's agricultural production is also expected to increase in the future (Schulte-Uebbing & de Vries, 2021). On the other hand, urbanization and urban sewage systems are projected to increase in the future (Strokal et al., 2021; van Puijenbroek et al., 2023). All these changes will influence N emissions to the air, and N and P flows to waters from human activities.

Existing scenarios: Scenario analysis is used to explore how the future may unfold in terms of global change drivers and to assess effective strategies to mitigate emissions to the air and waters. Scenarios can be predictive, explorative, and normative, depending on the research objective (Börjeson et al., 2006; Maier et al., 2016). Predictive scenarios often focus on forecasting to analyze "what will happen" if the likely development occurs in the future (Börjeson et al., 2006; Maier et al., 2016). As an example of explorative scenarios which can be used to answer the question "what could happen?" (Maier et al., 2016), Shared Socioeconomic Pathways (SSPs) provide five possible future trajectories regarding socioeconomic development (O'Neill et al., 2014). SSPs can be combined with Representative Concentration Pathways (RCPs), which cover the climate forcing dimensions of possible futures (van Vuuren et al., 2011). The SSP-RCP combinations can be the basis for identifying the impacts of changes in socioeconomic factors and climate on the quality of air (e.g., GHG emissions, Riahi et al. (2017)) and water (e.g., runoff, Bouwman et al. (2024)). On the other hand, normative scenarios explore the ways to reach a desired target to answer the question "how can a specific future be realized?" (Maier et al., 2016). In this case, backcasting is a common approach that provides potential solutions to meet the desired target (Li et al., 2019).

Backcasting and Indicator for Coastal Eutrophication Potential: Studies investigating future pollution trends associated with nutrient emissions mainly focus on forecasting (Grizzetti et al., 2021) or explorative scenarios (Grizzetti et al., 2023; Bouwman et al., 2024; Wang et al., 2024). Backcasting studies exploring options to reach a desirable future are scarce (Li et al., 2019; Van Eynde et al., 2025). For example, the Indicator for Coastal Eutrophication Potential (ICEP, Billen and Garnier (2007)) can be used to set a target for safe levels of nutrient fluxes at the river mouth to avoid coastal eutrophication. The ICEP is an elegant method to estimate the potential eutrophication risk in coastal waters based on nutrient fluxes and the molar Redfield ratio (C:N:P:Si = 106:16:1:20) (Redfield et al., 1963) (see Chapter 3 for equations to calculate N-ICEP and P-ICEP). When there is a nutrient imbalance according to the Redfield ratio, the ICEP value indicates the potential growth of the

harmful non-siliceous algae (e.g., cyanobacteria), which can cause eutrophication in coastal waters (Billen & Garnier, 2007). Accordingly, the risk of coastal eutrophication is considered low when the ICEP value is below zero (Billen & Garnier, 2007).

However, to what extent global change may affect nutrient pollution in coastal waters of Europe and to what extent pollution reductions are needed to prevent the risk of coastal eutrophication in the future is not well studied. To my knowledge, the existing studies are limited to identify basin-specific targets and required reductions in river exports of N and P to avoid coastal eutrophication in Europe in the future under global change (Knowledge gap 2). *This thesis fills this gap by quantifying the future river exports of N and P, and identifying required reductions to avoid coastal eutrophication in Europe under global change.*

1.1.6 EU policies for nutrient management

Existing policies: In response to the abovementioned challenges, the EU has implemented a number of frameworks and regulations since the 1990s to reduce nutrient pollution. Current policies of the EU have also driven great interest in renewable resources to contribute to sustainable agriculture and the circular economy. Below, I elaborate on the existing EU policies and ambitions that are, to my knowledge, directly or indirectly related to mitigating air and/or water pollution associated with nutrient emissions and implementing BBFs. A chronological summary of these policies is given in Table S1.2, indicating their relation to air and/or water pollution.

Air: EU policies for air pollution control target emissions from agriculture, industry, and energy sectors. The National Emissions Reduction Commitments Directive (NECD) and Industrial and Livestock Rearing Emissions Directive (IED) regulate NH_3, NO_x, and other key pollutants at national and sectoral levels (EU, 2016; 2024a). For example, the NECD sets reduction targets by 2030 compared to 2005, for five air pollutants, including NH_3 and NO_x that are related to the impacts on human health (EU, 2016). On the other hand, complementary frameworks like the Paris Agreement, the European Green Deal, and the Zero Pollution Action Plan reinforce emission reduction targets with climate ambitions (UNFCCC, 2015; EC, 2019; 2021a). For instance, the decarbonization or climate neutrality ambition of the EU Green Deal is to achieve net-zero GHG emissions by 2050 across the EU (EC, 2019). Despite the progress, a challenge remains in mitigating the emissions of NH_3 and NO_x mainly from agriculture due to insufficient monitoring and regulation (EEA, 2024b).

Water: EU policies for water pollution associated with nutrients focus largely on reducing nutrient losses from agriculture and urban sources while preserving biodiversity and ecosystems. The main instrument, the Water Framework Directive (WFD), aims to achieve "good chemical and ecological status" of all water bodies with support of the Nitrates Directive (ND) and Urban Wastewater Treatment Directive (UWWTD) (EC, 2000). Broader frame-

works like the Habitats Directive and Marine Strategy Framework Directive extend the ecological dimension by linking water quality to biodiversity and ecosystem conservation (EC, 1992; EU, 2008a), as the imbalanced nutrient flows have contributed to biodiversity loss and aquatic eutrophication (de Vries et al., 2021). Nevertheless, those frameworks have deficits and weak enforcement. For instance, while the chemical status of surface waters is assessed based on environmental quality standards for a Europe-wide list of priority substances (e.g., lead, mercury), the criteria for nutrients can be identified by countries (Poikane et al., 2019). The criteria of some of the EU member states are found to be insufficient to protect rivers from ecological deterioration (Poikane et al., 2021). On the other hand, the ND's limit on the application of animal manure and/or manure end products to agricultural soils might indirectly contribute to manure surplus, rather than providing in-depth solutions for livestock dense regions. Fortunately, the use of Recovered Nitrogen from manURE (ReNuRe, sometimes it is abbreviated as RENURE) proposal has been approved by the EU Nitrates Committee to allow the use of processed manure products under the ND (EC, 2025a).

Link to Sustainable Development Goals: These EU policies and targets have synergistic effects on Sustainable Development Goals (SDGs) (Dunne, 2010), and have the common intention to reduce pollution. For example, the WFD (EC, 2000) contributes to SDG 6 (clean water and sanitation (UN, 2015)) by aiming to achieve "good chemical and ecological status" of EU ground- and surface water bodies. The ND (EC, 1991a) and the Zero Pollution Action Plan (EC, 2021a) are linked to SDG 14 (life below water) and SDG 15 (life on land) by focusing on reducing nutrient pollution and protecting ecosystems. Moreover, the European Green Deal (EC, 2019) aligns with SDG 13 (climate action) and SDG 7 (affordable and clean energy) through targeting climate neutrality, renewable energy expansion, and emissions reduction. Furthermore, the Farm to Fork Strategy (EC, 2020) supports SDG 2 (zero hunger) and SDG 12 (responsible consumption and production) by promoting sustainable food systems, food security, and reduced environmental impacts of agriculture.

Despite the progress of the EU policies and action plans, water quality has not yet reached desirable levels (Nikolaidis et al., 2022; EEA, 2025e). Based on the recent analysis, the Green Deal targets on reducing the area of EU ecosystems where air pollution threatens biodiversity by 25% and mitigating nutrient losses by 50% are not likely to be met by 2030 (Grizzetti et al., 2023; EEA, 2025e). Obviously there is a need for new integrated strategies to reduce nutrient-driven air and water pollution synergistically, but the effects of these strategies have not been explored for the EU areas and future years (Knowledge gap 3a). *This thesis fills this gap by exploring synergetic solutions to mitigate future air and water pollution associated with nutrient emissions.*

1.1.7 Strategies to reduce nutrient losses in Europe

o Bio-based fertilizers

Definition: BBFs can be defined as "Fertilizers derived from biomass using the nutrient recovery and re-use technologies (physical, thermal, chemical and/or biological), in order to up concentrate nutrients from the initially treated biomass and hence improve its nutrient efficiency" (Sigurnjak et al., 2023). These BBF fertilizers are produced through enhanced nutrient recovery and recycling from organic waste (Case & Jensen, 2019; Chojnacka et al., 2020). BBFs are promising alternatives to non-renewable and energy-intensive synthetic fertilizers that cause a high environmental burden during their extraction and transformation (Sniatala et al., 2023). BBFs can largely reduce organic waste through upcycling, increase the uptake of micronutrients, improve crop quality, promote soil biodiversity (Kurniawati et al., 2023b), provide adjusted nutrient ratios to align better with crop needs (Chojnacka et al., 2020), and increase nutrient use efficiency (Jin et al., 2022). Moreover, they can contribute to nutrient exchange between regions (Tur-Cardona et al., 2018) and foster circular economy (Upadhyay et al., 2024).

Types and scope: The three largest groups of organic waste to produce BBFs are agricultural waste, human waste, and food waste (Chojnacka et al., 2020). Among the agricultural waste, livestock slurry/manure is the most well-known example for nutrient recovery, especially N, to produce BBFs (Huygens et al., 2020; Saju et al., 2023). Sewage sludge from domestic wastewater treatment plants is another significant source of nutrient recovery, and can be used in agriculture after stabilization, removal of toxic compounds, and pathogens (González-García et al., 2022; Chojnacka et al., 2023). Huygens et al. (2019) demonstrate that more than 90% P can be recovered from sewage sludge to produce safe alternatives for the phosphate rock (e.g., STRUBIAS materials: STRUvite, BIochar, or incineration AShes). Food waste from the production, processing, and consumption chain can also be valorized into BBFs (Hu et al., 2021; O'Connor et al., 2021).

Production processes: Technologies for producing BBFs through nutrient recovery from organic waste vary greatly from physicochemical to biological (see Table S5.4 for illustrative examples). For example, NH_3 stripping-scrubbing process is a common technology with clean end products having comparable characteristics as synthetic N fertilizers (Sigurnjak et al., 2019; Huygens et al., 2020). The end products are ammonium salts such as ammonium nitrate and ammonium sulfate that can be used as ReNuRe fertilizer (Vingerhoets et al., 2024). On the other hand, P recovery from treated municipal wastewater by struvite precipitation could increase the circularity of P, thereby reducing the dependence on external fertilizers (Huygens et al., 2019; Vingerhoets et al., 2023b).

Reduction impacts: Based on the production and application techniques, BBFs could reduce the emissions of GHG (Egene et al., 2022; Zilio et al., 2022; Clagnan et al., 2023; Hu et al., 2024) and NH_3(Wester-Larsen et al., 2022; Zilio et al., 2022), as well as NO_3^- leach-

ing (Zilio et al., 2022). Existing literature focusing on these impacts is often on a small scale, including laboratory (Egene et al., 2022; Wester-Larsen et al., 2022; Shrivastava et al., 2023) or field-scale (Zilio et al., 2022). These studies focus on specific impacts while ignoring synergies and trade-offs (e.g., pollution swapping, (Velthof et al., 2024)). None of the existing studies have thoroughly assessed the impacts of using BBFs in European agricultural systems on reducing N emissions to the air and nutrient loads to rivers and coastal waters (Knowledge gap 3b). *This thesis fills this gap by quantifying the effects of using BBFs alone and in combination with other nutrient management strategies on reducing future nutrient emissions to the air, rivers and coastal waters of Europe.*

o Other potential nutrient management strategies

N mitigation: Examples of strategies for reducing N losses from agricultural land include 4R nutrient management (right fertilizer type, right amount, right placement, and right time), crop rotation (e.g., legume-cereal) (Gu et al., 2023), and balanced fertilization (Grizzetti et al., 2023). Examples for reducing N losses from livestock systems are mainly manure management and application techniques, livestock density limits, and dietary additives (Ti et al., 2019; Bielza et al., 2025). However, management strategies aiming at one specific N emission might lead to trade-offs among other N emissions (Zhang et al., 2024a). Therefore, strategies to reduce air and water pollution associated with nutrient emissions should be synergetic. For example, synergy occurs when a mitigation measure for one nutrient emission decreases another.

P mitigation: A major management strategy for reducing P loss from agricultural land is balanced fertilization, namely the determination of P application rate based on crop P uptake and soil P status (van Doorn et al., 2023). This can be very complementary to BBFs, as they can provide adjusted nutrient (NPK) ratios to align better with crop requirements (Chojnacka et al., 2020). Van Eynde et al. (2025) investigated nine different P management strategies for the EU agricultural land, and indicated that current P inputs across the EU can be reduced by 21% without negative effects on crop production.

Mitigation of N and P together: The effectiveness of the mitigation measure for agricultural nutrient losses is often correlated with nutrient use efficiency in agro-food chain (Chatzimpiros & Harchaoui, 2023; You et al., 2023). Strategies such as increasing nutrient use efficiency in agriculture (Schulte-Uebbing & de Vries, 2021), reconnecting livestock with cropping systems (Billen et al., 2021), and dietary changes (Billen et al., 2021; Simon et al., 2024) mitigate both N and P losses. On the other hand, improving wastewater treatment systems, and nutrient recovery and recycling are significant potentials for mitigating nutrient losses to water systems, especially for P (Huygens et al., 2019; Zhu et al., 2023). This is because sewage treatment systems are often the main contributors of P losses to rivers and coastal waters of Europe (Vigiak et al., 2023). To my knowledge, studies addressing such management strategies lack an assessment of the effects of using the recovered P from treated sewage sludge as BBF on water pollution in the EU (Knowledge gap 3c). *This thesis*

fills this gap by quantifying the effects of using the recovered P from treated sewage sludge as BBF, alone and combined with other nutrient management strategies, on reducing future nutrient losses to rivers and coastal waters of Europe.

1.2 Knowledge gaps and research objectives

Here, I summarize the knowledge gaps that are identified in Section 1.1 into three main categories:

(1) Lack of an integrated model that quantifies the contribution of agriculture to nutrient emissions to the air, and surface waters (rivers and coastal waters) in a spatially explicit way across the EU and the UK basins;
(2) Limited knowledge of basin-specific targets and required reductions in river exports of N and P to avoid coastal eutrophication in Europe in the future under global change;
(3) Lack of EU-wide assessment of the effects of bio-based fertilizers that are produced through recovered nutrients on reducing future nutrient emissions to the air, rivers and coastal waters.

The overarching objective of this PhD thesis is, thus, to enhance our understanding of basin-specific nutrient emissions to the air and surface waters, and explore the role of BBFs and other nutrient management strategies to reduce future nutrient pollution under global change across the EU and the UK.

To this end, three research sub-objectives (SO) are formulated and achieved, as discussed in Chapters 2-4:

SO1: To quantify the contribution of agricultural activities to air, river and coastal water pollution simultaneously with a focus on nutrients in European basins (Knowledge Gap 1, Chapter 2);

SO2: To assess the future river exports of N and P, and identify required reductions to avoid coastal eutrophication in Europe under global change (Knowledge Gap 2, Chapter 3);

SO3: To quantify the effects of using recovered N from manure processing and recovered P from treated sewage sludge, increasing N and P use efficiencies in agriculture, and improving sewage treatments on reducing future nutrient emissions to the air, and losses to rivers and coastal waters of Europe (Knowledge Gap 3, Chapter 4).

Each SO forms a research chapter of this thesis. Table 1.3 presents an overview of the knowledge gaps, research sub-objectives, methodology used to close the knowledge gaps, and main outcomes of the research chapters of this PhD thesis.

Table 1.3 Overview of the chapters, knowledge gaps, research sub-objectives, methodology to fill the knowledge gaps, and main outcomes of this PhD thesis. MARINA-Nutrients is the Model to Assess River Inputs of pollutaNts to seAs for Nutrients.

Chapter	Knowledge gap (Section 1.1)	Research sub-objective (Section 1.2)	Closing the gap (Chapters 2-4)
2	(1) Lack of an integrated model that quantifies the contribution of agriculture to nutrient emissions to the air, and surface waters (rivers and coastal waters) in a spatially explicit way across the EU and the UK basins	SO1: To quantify the contribution of agricultural activities to air, river and coastal water pollution simultaneously with a focus on nutrients in European basins	Development and validation of the new MARINA-Nutrients for Europe, accounting for important sources of nutrient emissions to the air and surface waters (rivers and coastal waters) in a spatially explicit way (for 601 basins)
3	(2) Limited knowledge of basin-specific targets and required reductions in river exports of N and P to avoid coastal eutrophication in Europe in the future under global change	SO2: To assess the future river exports of N and P, and identify required reductions to avoid coastal eutrophication in Europe under global change	Projection of water pollution and reduction targets for nutrient exports by the top ten polluted rivers to avoid coastal eutrophication in Europe in the future under global change
4	(3) Lack of EU-wide assessment of the effects of bio-based fertilizers that are produced through recovered nutrients on reducing future nutrient emissions to the air, rivers and coastal waters	SO3: To quantify the effects of using recovered N from manure processing and recovered P from treated sewage sludge, increasing N and P use efficiencies in agriculture, and improving sewage treatments on reducing future nutrient emissions to the air, and losses to rivers and coastal waters of Europe	Integration of bio-based fertilizers that are produced through recovered nutrients into the MARINA-Nutrients model, development and application of new scenarios to simultaneously reduce nutrient emissions to the air and surface waters (rivers and coastal waters) in Europe

1.3 Thesis methodology and outline

This PhD thesis consists of five chapters. **Chapter 1** introduces the background information and justifies the overarching objective of this PhD research. Chapters 2-4 consist of the results corresponding to each research sub-objective. Figure 1.3 shows the outline of the thesis chapters with a focus on novelties and methods. Table S1.1 and Figure S1.1 (Appendix S1) respectively present the description and map of the study area.

Chapter 2 (SO1) addresses Knowledge gap 1 and is novel in two key aspects: (1) the development and validation of a new MARINA-Nutrients for Europe, the first consistent model integrating nutrient emissions to the air and surface waters (rivers and coastal waters) in a spatially explicit way, and (2) new insights into the contribution of agriculture to nutrient-driven air and water pollution across 601 basins in the EU. By applying the new MARINA-Nutrients for Europe, the annual N emissions to the air, inputs of N and P to rivers, and their exports to seas are quantified for the period of 2017-2020. The model accounts for agriculture-associated N emissions to the air; and agriculture, non-agriculture, and sewage-associated nutrient losses to rivers and coastal waters of Europe, considering retention and losses in soils (e.g., P accumulation) and rivers (e.g., damming, sedimentation). Based on the model results, the hotspots for air pollution, water pollution, or air and water pollution are identified (see Chapter 2 for the definition of hotspots). In addition, the relative contributions of sources to nutrient losses into rivers and seas are assessed per European sea (Arctic Ocean, Atlantic Ocean, Baltic Sea, Black Sea, Mediterranean Sea, and North Sea). This chapter provides a quantitative baseline for future scenario analysis.

Chapter 3 (SO2) addresses Knowledge gap 2 and presents: (1) new insights into future water pollution with nutrients across the EU under global change, and (2) the application of the backcasting approach to quantify the required reductions for exports of N and P by the top ten polluted rivers to avoid the risk of coastal eutrophication. A future baseline scenario is developed by combining the storylines of SSP5 (fossil-fuel-based development) and RCP8.5 (high-end climate change) to quantify the effects of global change on nutrient exports by rivers to coastal waters of Europe. Based on the future baseline (SSP5-RCP8.5), the actual exports of N and P by the top ten polluted rivers to coastal waters are projected for 2050 by applying the MARINA-Nutrients for Europe. The maximum allowable exports of N and P by the top ten polluted rivers are estimated by targeting an ICEP value of "0" to ensure low risks of coastal eutrophication. The difference between the projected actual and maximum allowable nutrient exports by the top ten polluted rivers is calculated by a backcasting approach to predict how much reductions will be required to avoid the risk of coastal eutrophication in 2050.

Chapter 4 (SO3) addresses Knowledge gap 3 and is novel in two key aspects: (1) integration of BBFs into the MARINA-Nutrients for Europe and the provision of new scenarios,

and (2) novel insights into the effects of BBFs on reducing future nutrient emissions to the air and surface waters across the EU in combination with other nutrient management strategies. The BBFs considered in this study are recovered N from manure (ReNuRe) and recovered P from treated sewage sludge (RPSS). To this end, six new scenarios are developed for 2050, including the implementation of BBFs alone and in combination with increasing nutrient use efficiencies in agriculture and with improving sewage treatments (see Chapter 4 for the descriptions of scenarios). Under the six new scenarios, N emissions to the air, inputs of nutrients to rivers and river exports of nutrients to coastal waters are projected for 2050 by applying the MARINA-Nutrients for Europe. the results are assessed relative to the period of 2017-2020 and the future baseline. This chapter provides a significant large-scale assessment of using BBFs which could guide the future research on BBFs and support the decision making process for their implementation.

Chapter 5 connects the three research chapters by discussing the main findings, novelties, modeling approaches, strengths, weaknesses, opportunities, and threats of this PhD research. In addition, this chapter presents reflection on the main perspectives for the implementation of BBFs and on their role in air and water pollution (reduction) including (1) environment, (2) agronomy, (3) technology, (4) policy and socioeconomic, and (5) scale and integration. Finally, the chapter provides a future outlook with recommendations for scientists and policymakers.

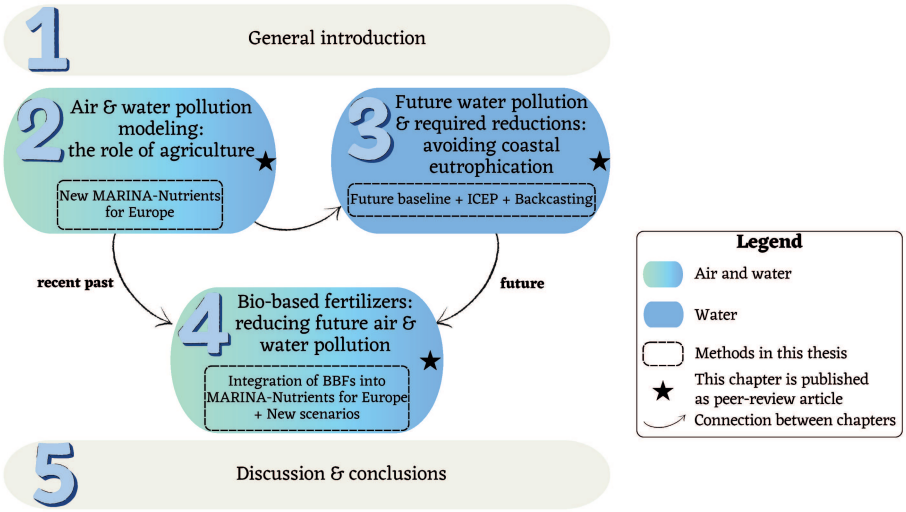


Figure 1.3 Overview of the PhD thesis structure with the focus of chapters, their methods and connections. MARINA-Nutrients is the Model to Assess River Inputs of pollutaNts to seAs for Nutrients, ICEP is an Indicator for Coastal Eutrophication Potential, and BBFs are bio-based fertilizers. Backcasting is explained in Section 1.1.5.



Chapter 2

Hotspots of nutrient losses to air and water: an integrated modeling approach for European river basins

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This paper is adjusted to the PhD thesis format.

Abstract

Nutrient pollution of air and water is a persistent problem in Europe. However, the pollution sources are often analyzed separately, preventing the formulation of integrative solutions. This study aimed to quantify the contribution of agriculture to air, river and coastal water pollution by nutrients. A new MARINA-Nutrients model was developed for Europe to calculate inputs of nitrogen (N) and phosphorus (P) to land and rivers, N emissions to air, and nutrient export to seas by river basins. Under current practice, inputs of N and P to land were 34.4 and 1.8 $Tg \cdot yr^{-1}$, respectively. However, only 12% of N and 3% of P reached the rivers. Agriculture was responsible for 55% of N and sewage for 67% of P in rivers. Reactive N emissions to air from agriculture were calculated at 4.0 $Tg \cdot yr^{-1}$. Almost two-fifths of N emissions to air were from animal housing and storage. Nearly a third of the basin area was considered as pollution hotspots and generated over half of N emissions to air and nutrient pollution in rivers. Over 25% of river export of N ended up in the Atlantic Ocean and of P in the Mediterranean Sea. These results could support environmental policies to reduce both air and water pollution simultaneously, and avoid pollution swapping.

2.1 Introduction

Intensive agriculture and high population density are often the main causes of air and water pollution with nutrients in Europe (Sutton et al., 2011a; Sutton et al., 2011b). Nitrogen and phosphorus inputs to land from applied fertilizers increased by 3% and 16% in the European Union (EU-28) from 2010 to 2020, respectively (FAOSTAT, 2022). Leip et al. (2011) showed that 60% of N applied to agricultural land in Europe is taken by crops and the rest is mainly lost to the environment. Half of the P imported by the EU is accumulated in agricultural land and the remainder is lost to the environment (van Dijk et al., 2016). Losses of N and P to the environment have substantially affected air (Westhoek et al., 2015; Giannakis et al., 2019; Xu et al., 2019) and water quality (de Vries & Schulte-Uebbing, 2020; Wolfram et al., 2021; de Vries et al., 2023). Giannakis et al. (2019) showed that a majority of EU countries have difficulties to meet their ammonia emission ceilings. Despite the policies and improved monitoring systems, water quality has worsened in Europe (Grizzetti et al., 2021; Wolfram et al., 2021).

Several studies have quantified N losses to air, often at the administrative scales (de Vries et al., 2011; Leip et al., 2011; Velthof et al., 2014). Other studies focused on N and P losses to rivers and coastal waters, often at the grid and/or basin scales (Mayorga et al., 2010; Beusen et al., 2016; Grizzetti et al., 2021). However, existing studies for Europe hardly focus on both air and water pollution simultaneously. Often, there is a mismatch in the spatial scales for air and water analyses. This results in a knowledge gap in terms of environmental impact assessment and management. For example, reducing one pollutant in surface waters might decrease (synergy) or increase (trade-off) other pollutants in the atmosphere or vice versa (e.g., pollution swapping). Air policies are typically at administrative levels (e.g., Directive 2016/2284/EU, EU (2016)) whereas water policies often cover basin scale (e.g., River Basin Management Plans required by the Directive 2000/60/EC, EC (2000)). A comprehensive assessment of nutrient pollution for both air and water does not exist at a basin scale. It is important to develop integrated environmental policies that tackle both air and water pollution with nutrients in Europe. For this, consistent assessments of air and water pollution are urgently needed to identify the contribution of sources to this pollution and support current debates on solving N issues that require integrated approaches to reduce pollution synergistically.

Models are useful tools to assess air and water quality (e.g., pollution and its sources) at different temporal-spatial scales, and explore solutions to support policymakers (Table S2.1). In contrast, modeling studies generally focus either on a particular pollution source or a receiving body (e.g., surface or ground waters) (Table S2.1). The MARINA model (Model to Assess River Inputs of pollutaNts to seAs) is an example of this (Strokal et al., 2016). Several versions of MARINA exist. Some focus on river exports of nutrients (Li et al., 2019; Chen et al., 2022; Wang et al., 2022a) and some on other pollutants (Strokal et al., 2021; Li et al., 2022b). The original versions of the model (MARINA 1.0, 2.0) were developed to quantify

annual nutrient inputs from land to rivers and sea at basin and sub-basin scales (Strokal et al., 2016; Wang et al., 2020a). MARINA-Nutrients is a deterministic, steady-state and lumped model based on a process-based and uncalibrated modeling approach to quantify N and P flows from land to rivers and sea (Table S2.2). However, such a model does not consider N emissions to air from agricultural activities in Europe (Table S2.2). MITERRA-Europe (integrated model to assess the implementation of agricultural measures on emissions to air and waters in Europe) is a deterministic and static nutrient cycling model that quantifies annual greenhouse gases (e.g., N_2O) and ammonia emissions to the atmosphere, and N and P flows from agriculture at a regional scale in Europe (Table S2.1 and Table S2.2) (Velthof et al., 2009). Linking these two models (MARINA-Nutrients and MITERRA-Europe) opens an opportunity to assess the significant sources of both air and water pollution in Europe in a spatially explicit way.

The sources of nutrient pollution in surface waters can be diffuse (e.g., leaching or runoff from soils) and point (e.g., pipes discharging to rivers) (Mayorga et al., 2010). The MARINA-Nutrients model distinguishes between diffuse and point sources of nutrients in rivers and coastal waters at the basin and sub-basin scales (Table S2.2) (Strokal et al., 2016; Wang et al., 2020a). The MITERRA-Europe model focuses on agricultural sources of air pollution including N emissions from animal houses and manure storage systems at the European scale (Table S2.2) (Oenema et al., 2009; Velthof et al., 2009). A detailed assessment of N losses to air, N and P losses to water from agricultural production at a basin scale can help develop basin-specific nutrient management plans with effective pollution reduction options.

This study aimed to quantify the contribution of agricultural activities to air, river and coastal water pollution simultaneously with a focus on nutrients in European basins. To this end, we soft-linked MARINA-Nutrients and MITERRA-Europe. The new MARINA-Nutrients model for Europe accounts for reactive N losses to air (e.g., NH_3, N_2O) from agriculture, and nutrient losses to rivers and coastal waters from agriculture, sewage and nature. The basin-scale modeling allows for spatially explicit analyses of water pollution and exploring specific measures. Our results could contribute to European environmental policies (e.g., River Basin Management Plans required by the Water Framework Directive, National Emission Ceilings Directive) to allocate effective nutrient management strategies and avoid pollution swapping by basin-scale analysis of air and water pollution.

2.2 Materials and methods

2.2.1 MARINA-Nutrients model for Europe

A new version of the MARINA-Nutrients model for European basins (Supplementary Text S2.1) was developed from the existing MARINA approaches (Strokal et al., 2016; Wang et al., 2020a) by adding air emissions from agriculture. Thus, the new MARINA-Nutrients quantifies the contribution of agricultural activities to air, river and coastal water pollution simultaneously through a soft-linking approach (Figure 2.1). Briefly, outputs of the MITERRA-Europe model were used as inputs to the MARINA-Nutrients model. The new model quantifies inputs of N and P to land and rivers, N emissions to air, and river exports of N and P to seas based on human activities (e.g., agriculture and sewage), hydrology (e.g., water discharges) and land use in European basins. Model outputs of the MARINA-Nutrients model are at the basin and sub-basin (only for the Danube River) scale (study area description in Supplementary Text S2.2 and Figure S2.1). N and P inputs to rivers and their river exports are in dissolved organic and inorganic forms.

Model inputs for calculating agriculture-associated N and P inputs to land were derived from the MITERRA-Europe model. MITERRA-Europe is an emission factor-based model that calculates N emissions to air, and N and P flows from agricultural activities as a function of e.g., land use, fertilizer use and livestock numbers (Table S2.2) (Velthof et al., 2009; de Vries et al., 2011). The results from the updated version of MITERRA-Europe were used to assess the current pollution in Europe. This updated version is based on the recent (2016–2018) European statistics and other datasets (e.g., FAOSTAT) (Duan et al., 2021).

The new MARINA-Nutrients model for Europe is the first trial for the river-basin-scale analysis of air and water pollution mainly from human activities. Our improvements are the integration of nutrient inputs from livestock systems (e.g., manure applied on agricultural land from stables and deposited on land during grazing), cropland and grassland as well as from human waste unconnected to sewage systems in Europe (Supplementary Text S2.1 for details). More details on the input data and model calculations are provided in Table S2.3, Table S2.4, Table S2.5, and Section 2.1.1.

o Quantifying N and P inputs to land

The MITERRA-Europe model outputs were used as inputs of N and P to agricultural land by adjusting the spatial level of detail to the basin scale (Figure 2.1). Inputs of N and P to agricultural land originate from applications of synthetic fertilizers and animal manure, grazing, atmospheric N deposition, biological N_2 fixation, organic matter leaching and P weathering (Figure 2.1). Losses of N and P during animal housing and manure storage (e.g., denitrification and leaching) were considered before manure application. The losses from animal housing and storage systems were calculated as a function of emission and leaching fractions by the MITERRA-Europe model (Velthof et al., 2009; Velthof et al., 2014). N inputs to non-agricultural land include at-

mospheric deposition, organic matter leaching, and biological N_2 fixation by natural vegetation; and P inputs to non-agricultural land include organic matter leaching and P weathering (Figure 2.1). Nutrient inputs to land from organic matter leaching and P weathering were calculated by the new MARINA-Nutrients model as a function of area, runoff and coefficients (Table S2.3). N inputs to non-agricultural land were derived from the IMAGE model (i.e. atmospheric deposition and biological N_2 fixation) (Table S2.5) (Beusen et al., 2022).

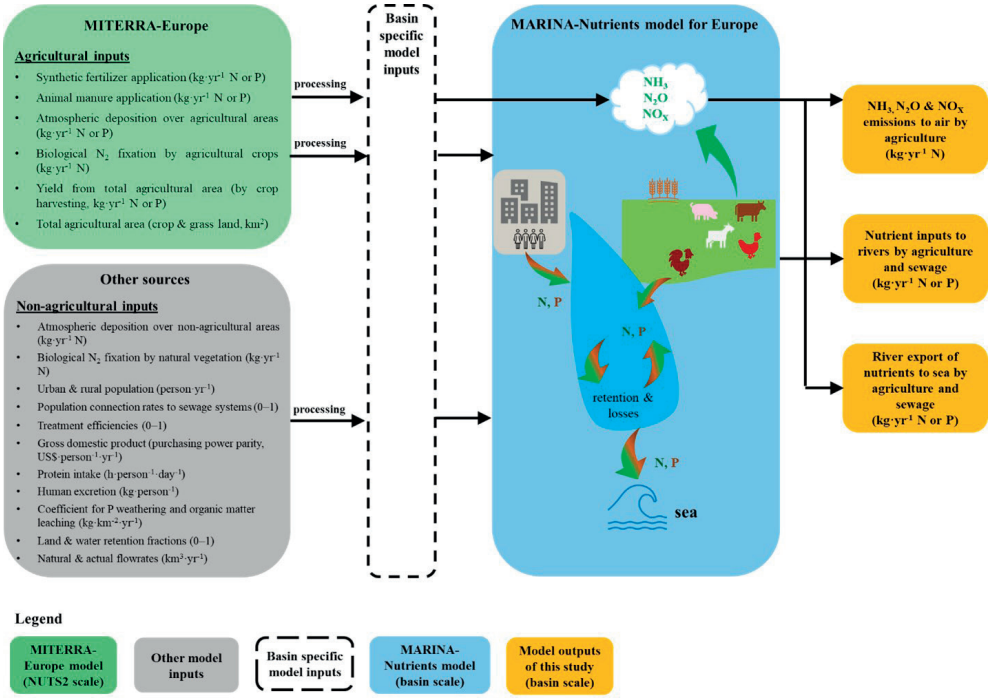


Figure 2.1 Framework of the new MARINA-Nutrients model for Europe. NUTS2: Nomenclature of Territorial Units for Statistics level 2 (Eurostat, 2022). N emissions to air are ammonia, nitrous oxide and nitrogen oxides (NO_x).

o *Quantifying N and P inputs to rivers*

Nutrient inputs to rivers from diffuse and point sources were calculated by the new MARINA-Nutrients model for Europe. The inputs of N and P to rivers from diffuse sources were calculated as a function of nutrient inputs to land that were corrected for crop uptake and animal grazing, and from point sources as a function of e.g., population, sewage connection rates and treatment efficiencies. Nutrient retentions and losses (e.g., denitrification) in soil are calculated as a function of runoff following a process-based approach of Strokal et al. (2016) and Li et al. (2022b). Nutrient inputs to rivers from agricultural and non-agricultural areas were quantified following Strokal et al. (2016) and uncalibrated modeling approach of Li et al. (2022b).

The model quantifies the following nutrient forms: dissolved inorganic N (DIN), dissolved organic N (DON), dissolved inorganic P (DIP) and dissolved organic P (DOP) to rivers. The sum of inorganic and organic forms is equal to the total dissolved N (TDN) and total dissolved P (TDP). The inputs of TDN and TDP to rivers were calculated considering retentions and losses on land based on the overall equations (Strokal et al., 2016; Li et al., 2022b):

$$RSdif_{F.y.j} = WSdif_{E.y.j} x\ G_{F.j} x\ FE_{ws.F.j} \quad (1)$$

$$RSpnt_{F.y.j} = RSpnt_{E.y.j} x\ FEpnt_{F.y} \quad (2)$$

where, $RSdif_{F.y.j}$ is the total input of nutrient form F (DIN, DON, DIP, DOP) to rivers by diffuse source y and basin j (kg·yr^{-1}), $WSdif_{E.y.j}$ is the total input of nutrient element E to agricultural or non-agricultural land in basin j from source y (kg·yr^{-1}), $G_{F.j}$ is the fraction of nutrient form F that is remained in soils of basin j after animal grazing and crop harvesting (0–1) only applied to agricultural land, and $FE_{ws.F.j}$ is the export fraction of nutrient form F entering rivers of basin j (0–1). The $FE_{ws.F.j}$ fraction is calculated as a function of annual runoff from land to streams and takes implicitly into account the retentions of nutrients in soils prior to their transport to rivers. $RSpnt_{F.y.j}$ is the total input of nutrient form F (DIN, DON, DIP, DOP) to rivers by point source y and basin j (kg·yr^{-1}), $RSpnt_{E.y.j}$ is the input of nutrient element E from point source y to rivers in basin j (kg·yr^{-1}), and $FEpnt_{F.y}$ is the fraction of nutrient form F entering rivers from point source y in basin j (0–1) (Table S2.3, Table S2.4, and Table S2.5 provide more details).

o *Quantifying N emissions to air*

N emissions to air from sewage systems and unconnected human waste were not considered in this study as the main focus was agriculture. The emissions of ammonia (NH_3), nitrous oxide (N_2O) and nitrogen oxides (NO_x) to air from animal and crop production systems were calculated as a function of land use, animal numbers, manure management and emission factors by the MITERRA-Europe model (Table S2.6) (Duan et al., 2021). We took the emissions from MITERRA-Europe in NUTS2 scale and aggregated them to the basin scale (Table S2.5). Total N emissions to air from agriculture were calculated based on the overall equation:

$$N_{total.y.j} = NH_{3.y.j} + N_2O_{y.j} + NO_{x.y.j} \quad (3)$$

where, $N_{total.y.j}$ is the total emission of N to air by agricultural source y and basin j (kg·yr^{-1} N), $NH_{3.y.j}$ is the emission of NH_3 to air by agricultural source y and basin j (kg·yr^{-1} N), $N_2O_{y.j}$ is the emission of N_2O to air by agricultural source y and basin j (kg·yr^{-1} N), and $NO_{x.y.j}$ is the emission of NO_x to air by agricultural source y and basin j (kg·yr^{-1} N) (Table S2.3, Table S2.4, and Table S2.5 provide more detail).

o *Quantifying river exports of N and P*

The new MARINA-Nutrients model also accounts for river exports of DIN, DON, DIP and DOP by the European basins as a function of nutrient inputs to rivers, retentions and losses in rivers. The overall equation is as follows (Strokal et al., 2016):

$$M_{F.y.j} = \left(RSdif_{F.y.j} + RSpnt_{F.y.j}\right) x\ FE_{riv.F.outlet.j} x\ FE_{riv.F.mouth.j} \quad (4)$$

where $M_{F.y.j}$ is the total river export of nutrient form F (DIN, DON, DIP, DOP) by source y and basin j (kg·yr^{-1}). $FE_{riv.F.outlet.j}$ is the fraction of nutrient form F exported to the outlet of basin j (0–1). $FE_{riv.F.mouth.j}$ is the fraction of nutrient form F exported from the basin j outlet to the river mouth (0–1). River retentions include damming, sedimentation (only for P) and water consumption calculated by equations 74–89 in Table S2.3 based on the data derived from HydroLAKES (Messager et al., 2016) (Table S2.3, Table S2.4, and Table S2.5 provide more details).

o *Defining hotspots for N and P losses*

Hotspots for total N emissions to air, nutrient inputs to rivers and nutrient exports by rivers to seas are defined following the approach of Wang et al. (2018) and Li et al. (2022b). First, the model results on the total N emissions to air, the inputs of TDN and TDP to rivers and the river exports of TDN and TDP to seas per km^2 of basin area were ranked from the lowest to the highest values. We had five groups for pollution levels: Group I (20% of the basins) with the lowest pollution levels and Group V (20% of the basins) with the highest pollution levels. Group V (top 20% of the basins) is considered a pollution hotspot. Air pollution hotspots represent the basins in Group V based on total N emissions to air (Figure 2.2). Water pollution hotspots represent the basins in Group V based on only TDN, only TDP or both TDN and TDP inputs to rivers (Figure 2.2). Air and water pollution hotspots represent the basins in Group V based on N losses to air and TDN inputs to rivers, N losses to air and TDP inputs to rivers or all three (Figure 2.2). The ranges of the groups are described in Section 2.3.

2.2.2 Model performance

The new MARINA-Nutrients model for Europe was validated by comparing modeled river exports with the measurements of DIN, DON, TDN, DIP, DOP and TDP at the river mouths (Figure 2.3). We derived the measured concentrations (mg·L^{-1}) of DIN, DON, TDN, DIP and TDP from the stations at or close to the river mouths for the period of 2000–2017 from the GEMStat database (UNEP, 2022). We included the observed yields of DIN, DON, DIP and DOP (kg·km^{-2}·yr^{-1}) that were used to validate a nutrient model including Europe (Global *NEWS-2* model) for the period of 1990–2000 (Mayorga et al., 2010). We also used the data from literature on concentrations (mg·L^{-1}) (Hesse & Krysanova, 2016) and loads (kg·yr^{-1}) (Ylöstalo et al., 2016; Cozzi et al., 2018; Räike et al., 2018; Vybernaite-Lubiene et al., 2018; Friedland et al., 2019; Räike et al., 2020) of DIN, DON, DIP, DOP, total N

(TN) and total P (TP) for various years (e.g., 1995 and 2001) and periods changing between 1980–2016. All observed values for 58 rivers were normalized to the same unit ($kg \cdot yr^{-1}$) for model validation. Details of the data are given in Table S2.7 and the validation results are indicated in Section 2.3.

2.3 Results and discussion

We start with the model results for nutrient inputs to land. Next, we present nutrient inputs to rivers, N emissions to air, and nutrient export to seas by basins. Due to the model inputs from different years (i.e. 2010, 2017 and 2020), the model results are averages for the period 2017–2020.

2.3.1 Nutrients on land from agriculture

Under the current practice, the new MARINA-Nutrients model estimated the total inputs of 34.4 $Tg \cdot yr^{-1}$ N and 1.8 $Tg \cdot yr^{-1}$ P to land in the EU-28. Agriculture was responsible for 84% of N and 96% of P on land (Table S2.8). In most of the basins, synthetic fertilizer application was the main contributor of N inputs to agricultural land with a range of 41–9260 $kg \cdot km^{-2} \cdot yr^{-1}$ N per basin. Animal manure (applied on agricultural land from stables and deposited on land during grazing) was the main contributor of P inputs to agricultural land with a range of 2–2190 $kg \cdot km^{-2} \cdot yr^{-1}$ P per basin. Most of the inputs to agricultural land were removed via crop harvesting (i.e. 62% of N and 72% of P). Only 12% of N and 3% of P inputs on agricultural land reached the rivers. N emissions to air from agricultural soil were 9% of N inputs on agricultural land. Most of the remaining P input is retained in the agricultural soil.

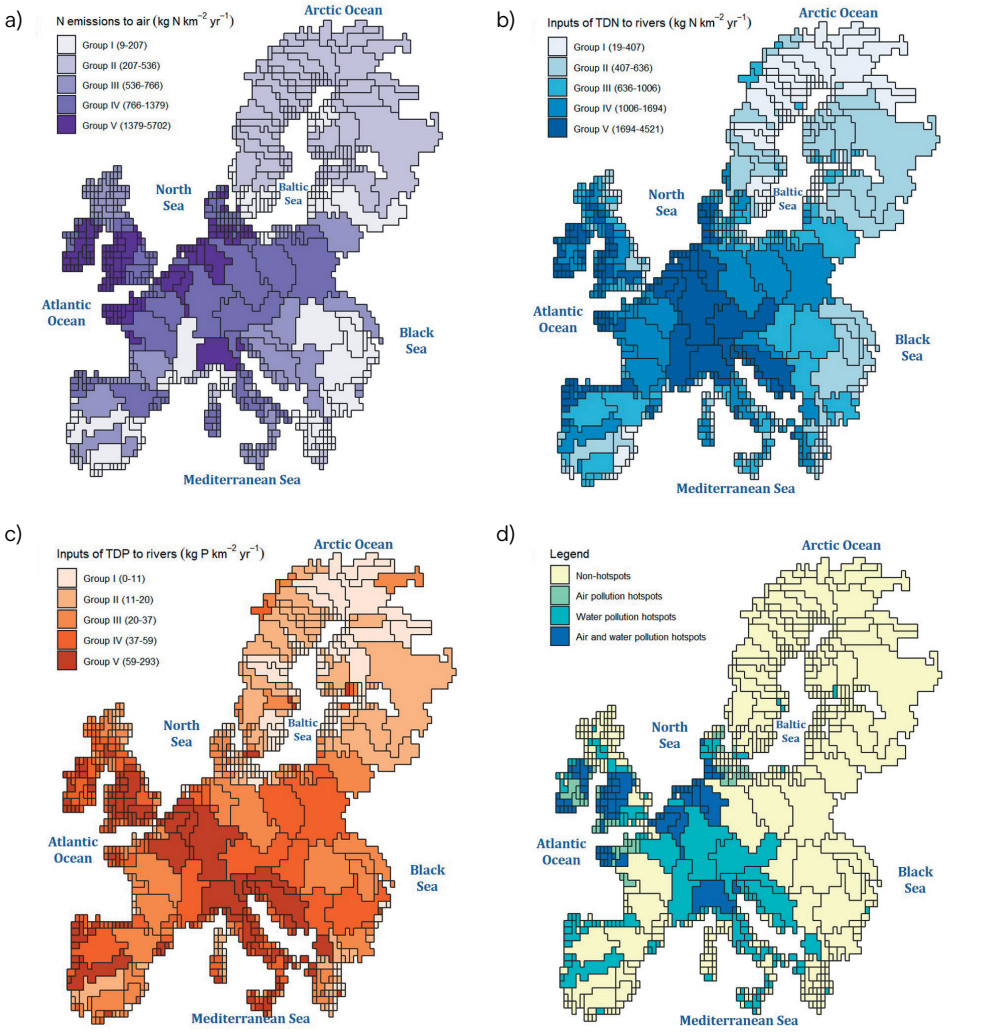


Figure 2.2 Nutrient losses to air and waters by the European basins. a) Reactive N emissions to air by basin (kg·km^{-2}·yr^{-1} N). b) Inputs of total dissolved N (TDN) to rivers by basin (kg·km^{-2}·yr^{-1} N). c) Inputs of total dissolved P (TDP) to rivers by basin (kg·km^{-2}·yr^{-1} P). Groups I to V were defined based on the pollution levels (20% of basins for each group) for N losses to air, and N and P losses to rivers by basins. d) Combined map of N emissions to air, inputs of TDN and TDP to rivers (kg·km^{-2}·yr^{-1} N or P).

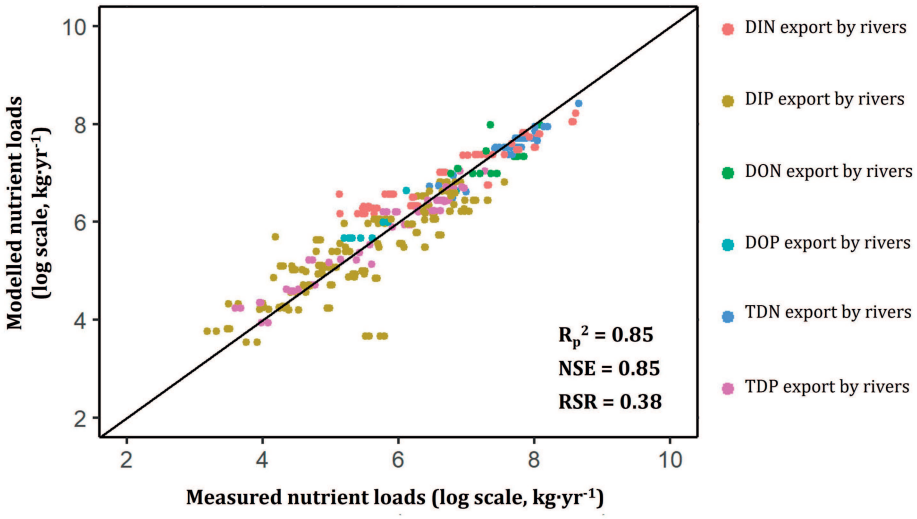


Figure 2.3 Measured versus modeled river exports of dissolved inorganic N (DIN), dissolved organic N (DON), total dissolved N (TDN), dissolved inorganic P (DIP), dissolved organic P (DOP) and total dissolved P (TDP) to the European seas (log10 scale; kg·yr^{-1}). Each dot represents an individual river mouth for 58 rivers. Measured loads indicate all the observed data (average per year) at or close to the river mouths in various years between 1990–2017 (Table S2.7 for details). Modeled nutrient loads are annual river exports of nutrients at the river mouths between 2017–2020.

2.3.2 Nutrients in rivers from agriculture and sewage

European rivers received 6.2 Tg·yr^{-1} of TDN and 0.25 Tg·yr^{-1} of TDP between 2017–2020 (Figure 2.2). Most of the TDN inputs were DIN (88%) and the rest was DON (12%). The DIP constituted 87% of TDP inputs to rivers while DOP constituted 13%. For the inputs of TDN and TDP to rivers per basin, the five groups have ranges changing between 19–4521 kg·km^{-2}·yr^{-1} N and 0–293 kg·km^{-2}·yr^{-1} P, respectively (Figure 2.2).

The contribution of sources to river pollution differed among nutrients and basins. Agriculture was responsible for 55% of N and sewage for 67% of P in rivers (Figure 2.4). From agriculture, synthetic fertilizer application was the largest source of the TDN inputs to rivers for most of Europe, while manure (applied on agricultural land from stables and deposited on land during grazing) was in western Europe (Figure 2.4). However, for the northern European basins, diffuse sources from non-agricultural areas (e.g., organic matter leaching and P weathering) were mainly responsible for the nutrient inputs to rivers (Figure 2.4). The diffuse sources from non-agricultural areas accounted for 13% and 32% of TDN and TDP inputs to rivers, respectively. Point sources were the main contributors to TDP inputs to rivers while sewage systems constituted 13% of the TDN inputs to rivers (Figure 2.4).

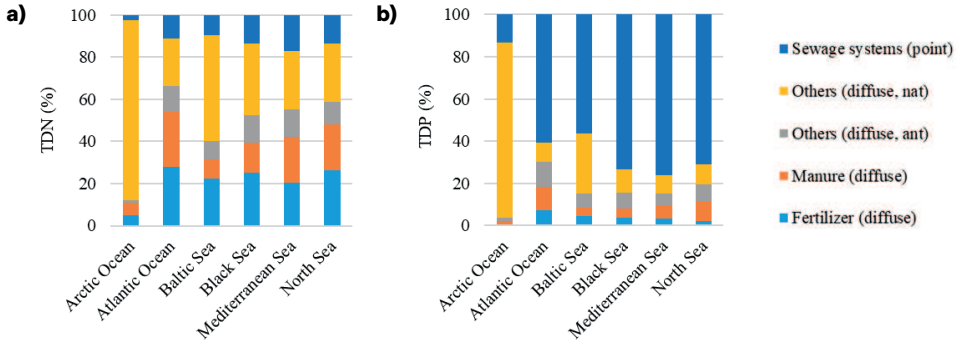


Figure 2.4 a) Relative contribution of sources to inputs of total dissolved N (TDN) to rivers per discharge sea (%). b) Relative contribution of sources to inputs of total dissolved P (TDP) to rivers per discharge sea (%). In legend, fertilizer means synthetic fertilizers applied; manure means that applied on agricultural land from stables and deposited on land during grazing; others (diffuse, ant) means other diffuse anthropogenic sources of N (i.e. atmospheric deposition and organic matter leaching over agricultural areas, and biological N_2 fixation by crops) and P (i.e. atmospheric deposition, organic matter leaching and P weathering over agricultural areas) in rivers; others (diffuse, nat) means other diffuse non-agricultural sources of N (i.e. atmospheric deposition and organic matter leaching over non-agricultural areas, and biological N_2 fixation by natural vegetation) and P (i.e. organic matter leaching and P weathering over non-agricultural areas) in rivers; and sewage systems means effluent from wastewater treatment plants.

2.3.3 Nitrogen emissions to air from agriculture

Reactive N emissions to air were 4.0 $Tg \cdot yr^{-1}$ N between 2017–2020 including the emissions from agricultural soil (2.5 $Tg \cdot yr^{-1}$ N) and animal housing and storage systems (1.5 $Tg \cdot yr^{-1}$ N) (Figure 2.2). Animal housing and storage systems were an important contributor by almost two-fifth (38%) of reactive N emissions to air from agriculture. Reactive N emissions consisted of NH_3, N_2O, and NO_x from animal housing and storage systems and agricultural soils. For the reactive N emissions to air per basin, the five groups have ranges changing between 9–5702 $kg \cdot km^{-2} \cdot yr^{-1}$ N (Figure 2.2). NH_3 emissions were the largest contributor to the reactive N emissions to air from housing and storage systems (37%), manure application (24%), synthetic fertilizer application (17%) and grazing (7%) (Table S2.9). N_2O and NO_x emissions contributed to 12% and 4% of the reactive N emissions, respectively.

2.3.4 Air and water pollution hotspots

Air and river pollution was concentrated in the basins of western, central and southern Europe (Figure 2.2). Basins with river inputs exceeding 1694 $kg \cdot km^{-2} \cdot yr^{-1}$ N for TDN and 59 $kg \cdot km^{-2} \cdot yr^{-1}$ P for TDP were considered water pollution hotspots (Figure 2.2). Basins with reactive N emissions to air from agriculture exceeding 1379 $kg \cdot km^{-2} \cdot yr^{-1}$ N were considered air pollution hotspots (Figure 2.2). As a result, 230 basins were assessed as pollution hotspots of air or water or both. Under the current practice, 31% of the European basin area, including 37% of the total agricultural land and 59% of the total population, was responsible for over half of the losses to air and rivers (Figure 2.2). These hotspots mainly resulted from agricultural activities and generated 53% of the total N emissions to air, 55% of TDN, and 57 of TDP losses to rivers between 2017–2020 (Figure 2.2). In Europe, 7% of the total basin area having 18% of the population was polluted by nutrient losses to air and water simultaneously (Figure 2).

2.3.5 River exports of nutrients

Under the current practice, 2.7 $Tg \cdot yr^{-1}$ of TDN and 0.11 $Tg \cdot yr^{-1}$ of TDP were exported by rivers to the European seas (Table S2.10, Figure 2.5 and Figure 2.6). For river exports of TDN and TDP, five groups have ranges changing between 12–2746 $kg \cdot km^{-2} \cdot yr^{-1}$ N and 0–150 $kg \cdot km^{-2} \cdot yr^{-1}$ P, respectively (Figure 2.5 and Figure 2.6). Hotspots for river exports (Group V) were responsible for two-fifths (40%) of TDN and nearly half (46%) of TDP exports to the European seas (Figure 2.5 and Figure 2.6). Over a fourth of TDN (28%) was exported to the Atlantic Ocean and TDP (27%) to the Mediterranean Sea, and the rest was exported to the Arctic Ocean, Baltic, Black and North seas by rivers (Figure 2.5 and Figure 2.6).

Source contribution to the river exports of nutrients varied to a large extent among the discharge seas (Figure 2.5 and Figure 2.6). For example, the application of synthetic fertilizer and animal manure (including manure deposited on land during grazing) contributed considerably to the DIN exports, whereas organic matter leaching over non-agricultural areas was the main source of the DON export (e.g., the Arctic Ocean) (Figure 2.5). Sewage systems accounted for two-thirds of the TDP export and were the major source of the DIP export (Figure 2.6). Except for the Arctic Ocean, organic matter leaching over non-agricultural areas was the main source of the DOP export by rivers (Figure 2.6).

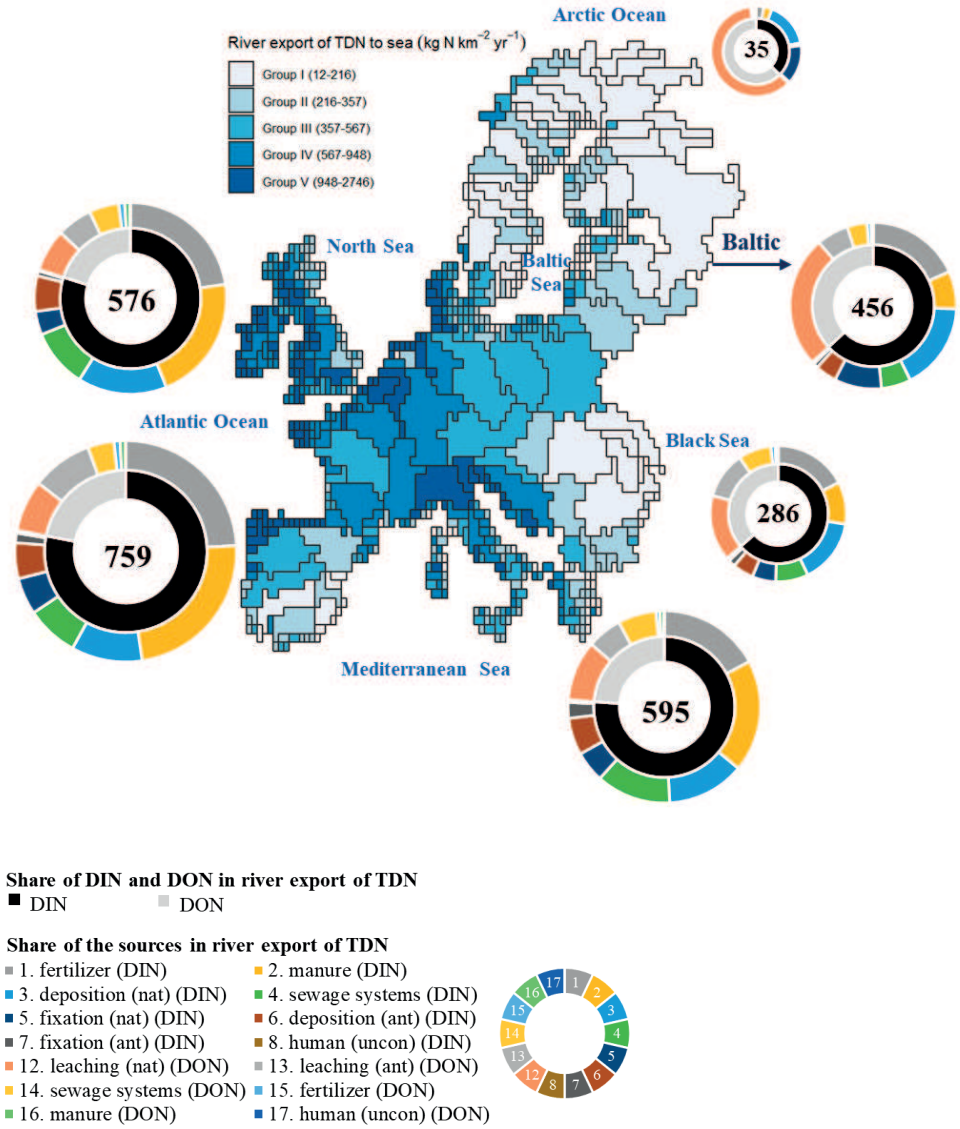


Figure 2.5 River export of total dissolved N (TDN) per basin (kg·km^{-2}·yr^{-1} N). Groups I to V were defined based on the pollution levels (20% of basins for each group) for TDN export by basins. Doughnut pie charts indicate the river exports of dissolved inorganic N (DIN) and dissolved organic N (DON) (inner doughnut), and their source attribution (outer doughnut) in percentages. Numbers in doughnut pie charts represent the river export of TDN to each sea (Gg·yr^{-1} N). Numbers in the legend indicate the color codes for source attribution. In this figure, ant means for anthropogenic sources of N in seas; nat means non-agricultural sources of N in seas; and uncon means wastewater from population not connected to sewage systems.

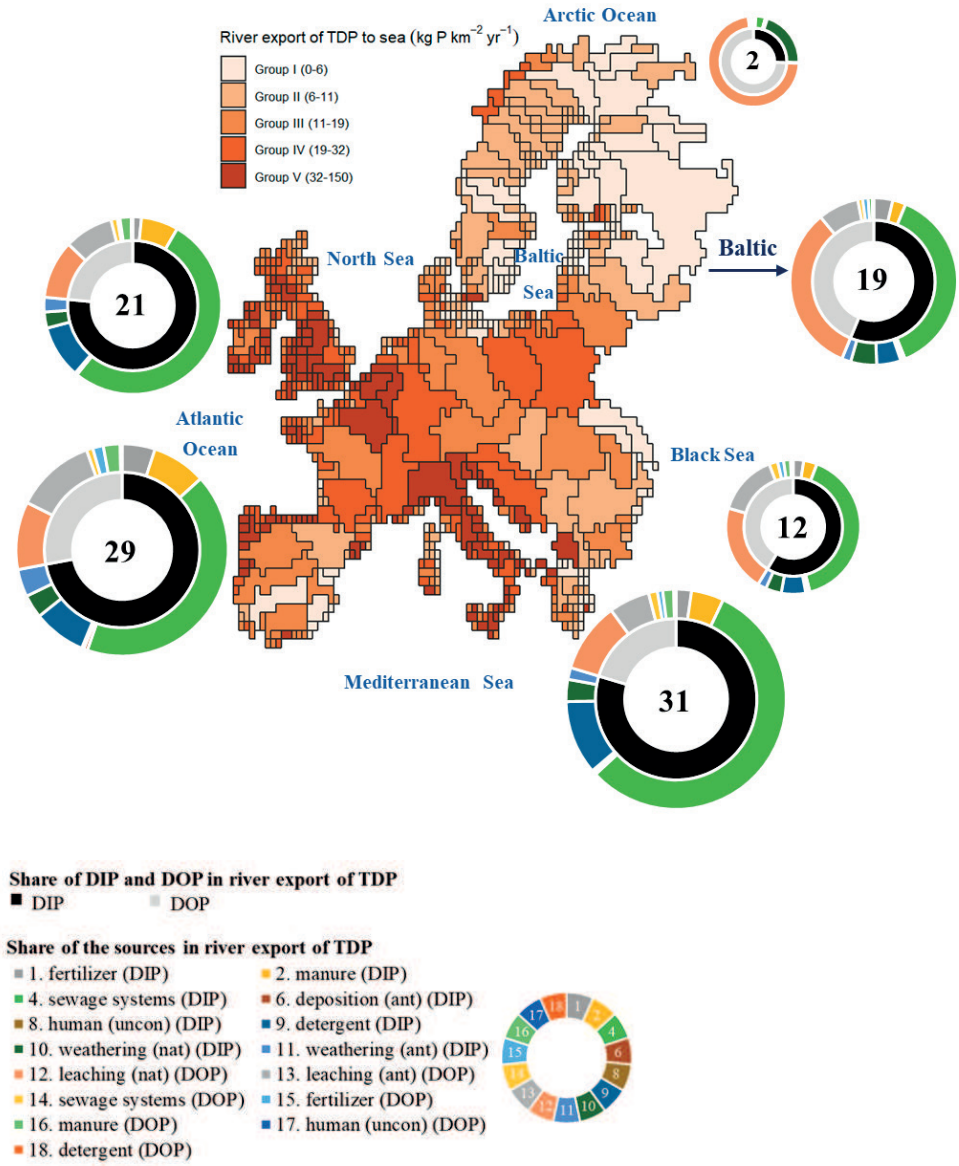


Figure 2.6 River export of total dissolved P (TDP) per basin (kg·km^{-2}·yr^{-1} P). Groups I to V were defined based on the pollution levels (20% of basins for each group) for river export of TDP by basins. Doughnut pie charts indicate the dissolved inorganic P (DIP) and dissolved organic P (DOP) exports (inner doughnut), and their source attribution (outer doughnut) in percentages. Numbers in doughnut pie charts represent the river export of TDP to each sea (Gg·yr^{-1} P). Numbers in the legend indicate the color codes for the source attribution. See Figure 2.5 for details of abbreviations.

2.3.6 Reflection on model advantages, limitations and uncertainties

The new MARINA-Nutrients model for Europe has a number of advantages. It provides a detailed assessment for the contribution of agriculture to air and water pollution in Europe in a spatially explicit way. The basin scale assessment of nutrient losses can help develop basin-specific nutrient management plans with effective pollution reduction options. This can contribute to analyzing integrated management options to reduce nutrient losses synergistically and help solving the N debate.

The model is restricted to dissolved organic and inorganic nutrient forms in rivers and seas taking into account the retentions and losses of these nutrients in the river networks. However, the model takes steady-state and process-based approaches, and does not consider particulate nutrients implying that the total pollution levels might be underestimated. Particulate nutrients in rivers are mainly caused by erosion (Beusen et al., 2005). We showed the contribution of agricultural production to river and coastal water pollution with the dissolved nutrient forms in European basins, which was the main objective of this study.

Consideration of physical and chemical factors that control the nutrient transport in the atmosphere (e.g., denitrification) and aquatic environment (e.g., denitrification, uptake by aquatic plants and sedimentation) are needed to fully understand the human impact on the atmospheric, aquatic and terrestrial ecosystems (Beusen et al., 2015). Our steady-state model, however, does not explicitly account for dynamic processes in the basins and rivers, and quantifies the nutrient retentions with a lumped approach. Nevertheless, we do account for the effect of climatic factors (e.g., rainfall) on the nutrient losses from land to surface waters. For instance, the fraction of nutrients exported from land to rivers in a basin is calculated as a function of surface runoff, which can differ among basins and is influenced by climate changes (footnotes of Table S2.3).

We account for N emissions to air from agriculture. However, N emissions to the atmosphere from other human activities (e.g., sewage systems and industries) (Strokal & Kroeze, 2014) as well as surface waters (Seitzinger & Kroeze, 1998; Seitzinger et al., 2000) should also be considered for a comprehensive analysis of agricultural contribution in future studies. Our study is the first attempt to integrate air and water pollution aspects and to quantify the contribution of agriculture to air and water pollution in the European basins. Future studies could build on this and apply our modeling tool to add industries and other sectors for N emissions to the air.

Some model input datasets are available on grids (e.g., atmospheric deposition over non-agricultural areas), and others are at national (e.g., sewage connection rates) or regional (e.g., synthetic fertilizer application) scales. We aggregated these model inputs into the basins by a process-based and lumped modeling approach. Through this approach, processes for retentions (e.g., P in soils) and losses (e.g., crop uptake) of nutrients in soils were modeled at the basin scale. Similarly, retentions (e.g., damming and sedimentation) and losses (e.g.,

water consumption) of nutrients in rivers were modeled at the basin scale. This approach may cause both under- or overestimation of the results and source attribution for air and water pollution in Europe.

As the data on dissolved nutrient forms were limited, the observations of TN and TP were also used in the validation. This could influence the validation results of dissolved nutrient export. In addition, both tidal effects and salt water intrusion can influence the measured nutrient concentrations and hence, the results of model validation. However, we focused on annual averages and did not consider the seasonality. Thus, we consider that the tidal effects do not have large influence on our model validation conclusions.

The most recently available data were used as model inputs. The data were available for different years. Thus, discrepancies in the temporal scale of the inputs might also affect model results. However, our conclusions are still relevant because the results are presented for the period of 2017–2020. This is the period for which most of the model inputs were derived.

Hotspots for air pollution, water pollution, and air and water pollution were defined by considering the top 20% of most polluted basins. This approach has been applied in other studies for nutrients (Wang et al., 2018; Li et al., 2022b). However, the approach does not consider explicitly the adverse impacts of nutrient pollution (e.g., nitrate pollution for drinking purposes and eutrophication in surface waters). Our approach aims to provide an indication of the pollution extent between the basins. Thus, ranking the basins from the highest to the lowest nutrient pollution levels of air and/or water fits the intended purpose of the hotspot analysis. This makes it easier to identify nutrient-related hotspots of both air and water pollution.

2.3.7 Comparisons of model outputs with measurements and other studies

We validated our model by comparing modeled river exports of DIN, DON, TDN, DIP, DOP and TDP with the measurements at or close to the river mouths. The model performance was assessed by the Pearson's coefficient of determination (R^2; 0 to 1), Nash-Sutcliffe efficiency (NSE; -∞ to 1), and the ratio of root mean square error (RMSE) and standard deviation of observations (RSR; 0 to +∞) according to Moriasi et al. (2007). R^2 indicates the degree of collinearity between modeled and observed data (Moriasi et al., 2015). NSE shows how well the modeled and observed data fits the 1:1 line (Moriasi et al., 2015). RMSE generally gives larger weight to high values than low values as errors in high values are usually greater in absolute value than errors in low values (Moriasi et al., 2015). Therefore, Moriasi et al. (2007) suggested normalizing RMSE by the standard deviation of the observations which is referred to as RMSE-observations standard deviation ratio (RSR). These indicators performed well in the new MARINA-Nutrients model: R^2 = 0.85, NSE = 0.85 and RSR = 0.38 (Moriasi et al., 2007; Moriasi et al., 2015) (Figure 2.3). We also calculated the statistical indicators for TDN and TDP, which are also promising. The statistics were: for DIN, R^2 =

0.88, NSE = 0.75 and RSR = 0.49; for TDN, R^2 = 0.89, NSE = 0.80 and RSR = 0.44; for DIP, R^2 = 0.72, NSE = 0.70 and RSR = 0.54; and for TDP, R^2 = 0.95, NSE = 0.93 and RSR = 0.26.We compared our model results with those of other modeling studies to build trust in the new MARINA-Nutrients for Europe (Strokal et al., 2016). The N inputs to agricultural land are lower for fertilizer, manure, biological fixation but higher for atmospheric deposition than the estimations of de Vries et al. (2021) for 2010 (Table S2.11). The differences can be explained by different estimation years and model approaches.

Spatial variabilities in river pollution hotspots of Europe coincide with existing studies. For instance, water pollution hotspots in western and central Europe are in line with the studies quantifying nutrient inputs to surface waters (de Vries et al., 2021; Li et al., 2022b).

A validation as we did for the river exports of nutrients is challenging for the N emissions to air because these emissions are not readily measured. We used existing modeling approach that has been evaluated in previous studies (de Vries et al., 2011; Kros et al., 2012). We built trust in our model by comparing results with those of other studies. Our model results for the emissions of NH_3, N_2O and NO_x to air are comparable with other modeling studies for Europe (Table S2.9) (Bouwman et al., 2006; de Vries et al., 2010; de Vries et al., 2011). Our modeled NH_3 (3401 $Gg{\cdot}yr^{-1}$ N) is higher than the other estimates (2848–3066 $Gg{\cdot}yr^{-1}$ N), N_2O (374 $Gg{\cdot}yr^{-1}$ N) is slightly lower (379–511 $Gg{\cdot}yr^{-1}$ N) and NO_x (155 $Gg{\cdot}yr^{-1}$ N) is within the range of other studies (77–219 $Gg{\cdot}yr^{-1}$ N) (Table S2.9). The differences between our and other studies could be associated with differences in emission factors used and estimation years.

The river export of TDN is comparable with other modeling studies (Table S2.12) (Mayorga et al., 2010; Beusen et al., 2016). However, the modeled TDP is lower than other modeling studies (Table S2.13) (Mayorga et al., 2010; Beusen et al., 2016). This could be due to different scope of some other studies (footnotes in Table S2.13). TDN and TDP were compared with TN and TP in some cases (footnotes in Table S2.12 and Table S2.13). This may also explain the differences between our results and other studies (Grizzetti et al., 2012; Beusen et al., 2015; Grizzetti et al., 2021).

2.3.8 Implications for future policies

Synergetic solutions are needed to simultaneously mitigate air and water pollution in Europe. To develop these solutions, we need a comprehensive assessment of N and P pollution for air and water. This new MARINA-Nutrients model for Europe quantifies nutrient losses to air and waters from human activities by accounting for direct and indirect losses from agricultural production systems (e.g., housing and storage, cropland, and grassland). The new model can provide full N and P cycles in agricultural systems as well as nutrient losses from sewage systems by addressing the two significant sources of nutrient pollution in Europe. Our basin-scale model allows for spatially explicit analysis of air and water pollution, and

hence to develop specific measures (e.g., river basin management plans). For example, in air and water pollution hotspots, the intensity of agricultural activities can be decreased, whereas we can focus on specific runoff-reducing measures in only water pollution hotspots. Our results could assist policymakers to formulate effective and integrative nutrient management strategies, prioritize measures and contribute to preventing their trade-offs by basin-scale analysis of air and water pollution in Europe. The new model is also applicable for the other world basins under different climatic conditions through the MARINA model family (e.g., China and Global).

Suggestions for future studies include considering the other sources (e.g., industrial wastewater and aquaculture) to quantify the nutrient losses to rivers and coastal waters, as well as the N emissions to air (e.g., transportation) by the model. This will provide a full analysis of N emissions to air and the inputs of N and P to rivers and coastal waters in Europe.

2.4 Conclusions

This study quantified annual inputs of N and P to land and rivers, N emissions to air, and river exports of N and P to seas by basins in Europe. For this purpose, we developed and applied a new version of the MARINA-Nutrients model to 601 European basins. Results showed that agriculture was responsible for 84% of N and 96% of P on land between 2017–2020. Synthetic fertilizer and manure applications (including manure deposited on land during grazing) were the largest contributors to the nutrients on land. Of these inputs, 12% of N and 3% of P reached the rivers. The sources of air and river pollution varied considerably among the basins. Agriculture was responsible for 55% of N and sewage for 67% of P in rivers among the sources considered in this study. Almost two-fifths of reactive N emissions to air were from animal housing and storage. Nearly a third of the basin area was responsible for over half of total N emissions to air and nutrient pollution in rivers. Over a fourth of river export of N ended up in the Atlantic Ocean and of P in the Mediterranean Sea. Our study can assist the formulation of effective nutrient management strategies by basin-scale analysis of air and water pollution to prevent pollution swapping in Europe.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Aslıhan Ural-Janssen: Conceptualization, Methodology, Validation, Visualization, Roles/Writing – original draft, Writing – review & editing. Carolien Kroeze: Conceptualization, Methodology, Supervision, Writing – review & editing, Funding acquisition. Jan Peter Lesschen: Methodology, Resources, Writing – review & editing. Erik Meers: Supervision, Writing – review & editing, Funding acquisition. Peter J.T.M. van Puijenbroek: Methodology, Resources, Writing – review & editing. Maryna Strokal: Conceptualization, Methodology, Supervision, Writing – review & editing, Funding acquisition, Project administration.

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Supplementary materials and data availability

Supplementary materials can be found online at https://doi.org/10.15302/J-FASE-2023526 and in this thesis under *S2: Supplementary materials for Chapter 2*. The main model results supporting Figures 2.2–2.6 generated in this study are deposited in the DANS Easy repository https://doi.org/10.17026/DANS-ZG6-7WZ4.



Chapter 3

Large reductions in nutrient losses needed to avoid future coastal eutrophication across Europe

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This paper is adjusted to the PhD thesis format including the adjustment of some river names.

Abstract

Rapid technological development in agriculture and fast urbanization have increased nutrient losses in Europe. High nutrient export to seas causes coastal eutrophication and harmful algal blooms. This study aims to assess the river exports of nitrogen (N) and phosphorus (P), and identify required reductions to avoid coastal eutrophication in Europe under global change. We modelled nutrient export by 594 rivers in 2050 for a baseline scenario using the new MARINA-Nutrients model for Europe. Nutrient export to European seas is expected to increase by 13-28% under global change. Manure and fertilizers together contribute to river export of N by 35% in 2050. Sewage systems are responsible for 70% of future P export by rivers. By 2050, the top ten polluted rivers for N and P host 42% of the European population. Avoiding future coastal eutrophication requires over 47% less N and up to 77% less P exports by these polluted rivers.

3.1 Introduction

Coastal eutrophication is a serious problem in Europe (Boesch, 2019). Eutrophication occurs by nutrient enrichment in aquatic environments from land-based diffuse (e.g., agricultural runoff) and point sources (e.g., sewage systems), and atmospheric deposition (EEA, 2019). Eutrophication directly affects the aquatic environment by e.g., the increased frequency of algal blooms (Glibert et al., 2018a), increased sedimentation, depletion of light, changes in aquatic vegetation (Njock et al., 2023) and indirectly by oxygen depletion (Glibert et al., 2018b) and fish kills (Kudela et al., 2018). Despite the environmental policies, rivers transport large amount of nutrients to coastal waters (Nikolaidis et al., 2022; Vigiak et al., 2023) and thus, eutrophication is still an issue in European seas (Jansson et al., 2019).

Human activities are main drivers of nutrient losses in surface waters (Poikane et al., 2020; Grizzetti et al., 2021), and thus coastal eutrophication. Intensive agriculture (e.g., use of synthetic fertilizers, animal manure) and domestic sewage systems have been major diffuse and point sources of nutrients in Europe, respectively (Agency et al., 2018; Ural-Janssen et al., 2023). Other pressures (e.g., morphological, hydrological) also play a role in the coastal eutrophication (Garnier et al., 2021). It is important to estimate the nutrient losses to rivers and seas to reduce or prevent the risk of coastal eutrophication in future.

Water quality models can help estimate nutrient losses to surface waters (Shan et al., 2023). Several models have been developed for Europe at different spatial scales. For example, GREEN is a statistical model estimating the annual loads of nitrogen (N) and phosphorus (P) to seas at a catchment scale around 200 km^2 (Grizzetti et al., 2012). Riverstrahler is a biogeochemical drainage network model calculating the nutrient export at the river outlet considering processes through the drainage network (e.g., retention, transfer and transformations) and in the riparian zone (e.g., denitrification) (Billen et al., 2018; Billen & Garnier, 2022). The MARINA-Nutrients model for Europe is a process based model quantifying the inputs of N and P to rivers, and their river export to seas at the river basin scale (Ural-Janssen et al., 2023), and developed based on the MARINA (Model to Assess River Inputs of pollutaNts to seAs) model approach. MARINA model has been implemented to predict future nutrient pollution in surface waters based on scenario analyses globally (Strokal et al., 2021) and for China (Wang et al., 2020a). These provide useful insights on the future trends of water pollution. However, such future analyses using the MARINA-Nutrients model are limited for European river basins.

Scenario analyses can help analyze future trends in coastal eutrophication. Shared Socioeconomic Pathways (SSPs) are environmental narratives that are widely used in modeling studies (van Vuuren et al., 2021; Beusen et al., 2022). The SSPs consist of five future trajectories based on various drivers (e.g., socioeconomic, technological) (Riahi et al., 2017). Changes in land use and climate are significant pressures driven by popula-

tion growth, socioeconomic development and dietary choices (van Vuuren et al., 2017). Changes in agricultural practices associated with land use alter nutrient runoff. SSPs are used alongside Representative Concentration Pathways (RCPs) to analyse the impacts of global change (Beusen et al., 2022; Li et al., 2022a). RCPs identify several alternatives for radiative forcing reaching 2.6-8.5 W m^{-2} by 2100 for future climate change (van Vuuren et al., 2011). These scenarios have been well applied to predict future N and P delivery to surface waters (Wang et al., 2020a; Beusen et al., 2022).

Most future studies on nutrients apply forecasting techniques to explore the future consequences of expected trends (van Puijenbroek et al., 2019; Hader et al., 2022; de Vries et al., 2023). Studies applying backcasting, by exploring ways to reach a desirable future, are scarce (Li et al., 2019). To our knowledge, no studies have been published on backcasting the future export of N and P by European rivers. Such backcasting exercises could be based on maximum allowable nutrient losses to seas, and support the formulation of effective nutrient management strategies that help avoid the risk of coastal eutrophication in future.

Models quantifying nutrient delivery to surface waters can provide important inputs to assess the risk of coastal eutrophication (Garnier et al., 2021; Billen & Garnier, 2022). Indicator for Coastal Eutrophication Potential (ICEP) is an approach to estimate the risk based on the Redfield ratio (Billen & Garnier, 2007). The Redfield ratio represents the C:N:P:Si ratio (106:16:1:20) for the growth of diatoms (Redfield et al., 1963). When there is excess N and/or P over silica (Si) in coastal waters, the non-siliceous algae might grow instead of diatoms (Billen & Garnier, 2007; Garnier et al., 2010). ICEP uses this imbalance to indicate the potential for coastal eutrophication, and the risk for harmful algal blooms (Billen & Garnier, 2007). Based on the limiting nutrient, N-ICEP or P-ICEP is calculated by an empirical formula based on the fluxes of N, P and dissolved silica (DSi) in coastal waters (Billen & Garnier, 2007). The ICEP value indicates coastal eutrophication potential (i.e. high or low) (Billen & Garnier, 2007), and hence can support the future scenario analyses to estimate the risk of coastal eutrophication in European seas.

This study aims to assess the future river exports of N and P, and identify required reductions to avoid coastal eutrophication in Europe under global change. We used the MARINA-Nutrients model for Europe (Ural-Janssen et al., 2023) to estimate the nutrient exports by 594 European rivers and ICEP by 2050 with a baseline scenario combining SSP5 and RCP8.5 (SSP5-RCP8.5) with the largest socioeconomic and climate change to observe the effect on the nutrient losses to waters. The year 2050 was chosen arbitrary as a medium-term between short-term (2030) and long-term (2100) future. In addition, many versions of the MARINA model have been applied to 2050 and already provided data for our model application (Strokal et al., 2017; Li et al., 2019; Wang et al., 2020a; Strokal et al., 2021; Li et al., 2023). We combine forecasting and backcasting. First, we quantified the nutrient

inputs to rivers and their export by rivers to the coastal waters of Europe for 2050 under the SSP5-RCP8.5 scenario. Second, we calculated the maximum allowable river export of nutrients to avoid coastal eutrophication based on the ICEP approach (Billen & Garnier, 2007; Garnier et al., 2010). Finally, we identified required reductions to avoid the risks of coastal eutrophication for the top ten polluted rivers of Europe regarding N and P.

3.2 Materials and methods

3.2.1 MARINA-Nutrients model for Europe

The MARINA-Nutrients model for Europe is developed to quantify N emissions to the atmosphere from agriculture, inputs of the N and P to rivers and river exports of N and P to seas from diffuse (i.e. agricultural and non-agricultural areas) and point (i.e. sewage systems) sources at the basin scale (Ural-Janssen et al., 2023). Diffuse sources include various land-based sources of nutrients per basin (and sub-basins of the Danube River, see Supplementary Text S3.1) while accounting for nutrient retention on land, and parameterized export processes of nutrients as a function of runoff (Ural-Janssen et al., 2023). The model has been validated for the river exports of N and P based on long term observations, and applied for current pollution in Europe in our pervious study (e.g., Pearson's coefficient of determination (R^2) = 0.72 - 0.95 depending on nutrient form, R^2 = 0.85 and Nash-Sutcliffe efficiency (NSE) = 0.85 for all nutrient forms of nutrient exports by 58 European rivers, indicating good performance (Ural-Janssen et al., 2023)). In addition, uncertainty assessments (e.g., sensitivity analyses) associated to the simulated river exports of nutrients by the MARINA models were applied for various model inputs as well as parameters in other studies (Strokal et al., 2016; Chen et al., 2019b; Wang et al., 2020a). Based on those results, the outputs of the MARINA models are sensitive mostly to changes in the following model inputs: water discharges, manure production and synthetic fertilizer application (Strokal et al., 2016; Wang et al., 2020a). We believe that this also holds for the MARINA-Nutrients model for Europe because we build on the modeling approach of the existing MARINA models. Here, we applied the model for future analyses of nutrient pollution in rivers and coastal waters of Europe.

The MARINA-Nutrients model for Europe calculates the river exports of N and P in dissolved inorganic (i.e. DIN, DIP) and organic (i.e. DON, DOP) forms as a function of nutrient inputs to agricultural and non-agricultural areas, nutrient inputs to rivers from sewage systems, and export fractions of those inputs reaching the coastal waters (Ural-Janssen et al., 2023). Sources of nutrient inputs to agricultural areas are the application of synthetic fertilizers, application of animal manure and manure deposited on land during grazing, biological N_2 fixation by crops, atmospheric deposition and human waste from population not connected to sewage treatment systems. Sources of nutrient inputs to non-agricultural areas are biological N_2 fixation by natural vegetation and atmospheric

deposition. Parameterized export processes are organic matter leaching from agricultural and non-agricultural soils, and P weathering from agricultural and non-agricultural soils following (Strokal et al., 2016). Point sources of nutrient inputs to rivers include urban and rural wastewater treatment systems, except industrial wastewaters.

The model accounts for hydrology (e.g., natural and actual water discharges), basin characteristics (e.g., land use), retention and losses of nutrients on land and rivers, and the distance between the source and river mouth (i.e. coastal waters) (Ural-Janssen et al., 2023). The model quantifies the river exports of N and P for all mentioned forms by the equations in Box 3.1 and Box S3.1 (Ural-Janssen et al., 2023).

Box 3.1 Main equations for quantifying the river exports of N and P to seas by the MARINA-Nutrients model for Europe (further equations in Box S3.1 and descriptions in Box S3.2).

$M_{F.y.j}$: River export of nutrients by source y and basin j

$$M_{F.y.j} = \left(RSdif_{F.y.j} + RSpnt_{F.y.j}\right) \times FE_{riv.F.outlet.j} \times FE_{riv.F.mouth.j} \quad (1)$$

$$RSdif_{F.y.j} = WSdif_{E.y.j} \times G_{F.j} \times FE_{ws.F.j} \quad (2)$$

$$RSpnt_{F.y.j} = RSpnt_{E.hum.con.j} \times FEpnt_{F.hum.con.j} \quad (3)$$

$$G_{F.j} = 1 - (WSdif_{E.ex.j}/WSdif_{F.gross.j}) \quad (4)$$

$$FE_{ws.F.j} = CR_F \times Rnat_j \quad (5)$$

$$RSpnt_{E.hum.con.j} = RSpnt_{E.hum.con.urb.j} + RSpnt_{E.hum.con.rur.j} \quad (6)$$

$$RSpnt_{E.hum.con.urb.j} = Eexc_{hum.con.urb.j} \times (1 - hw_{frem.E.j}) \quad (7)$$

$$RSpnt_{E.hum.con.rur.j} = Eexc_{hum.con.rur.j} \times (1 - hw_{frem.E.j}) \quad (8)$$

$$FEpnt_{DIN.hum.con.j} = 0.485 + 0.225 \times (hw_{frem.N.j} / 0.86), \text{ constant parameters for DON, DIP and DOP} \quad (9)$$

$$FE_{riv.F.outlet.j} = \left(1 - D_{F.j}\right) \times \left(1 - L_{F.j}\right) \times \left(1 - FQrem_j\right) \text{ for DIN and DIP} \quad (10)$$

$$FE_{riv.F.outlet.j} = \left(1 - FQrem_j\right) \text{ for DON and DOP} \quad (11)$$

$$FE_{riv.F.mouth.juT} = {}^{juT}FE_{riv.F.outlet.jmC} \times {}^{juT}FE_{riv.F.outlet.jdC} \quad (12)$$

$$FE_{riv.F.mouth.jmT} = {}^{jmT}FE_{riv.F.outlet.jdC} \quad (13)$$

$$FE_{riv.F.mouth.jmC} = {}^{jmC}FE_{riv.F.outlet.jdC} \quad (14)$$

$FE_{riv.F.mouth.jdC} = 1$ as the outlets of down-stream sub-basins discharge directly to seas

In Box 3.1: $M_{F.y.j}$ is the total river export of nutrient form F (generic for DIN, DON, DIP, DOP) by source y and basin j (kg yr^{-1}). River export of nutrients is quantified as a function of nutrient inputs to rivers that are corrected for removal (e.g., water consumption) and retention (e.g., sedimentation, damming) in rivers, and the travelling distance of nutrients to the river mouth (Eq. 1). $RSdif_{F.y.j}$ is the total input of nutrient form F (DIN, DON, DIP, DOP) to rivers by diffuse source y and basin j (kg yr^{-1}). Nutrient inputs to rivers from diffuse sources are calculated as a function of nutrient inputs to agricultural and non-agricultural land that are corrected for removal (via crop harvesting and animal grazing) and retention

(e.g., accumulation) in soils and runoff from land to streams (Eq. 2). $RSpnt_{F.y.j}$ is the total input of nutrient form F (DIN, DON, DIP, DOP) to rivers by point source y and basin j (kg yr^{-1}). Nutrient inputs to rivers from point source are quantified as a function of population that is connected to sewage systems, human N and P excretion rates, removal efficiencies of nutrients during wastewater treatment and the fraction of nutrients entering rivers by the effluent from sewage treatment systems (Eq. 3). $FE_{riv.F.outlet.j}$ is the fraction of nutrient form F (DIN, DON, DIP, DOP) exported to the outlet of basin j (0-1). This export fraction accounts for aquatic retention (e.g., sedimentation) and losses in water systems (e.g., denitrification and water removals) (Strokal et al., 2016). $FE_{riv.F.mouth.j}$ is the fraction of nutrient form F (DIN, DON, DIP, DOP) exported from the basin j outlet to the river mouth (0-1). $WSdif_{E.y.j}$ is the total input of nutrient element E (generic for N, P) to agricultural or non-agricultural land in basin j from source y (kg yr^{-1}). $G_{F.j}$ is the fraction of nutrient form F (DIN, DON, DIP, DOP) that is remained in soils of basin j after nutrient export from agricultural land by animal grazing and crop harvesting (0-1). The fraction is calculated as the ratio of nutrient inputs remained on agricultural land after animal grazing and crop harvesting ($WSdif_{F.gross.j} - WSdif_{E.ex.j}$) to the total inputs of nutrients to agricultural land ($WSdif_{F.gross.j}$). $FE_{ws.F.j}$ is the export fraction of nutrient form F (DIN, DON, DIP, DOP) entering rivers of basin j (0-1). The export fraction is calculated as a function of annual runoff from land to streams ($Rnat_j$) and implicitly accounts for nutrient retention in soils (e.g., denitrification, temporary accumulation in soils/aquifers) into account prior to nutrient transport to rivers (Strokal et al., 2016). $WSdif_{E.ex.j}$ is the export (ex) of nutrient element E (N, P) from agricultural land via crop harvesting and animal grazing in basins j (kg yr^{-1}). $WSdif_{F.gross.j}$ is the sum of all the inputs of nutrient form F (DIN, DON, DIP, DOP) to agricultural land in basin j (kg yr^{-1}). CR_F is the uncalibrated export coefficient of nutrient form F (DIN, DON, DIP, DOP) for runoff exporting nutrients from land to rivers (unitless). $Rnat_j$ is the annual surface runoff from land to streams in basin j (m yr^{-1}). $RSpnt_{E.hum.con.j}$ is the input of nutrient element E (N, P) to rivers from population connected to sewage systems in basin j (kg yr^{-1}). $FEpnt_{F.hum.con.j}$ is the fraction of nutrient form F (DIN, DON, DIP, DOP) entering rivers from population connected to sewage systems in basin j (0-1) (Table S3.2 for details). $Eexc_{hum.con.urb.j}$ is the nutrient element E (N, P) in human excretion from urban population connected to sewage systems in basin j (kg yr^{-1}). $Eexc_{hum.con.rur.j}$ is the nutrient element E (N, P) in human excretion from rural population connected to sewage systems in basin j (kg yr^{-1}). $hw_{frem.E.j}$ is the removal fraction of nutrient element E (N, P) during wastewater treatment in basin j (same for urban and rural treatment systems) (0-1). $D_{F.j}$ is the fraction of nutrient form F (DIN, DIP) retained in reservoirs and lakes in basin j (0-1). $L_{F.j}$ is the fraction of nutrient form F (DIN, DIP) retained in and/or lost from water systems (e.g., denitrification, sedimentation) in basin j (0-1). $FQrem_j$ is the fraction of nutrient form F (DIN, DON, DIP, DOP) removed from water systems in basin j by water consumption (0-1). Eqs. 12-14 are only used for the sub-basins of the Danube River (see Supplementary Text S3.1). $FE_{riv.F.mouth.juT}$ is the fraction of nutrient form F (DIN, DON, DIP, DOP) exported from the outlet of upstream tributary (juT) to the river mouth (0-1).

$^{juT}FE_{riv.F.outlet.jmC}$ and $^{juT}FE_{riv.F.outlet.jdC}$ are the fractions of nutrient form F (DIN, DON, DIP, DOP) exported from the outlet of upstream tributary (upper case: juT) to the outlets of the main channel in middlestream (lower case: jmC) and in downstream (lower case: jdC) sub-basins (0-1). $FE_{riv.F.mouth.jmT}$ is the fraction of nutrient form F (DIN, DON, DIP, DOP) exported from the outlet of middlestream tributary (jmT) to the river mouth (0-1). $^{jmT}FE_{riv.F.outlet.jdC}$ is the fraction of nutrient form F (DIN, DON, DIP, DOP) exported from the outlet of middlestream tributary (upper case: jmT) to the outlet of the main channel in downstream (lower case: jdC) sub-basins (0-1). $FE_{riv.F.mouth.jmC}$ is the fraction of nutrient form F (DIN, DON, DIP, DOP) exported from the outlet of middlestream main channel (jmC) to the river mouth (0-1). $^{jmC}FE_{riv.F.outlet.jdC}$ is the fraction of nutrient form F (DIN, DON, DIP, DOP) exported from the outlet of the main channel in middlestream (upper case: jmC) to the outlet of the main channel in downstream (lower case: jdC) sub-basin (the outlet of this downstream subbasin is the river mouth) (0-1).

3.2.2 Description of baseline scenario for 2050 and model inputs

We selected the SSP5-RCP8.5 as a baseline scenario for the year 2050 to identify required reductions in the river export of nutrients. SSP5 includes relatively rapid economic development, high fossil fuel dependency, high-tech increased agricultural production and local environmental management strategies (O'Neill et al., 2017). Our SSP5 scenario assumes a high economic development with high urbanization and low population growth (Strokal et al., 2021). We coupled the SSP5 with the RCP8.5 scenario considering high-end climate change (van Vuuren et al., 2011). Scenario considerations are indicated in Table S3.1 (Table S3.2 for model inputs). We estimated the inputs of N and P to rivers, and their export by rivers to seas for 2050 based on the SSP5-RCP8.5 scenario in a pessimistic future by using the MARINA-Nutrients model for Europe.

Model inputs for agricultural activities (e.g., nutrient inputs to land from fertilizers) were derived from the MITERRA-Europe model at regional scale and processed to a basin scale (see Ural-Janssen et al. (2023) for further details). MITERRA-Europe is an integrated model to assess the implementation of agricultural measures on emissions to air and waters in Europe (Velthof et al., 2009). The model estimates N emissions to air (e.g., N_2O, NH_3) from cropland, grassland, and animal housing and storage systems, and N and P balances in agriculture at NUTS2 scale based on emission factors and mass balance approach (Velthof et al., 2009). We used nutrient inputs to agricultural land by e.g., synthetic fertilizers and manure application from the MITERRA-Europe model (see Ural-Janssen et al. (2023) for further details). Current model inputs for agriculture for the year 2017 (Duan et al., 2021) were projected for 2050 as a function of the percent changes between 2015-2050 in the IMAGE model based on the SSP5-RCP8.5 scenario (Beusen et al., 2022). We chose the year 2015 to represent our current model results (2017-2020) and 2050 for future. Main assumptions in the SSP5-RCP8.5 scenario of Beusen et al. (2022) for 2050 are (1) calculation of overall N use efficiency (NUE = N yield / total N input) based on the NUE

in 2015 and the change between 1980-2015, which is corrected for the future N yield change relative to the historical yield change; (2) for a given NUE, calculation of fertilizer use on croplands by the difference between total N inputs and manure, atmospheric deposition and biological N_2 fixation; (3) substitution of synthetic fertilizers by recycled N and P from human urine obtained from new sewage systems installed after 2015 and (4) excluding the pastoral grasslands from the calculation of fertilizer input considering the use of synthetic fertilizer in pastoral areas is negligible. We did not consider the human urine as a source of fertilizer in agriculture in our scenario application. Model inputs for non-agricultural sources (i.e. atmospheric deposition over non-agricultural areas and biological N_2 fixation by natural vegetation) were derived from the IMAGE model at 0.5° resolution and processed to basin scale for current (2015) and 2050 based on the SSP5-RCP8.5 scenario (Beusen et al., 2022).

Model inputs for sewage systems were derived from Ural-Janssen et al. (2023) for current (2020) and from Strokal et al. (2021) for 2050 at basin scale based on the SSP5 scenario. Model inputs for hydrology were derived from the Variable Infiltration Capacity (VIC) model (i.e. natural water discharges) (van Vliet et al., 2016) and calculated as a function of sectoral water consumptions from Khan et al. (2022) (i.e. actual water discharges) at 0.5° resolution and processed to basin scale for current (the most recent data on natural water discharges was for 2010) and future (2050) based on the RCP8.5 scenario.

3.2.3 Required reductions in river export of nutrients to avoid coastal eutrophication

o Indicator for coastal eutrophication potential

We used the Indicator for Coastal Eutrophication Potential (ICEP) approach of Billen and Garnier (2007) to estimate the risk of eutrophication in the coastal waters of Europe by 2050 (Figure 3.1). ICEP is calculated by fluxes of N, P and Silica (Si) based on the Redfield ratio (C:N:P:Si = 106:16:1:20 (Redfield et al., 1963)) (Billen & Garnier, 2007). The Redfield ratio is the requirement for the growth of diatoms. When there is excess N and P fluxes over Si, the harmful non-siliceous algae (e.g., cyanobacteria) might develop instead of diatoms (Billen & Garnier, 2007). The ICEP value is quantified based on the following Eqs. 15 and 16 (Billen & Garnier, 2007):

$$ICEP_N = \left[\frac{TDN_{flux}}{(14\ x\ 16)} - \frac{DSi_{flux}}{(28\ x\ 20)}\right] x\ 106\ x\ 12 \text{ when N:P ratio < 16 (N limiting)} \quad (15)$$

$$ICEP_P = \left[\frac{TDP_{flux}}{31} - \frac{DSi_{flux}}{(28\ x\ 20)}\right] x\ 106\ x\ 12 \text{ when N:P ratio > 16 (P limiting)} \quad (16)$$

where $ICEP_N$ is the Indicator for Coastal Eutrophication Potential for N (kg C-eq. km^{-2} day^{-1}), TDN_{flux} is the total dissolved N flux (the sum of DIN and DON) to seas by rivers (kg km^{-2} day^{-1}) and DSi_{flux} is the total dissolved Si flux to seas by rivers (kg km^{-2} day^{-1}). $ICEP_P$ is

the Indicator for Coastal Eutrophication Potential for P (kg C-eq. km^{-2} day^{-1}) and TDP_{flux} is the total dissolved P flux (the sum of DIP and DOP) to seas by rivers (kg km^{-2} day^{-1}). The potential risk of coastal eutrophication is low when the ICEP value is below zero and vice versa (Billen & Garnier, 2007).

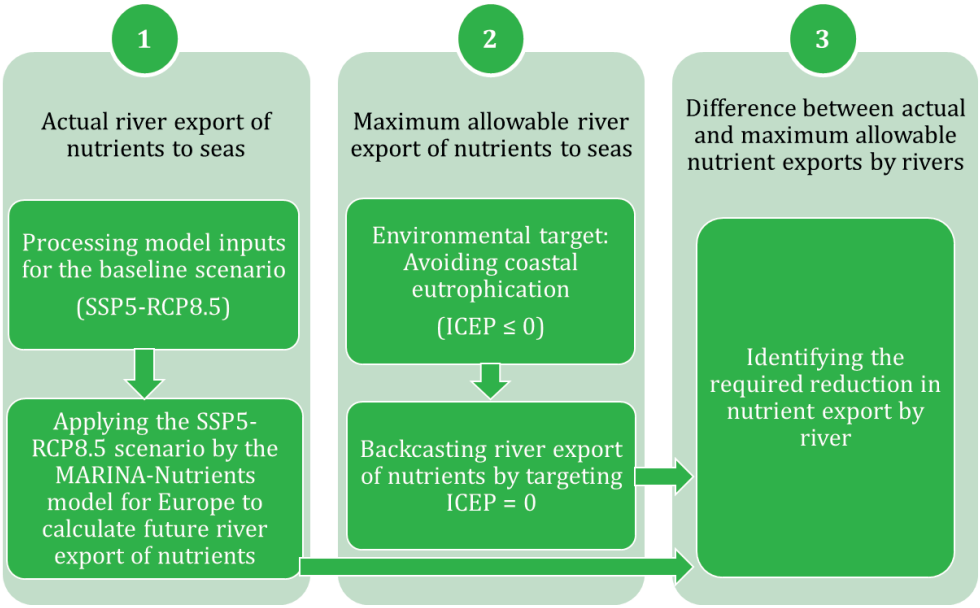


Figure 3.1 Stepwise methodology as applied in this study. SSP5 is short for Shared Socioeconomic Pathway 5 (fossil fueled development) and RCP8.5 is short for Representative Concentration Pathways (high-end climate change) (Section 2.2.1 for a description of the MARINA-Nutrients model for Europe).

o *Backcasting calculations*

We applied backcasting in a stepwise approach (Figure 3.1). First, we quantified the maximum allowable river exports of N and P to seas by targeting the ICEP as "0" to ensure low risks of coastal eutrophication following the approach of Li et al. (2019). We used an uncertainty range of "-1" and "+1" (kg C-eq. km^{-2} day^{-1}) as in Li et al. (2019). For backcasting the maximum allowable river export of total dissolved nitrogen (TDN) and total dissolved phosphorus (TDP), Eqs. 17 and 18 are as follows (Li et al., 2019):

$$TDN_{max} = \left[\frac{ICEP_N}{(106\ x\ 12)} + \frac{DSi_{flux}}{(28\ x\ 20)}\right] x\ 14\ x\ 16 \quad (17)$$

$$TDP_{max} = \left[\frac{ICEP_P}{(106\ x\ 12)} + \frac{DSi_{flux}}{(28\ x\ 20)}\right] x\ 31 \quad (18)$$

where $ICEP_N$ = 0 (–1 and +1), $ICEP_P$ = 0 (–1 and +1), TDN_{max} and TDP_{max} are the maximum allowable river export of TDN and TDP (kg km^{-2} day^{-1}), and DSi_{flux} is total dissolved Si flux to seas by rivers (kg km^{-2} day^{-1}) for 2050 (Table S3.2 for model inputs).

Second, we calculated the required reductions in nutrient exports based on the SSP5-RCP8.5 scenario for 2050 relative to the maximum allowable nutrient exports by rivers. We did backcasting calculations for the top ten polluted rivers per nutrient. Top ten rivers were chosen based on their pollution levels for N (i.e. the Danube, Rhine, Wisla, Elbe, Odra, Seine, Tejo, Nemanus, Daugava, Ebro) and P (i.e. the Danube, Rhine, Wisla, Seine, Odra, Tejo, Dnestr, Ebro, Daugava, Pregolya).

3.3 Results and discussion

3.3.1 Current and future river export of nutrients: Totals by form

In this section, we describe the model results for future river exports of N and P to the coastal waters of Europe based on the SSP5-RCP8.5 scenario. We estimated nutrient inputs in European seas by sources and basins for the year 2050. We compared the future (2050) river export of nutrients with the current (2017-2020) nutrient export from Ural-Janssen et al. (2023).

River exports of total dissolved N (TDN) and total dissolved P (TDP) to European seas are projected to increase by 13% and 28%, respectively, between today and 2050 under global change (Figure 3.2). Today (2017-2020), rivers export 2690 Gg of TDN and 128 Gg of TDP to the coastal waters. By 2050, these amounts are projected to be 3044 Gg for TDN and 163 Gg for TDP (Figure 3.2). Less than half of TDN in the coastal waters (39%) is from synthetic fertilizers and animal manure (applied on agricultural land from housing systems and deposited on land during grazing) today (Figure 3.2). Sewage systems are responsible for over half of TDP in the coastal waters (60%) (Figure 3.2). In the future, synthetic fertilizers and manure (applied on agricultural land from housing systems and deposited on land during grazing) will contribute to river export of TDN by 35% (with an increase of 15% from manure) (Figure 3.2). This is due to the fact that our assumptions do not include the implementation of the EU policies (e.g., Green Deal Farm to Fork Strategy) (Section 3.2.2 for scenario description). Sewage systems are expected to be responsible for 70% of future TDP export by rivers with an increase of 49% relative to today (Figure 3.2).

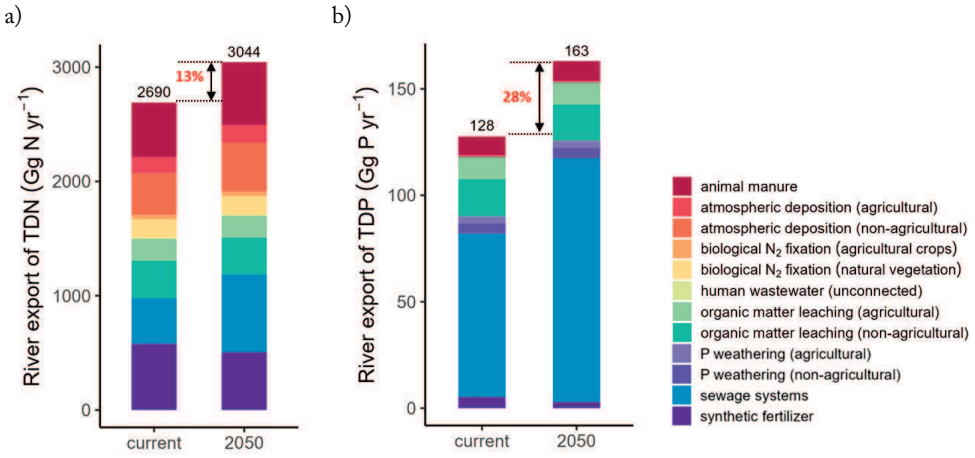


Figure 3.2 Current (2017-2020) and future (2050) river export of nutrients by source to the coastal waters of Europe based on the SSP5-RCP8.5 scenario. (a) River export of total dissolved nitrogen (TDN, Gg N yr^{-1}). (b) River export of total dissolved phosphorus (TDP, Gg P yr^{-1}). SSP5 is short for Shared Socioeconomic Pathway 5 (fossil fueled development) and RCP8.5 is short for Representative Concentration Pathways (high-end climate change). Source: MARINA-Nutrients model for Europe (Section 3.2.1).

We found that socioeconomic change (related to the SSP5) has a large impact on nutrient losses to rivers, and hence on the river exports of TDN and TDP. Climate change (related to the RCP8.5) will alter runoff and river discharges. Lower natural water discharges are projected for most of the basins (70%) for 2050 compared to the current level (2017-2020). This can affect the nutrient retention on land and in rivers, and nutrient losses to rivers and to seas. Global change will lead to the increased river export of TDN and TDP by 2050 is mainly by sewage systems, followed by animal manure (applied on agricultural land from housing systems and deposited on land during grazing) among the considered sources in this study. This is due to the increased urbanization (moving towards cities and increasing population connected to sewage systems) but moderate improvement of the urban wastewater treatment plants (Strokal et al., 2021), and the increased livestock production and thus, manure production and application (Beusen et al., 2022).

Future river exports of nutrients show large variability among the basins relative to 2017-2020 (Figure 3.3). Nearly two-thirds of the European rivers are projected to export more nutrients in 2050 than today (59% for TDN and 67% for TDP). For TDN export (kg N km^{-2} yr^{-1} per basin) a few rivers (~2%) are projected to have large increases by more than 67%, 6% of the rivers will have moderate increases and half of the rivers will have small increases by up to 33% (Figure 3.3). The pollution is projected to be worse for the river export of TDP (kg P km^{-2} yr^{-1} per basin) (Figure 3.3). 5% of the rivers will have large increases by more than 67%, 18% of the rivers will have moderate increases and 43% of the rivers will have increases by up to 33% compared to the current (2017-2020) TDP export by rivers

(Figure 3.3). About one-third of the rivers are projected to export less TDP and TDN in 2050 than today (Figure 3.3). These reductions are generally calculated for relatively small rivers, covering a relatively small part of the total study area.

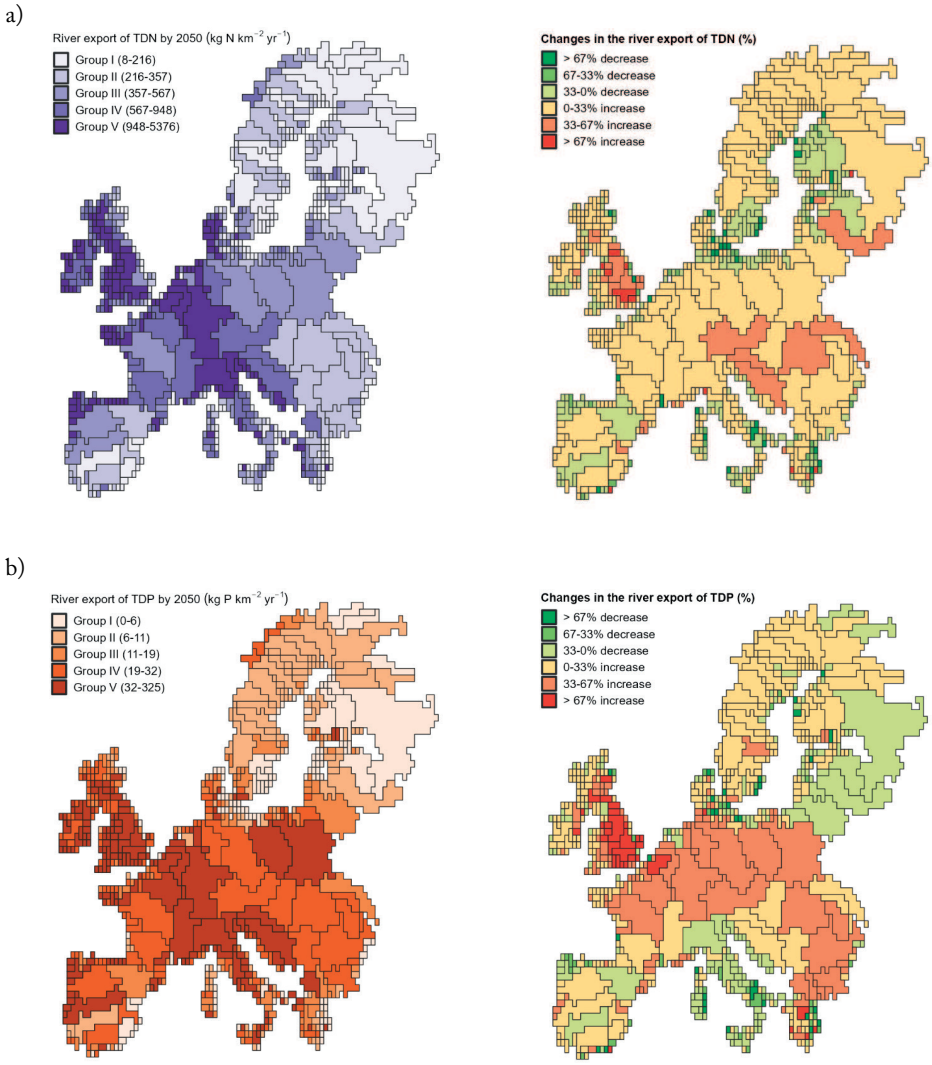


Figure 3.3 Future (2050) river export of nutrients to the coastal waters of Europe based on the SSP5-RCP8.5 scenario. (a) River export of total dissolved nitrogen (TDN, kg N km^{-2} yr^{-1}) and percent changes relative to 2017-2020 (%). (b) River export of total dissolved phosphorus (TDP, kg P km^{-2} yr^{-1}) and percent changes relative to 2017-2020 (%). Groups I-V are defined based on the current river export of TDN and TDP to seas (each group has 20% of basins, from highest to lowest river exports). SSP5 is short for Shared Socioeconomic Pathway 5 (fossil fueled development) and RCP8.5 is short for Representative Concentration Pathways (high-end climate change). Source: MARINA-Nutrients model for Europe (Section 3.2.1).

The relative changes between current and future river export of nutrients vary among the basins and nutrient forms (i.e. DIN, DON, DIP, DOP) (Figure 3.4 and Figure 3.5). We calculated increases in the total river export of nutrient forms to seas for 2050 with the SSP5-RCP8.5 scenario compared to the current situation, except for DOP. The largest exports of all nutrient forms are often by the rivers located in western, central and southern Europe (Figure 3.4 and Figure 3.5). The dominant source of nutrient pollution in rivers varies based on the nutrient form. Below, we describe the changes in future river export of each nutrient form compared to today, and their largest contributor among the sources considered in this study.

In Europe, river export of DIN and DON is projected to increase by 13% and 12% by 2050 relative to today, respectively (Figure 3.4). The future DIN export by rivers to seas is projected to be by up to 4949 kg N km^{-2} yr^{-1} while the maximum current DIN export is 2510 kg N km^{-2} yr^{-1} (Figure 3.4). Spatial variability for the magnitude of the DIN export by rivers is similar for both current (2017-2020) and future (2050) years (Figure 3.4), while source attribution differs among the basins and seas (Figure 3.6). Animal manure (application on agricultural land from housing systems and deposited on land during grazing) is expected to be an important source and contribute to the river export of DIN by 23% (e.g., up to 71% per basin) in 2050, followed by synthetic fertilizer application (22%) and sewage systems (20%) (Figure 3.6). The future DON export by rivers to seas is projected to vary between 4-744 kg N km^{-2} yr^{-1} (Figure 3.4). More basins are expected to shift to the largest river export range (Group V) (Figure 3.4). Organic matter leaching over non-agricultural areas is projected to dominate the river export of DON by 41% (e.g., up to 77% per basin) in 2050 (Figure 3.6).

River export of DIP is projected to increase by 37%, while the DOP export by European rivers will slightly decrease (1%) in 2050 (Figure 3.5). The changes in the magnitude of the DIP export by rivers to seas vary among both temporal and spatial scales (Figure 3.5). The future DIP export by rivers to seas is projected to be up to 316 kg P km^{-2} yr^{-1}, more than duplicate of the maximum current DIP export (146 kg P km^{-2} yr^{-1}) (Figure 3.5). Sewage systems are expected to remain as a significant contributor to the DIP export by 86% (e.g., up to 97% per basin) in 2050 (Figure S3.1). The ranges for future DOP export by rivers to seas are expected not to change relative to today (Figure 3.5). Similar to DON, organic matter leaching over non-agricultural areas is projected to dominate the river export of DOP by 53% in 2050 (Figure S3.1).

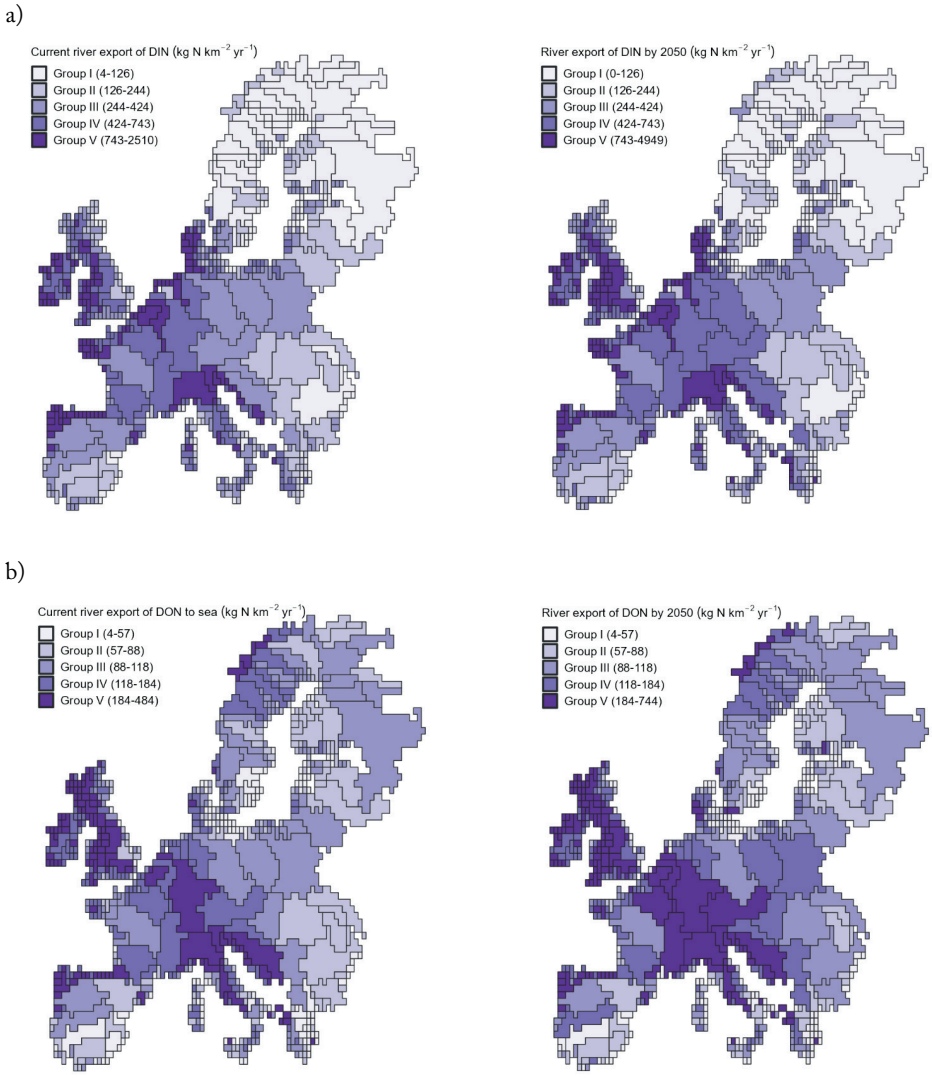


Figure 3.4 Current (2017-2020) and future (2050) river export of nitrogen to the coastal waters of Europe based on the SSP5-RCP8.5 scenario. (a) River export of dissolved inorganic nitrogen (DIN, kg N km^{-2} yr^{-1}). (b) River export of dissolved organic nitrogen (DON, kg N km^{-2} yr^{-1}). Groups I-V are defined based on the current river export of DIN and DON to seas (each group has 20% of basins, from highest to lowest river exports). SSP5 is short for Shared Socioeconomic Pathway 5 (fossil fueled development) and RCP8.5 is short for Representative Concentration Pathways (high-end climate change). Source: MARINA-Nutrients model for Europe (Section 3.2.1).

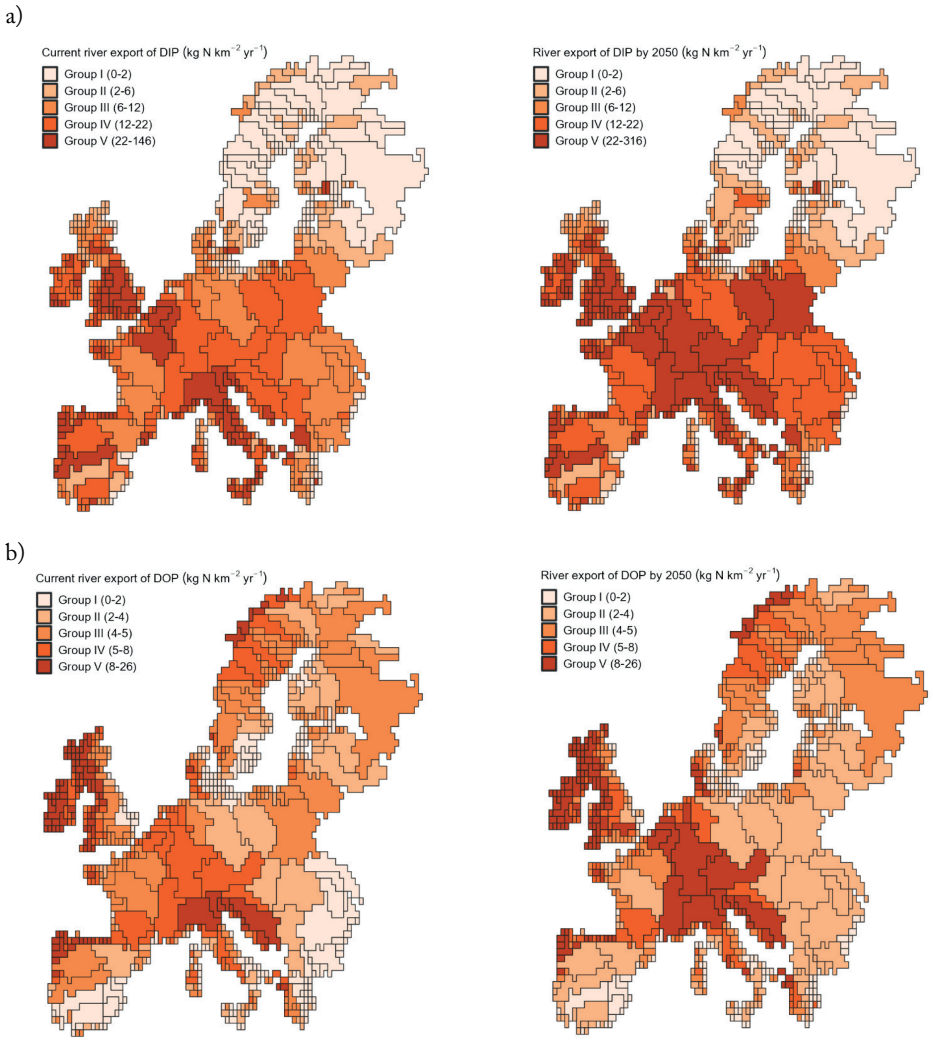


Figure 3.5 Current (2017-2020) and future (2050) river export of phosphorus to the coastal waters of Europe based on the SSP5-RCP8.5 scenario. (a) River export of dissolved inorganic phosphorus (DIP, kg P km^{-2} yr^{-1}). (b) River export of dissolved organic phosphorus (DOP, kg P km^{-2} yr^{-1}). Groups I-V are defined based on the current river export of DIP and DOP to seas (each group has 20% of basins, from highest to lowest river exports). SSP5 is short for Shared Socioeconomic Pathway 5 (fossil fueled development) and RCP8.5 is short for Representative Concentration Pathways (high-end climate change). Source: MARINA-Nutrients model for Europe (Section 3.2.1).

3.3.2 River export of nutrients by 2050: Source attribution

In this section, we describe the river export of nutrients to seas/oceans for 2050 by nutrient form and source. The Atlantic Ocean is projected to receive the largest river export of nutrients for all forms in 2050 (e.g., 28% of DIN, 27% of DIP) (Figure 3.6 and Figure S3.1). The Mediterranean and North Seas are expected to receive similar river export of nutrients (e.g., 23% of

DIN, 24-23% of DIP respectively) (Figure 3.6 and Figure S3.1). The Baltic and Black Seas are projected to receive similar river export of DIN and DIP, but different DON (21% and 14% of the total, respectively) and DOP (24% and 14% of the total, respectively) exports (Figure 3.6 and Figure S3.1). The least nutrients for all forms will be exported by rivers to the Arctic Ocean due to less human activities in those river basins (Figure 3.6 and Figure S3.1).

The relative share of sources of nutrient exports differs among the seas. For example, animal manure (applied on agricultural land from housing systems and deposited on land during grazing) is expected to be a major source for the river export of DIN to seas by 2050 (e.g., Atlantic Ocean, Mediterranean and North Seas), followed by the application of synthetic fertilizers and sewage systems except for the Arctic Ocean (Figure 3.6). Sewage systems are expected to remain as the main source of the DIP export by rivers in 2050 except for the Arctic Ocean (Figure S3.1). Most of the DIN and DIP exports to the Arctic Ocean will be from non-agricultural sources (e.g., atmospheric deposition over non-agricultural areas, P weathering over non-agricultural soils) (Figure 3.6 and Figure S3.1). River export of DON and DOP will be mainly from non-agricultural sources (e.g., 41% of DON and 53% of DOP exports by the organic matter leaching over non-agricultural areas) (Figure 3.6 and Figure S3.1).

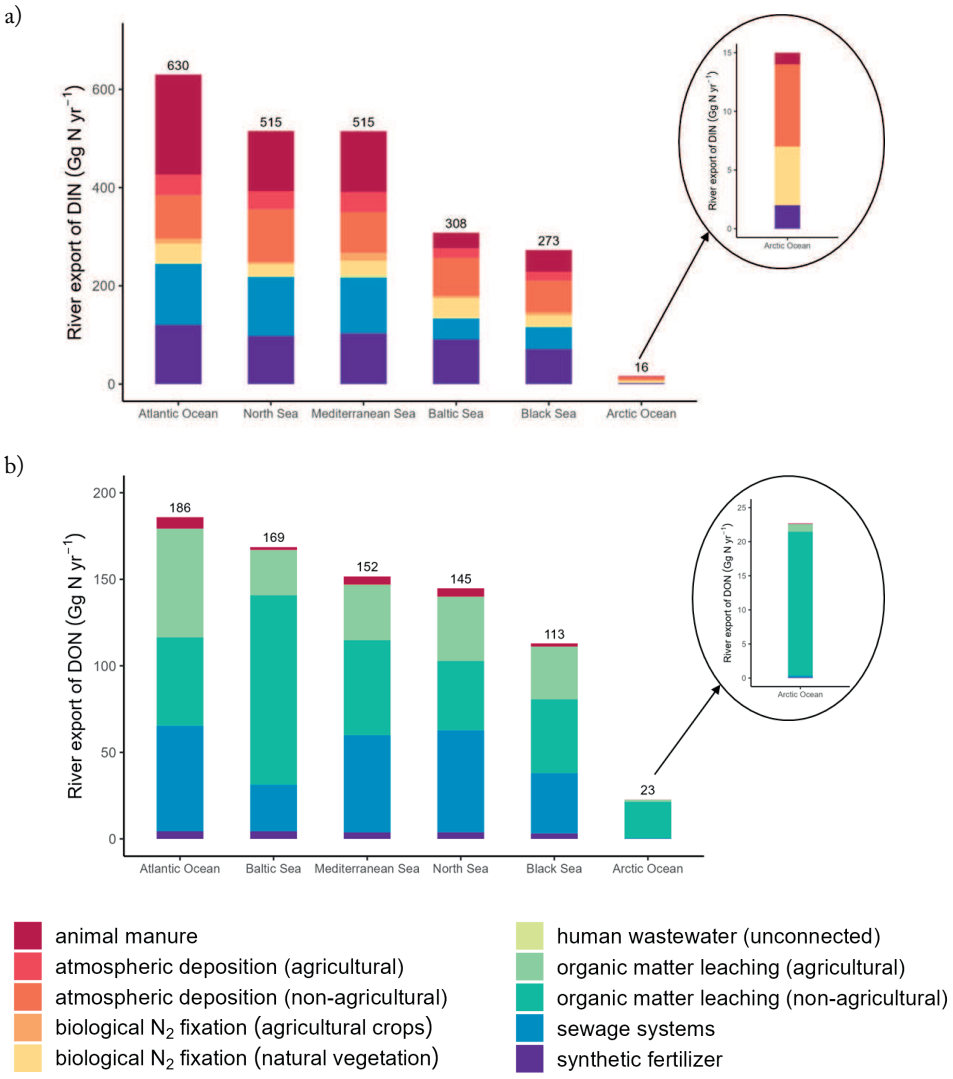


Figure 3.6 Future (2050) river export of nitrogen by source to the European seas based on the SSP5-RCP8.5 scenario. (a) River export of dissolved inorganic nitrogen (DIN, Gg N yr^{-1}). (b) River export of dissolved organic nitrogen (DON, Gg N yr^{-1}). Colors represent sources of the river export. SSP5 is short for Shared Socioeconomic Pathway 5 (fossil fueled development) and RCP8.5 is short for Representative Concentration Pathways (high-end climate change). Source: MARINA-Nutrients model for Europe (Section 3.2.1).

3.3.3 Required reductions to avoid coastal eutrophication

In this section, we present the maximum allowable nutrient exports by rivers and required reductions to avoid the risks of coastal eutrophication in Europe. First, we describe the maximum allowable river export of nutrients that is calculated by the ICEP approach for the top ten most polluted rivers (Section 3.2.3 for the approach). Second, we present the required reductions to meet the allowable nutrient exports to seas and reduce coastal eutrophication risk in European seas by 2050.

o Maximum allowable river export of nutrients to seas

We used Eqs. 17 and 18 to calculate maximum allowable river exports of nutrients. More than half of the rivers (53%) are expected to exceed the maximum allowable levels for TDN export, while 18% exceed the allowable levels for TDP export by 2050 based on the SSP5-RCP8.5 scenario. We selected the top ten most polluted rivers from the analyses of Figure 3.2-Figure 3.6 (Figure 3.7). The number of people living in these river basins and affected by coastal eutrophication from 2050 onwards is projected to be 42% of the European population in the study area.

The maximum allowable levels reflect the river export of TDN and TDP to ensure ICEP = 0 (with an uncertainty range of -1 to +1), indicating low risk for harmful algal blooms. The calculated maximum allowable river exports of TDN are 112 (60 to 163) Gg yr^{-1} for Danube, 14 (3-25) Gg yr^{-1} for Rhine, 34 (21-46) Gg yr^{-1} for Wisla, 43 (33-52) Gg yr^{-1} for Elbe, 21 (13-28) Gg yr^{-1} for Odra, 18 (13-23) Gg yr^{-1} for Seine, 6 (1-12) Gg yr^{-1} for Tejo, 10 (4-16) Gg yr^{-1} for Nemanus, 5 (-2-11) Gg yr^{-1} for Daugava and 3 (-3-8) Gg yr^{-1} for Ebro (Figure 3.7). The maximum allowable river exports of TDP are 15 (8-22) Gg yr^{-1} for Danube, 2 (0.4-3) Gg yr^{-1} for Rhine, 5 (3-6) Gg yr^{-1} for Wisla, 2 (2-3) Gg yr^{-1} for Seine, 3 (2-4) Gg yr^{-1} for Odra, 1 (0.1-2) Gg yr^{-1} for Tejo, 0.3 (-0.3-1) Gg yr^{-1} for Dnestr, 0.4 (-0.4-1) Gg yr^{-1} for Ebro, 0.6 (-0.3-1) Gg yr^{-1} for Daugava and 0.4 (0.2-0.6) Gg yr^{-1} for Pregolya (Figure 3.7). These levels are considerably lower than the actual nutrient exports projected for 2050 by the MARINA-Nutrients model based on the SSP5-RCP8.5 scenario (Figure 3.7 and Section 3.3.2).

o Required reductions to meet the allowable nutrient exports to seas

To meet the maximum allowable nutrient export levels by 2050, river export of nutrients needs to be reduced considerably. Avoiding future coastal eutrophication requires over 47% less N and up to 77% less P export by the top ten polluted rivers to seas relative to the SSP5-RCP8.5 scenario levels (Figure 3.7). River exports of TDN and TDP need to be reduced by 69-26%, 91-74% and 64-29% for Danube, Rhine and Wisla, respectively (Figure 3.7). More than 47% of TDN and more than 12% of TDP exports need to be reduced for the rest of the top ten rivers to limit ICEP to 0 (Figure 3.7).

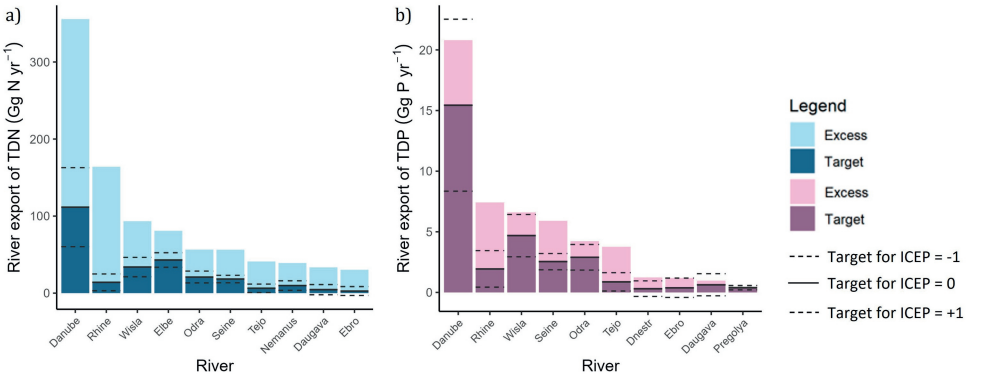


Figure 3.7 Future (2050) nutrient export by the top ten polluted rivers of Europe based on the SSP5-RCP8.5 scenario. (a) River export of total dissolved nitrogen (TDN, Gg N yr^{-1}). (b) River export of total dissolved phosphorus (TDP, Gg P yr^{-1}). Targets "0", "-1" and "+1" (kg C-eq. km^{-2} day^{-1}) are for (a) $ICEP_N$ and (b) $ICEP_P$. SSP5 is short for Shared Socioeconomic Pathway 5 (fossil fueled development) and RCP8.5 is short for Representative Concentration Pathways (high-end climate change). Source: MARINA-Nutrients model for Europe (Section 3.2.1).

3.3.4 Comparisons of model outputs with literature

Our study focuses on future analyses of river exports of nutrients to the European coastal waters. Model validation for the current period was done in our earlier study (Ural-Janssen et al., 2023). To build trust in our future analyses, we compared our model results with available future modeling studies for Europe. In general, our model projections for the river export of nutrients to seas by 2050 are in line with other studies. Many coastal waters are expected to be affected by an increase riverine nutrient delivery in Europe (e.g., up to 40% increase in TDN export to the Black Sea, up to 61% increase in TDP export to the North Sea) relative to today. This is in line with the expected increase in global river exports of N and P by 2050 based on the SSP5-RCP8.5 (Beusen et al., 2022).

The increased nutrient transport to coastal waters by 2050 will be mainly caused by urbanization (e.g., 71% increase for river export of TDN from sewage systems) based on our baseline (SSP5-RCP8.5) scenario. This is mainly due to the river export of nutrients from higher amount of wastewater produced with moderately improved treatment efficiencies (Strokal et al., 2021) (Table S3.1 and Section 3.2.2 for details). There will be a shift from synthetic fertilizers to animal manure and that is in line with the SSP5-RCP8.5 scenario assumptions (e.g., reducing synthetic fertilizer application by recycling nutrients from human urine Section 3.2.2). Diffuse sources of river exports of N (e.g., atmospheric deposition over non-agricultural areas, biological N_2 fixation by natural vegetation) and P (e.g., P weathering from non-agricultural soils) from non-agricultural areas are expected not to change significantly by 2050, similar to global as indicated by Beusen et al. (2022).

3.3.5 Uncertainties and limitations

We discuss the uncertainties and limitations in (1) data, (2) the model and (3) the scenarios. First, we reflect on the limitations related to data availability. Our backcasting approach could only be applied to a limited number of rivers due to data availability for DSi (e.g., regular DSi measurements were not available for European rivers). The DSi data was derived from the Global *NEWS2* model for 2050 based on Global Orchestration scenario (Seitzinger et al., 2010) as this was the closest option to the SSP5-RCP8.5 scenario. The data was not available for all the river basins considered in this study. Thus, we only considered the DSi fluxes of the large and most polluted rivers with N and P in backcasting of maximum allowable nutrient exports by rivers to assess the risk of coastal eutrophication in Europe by 2050. The DSi fluxes to seas might vary depending on socioeconomic (e.g., dam constructions (Seitzinger et al., 2010)) and climate (e.g., precipitation (Seitzinger et al., 2010) and temperature (Zhang et al., 2023)) changes. Thus, further research on DSi observations and on how the controlling factors could affect the DSi fluxes in future is required.

Second, our model has limitations. Our model inputs and coefficients are from various sources (e.g., MITERRA-Europe model, literature) and this can increase uncertainties. As we do large-scale modeling, some parameters are basin-scale specific (e.g., nutrient removal efficiency in sewage treatment systems), not site specific. The MARINA-Nutrients model for Europe does not account for the inputs of N and P from the industrial wastewater treatment systems (only domestic wastewaters) and N input from atmospheric deposition onto coastal waters. Thus, we underestimate the future river export of nutrients and in turn the required reduction to reach the maximum allowable nutrient exports by the top ten polluted rivers to seas. Nevertheless, our study shows that even with the considered sources, nutrient exports by the top ten polluted rivers will exceed the maximum allowable levels by 2050. Another limitation is not considering particulate forms of nutrients. Our model does not account for particulate N (PN) and P (PP), and nutrient losses associated to erosion explicitly. River export of PN and PP were estimated at around 20% and 50% of the total N and total P for Europe in 2000 by Mayorga et al. (2010). Therefore, the contribution of the river export of PP to the total P (TP = DIP + DOP + PP) pollution in the European seas is as much as the TDP export to seas. P loss from soil erosion is a significant source of PP inputs to rivers and in turn, river export of TP to seas (Alewell et al., 2020; Panagos et al., 2022). However, we need to realize that not all particulate N and P is biologically available, so it could be argued that this omission affects our results and conclusions to only a limited extent. Moreover, we consider the common pathways (e.g., runoff, discharges of sewage effluents after treatment) but our process-based model does not account for the dynamics in retention and release processes of nutrients between soil and sediments, and waters (Strokal et al., 2016). For example, the effect of animal manure (applied on agricultural land and deposited during grazing) on the river export of P might be delayed by dynamic processes (e.g., P accumulation) (Strokal & de Vries, 2012). Similarly, biogeochemical processes in soil and waters can alter the N export by rivers (Beusen et al., 2022). For example, more denitrification results

in more N removal from the soil and waters, and thus less N available for export to the river mouth. Denitrification is influenced by several factors such as temperature; precipitation; soil microbial biomass, carbon, N and clay contents; soil texture and pH in soils (Li et al., 2022c); temperature, and availability of N and C in rivers (Deng et al., 2020). These all can influence the process of denitrification, and therefore the availability of N in the river. In the MARINA-Nutrients model for Europe, processes of nutrient retention and losses are aggregated to basin scale (Ural-Janssen et al., 2023) which might cause over/underestimation of future nutrient exports to seas. Ideally, more in-depth analyses that should comprise all the relevant sources of nutrient exports by rivers, account for losses in all nutrient forms, and consider all the relevant processes (e.g., morphological and hydrological) are needed for a comprehensive assessment of the future coastal eutrophication in Europe. We nevertheless consider that the level of detail of the MARINA-Nutrients model for Europe fit for purpose in this study: to explore large-scale future trends for relatively long-term future.

Third, scenario related limitations might associate with our baseline scenario assumptions. We chose a SSP-RCP combination with the largest changes (i.e. SSP5: fossil fueled development and RCP8.5: high-end climate change). We derived the actual model inputs to calculate nutrient exports from point sources for 2050 based on the SSP5 scenario from (Strokal et al., 2021), while calculating the model inputs for diffuse sources as a function of the percent changes (per basin) between 2015-2050 in the IMAGE model based on the SSP5-RCP8.5 scenario (Beusen et al., 2022). Different from the SSP5 assumptions by Beusen et al. (2022), we do not consider the human urine as a source of fertilizer in agriculture. Our choice to use SSP5-RCP8.5 as a basis determines the required reductions to meet the allowable river export levels to avoid coastal eutrophication. Other SSP-RCP combinations may result in other required reductions. This should be realized when interpreting our results.

3.4 Conclusions

We estimated the delivery of dissolved N and P by rivers to the seas of Europe (e.g., Arctic Ocean, Atlantic Ocean, Baltic Sea, Black Sea, Mediterranean Sea, North Sea) for the year 2050 based on the SSP5-RCP8.5 scenario. We modelled future nutrient pollution in the coastal waters of Europe by sources and basins. River exports of TDN and TDP are projected to increase by 13% and 28%, respectively, under global change relative to today. Manure and fertilizers contribute to river export of TDN by 35% in 2050. Sewage systems are responsible for 70% of future TDP export by rivers. As a result, more eutrophication is expected in the coastal waters of Europe by 2050. The top ten polluted rivers (N and P) are expected to host 42% of the European population in 2050. This implies that at least 42% of the population in the study area may be affected by coastal eutrophication from 2050 onwards.

We quantified the required reductions in river pollution to avoid the potential risks of coastal eutrophication under global change. For this, we set maximum allowable nutrient exports by rivers to seas using the ICEP approach. We quantified the maximum allowable TDN and TDP fluxes from the top ten most polluted rivers based on the ICEP = 0 (with an uncertainty range of -1 to +1). Then, we compared the maximum allowable nutrient export levels to the results of SSP5-RCP8.5 scenario to identify the required reductions to avoid future coastal eutrophication in Europe. To avoid the risk of coastal eutrophication, we urgently need to reduce nutrient delivery by rivers to seas. Avoiding future coastal eutrophication requires over 47% less N and up to 77% less P exports by the top ten polluted rivers (e.g., Danube, Rhine, Wisla).

Recycling or upcycling nutrients in agriculture from organic waste could be a promising option to reduce river export of nutrients and avoid future coastal eutrophication, but needs to be studied comprehensively to better understand the synergies and to avoid trade-offs (e.g., pollution swapping). For instance, large amount of nutrients from urban wastewater treatment systems should be recycled more and effectively in agriculture to prevent high nutrient exports by rivers from sewage systems to seas. Our study shows how backcasting approach can be used to define desired futures for reduced pollution and low risks of coastal eutrophication. This could support the implementation of effective environmental policies to avoid coastal eutrophication in the future.

Our baseline scenario should be interpreted as a pessimistic future. Considering the abovementioned limitations of this study, future studies can build on the SSP5-RCP8.5 scenario by incorporating the EU policies and targets (e.g., Green Deal, Farm to Fork) to illustrate the effect of their implementation on the riverine nutrient transport and eutrophication in European seas by 2050.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Aslıhan Ural-Janssen: Conceptualization, Methodology, Visualization, Writing – original draft, Writing – review & editing. Carolien Kroeze: Conceptualization, Funding acquisition, Methodology, Supervision, Writing – review & editing. Erik Meers: Funding acquisition, Supervision, Writing – review & editing. Maryna Strokal: Conceptualization, Funding acquisition, Methodology, Project administration, Supervision, Writing – review & editing.

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Data availability

The main model results supporting Figures 3.2-3.7 generated in this study are deposited in the DANS Easy repository https://doi.org/10.17026/PT/OUJR9H.

Supplementary data

Supplementary data can be found online at https://doi.org/10.1016/j.marenvres.2024.106446 and in this thesis under *S3: Supplementary materials for Chapter 3*.



Chapter 4

Mitigating nutrient losses in Europe: Synergistic solutions for air and water pollution by 2050

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This paper is adjusted to the PhD thesis format.

Abstract

Agriculture and urbanization often cause nitrogen (N) and phosphorus (P) losses, and associated environmental impacts in Europe. Here, we aim to quantify the effects of using recovered N from manure processing and recovered P from treated sewage sludge, increasing N and P use efficiencies in agriculture, and improving sewage treatments on reducing future nutrient emissions to the air, and losses to rivers and seas of Europe. Exploring synergistic options we show that 30% less N is expected in the air via ReNuRe products and higher N use efficiency by 2050. Rivers are projected to receive 23-68% less nutrients relative to the future baseline scenario. These rivers may export up to two-thirds less nutrients relative to the future baseline. Recovered P from treated sewage sludge can fulfill P fertilizer need in Europe in 2050. Our analysis supports the adoption of bio-based fertilizers to mitigate nutrient losses and contributes to circular economy.

4.1 Introduction

Intensive agriculture and urbanization are important sources of nutrient pollution and associated impacts in Europe (Grizzetti et al., 2021; Strokal et al., 2021; Ural-Janssen et al., 2023). Nitrogen often is released to the air from agricultural activities in the form of NH_3, N_2O, and NO_x (de Vries et al., 2024). Nutrients are transported to rivers and coastal waters from agricultural via runoff as diffuse sources and from urban areas via sewage systems as point sources (Strokal et al., 2016). European rivers exported annually 2690 Gg of N and 128 Gg of P to coastal waters during 2017-2020 (Ural-Janssen et al., 2023). This challenges nutrient pollution reduction (Vigiak et al., 2023; de Vries et al., 2024), and the recovery of European freshwater biodiversity (Haase et al., 2023). In the future, rivers may export more nutrients because of population growth, increased urbanization, and agricultural intensification unless policy interventions are implemented effectively (Ural-Janssen et al., 2024). This calls for sustainable solutions to mitigate nutrient losses and their environmental impacts.

Several European policies have been implemented for nutrient losses including the National Emission Ceilings Directive (EC, 2001) with emission targets for NH_3 and NO_x, the Nitrates Directive (EC, 1991a) and the Water Framework Directive (EC, 2000) with N and P limits for their loadings to waterbodies, the Paris Agreement (UNFCCC, 2015) with targets for reducing GHG emissions and the Waste Framework Directive (EU, 2008b) with recommendations on recycling and recovery instead of disposal. The Green Deal Farm-to-Fork (F2F) strategy aims for sustainable food production by reducing food waste, enhancing circularity via waste recycling and improving nutrient use efficiency to mitigate nutrient losses by at least 50% without deteriorating soil fertility, claiming that this will reduce fertilizer use by at least 20% in 2030 (EC, 2020).

Upcycling organic wastes to BBFs could substitute synthetic fertilizers while maintaining the crop yield efficiency (Chojnacka et al., 2020). ReNuRe is one of the BBFs, being a clean end product with comparable characteristics as synthetic N fertilizers (Huygens et al., 2020). Ammonium salts produced through stripping-scrubbing process can be used as ReNuRe fertilizer (Vingerhoets et al., 2024). Vingerhoets et al. (2023b) showed that a combination of N recovery from manure by ammonia stripping-scrubbing, and P recovery from municipal wastewater by struvite precipitation could increase the circularity of N by 15% and of P by 24%, thereby reducing the dependence on external fertilizers. Existing studies mainly assess the characterization (Case & Jensen, 2019; Morey et al., 2023) and agronomic efficiency of BBFs (Sigurnjak et al., 2019; Breda et al., 2020; Ye et al., 2020; Saju et al., 2023). However, their environmental effects on surface waters can fully or partly offset the benefits of nutrient recycling. Studies evaluated their environmental impacts on field levels (Aguirre-Villegas et al., 2019; Wester-Larsen et al., 2022; Albert & Bloem, 2023) while focusing on specific impacts by ignoring issues like pollution swapping and impacts of site conditions. Other studies applied life cycle assessments to evaluate the

environmental impacts of BBFs (Ravi et al., 2022; Rydgård et al., 2024) or the processes to create them (Yoshida et al., 2018). Nevertheless, these studies often ignore the fact that the nutrient use efficiency of applied products is highly controlled by site-specific agro-ecological conditions (You et al., 2023). Thus, there is a need for a comprehensive study assessing the effects of BBFs on air and water pollution to support their implementation.

Environmental models are useful tools to explore future trends in air and water quality in response to projected changes in pressures and drivers, knowing that nutrient management in agricultural systems have a strong influence on the emissions of nutrients to air and waters (Young et al., 2021). Several models exist for large-scale simulations of nutrient flows from agriculture, including MITERRA-Europe (Velthof et al., 2009), INTEGRATOR (de Vries et al., 2011; 2021), GREEN (Grizzetti et al., 2012), IMAGE-GNM (Beusen et al., 2015) and MARINA (Strokal et al., 2016). These models have been used to simulate the flows of N and P in agroecosystems, and to evaluate technical or social innovations affecting the nutrient use efficiency. The models usually take a process-based approach that is combined with empirical mass balance approaches to assess annual fluxes at high spatial resolution. De Vries et al. (1998) showed that model simplification, in terms of less detailed formulations of processes (process aggregation) at low temporal resolution (temporal aggregation), is an adequate step in the upscaling of modeling results from a local to a regional scale when interested in annual average fluxes. Both MITERRA-Europe (Velthof et al., 2009) and INTEGRATOR (de Vries et al., 2011; 2021) quantifies N losses to air and water across Europe, and have been applied at various resolutions. Moreover, GREEN simulates river exports of total N and P to European seas at basin scale (Grizzetti et al., 2012). Furthermore, IMAGE-GNM simulates N and P flows to freshwaters at a 0.5° grid cell resolution globally (Beusen et al., 2022).

The MARINA-Nutrients model for Europe (Ural-Janssen et al., 2023) combines both air and water emissions and simulates N emissions to the air, and N and P flows from land to rivers and by rivers to coastal waters. It is a European version of the MARINA model family (Micella et al., 2024c) running at the sub-basin scale for dissolved organic and inorganic nutrients, which makes the model different from the abovementioned examples. This model is chosen in this study because it addresses air (agriculture-associated), river, and coastal water (agriculture and sewage-associated) pollution for European basins, supporting the identification and assessment of policy-relevant strategies on reducing air and water pollution under global change. The model has been validated and used to identify required reductions to avoid future coastal eutrophication in Europe under global change (Ural-Janssen et al., 2023; 2024). In this study, we use the model to reveal new insights on the effects of BBFs on air and water pollution control in the future when considering socioeconomic developments and climate change. Scenarios such as Shared Socioeconomic Pathways (SSPs, O'Neill et al. (2014)) and Representative Concentration Pathways (RCPs, van Vuuren et al. (2011)) provide various storylines to project future changes in socioeconomic developments and hydrology.

This study aims to quantify the effects of using recovered N from manure processing and recovered P from treated sewage sludge, increasing N and P use efficiencies in agriculture, and improving sewage treatments on reducing future nutrient emissions to the air, and losses to rivers and coastal waters of Europe. To our knowledge, no studies have thoroughly assessed the impact of using BBFs in European agroecosystems on N emissions to the air and nutrient loads to rivers and coastal waters, while accounting for variation in crop and soil types, climates, and management practices. This work was designed to fill these gaps by providing an in-depth, quantitative assessment across Europe at the basin scale under global change. We used the MARINA-Nutrients model for Europe with a baseline scenario from Ural-Janssen et al. (2024), and incorporated six alternative scenarios on nutrient management strategies.

4.2 Materials and methods

4.2.1 MARINA-Nutrients model for Europe ·

Our model is process-based, steady-state with a lumped approach to estimate N emissions to the air from agriculture, inputs of DIN, DIP, DON, DOP to rivers, and their river exports to seas per basin (and sub-basins of the Danube River) for 594 European rivers while accounting for nutrient retention in and export from land (e.g., P accumulation in soils, crop uptake) and rivers (e.g., damming, water removal) (Ural-Janssen et al., 2023). The model considers the following pollution sources:

- Agricultural sources include animal housing and manure storage systems, the application of synthetic fertilizers and untreated manure on land, manure deposited on land during grazing, biological N_2 fixation by crops, atmospheric N and P deposition on agricultural land, leaching of organic matter and weathering of P-contained minerals from agricultural areas to surface waters;
- Non-agricultural sources encompass human waste from populations not connected to sewage systems, biological N_2 fixation by natural vegetation, atmospheric N deposition on non-agricultural land, leaching of organic matter and weathering of P-contained minerals from non-agricultural areas;
- Urbanization-related sources include urban and rural sewage systems (human waste that is collected and treated).

We integrate the BBFs produced by N recovery from manure processing and by P recovery from treated sewage sludge into the model quantifying:

1. N emissions to the air from manure processing systems (AD+AS) and from the application of ReNuRe products on agricultural land;
2. DIN and DON inputs to rivers and their exports to seas from the application of ReNuRe products on agricultural land;
3. DIP and DOP inputs into rivers and their exports to seas from the application of recovered P from treated sewage sludge (RPSS) on agricultural land.

N emissions to the air from agricultural sources are quantified as:

$$Nemission_{f.a.y.j} = Nexcretion_{a.y.j} \times EF_{f.a.y.j} \quad (1)$$

$$Nemission_{f.y.j} = WSdif_{N.y.j} \times EF_{f.y.j} \quad (2)$$

where $Nemission_{f.a.y.j}$ represents N emissions to the air by form f (NH_3, N_2O, NO_x), animal type a (dairy cattle, other cattle, pig, poultry, other animals), source y (animal housing, manure storage) and basin j (kg N yr^{-1}). These emissions are quantified as a function of animal N excretions ($Nexcretion_{a.y.j}$, kg N yr^{-1}) and EFs ($EF_{f.a.y.j}$, 0-1) per N form f, animal type a, source y and basin j. $Nemission_{f.y.j}$ represents N emissions to the air by form f (NH_3, N_2O, NO_x), by source y (synthetic fertilizer, untreated manure, ReNuRe products and effluent from manure processing, manure deposited on land during grazing, atmospheric deposition and biological N_2 fixation) and basin j (kg N yr^{-1}). These emissions are quantified as a function of N inputs to agricultural land ($WSdif_{N.y.j}$, kg N yr^{-1}) and EFs ($EF_{f.y.j}$, 0-1) per form, source y and basin j.

Nutrient inputs to rivers from agricultural diffuse sources are quantified as:

$$RSdif_{f.y.ag.j} = WSdif_{f.y.j} \times G_{f.j} \times FE_{ws.f.j} \quad (3)$$

$$RSdif_{f.y.nonag.j} = WSdif_{f.y.j} \times FE_{ws.f.j} \quad (4)$$

where $RSdif_{f.y.ag.j}$ represents annual nutrient inputs to rivers by form f (DIN, DON, DIP, DOP), diffuse source y from agricultural (ag) areas in basin j (kg N or P yr^{-1}). Nutrient inputs to rivers from agricultural diffuse sources are quantified as a function of nutrient inputs to land ($WSdif_{f.y.j}$, kg N or P yr^{-1}) that are corrected for nutrient export from agricultural land (via crop harvesting and animal grazing) ($G_{f.j}$, 0-1), and runoff from land to streams ($FE_{ws.f.j}$, 0-1) (Ural-Janssen et al., 2023).

Nutrient inputs to rivers from point sources are quantified as:

$$RSpnt_{f.y.j} = Pop_j \times fr_{pop.sew.j} \times WScap_{e.j} \times (1 - hw_{frem.e.j}) \times FEpnt_{f.j} \quad (5)$$

where $RSpnt_{f.y.j}$ represents annual nutrient inputs to rivers by form f (DIN, DIP, DON, DOP) point source y in basin j (kg N or P yr^{-1}). Nutrient inputs to rivers from point sources are quantified as a function of population (Pop_j, person) that is connected to sewage systems ($fr_{pop.sew.j}$, 0-1), per capita human N and P excretion rates ($WScap_{e.j}$, kg N or P $person^{-1}$ yr^{-1}), removal efficiencies of nutrients during wastewater treatment ($hw_{frem.e.j}$, 0-1) and the fraction of nutrients entering rivers by the effluent from wastewater treatment systems ($FEpnt_{f.j}$, 0-1).

River exports of nutrients from diffuse and point sources are quantified as:

$$M_{f.y.j} = \left(RSdif_{f.y.j} + RSpnt_{f.y.j}\right) \times FE_{riv.f.outlet.j} \times FE_{riv.f.mouth.j} \quad (6)$$

where $M_{f.y.j}$ represents annual exports of nutrients to seas by form f (DIN, DON, DIP, DOP), diffuse and point source y and basin j (kg N or P yr^{-1}). River exports of nutrients are quantified as a function of nutrient inputs to rivers that are corrected for removal (e.g., water consumption) and retention (e.g., damming, sedimentation) in rivers ($FE_{riv.f.outlet.j}$, 0-1), and the traveling distance of the nutrients to the river mouth ($FE_{riv.f.mouth.j}$, 0-1). TDN and TDP consist of the dissolved inorganic and organic forms (TDN = DIN + DON, TDP = DIP + DOP). Further details for model calculations are in Table S4.1, Table S4.2, and Table S4.3, and for uncertainty and limitations in Supplementary Text S4.2.

4.2.2 Scenario development

The future baseline scenario is taken from our previous study (Ural-Janssen et al., 2024) and the combination of SSP5-RCP8.5. This scenario assumes global warming with high economic developments, rapid urbanization, and intensified agriculture (Supplementary Text S4.1). Here, we develop six alternative scenarios that differ in replacing synthetic fertilizers with BBFs and enhancing nutrient use efficiency to reduce nutrient losses in Europe for 2050: two for N, and four for P (Figure 4.1 and Table S4.4).

Two N-scenarios include:

- *The ReNuRe scenario* includes N recovery from manure via AD followed by an AS process. Combining both technologies, the raw animal slurry is converted into an ammonium salt that can be used as ReNuRe fertilizer (Sigurnjak et al., 2019), and an organic matter-rich effluent. This scenario replaces synthetic N fertilizers with the ReNuRe products produced by manure processing while maintaining the effective N inputs as in the future baseline. For this, N excretions produced by dairy cattle, other cattle, and pigs are treated via AD+AS processes (Table S4.4, Figure S4.1 and Figure S4.2). AD increases inorganic content by breaking down organic components relative to untreated manure (Meers et al., 2020). AS removes NH_3 from an NH_3-rich waste stream in a stripping reactor by using an airflow, and transfers NH_3 from liquid to the gas phase with a removal of 56% (Vingerhoets et al., 2024) and captures the released NH_3 by a strong acid (e.g., sulfuric or nitric acid) (Sigurnjak et al., 2019). Excretions produced by poultry and other animals (i.e. sheep, goat) are considered to be applied as untreated manure on agricultural areas.
- *The ReNuRe-NUE scenario* expands the ReNuRe scenario with an assumed increase in NUE on agricultural land by 2050. NUE is considered as the overall system efficiency according to Bouwman et al. (2009), which is the ratio of N yield from crop harvesting and animal grazing to the total N input to agricultural areas (synthetic fertilizers, animal manure, atmospheric N deposition and biological N_2 fixation). We increased the NUE to 0.75 for the basins with NUE lower than 0.75 (Figure S4.3) following Schulte-Uebbing

and de Vries (2021), being a maximum feasible NUE for maintaining crop production while respecting environmental thresholds (i.e. N concentration in surface water to limit eutrophication). Based on the increased NUE and assuming a similar N yield as in the future baseline, we maintain the total effective N input and subsequently reduce the synthetic N fertilizer use. We assume that manure processes do not have an impact on P amount in animal manure and thus, the amount of P does not change in N-scenarios.

Four P-scenarios include:

- *The Pmed scenario* combines P recovery from treated sewage sludge and medium improvement of the WWTPs for better nutrient removal. This scenario replaces synthetic P fertilizers with RPSS while maintaining the total P inputs as in the baseline. The WWTP technologies per basin are assumed to be upgraded by one level: primary to secondary, and secondary to tertiary. Moreover, considering recently proposed technologies (> 90% of P recovery (Huygens et al., 2019)), we assume 90% of P recovery from these WWTPs and quantify the amount of RPSS accordingly. Finally, we identify the amount of synthetic P fertilizers that can be replaced by RPSS relative to the future baseline.
- *The Padv scenario* is similar to that of Pmed but here we assume that all the WWTPs lower than tertiary in basins will be upgraded to tertiary treatment.
- *The Pmed-PUE scenario* assumes further improvements in PUE on agricultural land in addition to the Pmed scenario. PUE is considered as the overall system efficiency according to Bouwman et al. (2009), which is the ratio of P yield from crop harvesting and animal grazing to the total P input to agricultural areas (synthetic fertilizers, animal manure, and atmospheric deposition). To increase the PUE in this scenario, we follow the approach of the NUE as the proxy in which we increase the PUE to 0.75 for the basins with the PUE lower than 0.75 (Figure S4.3). Based on the increased PUE and assuming a similar P yield as in the future baseline, we maintain the total P input and subsequently reduce the synthetic P fertilizer application.
- *The Padv-PUE scenario* includes all assumptions of the Padv and Pmed-PUE scenarios.

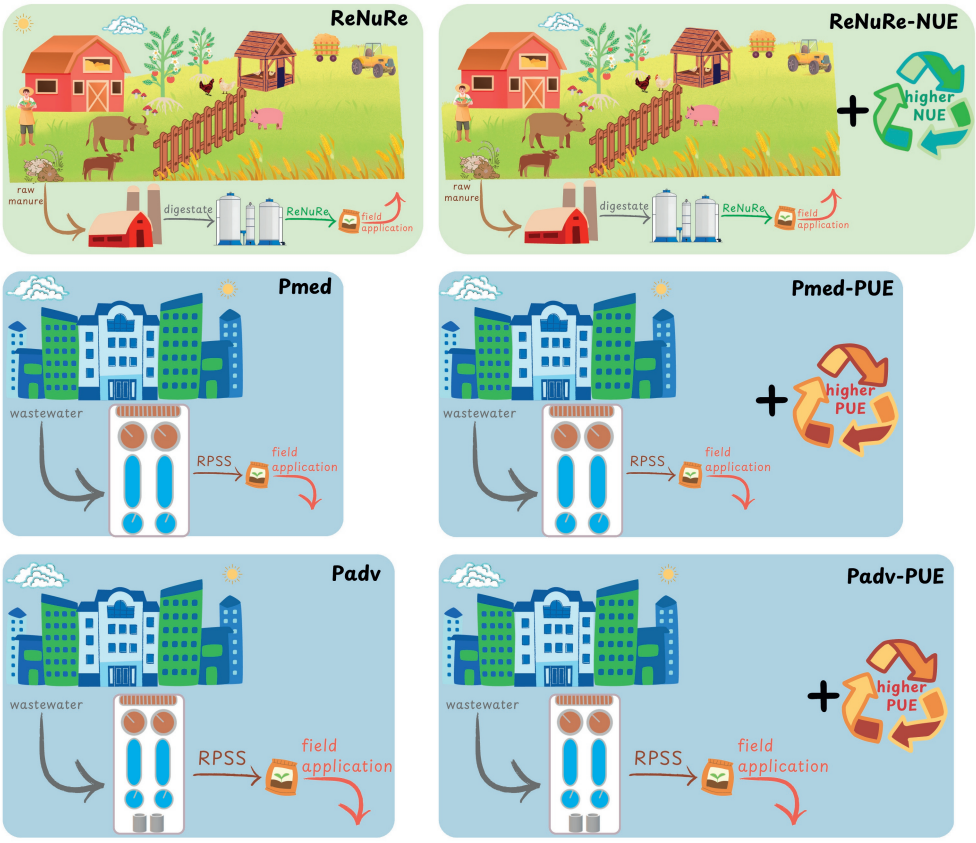


Figure 4.1 Schematic overview of the six alternative scenarios for 2050. These scenarios are ReNuRe (ReNuRe products), ReNuRe-NUE (ReNuRe products + increased NUE), Pmed (RPSS + medium upgrade of WWTPs), Pmed-PUE (RPSS + medium upgrade of WWTPs + increased PUE), Padv (RPSS + advanced upgrade of WWTPs) and Padv-PUE (RPSS + advanced upgrade of WWTPs + increased PUE). ReNuRe stands for REcovered Nitrogen from manURE, NUE is short for nitrogen use efficiency, RPSS is short for recovered phosphorus from treated sewage sludge, and PUE is short for phosphorus use efficiency. The treatment box in the Pmed and Pmed-PUE scenarios represents secondary treatment, and the treatment box in the Padv and Padv-PUE scenarios represents tertiary treatment. Source: Section 4.2.2 and Table S4.4.

4.3 Results

4.3.1 Reducing future N emissions to the air

In the baseline scenario, total N emissions to the air from agriculture will increase by 16% by 2050 compared to 2017-2020 (Figure S4.4). In contrast, alternative scenarios project reduced N emissions by 2050: 10% and 19% under the ReNuRe and ReNuRe-NUE scenarios, respectively (Figure S4.4). Compared to the 2050 baseline, ReNuRe projects a 22% reduction and ReNuRe-NUE projects a 30% reduction in N emissions for the whole study area (Figure S4.4).

Reduction potentials differ among NH_3, N_2O, and NO_x (Figure 4.2 and Figure S4.4). Between 2050 and the period of 2017-2020, the total NH_3 emissions are projected to be 10% and 19% lower under the ReNuRe and ReNuRe-NUE scenarios, respectively (Figure S4.4). For total N_2O, these reductions range from 7% to 20% under ReNuRe and from 12% to 30% under ReNuRe-NUE. Compared to the 2050 baseline, the ReNuRe and ReNuRe-NUE scenarios project 23-31% reductions for the NH_3, 8-21% reductions for N_2O and 21-37% reductions for NO_x (Figure 4.2).

Projected reductions in N emissions in the ReNuRe scenario are associated with manure processing (AD+AS) after the animal housing systems, and the application of ReNuRe products (Figure 4.2). AS process has very low NH_3 emissions (no N_2O and NO_X) compared to manure storage for dairy cattle, other cattle, and pigs. The use of such closed-loop management systems reduces N emissions compared to conventional practices, providing better nutrient recycling. Moreover, ReNuRe products release less N emissions into the air relative to the untreated manure when applied on the field. Larger reductions in the ReNuRe-NUE scenario are mainly from the reduction in synthetic fertilizers (Figure 4.2). Increasing the NUE lowered the total N input to agricultural land to 25,750 Gg N yr^{-1} compared to the future baseline (31,705 Gg N yr^{-1}, Figure S4.5). Applying ReNuRe products in combination with increasing NUE seems to be effective in reducing N emissions from agriculture.

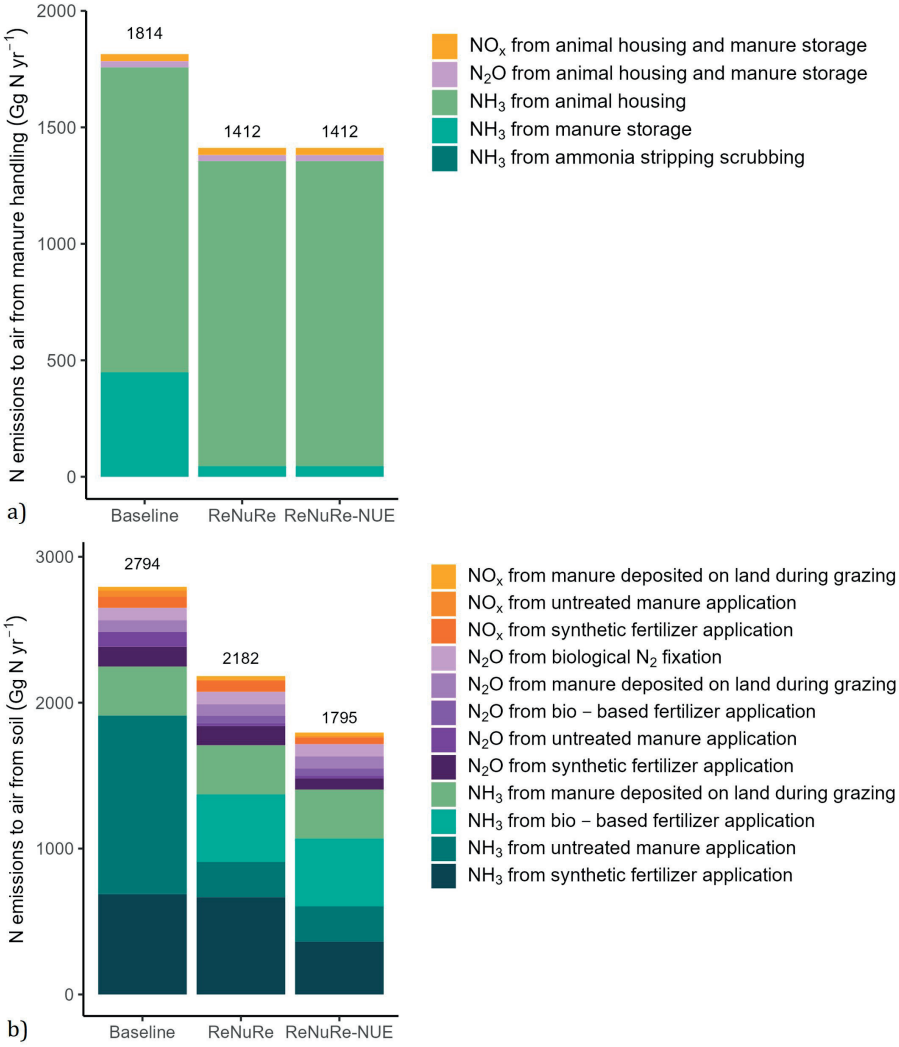


Figure 4.2 Nitrogen (N) emissions to the air from agricultural systems under the baseline and two alternative scenarios in 2050 (Gg N yr^{-1}). The baseline is based on the combination of SSP5-RCP8.5 (Shared Socioeconomic Pathway 5 and Representative Concentration Pathway 8.5). The two alternative scenarios are ReNuRe (ReNuRe products) and ReNuRe-NUE (ReNuRe products + increased NUE). ReNuRe stands for REcovered Nitrogen form manURE and NUE is short for nitrogen use efficiency. a) N emissions to the air from manure handling. b) N emissions to the air from agricultural soils. Source: The MARINA-Nutrients model for Europe.

4.3.2 Reducing future nutrients in rivers

Between 2050 and the period of 2017-2020, TDN and TDP in European rivers are projected to increase by 11% and 30%, respectively under the baseline scenario (Figure 4.3). This is mainly associated with more nutrients from point sources (sewage systems) as a result of future urbanization (Ural-Janssen et al., 2024). However, alternative scenarios project different trends. For TDN, the most effective scenario is combination of adopting ReNuRe products with increased NUE (ReNuRe-NUE) projecting a 14% reduction by 2050 relative to the period of 2017-2020 (Figure 4.3). For TDP, the Pmed and Padv-PUE scenarios project 39-58% reductions relative to 2017-2020 (Figure 4.3). Compared to the future baseline, ReNuRe-NUE projects 23% reductions for TDN and Padv-PUE projects 68% reductions for TDP in 2050 (Figure 4.3). By upgrading the WWTPs in the Pmed (one level up) and Padv (to tertiary treatment) scenarios could help reduce not only TDP (53-65%) but also TDN (11-13%) in rivers relative to the baseline (Figure 4.3).

Compared to the future baseline, 46% less DIN and 12% less DON inputs to all studied rivers from agriculture are projected in the ReNuRe-NUE scenario (Figure 4.3 and Figure S4.6). This is due to the decline in synthetic fertilizer use (39%) and atmospheric N deposition (24%) compared to the future baseline (Figure S4.5). Lower fertilizer use is associated with higher NUE, and less atmospheric N deposition is associated with reduced volatilization from manure storage, processing, and application (Figure 4.2).

In P-scenarios, RPSS can fulfill fertilizer needs in Europe for 2050 and reduce DIP and DOP in rivers (Figure 4.3 and Figure S4.5). DIP and DOP pollution in European rivers can be reduced by 25% and 8%, respectively, by implementing RPSS and increased PUE compared to the future baseline (Figure S4.6). This is because of the reduced fertilizer application under the Pmed-PUE and Padv-PUE scenarios relative to the future baseline (Figure S4.5).

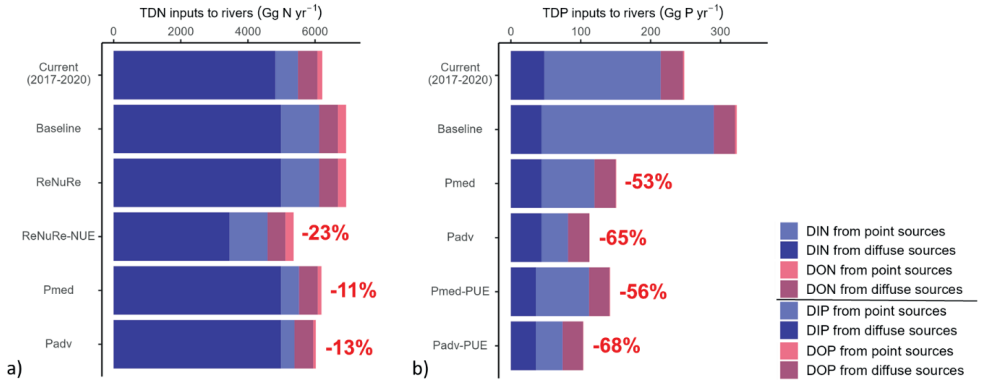


Figure 4.3 Inputs of nutrients to rivers from diffuse (agricultural, non-agricultural) and point (sewage systems) sources under current (2017-2020), the baseline, and six alternative scenarios in 2050 (Gg N or P yr^{-1}). a) Total dissolved nitrogen (TDN) consisting of dissolved inorganic and organic forms. b) Total dissolved phosphorus (TDP) consisting of dissolved inorganic and organic forms. The percentages are estimated relative to the baseline. The baseline is based on the combination of SSP5-RCP8.5 (Shared Socioeconomic Pathway 5 and Representative Concentration Pathway 8.5). The six alternative scenarios are ReNuRe (ReNuRe products), ReNuRe-NUE (ReNuRe products + increased NUE), Pmed (RPSS + medium upgrade of WWTPs), Pmed-PUE (RPSS + medium upgrade of WWTPs + increased PUE), Padv (RPSS + advanced upgrade of WWTPs) and Padv-PUE (RPSS + advanced upgrade of WWTPs + increased PUE). ReNuRe stands for REcovered Nitrogen from manURE, NUE is short for nitrogen use efficiency, RPSS is short for recovered phosphorus from treated sewage sludge, WWTPs is short for wastewater treatment plants, and PUE is short for phosphorus use efficiency. Source: The MARINA-Nutrients model for Europe.

There is a large spatial variability in future nutrient inputs to rivers (Figure S4.8), and their reductions relative to the future baseline. In a third of the studied basins, TDN inputs to rivers could be reduced by more than 46% under the ReNuRe-NUE scenario, while TDP inputs to rivers could decline by more than 68% under the Padv-PUE scenario compared to the future baseline (Figure 4.4). The simultaneous decrease in TDN and TDP inputs to rivers is mainly projected for eastern and southern basins (Figure 4.4), and basins with the highest inputs are expected in central and western Europe with intensively farmed regions and dense population (Figure S4.8). The main drivers of the reductions are less fertilizer inputs to agricultural land (Figure S4.5) due to the increased overall nutrient use efficiencies, and less nutrient losses from point sources (Figure S4.7) due to the improved wastewater treatment technologies relative to the future baseline.

Adopting ReNuRe products and improving NUE is a promising solution for reducing future N losses to air and waters from agriculture synergistically. For a third of the studied basins, more than 58% of TDN inputs to rivers and more than 34% of NH_3 emissions to the air are expected to reduce under the ReNuRe-NUE scenario (Figure 4.4). The decreasing N losses

are projected for most of the basins across Europe implying that combining the use of BBFs with the measures increasing NUE on agricultural land can reduce N emissions to the air and water simultaneously (Figure 4.4). Nevertheless, certain relatively small basins in western Europe are projected to release the highest levels of NH_3 (Group 5, Figure S4.8) primarily due to intensive livestock regions.

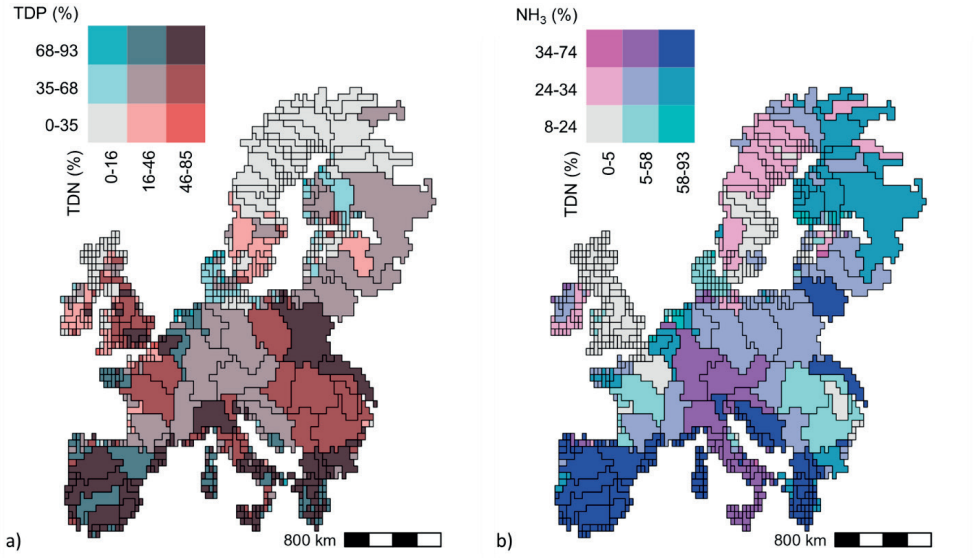


Figure 4.4 Air and water pollution reductions in alternative scenarios relative to the baseline scenario in 2050 (%). a) Future reductions in total dissolved N (TDN) vs. future reductions in total dissolved phosphorus (TDP) inputs to rivers from agricultural, non-agricultural and urbanization-related sources under the ReNuRe-NUE (ReNuRe products + increased NUE) and Padv-PUE (RPSS + advanced upgrade of WWTPs + increased PUE) scenarios, respectively, relative to the baseline (%). b) Future reductions in ammonia (NH_3) emissions to the air vs. future reductions in total dissolved nitrogen (TDN) inputs to rivers from agriculture under the ReNuRe-NUE scenario (ReNuRe products + increased NUE) relative to the baseline (%). The baseline is based on the combination of SSP5-RCP8.5 (Shared Socioeconomic Pathway 5 and Representative Concentration Pathway 8.5). ReNuRe stands for REcovered Nitrogen from manURE, NUE is short for nitrogen use efficiency, RPSS is short for recovered phosphorus from treated sewage sludge, WWTPs is short for wastewater treatment plants, and PUE is short for phosphorus use efficiency. Percentage reductions were calculated using loads for NH_3 emissions, TDN and TDP inputs to rivers. Source: The MARINA-Nutrients model for Europe.

4.3.3 Reducing future river exports of nutrients to seas

Between 2050 and the period of 2017-2020, river exports of TDN and TDP to European seas are estimated to increase by 14% and 28%, respectively (Figure S4.9). This is mainly driven by increased sewage systems and moderate treatment improvement (Ural-Janssen et al., 2024). Alternative scenarios project different trends. Compared to the period of 2017-2020, the ReNuRe-NUE projects a 9% reduction in the river export of TDN by 2050, which

is much higher for TDP (36-51%) under the P-scenarios (Figure S4.9). Compared to the future baseline, rivers may export up to two-thirds less nutrients to all studied seas in 2050 (Figure S4.9). This depends on the scenarios (Figure 4.5 and Figure S4.8). For instance, 62% less TDP export by European rivers is expected due to the implementation of upgraded treatment technologies and the use of RPSS together with higher PUE (Figure S4.9). When ReNuRe products are combined with higher NUE, 20% less TDN export by rivers is projected compared to the future baseline (Figure S4.9).

Total DIN export by rivers from agriculture and sewage systems is projected to reduce by 14-33% under the Pmed and ReNuRe-NUE scenarios, respectively, relative to the future baseline (Figure 4.5). The reduced DIN export under the P-scenarios mainly occurs as a result of upgraded WWTPs and associated nutrient removal (Figure S4.7). Relatively lower reduction in the ReNuRe-NUE, but higher reductions in the Pmed and Padv scenarios are estimated for DON export by all studied rivers (Figure 4.5).

Among P forms, overall more than 60% reductions are projected for total DIP export by all studied rivers, up to 78% under the Padv-PUE scenario relative to the future baseline (Figure 4.5). Relatively lower reductions in total DOP export by rivers (11-20%) are estimated (Figure 4.5). All reductions are caused by improving WWTPs (69-85% less TDP inputs to rivers, Figure S4.7) and increasing PUE (less fertilizer inputs to agricultural land, Figure S4.5) relative to the future baseline.

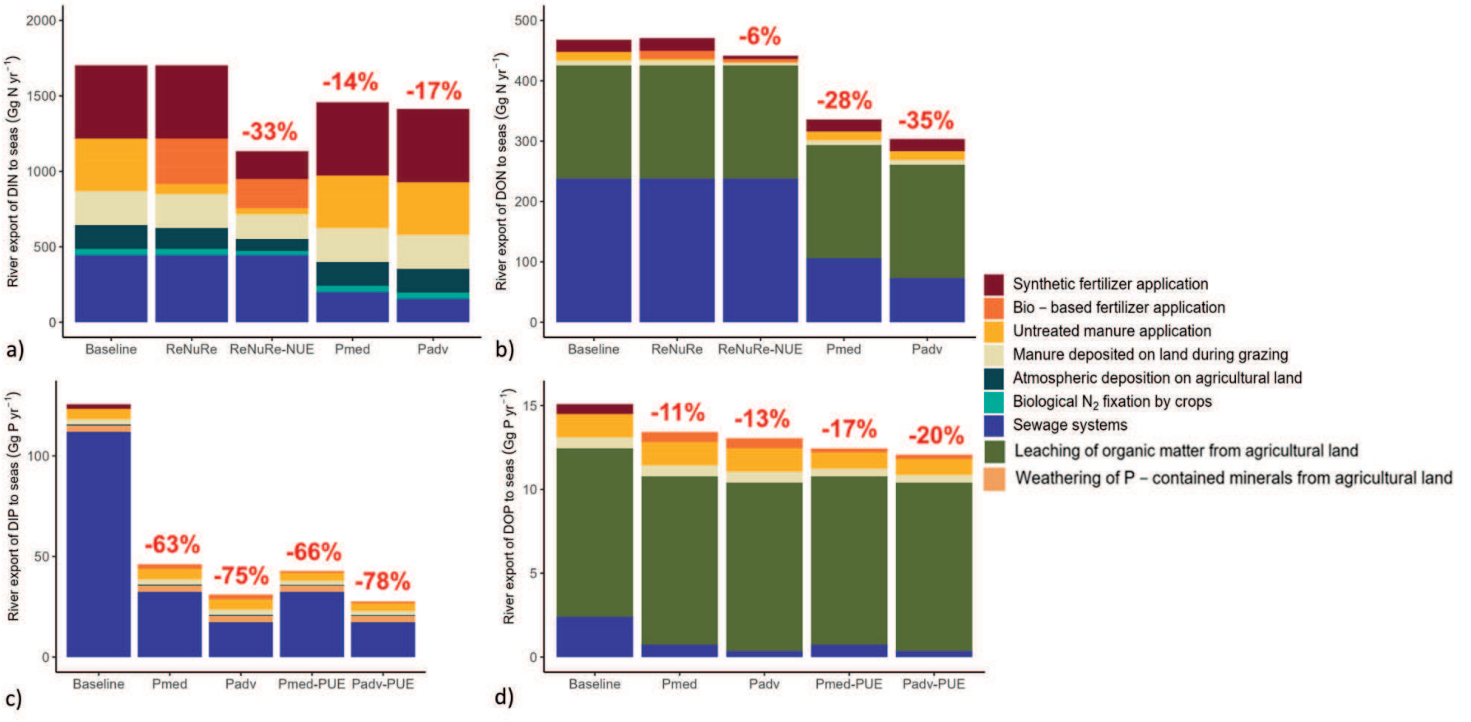


Figure 4.5 River exports of nutrients to seas from agriculture and sewage systems under the baseline and six alternative scenarios in 2050 (Gg N or P yr^{-1}). a) Dissolved inorganic nitrogen (DIN). b) Dissolved organic nitrogen (DON). c) Dissolved inorganic phosphorus (DIP). d) Dissolved organic phosphorus (DOP). The percentages are estimated relative to the baseline. The baseline is based on the combination of SSP5-RCP8.5 (Shared Socioeconomic Pathway 5 and Representative Concentration Pathway 8.5). The six alternative scenarios are ReNuRe (ReNuRe products), ReNuRe-NUE (ReNuRe products + increased NUE), Pmed (RPSS + medium upgrade of WWTPs), Pmed-PUE (RPSS + medium upgrade of WWTPs + increased PUE), Padv (RPSS + advanced upgrade of WWTPs) and Padv-PUE (RPSS + advanced upgrade of WWTPs + increased PUE). ReNuRe stands for REcovered Nitrogen from manURE, NUE is short for nitrogen use efficiency, RPSS is short for recovered phosphorus from treated sewage sludge, WWTPs is short for wastewater treatment plants, and PUE is short for phosphorus use efficiency. Source: The MARINA-Nutrients model for Europe.

4.4 Discussion

4.4.1 Comparisons

Minimum one-third of surface waters (e.g., 36% of rivers, 31% of coastal waters) of the European Union are reported eutrophic (EC, 2021b). Thus, achieving the targets of "good ecological and chemical status" according to the Water Framework Directive is still challenging without effective measures (EC, 2021b). In our study, we found that nutrient exports by the European rivers to the coastal waters could be reduced by 14-51% in 2050 relative to the current period (2017-2020), when ReNuRe products and RPSS are widely implemented and combined with management practices leading to higher nutrient use efficiencies in the field (Figure S4.9).

Moreover, gaseous N losses could be reduced up to 19% by 2050 compared to 2017-2020, when ReNuRe products combined with higher NUE (Figure S4.4). On the other hand, Rashid et al. (2025) found that when ammonium sulfate is applied as BBF with equivalent total N input, gaseous N losses were comparable to those from conventional fertilizers. They used the Daisy model to simulate N dynamics, including gaseous losses (e.g., NH_3, N_2O) based on fertilizer type, application rate, and environmental conditions. This could be explained by the choices of EFs for production and application of BBFs. Our study considers the ReNuRe products produced by AD and AS which prevents gaseous N losses, and lower EFs for their land application relative to synthetic fertilizers and untreated manure regardless of application type and environmental conditions (Table S4.3).

The use of BBFs did not negatively affect the N and P yields – defined as nutrient exported from agricultural land through crop harvesting and grazing since the total effective nutrient input level remained the same or the decline was compensated with an increase in nutrient use efficiency. Our findings align with literature regarding the agronomic performance of BBFs (Sigurnjak et al., 2019; Huygens et al., 2020; Ye et al., 2020; Rashid et al., 2025). On the other hand, while mineral-like BBFs can balance N supply with plant uptake and reduce N losses, they do not contribute to long-term soil fertility. Whereas, BBFs with higher organic N content like anaerobic digestate can enhance N supply capacity and soil fertility over time (Rashid et al., 2025). Therefore, future research should understand these implications and long-term benefits based on regional needs and barriers.

4.4.2 Future implications

Our model results account for spatially explicit analysis of air and water pollution, and contribute to developing synergistic solutions to mitigate nutrient losses from agriculture and sewage systems. Some nutrient management measures might have trade-offs known as pollution swapping (Velthof et al., 2014). However, synergetic options still receive relatively less attention in research and policy (Kanter et al., 2020). We show that the F2F target "reduction of nutrient losses by at least 50%" (EC, 2020) could be achieved for P inputs to rivers

under the P-scenarios, and more than 20% decline in N fertilizer use can be possible by 2050 under the ReNuRe-NUE scenario providing reduced N emissions to the air and waters. Our analysis also supports the River Basin Management Plans (EC, 2000) by basin-specific analysis for opportunities reducing air and water pollution simultaneously, and minimizing the risk of pollution swapping to help solve N crisis in Europe.

We chose animal manure and sewage sludge as relatively larger organic waste streams to produce BBFs. ReNuRe products produced from manure processing are promising alternatives to synthetic N fertilizers mainly due to the reduced NH_3 emissions. On the other hand, P recovery and recycling are inevitable to sustain agricultural production while maintaining water quality, in particular in areas where soil P pools are below agronomic thresholds. The Urban Waste Water Treatment Directive urges to re-use the sludge arising from WWTPs whenever appropriate (EC, 1991b), although using the recovered products derived from treated sewage sludge remains restricted/prohibited in most EU countries. High P recovery from municipal sewage sludge-derived ash is a sustainable alternative for circular economy (Liu et al., 2021; Fournie et al., 2022; Zhu et al., 2023). Our study shows that RPSS can provide a circular solution to meet P fertilizer demand in Europe for 2050. This helps close nutrient cycles at the basin scale and contributes to the EU zero pollution goals and legislation for P recovery. However, limitations such as heavy metal contamination (Husek et al., 2022) and regional surplus/deficiency (Panagos et al., 2022) should be considered while selecting processes. Thus, nutrient recovery and recycling from sewage sludge after sufficient treatment and heavy metal removal not only promises economic benefits, but also leads to reduced nutrient inputs to rivers and coastal waters from point sources.

BBFs have a large potential for resource recovery and circular economy, especially for manure-surplus countries, though their benefits are largely controlled by the enhanced use efficiency for both N and P. Their adoption with higher nutrient use efficiency in agriculture supports the F2F strategy and the United Nations Sustainable Development Goals (SDGs 2 and 6) by simultaneously mitigating air and water pollution, and improving agronomic efficiency. However, even if the environmental benefits of BBFs are underscored, marketing and usage are highly influenced by their cost and social perception (Moshkin et al., 2023; Garmendia-Lemus et al., 2024). ReNuRe products offer potential solution by providing the use of local nutrients and reducing dependency on imported fertilizers (Vingerhoets et al., 2025). Future studies should delve into cost-benefit analyses for the production and use of different BBFs, and explore the intricate relationship between stakeholders vs. agronomic and environmental impacts for a comprehensive assessment of implementing BBFs and introducing them to the market.

4.5 Conclusions

We analyze the potential of synergetic options for reducing future nutrient losses from agriculture and urbanization-related sources in Europe under six scenarios considering improved nutrient recovery, recycling, and efficiency. Our study is the first attempt to model the effects of adopting BBFs and improving nutrient use efficiencies in agriculture on N emissions to the air and nutrient inputs to waters, and the effects of enhancing sewage treatments on nutrient inputs to waters by 2050 while accounting for the spatial drivers of nutrient pollution. Adopting ReNuRe products and increasing NUE is found to be synergistic option to reduce N emissions to the air, and N inputs to rivers and seas of Europe. By combining optimized use of ReNuRe products with higher NUE, 30% less N in the air and 23% less N inputs to rivers are expected by 2050 compared to the future baseline. By combining enhanced sewage treatment, use of RPSS and higher PUE, 68% less P inputs to rivers are projected relative to the future baseline. Consequently, these rivers are predicted to export up to two-thirds less nutrients to seas compared to the future baseline. Besides, the recovered P from treated sewage sludge can provide P fertilizer demand in Europe by 2050. Our findings help achieve the F2F target on mitigating nutrient losses by 50% and reducing fertilizer use by 20%, and support circular economy.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Aslıhan Ural-Janssen: Writing – review & editing, Writing – original draft, Visualization, Methodology, Conceptualization. Erik Meers: Writing – review & editing, Supervision, Funding acquisition. Gerard H. Ros: Writing – review & editing, Methodology, Conceptualization. Ruben Vingerhoets: Writing – review & editing. Maryna Strokal: Writing – review & editing, Supervision, Project administration, Methodology, Funding acquisition, Conceptualization.

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Data availability

The main model results supporting Figures 4.2-4.5 generated in this study are deposited in the DANS Easy repository https://doi.org/10.17026/PT/AWTUAM.

Supplementary materials

Supplementary material can be found online at https://doi.org/10.1016/j.resconrec.2025.108472 and in this thesis under *S4: Supplementary materials for Chapter 4*.



Chapter 5

Synthesis

In this chapter, I synthesize my PhD research and reflect on the development and application of the new MARINA-Nutrients for Europe, which integrates air and water pollution associated with nutrient emissions. MARINA-Nutrients stands for Model to Assess River Inputs of pollutaNts to seAs for Nutrients. This European version is designed to simultaneously quantify nitrogen (N) emissions to the air, inputs of nutrients to rivers and river exports of nutrients to coastal waters distinguishing contributions by source and basins annually for the recent past and future. The new MARINA-Nutrients for Europe also aims to identify the required reductions for coastal water pollution and accounts for the effects of bio-based fertilizers (BBFs) in combination with other nutrient management strategies on nutrient emissions to the air, rivers and coastal waters.

Below, Section 5.1 summarizes this PhD dissertation followed by the main findings of the research chapters and their synthesis. Section 5.2 describes the new MARINA-Nutrients for Europe in comparison with illustrative examples of existing models for nutrient emissions to air and/or water systems (rivers and coastal waters), and my contributions to the family of the MARINA models. Section 5.3 shifts the focus to results mainly from Chapters 2-4 and discusses them compared to the results of other modeling studies. Section 5.4 reflects on the strengths, weaknesses, opportunities and threats of this PhD research (using the SWOT approach). Section 5.5 shifts the focus to perspectives on the implementation of BBFs in the EU and their role in air and surface water pollution reduction. Finally, Section 5.6 presents a future outlook by providing recommendations for science and policy for improving air and water quality in Europe in the future.

5.1 Research findings and novelties

Section 5.1 summarizes the story of this PhD research followed by the knowledge gaps addressed, the objectives and highlights of this thesis (Section 5.1.1 and Table S5.1). Then, it presents the key findings of each research chapter (Section 5.1.2), and the synthesis (Section 5.1.3).

5.1.1 This PhD research in a nutshell

Intensive agriculture and urbanization are important sources of nutrient pollution and associated impacts in Europe (Grizzetti et al., 2023). Air quality often suffers from reactive N emissions of agriculture, industry and transportation sectors (EEA, 2024b; 2025c). Those emissions are deposited on land as atmospheric N deposition, and then reach rivers and coastal waters (de Vries et al., 2024). Not only N, but also high phosphorus (P) inputs to rivers and coastal waters from human activities on the land cause eutrophication issues (Vigiak et al., 2023) and associated impacts (EC, 2021b; Haase et al., 2023). Integrated assessments of nutrient emissions to both air and surface waters are, so far, limited, especially on the basin scale for the European Union (EU) and the United Kingdom (UK). This prevents a

better understanding air and water pollution associated with nutrient emissions, their future trends and reduction needs to avoid future pollution and impacts. On the other hand, the EU Water Framework Directive requests River Basin Management Plans from the Member States (EC, 2000). Despite the progress of the EU policies, air, river and coastal water pollution associated with nutrient emissions is still a considerable issue (EEA, 2025e; Grizzetti et al., 2025) with consequences on human health and the environment (Table S1.1). Therefore, nutrient management strategies to reduce both air and water pollution simultaneously are needed, but their effects have not been explored enough for European basins and future years.

Increasing population and demand for agricultural productivity urge the renewable alternatives such as BBFs (see Section 1.1.7 for definition) to replace nonrenewable synthetic fertilizers. BBFs could potentially reduce nutrient pollution through nutrient recovery from the processing of organic waste streams such as animal manure and municipal wastewater/sewage sludge while increasing the circularity of nutrients and closing their loops (Vingerhoets et al., 2023b). However, the environmental impacts of BBFs are often analyzed at small scales (e.g., laboratory, field), and hardly consider both air and water pollution associated with nutrient emissions. Much less is known about the role of BBFs in mitigating nutrient emissions to the air and surface waters, in particular rivers and coastal waters, at the EU level while accounting for socioeconomic developments and climate change in the future. In addition, the role of BBFs in combination with other nutrient management options is exploited to a limited extent for the EU and the UK basins. Consequently, there is a lack of a comprehensive policy for the adoption and implementation of these products in the EU (see Chapter 1 for more information).

Therefore, this PhD research aimed to enhance our understanding of basin-specific nutrient emissions to the air and surface waters, and explore the role of BBFs and other nutrient management strategies to reduce future nutrient pollution under global change across the EU and the UK. To this end, three research sub-objectives are formulated and achieved in response to the knowledge gaps that are discussed in Chapter 1 and addressed in Chapters 2-4 (Table S5.1). The key findings of Chapters 2-4 are elaborated below (Table S5.1 and Figure 5.1).

5.1.2 Key findings of each chapter

Integrated air and water pollution modeling - hotspots of nutrient emissions

Chapter 2 is novel in providing a new integrated model to quantify nutrient emissions to the air and surface waters (rivers and coastal waters) at a basin scale for the EU and the UK. This chapter also presents fundamental insights into the contribution of agriculture to air, river, and coastal water pollution simultaneously by nutrients. The new European version of the MARINA-Nutrients is developed through soft linking the MARINA (Model to Assess

River Inputs of pollutaNts to seAs) and MITERRA-Europe (Integrated Modeling of Nutrient Flows at Regional Level) models. This is done by integrating and improving the existing modeling approaches, which then provide MARINA-Nutrients to simulate N emissions to the air from agriculture. The MARINA-Nutrients for Europe quantifies the emissions of ammonia (NH_3), nitrous oxide (N_2O), and nitrogen oxides (NO_x) to the air from agricultural sources, and the inputs of dissolved inorganic and organic N (DIN, DON) and P (DIP, DOP) to rivers and their exports by rivers to coastal waters from agricultural (e.g., synthetic fertilizers and manure), non-agricultural (e.g., biological N_2 fixation by natural vegetation) and sewage-associated sources (e.g., treatment systems). By applying MARINA-Nutrients to 601 river basins (593 individual basins and 8 sub-basins of the Danube River) in the EU and the UK, the annual loads of aforementioned nutrient emissions and losses are quantified for the period of 2017-2020. The basins with the top 20% of the highest N emissions to the air are identified as "air pollution hotspots". The basins with the top 20% of the highest inputs of N and/or P to rivers are identified as "water pollution hotspots", and the overlapping basins as "air and water pollution hotspots" (the 80th percentile based on loads per km^2 of basin area in kg km^{-2} yr^{-1}).

Model results reveal that almost two-fifths of reactive N emissions to the air from agriculture are from animal housing and storage. Moreover, agriculture –mainly the application of synthetic fertilizers and manure– and sewage systems are responsible for 55% of N and 67% of P in rivers, respectively, at the EU level. Nearly a third of the European basin area is found as pollution hotspots (air, water, or both), and produces over half of N emissions to the air and nutrients in rivers. Results show that 59% of the total population in the study area lives in those hotspots. Based on these research insights, the new MARINA-Nutrients model for Europe enables a better understanding of the contributions of agricultural activities to nutrient emissions to the air, rivers and coastal waters at the basin scale. This Chapter provides the model development and baseline that are subsequently used for future assessments in Chapters 3-4.

Future water pollution and required reductions for avoiding coastal eutrophication

Chapter 3 presents new insights on the trends and required reductions in river exports of nutrients to avoid coastal eutrophication in Europe by 2050 under global change. This chapter provides an illustrative example of applying a backcasting approach to identify the required reductions in exports of N and P by the top ten polluted rivers to prevent future coastal eutrophication. First, the river exports of N and P are projected by the MARINA-Nutrients model (from Chapter 2) for 2050 under a baseline scenario combining Shared Socioeconomic Pathway 5 (SSP5, fossil-fueled development) and Representative Concentration Pathway 8.5 (RCP8.5, high-end climate change). Second, the maximum allowable nutrient export levels to avoid coastal eutrophication are calculated by a backcasting approach and using an Indicator for Coastal Eutrophication Potential (ICEP). The ICEP value is set as 0 with an uncertainty range (-1 and +1) to ensure low risks of coastal eutrophication.

The differences between the projected nutrient exports and the maximum allowable nutrient exports in 2050 are identified as the required reductions in nutrient exports by the top ten polluted rivers of Europe.

Results reveal the following insights. Nutrient export to European seas is expected to increase by 13-28% by 2050 under socioeconomic developments and climate change. The application of synthetic fertilizers and manure is expected to contribute by 35% to the river export of N in 2050 relative to the period of 2017-2020. The remainder is mainly from sewage systems (22%), atmospheric deposition over agricultural areas (14%), and leaching of organic matter (10%) from agricultural areas. On the other hand, sewage systems are projected to account for 70% of P export by rivers in 2050. Avoiding future coastal eutrophication requires over 47% less N and up to 77% less P exports individually by the top ten polluted rivers in 2050 relative to the period of 2017-2020. The top ten polluted rivers based on their projected export loads of N are the Danube, Rhine, Wisla, Elbe, Odra, Seine, Tejo, Nemanus, Daugava, Ebro, and export loads of P are the Danube, Rhine, Wisla, Seine, Odra, Tejo, Dnestr, Ebro, Daugava, Pregolya. The basins of these polluted rivers are expected to host 42% of the population by 2050, implying that at least 42% of the population in the study area might be affected by coastal eutrophication in 2050. This chapter demonstrates where and how much nutrient pollution reductions are needed, and hence could support the implementation of effective strategies to prevent coastal eutrophication in European seas in the future.

Synergetic solutions for reducing future air and water pollution with nutrients

Chapter 4 is novel as it is the first attempt to model the effects of using BBFs alone and in combination with other nutrient management strategies on reducing future nutrient emissions to the air and surface waters (rivers and coastal waters) in a spatially explicit way in the EU and the UK. This chapter provides new insights into synergetic solutions that are combined nutrient management strategies for mitigating nutrient emissions to the air, rivers, and coastal waters simultaneously in 2050 under global change. Synergy occurs when a strategy for reducing one emission decreases another. Six new scenarios are developed considering the use of BBFs, increasing nutrient use efficiencies in agricultural land and improving sewage treatments, in particular upgrading the treatment technologies. In two scenarios, REcovered Nitrogen from manURE (ReNuRe, sometimes it is abbreviated as RENURE) products produced by manure processing; and in other four scenarios, Recovered P from treated Sewage Sludge (RPSS) are considered as BBFs to substitute synthetic fertilizers while maintaining the crop yield efficiency. The scenarios are ReNuRe (ReNuRe products), ReNuRe-NUE (ReNuRe products + increased N use efficiency), Pmed (RPSS + medium upgrade of sewage treatments), Pmed-PUE (RPSS + medium upgrade of sewage treatments + increased P use efficiency), Padv (RPSS + advanced upgrade of sewage treatments) and Padv-PUE (RPSS + advanced upgrade of sewage treatments + increased P use efficiency). These scenarios are executed by the MARINA-Nutrients model from Chapter 3.

Results reveal the following insights. Effects of the considered BBFs differ among N and P mitigation potentials and among air and surface water pollution reductions in the future. Adopting ReNuRe products and increasing N use efficiency in agricultural land is found to be a synergistic option to reduce N emissions to the air (30%), and N inputs to rivers (23%) and N export by rivers to coastal waters (20%) of Europe by 2050 compared to the future baseline from Chapter 3. By combining improved sewage treatment, use of RPSS and higher P use efficiency, 68% less P inputs to rivers and 62% less P export by rivers to coastal waters are projected relative to the future baseline. Besides, the RPSS can fulfill the P fertilizer demand of the EU and the UK by 2050. This chapter demonstrates that BBFs are alternatives to synthetic fertilizers as well as promising for reducing air and surface water pollution synergistically in combination with other nutrient management strategies.

5.1.3 Synthesis of the main findings

Agriculture plays an important role in emitting N for most basins in the EU and the UK (Chapter 2, Figure 5.1). For example, 55% of N in rivers in the study area is from agriculture (period of 2017-2020). For P in rivers, sewage systems are responsible for 67% of the emissions (Chapter 2, Figure 5.1). In the future, nutrient export to EU seas is expected to increase by 13-28% under socioeconomic drivers and climate change (Chapter 3, Figure 5.1). To avoid future coastal eutrophication, over 47% of N and up to 77% of P exports by rivers need to be reduced (Chapter 3, Figure 5.1). This is per basin for the top ten polluted rivers based on their nutrient export loads to coastal waters of Europe (Chapter 3). The use of BBFs solely does not reduce nutrient inputs to rivers since the level of total effective nutrient input to agricultural land is assumed to remain the same as in the future baseline scenario (Chapter 4). Therefore, more efforts are needed to reduce inputs of N and P to rivers and their exports to coastal waters. Synergetic solutions should be the combination of using BBFs (ReNuRe and RPSS), increased nutrient use efficiencies and improved treatment of wastewater in cities. Such synergetic solutions are quantified to reduce up to two-thirds of river export of P relative to the future baseline scenario, and N export by 20% (Chapter 4).

On the other hand, the use of BBFs alone helps reduce future N emissions to the air by a total of 22% for 601 basins (Chapter 4). By 2050, it might be possible to reduce N emissions to the air from agriculture by 30% when applying BBFs (ReNuRe products) in combination with higher N use efficiencies in agriculture (Chapter 4, Figure 5.1). This is for the entire study area. Basin-specific reductions differ for N emissions to the air, rivers and coastal waters depending on the contribution of agriculture to N pollution in basins.

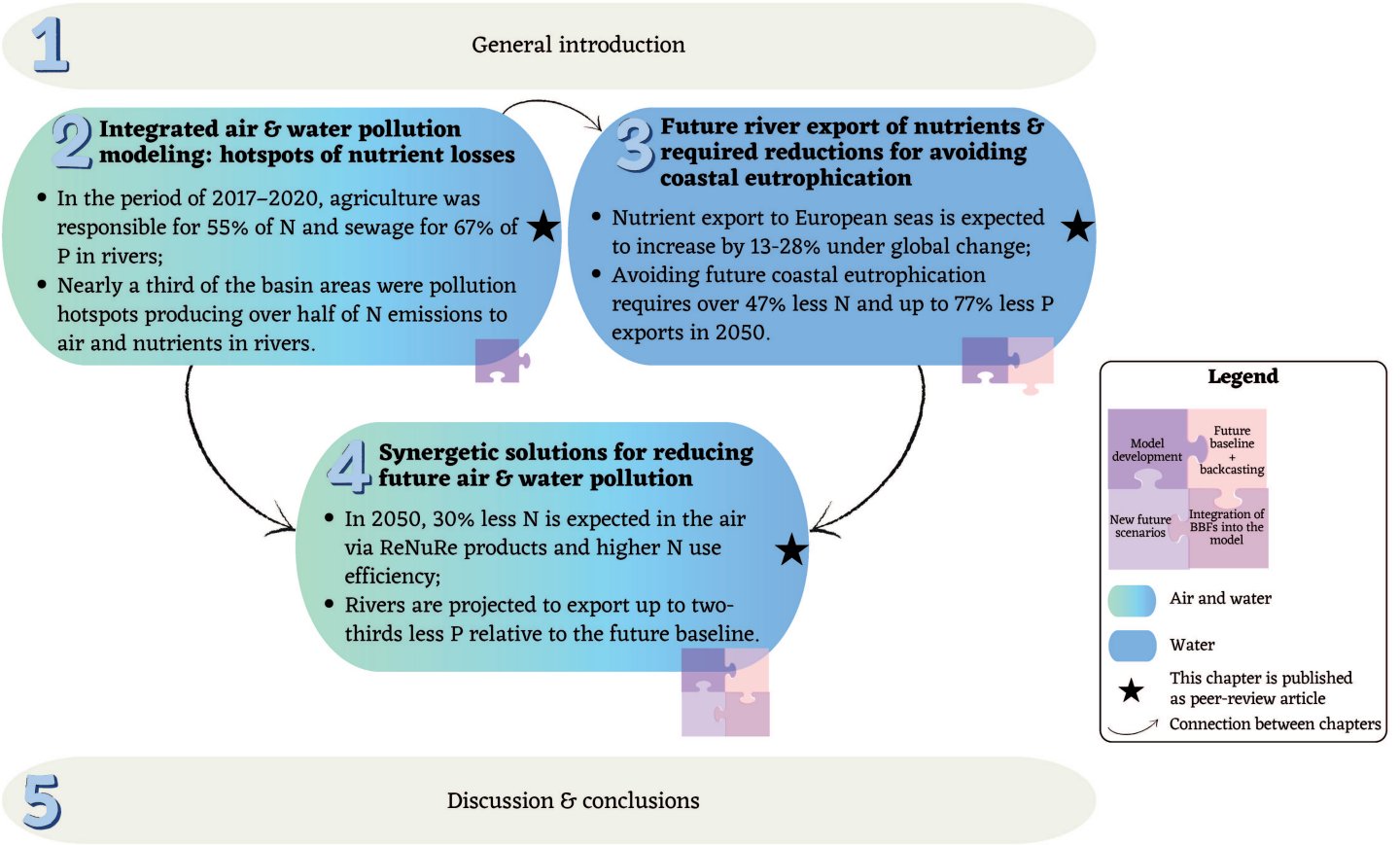


Figure 5.1 Overview of the key findings of the PhD thesis from Chapters 2-4. N is nitrogen, P is phosphorus, and ReNuRe stands for REcovered Nitrogen from manURE.

5.2 Integrated air and water pollution modeling

Section 5.2 is about the model developed in this thesis, and discusses my contribution to the existing models. Section 5.2.1 describes the new MARINA-Nutrients for Europe in comparison with several existing models that are selected based on four criteria: (1) large-scale models, (2) including agricultural sources, (3) including N and/or P, and (4) including air and/or water. Section 5.2.2 summarizes the family of MARINA models and my contribution to it.

5.2.1 New MARINA-Nutrients for Europe (this thesis)

Model development: The new MARINA-Nutrients for Europe is developed in this PhD thesis mainly to quantify nutrient emissions to the air and surface waters (rivers and coastal waters), identify required nutrient reductions and explore the impacts of using BBFs and other nutrient management strategies in the future. This European version of the MARINA-Nutrients is developed for 601 basins including eight sub-basins of the Danube River by soft coupling and improving the approaches of two existing models: MARINA (Strokal et al., 2016; Wang et al., 2020a) and MITERRA-Europe (Velthof et al., 2009). An existing MARINA modelling approach is process-based, deterministic and static for surface waters at the (sub)basin scale. MITERRA-Europe is a deterministic and static model for the emissions of GHG, NH_3 and NO_x to the atmosphere, and losses of N and P to ground- and surface waters from agriculture at an administrative scale. The MITERRA-Europe model uses a mass-balance approach to quantify the fluxes of N and P in agricultural systems. By soft coupling, the outputs of MITERRA-Europe are used as inputs to the MARINA-Nutrients after adjusting the spatial level of detail to the basin and (sub)basin scale (see Chapter 2 for details).

Model characteristics: The new MARINA-Nutrients for Europe integrates agriculture-associated air pollution and simulates the emissions of NH_3, N_2O and NO_x to the air, besides the flows of N and P from land to rivers and by rivers to coastal waters (see the model improvements, below). The model quantifies inputs of dissolved inorganic and organic N (DIN, DON) and P (DIP, DOP) to rivers and their exports by rivers to coastal waters from diffuse (agricultural and non-agricultural) and point (sewage-related) sources. Diffuse sources discharge nutrients from agricultural and non-agricultural land to rivers via runoff. Agricultural diffuse sources include the application of synthetic fertilizers and manure on land, manure deposited on land during grazing, biological N_2 fixation by crops, atmospheric N and P deposition on agricultural land, leaching of organic matter, weathering of P-contained minerals from agricultural areas, and human waste that stays on land from population not connected to sewage systems. Non-agricultural diffuse sources encompass biological N_2 fixation by natural vegetation, atmospheric N deposition on non-agricultural land, leaching of organic matter and weathering of P-contained minerals from non-agricultural areas. Point sources are the effluents of treated municipal wastewater to rivers. Sewage systems collect human waste from urban and rural populations. This collected waste goes to treatment facilities.

Depending on treatment efficiency, some amount of nutrients is discharged to nearby water systems via pipes. The model also accounts for nutrient retention in (e.g., P accumulation in soils) and loss from land (e.g., uptake by crops) before discharging nutrients into rivers, and nutrient retention in (e.g., damming, reservoirs) and loss from rivers (e.g., water removal) before discharging nutrients into coastal waters.

Model improvements: The improvements of the MARINA-Nutrients for Europe relative to the existing MARINA model versions (Strokal et al., 2016; Chen et al., 2019b; Wang et al., 2020a; Wang et al., 2024) are summarized below (see more in Chapters 2 and 4):

- Integration of atmospheric P deposition over agricultural areas from MITERRA-Europe (Chapter 2);
- Integration of NH_3, N_2O and NO_x emissions to the air from animal housing and manure management systems from MITERRA-Europe while accounting for five animal types (dairy cattle, other cattle, pig, poultry, other animals such as sheep and goat), except for NO_x, which is quantified as total from animal housing and manure storage. All the integrated data are processed to the basin scale (Chapter 2);
- Integration of NH_3, N_2O and NO_x emissions to the air from agricultural soils due to the application of synthetic fertilizers and animal manure, grazing, and biological N_2 fixation by crops (only for N_2O) while accounting for variation in crop and soil types in the EU. MITERRA-Europe also accounts for indirect N_2O emissions through ammonia volatilization and N leaching, which are included in N_2O emissions from agricultural soils. All the integrated data are processed to the basin scale (Chapter 2);
- Quantification of human waste that does not go to sewage systems as a land-based diffuse source for the inputs of N and P to rivers (Chapter 2);
- Quantification of N emissions to the air from the production of BBFs, namely ReNuRe, through manure processing (anaerobic digestion and ammonia stripping-scrubbing) and from their application on agricultural soils (Chapter 4);
- Quantification of nutrient inputs to rivers and exports by rivers to coastal waters from the application of two BBFs, ReNuRe and RPSS, on agricultural soils to substitute synthetic fertilizers (Chapter 4).

Model applications: In this thesis, the MARINA-Nutrients for Europe is applied in the recent past (the period of 2017-2020, Chapter 2) and future (2050, Chapter 4). Moreover, the model is applied with a backcasting approach to identify required reductions in nutrient exports by the top ten polluted rivers to avoid coastal eutrophication in Europe by 2050 under global change drivers including socioeconomic developments and climate change (Chapter 3). Furthermore, the model is applied to quantify nutrient losses from the production and application of BBFs to reveal novel insights on the effects of BBFs alone and in combination with other nutrient management strategies in reducing air and surface water pollution in 2050 (Chapter 4).

Comparisons with existing models: Key examples of the existing models include GAINS (Amann et al., 2011) for air, GREEN (Grizzetti et al., 2008) for water, and IMAGE (Stehfest et al., 2014) and MAgPIE (Dietrich et al., 2022) for both air and water. These models have similarities and differences with the MARINA-Nutrients for Europe regarding four aspects (Table 5.1):

- having a large scale,
- including agricultural sources,
- including N and/or P, and
- including air and/or water.

Similar to the model of this thesis, both GAINS and IMAGE are process-based models, yet their designs differ (Table 5.1). GAINS (Greenhouse gas - Air pollution Interactions and Synergies) simulates the global emissions of GHGs, NH_3, NO_x and others, and estimates mitigation potentials and their costs by using external activity data for the macroeconomic and sectoral (e.g., agriculture, energy) developments (Amann et al., 2011; JRC, 2024). GAINS operates for 5-year intervals up to 2070 for the EU (JRC, 2024). IMAGE (Integrated Model to Assess the Global Environment), by contrast, is a global integrated model that is developed to better understand the environmental consequences of human activities (Stehfest et al., 2014). IMAGE simulates the annual emissions of GHGs, NH_3, NO_x, SO_2 and others to the air, and the flows of N and P to surface waters from anthropogenic and natural sources (Stehfest et al., 2014). On the other hand, GREEN (Geospatial Regression Equation for European Nutrient losses) is a statistical water quality model that focuses on nutrient inputs to rivers and seas of Europe by hydrological catchments, mainly from agriculture and sewage systems (Grizzetti et al., 2008; Vigiak et al., 2023) such as the MARINA-Nutrients for Europe (Table 5.1).

Different from the model of this thesis, MAgPIE (Model of Agricultural Production and its Impact on the Environment) is a global partial equilibrium land use model that accounts for various land-based GHG emissions (Popp et al., 2014), and regional N budgets and losses (e.g., leaching, volatilization) from agricultural production (Bodirsky et al., 2014). Both IMAGE and MAgPIE are dynamic integrated models that run at 0.5° resolution and apply scenario analyses to project future emissions to the air and waters under changing climate and socioeconomic conditions up to 2100 (Table 5.1). The MARINA-Nutrients model for Europe, by contrast, runs at basin and sub-basin (for large river basins such as Danube) scales to better capture upstream–downstream interactions in regions with high variability in terms of agriculture and population density. This European version of MARINA-Nutrients for the first time accounts for N emissions to the air from agricultural sources (Table 5.2). Furthermore, the model developed in this thesis differs from the aforementioned models by the inclusion of BBFs and analysis of their effects on future nutrient emissions to the air, rivers and coastal waters.

Table 5.1 Main characteristics of the existing air and/or water pollution models included in this chapter for comparison with the MARINA-Nutrients for Europe (the model of this thesis, in bold). MARINA-Nutrients is short for Models to Assess River Inputs of pollutaNts to seAs for Nutrients. EU is European Union, UK is United Kingdom, NH_3 is ammonia, N_2O is nitrous oxide, NO_x is nitrogen oxides, N is nitrogen, P is phosphorus, DIN is dissolved inorganic N, DON is dissolved organic N, DIP is dissolved inorganic P, DOP is dissolved organic P, GHG is greenhouse gas, SO_2 is sulfur dioxide, BC is black carbon, OC is organic carbon, $PM_{2.5}$ is particulate matter that is 2.5 microns or less in diameter and PM_{10} is those with a diameter of 10 microns or less, NMVOCs is non-methane volatile organic compounds, and NO_3^- is nitrate.

						Pollutants			Sources		
Model name	**Approach**	**Spatial extent**	**Spatial resolution**	**Temporal extent**	**Temporal resolution**	**Air Emissions**	**Water Inputs to rivers**	**River exports**	**Agricultural**	**Non-agricultural**	**Sewage**
MARINA-Nutrients	**Static, uncalibrated**	**EU and the UK**	**Basin**	**Annual**	**2017-2020, 2050**	**NH_3, N_2O, NO_x**	**DIN, DON, DIP, DOP**	**DIN, DON, DIP, DOP**	**Yes**	**Yes**	**Yes (domestic)**
GAINS[a]	Static, uncalibrated	World, EU	10–50 km grid, 1-10 km grid	5-year intervals	1990-2050, to 2070 for the EU	GHG, NH_3, NO_x, SO_2, BC, OC, $PM_{2.5}$, PM_{10}, NMVOCs	None	None	Yes	Yes	No
GREEN[b]	Static, calibrated	Europe	Catchment of 7 km^2 average area	Annual	1990-2050	None	TN, TP	TN, TP	Yes	Yes	Yes (domestic, industrial)
IMAGE[c]	Dynamic, calibrated	World	0.5°x0.5° grid	Annual	1970-2100	GHG, NH_3, NO_x, SO_2, BC, OC, NMVOCs	TN, TP	TN, TP	Yes	Yes	Yes (domestic, industrial)
MAgPIE[d]	Dynamic, calibrated	World	0.5°x0.5° grid	5-year intervals	1995-2100	GHG, NH_3, NO_x	NO_3^- leaching	None	Yes	No	Yes (domestic)

a (Amann et al., 2011; JRC, 2024).
b (Grizzetti et al., 2008; Grizzetti et al., 2025).
c (Stehfest et al., 2014).
d (Bodirsky et al., 2014; Dietrich et al., 2022).

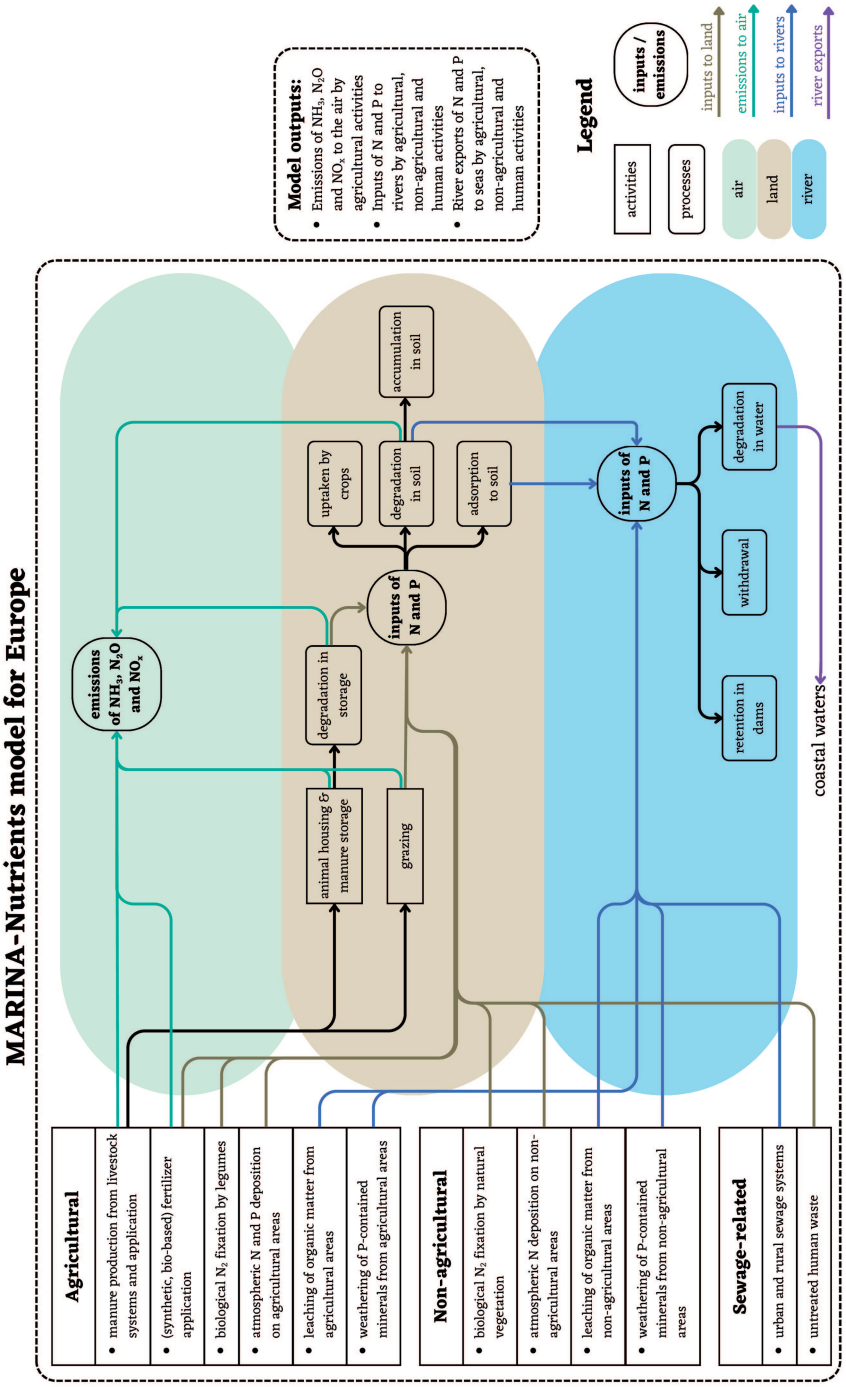


Figure 5.2 Overview of the new MARINA-Nutrients for Europe developed and applied in this thesis (Chapters 2-4). MARINA-Nutrients is Model to Assess River Inputs of polluta**N**ts to se**A**s for Nutrients, N_2 is dinitrogen, N is nitrogen, P is phosphorus, NH_3 is ammonia, N_2O is nitrous oxide, and NO_x is nitrogen oxides. The MARINA-Nutrients for Europe does not account for atmospheric N deposition on waters (see more in Chapter 2 and Section 5.2).

5.2.2 My contribution to the family of MARINA models

History of MARINA models: The first version of MARINA was developed for China and focused on the river exports of nutrients to coastal waters (Strokal et al., 2016). Since then this version has been called MARINA-Nutrients, and updated and applied for rivers and/or coastal waters at various scales such as China (Chen et al., 2019b; Wang et al., 2020a; Chen et al., 2022) and world (Wang et al., 2024). The model has also been applied to quantify the nutrient inputs to lakes (Wang et al., 2019; Yang et al., 2019). Beyond nutrients, the family of the MARINA models has been developed over more than 10 years to address complex water-quality issues worldwide (Micella et al., 2024c). The family includes many versions that have been published over 25 papers (Table 5.2), and several are for nutrients (Table 1.2). During my PhD, the family of the MARINA models has extended including antibiotics (Zhang et al., 2025), plastics (Strokal et al., 2022; Li et al., 2023), and multi-pollutants (e.g., chemicals, pathogens, emerging contaminants) (Strokal et al., 2021; Li et al., 2022b; Strokal et al., 2023; Bak et al., 2024; Li et al., 2024; Micella et al., 2024a; Micella et al., 2024b; Zhang et al., 2024b). The MARINA models operate at the sub-basin scale with a lumped, uncalibrated approach that enables the integration of diverse pollutant sources and pathways, and future projections (Strokal et al., 2019).

My contribution to the family of MARINA models (Table 5.2): In this PhD, a new version of MARINA-Nutrients is developed for 601 basins in the EU and the UK for air and surface waters (Chapters 2-4). This version differs from other versions of MARINA in several aspects (Table 5.2). First of all, the existing MARINA models are limited to water pollution and do not include emissions to the air. The new MARINA-Nutrients for Europe quantifies annual emissions of NH_3, N_2O and NO_x from crop and livestock production. Second, most applications of MARINA are for the basins of China and the globe, and the existing models have never been applied to the EU level separately. Third, some global applications of MARINA for multiple pollutants including nutrients are limited to a specific pollution source. For example, Li et al. (2022b) simulate the inputs of nutrients and a pathogen to rivers from global livestock production, while Strokal et al. (2021) focusing on the impacts of urbanization on the inputs of nutrients, microplastics, a chemical and a pathogen to rivers worldwide. The MARINA-Nutrients for Europe considers agricultural, non-agricultural and sewage-related sources. Fourth, the versions of MARINA for China and the globe include animal manure discharge as a point source (Strokal et al., 2016; Chen et al., 2019b; Wang et al., 2020a; Chen et al., 2022; Li et al., 2022b; Micella et al., 2024a; Micella et al., 2024b; Bak et al., 2025). The MARINA-Nutrients for Europe does not consider this source, as it is not the case for the EU and the UK.

Table 5.2 Main characteristics of MARINA models and their applications that are relevant to this thesis, along with my contribution (highlighted in bold). MARINA is short for Models to Assess River Inputs of pollutaNts to seAs. The first version of the MARINA-Nutrients model (China 1.0) forms the basis for the model development in this PhD research. For the versions of MARINA-Multi that include multiple pollutants, only nutrient forms and their sources are given. EU is the European Union, UK is the United Kingdom, NH_3 is ammonia, N_2O is nitrous oxide, NO_x is nitrogen oxides, N is nitrogen, P is phosphorus, DIN is dissolved inorganic N, DON is dissolved organic N, DIP is dissolved inorganic P, and DOP is dissolved organic P.

Model version	Spatial extent	Spatial resolution	Temporal extent	Temporal resolution	Pollutants: Air	Pollutants: Water		Sources: Agricultural production		Sources: Non-agricultural (diffuse)	Sources: Sewage (point)
					Emissions	Inputs to rivers	River exports	Crop (diffuse)	Livestock (diffuse & point)		
	Similarities				**Differences**						
MARINA-Nutrients for Europe	**EU and the UK**	**Basin, sub-basin**	**Annual**	**2017-2020, 2050**	**NH_3, N_2O, NO_x**	**DIN, DON, DIP, DOP**	**DIN, DON, DIP, DOP**	**Yes**	**Yes (diffuse)**	**Yes**	**Yes**
MARINA-Nutrients (Global 1.0)[a]	World	Sub-basin	Annual	2010, 2050	None	DIN, DON	DIN, DON	Yes	Yes (diffuse)	Yes	Yes
MARINA-Multi (Global 1.0)[b]	World	Sub-basin	Annual	2010, 2050, 2100	None	DIN, DON, DIP, DOP	No	No	No	No	Yes
MARINA-Multi (Global 2.0)[c]	World	Sub-basin	Annual	2010	None	DIN, DON, DIP, DOP	No	No	Yes	No	No
MARINA-Multi (Global 4.0)[d]	World	Sub-basin	Annual	2010, 2050, 2100	None	DIN, DON, DIP, DOP	DIN, DON, DIP, DOP	Yes	Yes	Yes	Yes

a (Wang et al., 2024).
b (Strokal et al., 2021).
c (Li et al., 2022b).
d (Micella et al., 2024a; Micella et al., 2024b; Bak et al., 2025).

5.3 Comparison of thesis results with other modeling studies for Europe

Section 5.3 discusses the results of this thesis in comparison with those of other modeling studies focusing on nutrient emissions to the air and/or surface waters (rivers and coastal waters) in Europe.

Chapter 2 focuses on the contribution of agriculture to air, river, and coastal water pollution simultaneously by nutrient emissions, and hotspots of air and/or water pollution across the EU and the UK basins (Ural-Janssen et al., 2023). The water pollution hotspots based on nutrient inputs to rivers for recent past (the period of 2017-2020) mainly in western and central Europe coincide with other modeling studies (de Vries et al., 2021; Li et al., 2022b; Micella et al., 2024a). The results of the MARINA-Nutrients model for Europe show that 2.7 Tg yr^{-1} of total dissolved N (TDN) and 0.11 Tg yr^{-1} of total dissolved P (TDP) were exported by rivers to European seas for the period of 2017-2020, comparable to the results of GREEN (Grizzetti et al., 2021). The GREEN model simulated the loads of total N (TN) and total P (TP) to European seas by 3.3-4.1 Tg N yr^{-1} and 0.26-0.30 Tg P yr^{-1}, respectively, for the period of 2005-2012 (Grizzetti et al., 2021). The differences arise mainly from considering the dissolved (this thesis) versus total (GREEN) nutrients and different periods, as the nutrient exports have decreased due to the policy measures (Grizzetti et al., 2021; Vigiak et al., 2023). On the other hand, the simulated emissions of NH_3, N_2O and NO_x to the air are comparable with other modeling studies for Europe (Bouwman et al., 2006; de Vries et al., 2010; de Vries et al., 2011). The differences could be associated with different emission factors (EFs) used and simulation years.

Chapter 3 focuses on future trends of river exports of nutrients to coastal waters and required reductions to avoid coastal eutrophication in Europe (Ural-Janssen et al., 2024). In general, the modeled trends of the total river export of nutrients by the EU and the UK basins in 2050 are in line with other modeling studies including Europe. Many coastal waters in Europe are expected to be affected by an increase in river nutrient exports, which is in line with the projected increase in global river exports of N and P by 2050 based on the SSP5-RCP8.5 (Beusen et al., 2022; Bak et al., 2025). The increased nutrient transport to coastal waters by 2050 will be mainly caused by urbanization (e.g., 71% more river export of TDN from sewage systems) as a result of nutrient inputs to rivers from sewage systems. With the increased urbanization and moderate improvements of sewage treatment systems in SSP5, a higher amount of wastewater will be produced with moderately improved treatment efficiencies (Strokal et al., 2021). There will be a shift from synthetic fertilizers to animal manure and that is in line with the SSP5-RCP8.5 scenario assumptions (e.g., reducing synthetic fertilizer application by recycling nutrients from human urine, Beusen et al. (2022)).

Chapter 4 focuses on reducing future nutrient emissions to the air, rivers and coastal waters of Europe through the application of BBFs (ReNuRe and RPSS) and other nutrient management strategies (Ural-Janssen et al., 2025). The total N emissions to the air are projected to reduce up to 19% in 2050 compared to the period of 2017-2020 when ReNuRe products are combined with higher NUE. On the other hand, Rashid et al. (2025) found that when ammonium sulfate is applied as BBF with equivalent total N input, gaseous N losses were comparable to those from conventional fertilizers. This could be explained by the choices of EFs for the production and application of BBFs. They used the Daisy model to simulate N dynamics, including gaseous losses (e.g., NH_3, N_2O) based on fertilizer type and environmental conditions across ten European cropping systems (Rashid et al., 2025). In this thesis, the ReNuRe products are assumed to be produced by anaerobic digestion followed by ammonia stripping-scrubbing to prevent gaseous losses from manure storage. Moreover, lower EF for the application of BBFs than synthetic fertilizers and manure is considered regardless of application method and environmental conditions (see more in Chapter 4). Furthermore, the Padv-PUE scenario, which considers using the RPSS as BBFs in combination with improving wastewater treatment plants (WWTPs) and increasing P use efficiency (PUE) in agricultural land is expected to reduce P inputs to rivers by 58% and P exports by rivers to coastal waters by 51% in 2050 relative to the period of 2017-2020. Similarly, Pistocchi et al. (2023) show that stricter rules are required for the collection and treatment of European wastewaters in order to further reduce nutrient emissions from domestic wastewater treatment systems. On the other hand, the projected river exports of nutrients by the MARINA-Nutrients for Europe have been compared with the results of GREEN for model intercomparisons to better understand uncertainties in the effects of water pollution reduction interventions for future nutrient loadings into European seas (Strokal et al., 2025). The comparison indicates promising reduction potentials for nutrient loadings in most coastal waters of Europe (Strokal et al., 2025).

5.4 Reflection on strengths, weaknesses, opportunities, and threats

Below, the strengths (Section 5.4.1), weaknesses (Section 5.4.2), opportunities (Section 5.4.3), and threats (Section 5.4.4) of this thesis are discussed following the SWOT approach (Stewart et al., 1965) (Figure 5.3). This reflection includes not only modeling approaches but also the way of integration, evaluation and how results are put into a broader context.

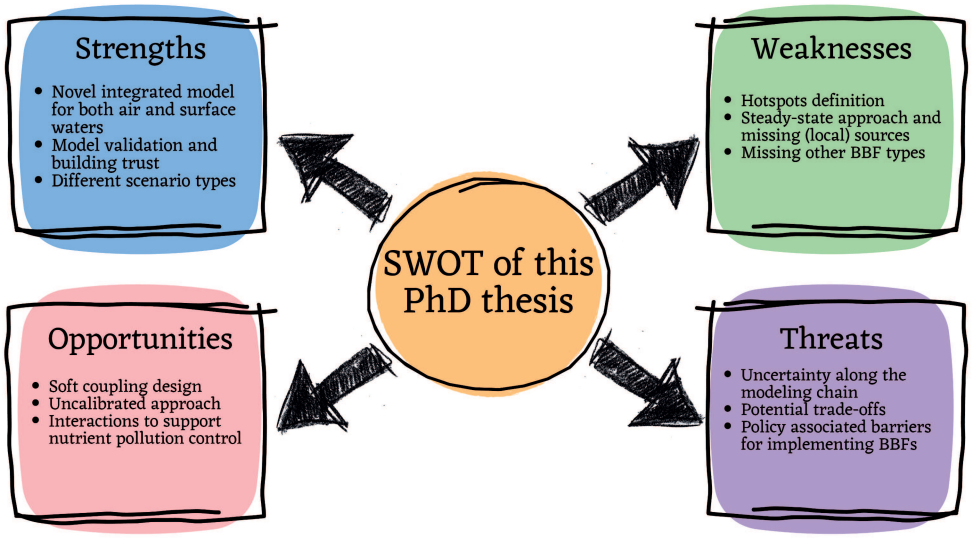


Figure 5.3 Overview of the main strengths, weaknesses, opportunities and threats (SWOT) of this PhD thesis. BBF is short for a bio-based fertilizer and MITERRA-Europe is the Integrated Modeling of Nutrient Flows at Regional Level for Europe.

5.4.1 Strengths

The three main strengths of this thesis include (1) presenting a novel integrated model to quantify the effects of BBFs on nutrient emissions to the air and surface waters (Chapters 2 and 4), (2) model validation and building trust approaches (Chapters 2-4), and (3) different scenario types (Chapters 3-4, Figure 5.3).

- ***Novel integrated model for both air and surface waters***

Modeling environmental effects of BBFs: This PhD study is the first attempt to model the effects of using BBFs alone and in combination with other nutrient management strategies on reducing nutrient emissions to the air, rivers and coastal waters in 2050 under socioeconomic developments and climate change. The model accounts explicitly for the spatial drivers of this pollution across the EU and the UK basins. Nutrient pollution is not an isolated environmental issue but part of a web of interactions across land, air and water systems. Management strategies or policies focusing on one receiving environment (e.g., air, water) and/or one pollutant (e.g., nutrient form) may not be effective or sufficient to reduce nutrient pollution (Kanter et al., 2020), and may cause issues such as pollution swapping (Velthof et al., 2024). Thus, a holistic governance, multisector approach and strategies are needed to reduce this pollution synergistically (Möhring et al., 2023), while mitigating potential trade-offs from separate management strategies and policies (Kanter et al., 2020). Models focusing either on N or P, and either air or water pollution cannot provide a comprehensive assessment of synergistic or antagonistic effects of single or integrated mitigation strategies for nutrient emissions. On the other hand, integrated models help explore strategies that can

reduce nutrient emissions to the air and waters simultaneously, and thus help avoid trade-offs such as pollution swapping (Velthof et al., 2024). Therefore, the MARINA-Nutrients model developed in this thesis allows exploring the effects of not only BBFs, but also other nutrient management strategies to reduce nutrient emissions to the air, rivers, and coastal waters of Europe simultaneously (see more in Chapter 4).

The integrated MARINA-Nutrients for Europe: It is a lumped process-based model that runs at the basin and sub-basin (for larger rivers such as the Danube River) resolution. Strokal et al. (2019) delineated 10,226 world basins and sub-basins based on the direction of water flow using a land mask and water bodies from a hydrological Variable Infiltration Capacity (VIC) model. The basin resolution offers two main advantages. First, by operating at the basin scale, the MARINA-Nutrients model avoids the heavy data and computational demands of more detailed resolutions, such as 0.5° grids (e.g., IMAGE, Stehfest et al. (2014) and MAgPIE, Bodirsky et al. (2014)). Yet, by operating at the sub-basin scale for large river basins (the Danube basin in the thesis study area), the MARINA-Nutrients for Europe links upstream human activities and downstream water pollution, while accounting for sub-basin contributions to this pollution. This supports spatially explicit analysis of how agriculture and sewage systems contribute to nutrient emissions to the air, and losses to rivers and coastal waters across the EU and the UK, which is an important objective of this thesis (Chapters 2 and 4). Second, the (sub)basin resolution is particularly useful for European assessments, since the Water Framework Directive requests periodic River Basin Management Plans from the Member States (EC, 2000). Therefore, the model of this thesis supports the development of these plans by providing a comprehensive assessment of air and surface water pollution associated with nutrient emissions as well as mitigation strategies. Last but not least, the consistency across scales provides comparability within the MARINA model family, offering opportunities for model intercomparison purposes to build trust in model results across scales and pollutants (Table 1.2 and Table 5.2).

o Model validation and building trust

Model validation: Validation refers to the process of evaluating how well the outputs of a model align with existing observations (Moriasi et al., 2007). In this thesis, the MARINA-Nutrients for Europe is developed by soft linking the two existing models, MARINA and MITERRA-Europe(see more in Section 5.2, and Chapters 2 and 4). The MITERRA-Europe modeling approach for air emissions has been evaluated in earlier studies (Velthof et al., 2009; de Vries et al., 2011; Kros et al., 2012; Hou et al., 2017). The model of this thesis is validated for the river exports of N and P at the river mouths (Ural-Janssen et al., 2023). This is done for nutrient exports by 58 rivers in Europe due to the monitoring data availability. The observational data are collected from the Global Environment Monitoring System for Freshwater Database (GEMStat) (UNEP, 2022) and from the literature (Mayorga et al., 2010; Hesse & Krysanova, 2016; Ylöstalo et al., 2016; Cozzi et al., 2018; Räike et al., 2018; Vybernaite-Lubiene et al., 2018; Friedland et al., 2019; Räike et al., 2020) for various years

(e.g., 1995, 2001) and periods (between 1980 and 2017). The monitoring locations closest to the river mouths are chosen to compare the loads of nutrient exports at the river mouths. The observational data including measured concentrations (mg l^{-1}) are converted to observed loads (kg yr^{-1}) and yields (kg km^{-2} yr^{-1}) to compare the simulated loads of nutrient exports at the river mouths. To evaluate model performance, statistical indicators widely used in hydrological and water quality model assessments, namely the Pearson's coefficient of determination (R_P^2, 0–1), Nash-Sutcliffe efficiency (R_{NSE}^2, 0–1), and the root mean square error to standard deviation ratio (R_{RSR}^2) are applied following (Moriasi et al., 2015). The model results for dissolved inorganic N (DIN), dissolved organic N (DON), total dissolved N (TDN), dissolved inorganic P (DIP), dissolved organic P (DOP), and total dissolved P (TDP) are validated. The MARINA-Nutrients for Europe performed well for river exports of nutrients to coastal waters for the period of 2017-2020 based on the aforementioned indicators: R_P^2 = 0.85, R_{NSE}^2 = 0.85 and R_{RSR}^2 = 0.38 (see more in Chapter 2). Not only validation is performed, but also the "building trust" approach is applied to evaluate the model developed in this thesis.

Building trust: This European version of MARINA-Nutrients is largely based on modeling approaches for nutrients that have been evaluated in earlier studies (see references in this paragraph). The model builds on the family of MARINA models, which have a lot of scientific trust in model development and evaluation for nutrients e.g., nationally (China) (Strokal et al., 2016; Chen et al., 2019b; Wang et al., 2020a; Chen et al., 2022) and globally (Strokal et al., 2021; Li et al., 2022b; Bak et al., 2024; Micella et al., 2024a). In line with the development of the MARINA models, the "building trust" approach with six strategies was developed by Strokal (2016) to evaluate these large-scale models, and extended to 13 strategies by Strokal et al. (2024). This approach aims to address uncertainties in model inputs, approaches, and outputs by covering the entire model development chain to evaluate large-scale water quality models (Strokal et al., 2024). The approach has been widely used for the applications of other MARINA versions when the observational data are limited or scarce (Strokal et al., 2021; Li et al., 2022b; Micella et al., 2024a; Micella et al., 2024b). In this thesis, the three strategies of the "building trust" approach are applied to evaluate the plausibility of the modeling approaches and results of the MARINA-Nutrients for Europe. These are the comparisons of model outputs and model trends among models to build trust in model results, as well as expert knowledge to verify some of the model parameters (see more in Chapters 2-4 and their supplementary materials: S2-S4) (Strokal et al., 2024). Validation and comparisons of model outputs with the results of other models are applied in Chapter 2. Comparisons of model trends with other studies are applied in Chapters 3-4, while expert knowledge is consulted during model development in Chapter 4 to verify some of the model parameters (e.g., EFs).

o *Different scenario types*

Scenarios: They are widely applied to explore alternative futures, while bridging science and policy through the assessment of effective solutions for environmental issues. Types of scenarios include predictive (e.g., trend or what-if), explorative (e.g., framed or unframed), and normative (e.g., preserving or transforming), depending on purpose (Börjeson et al., 2006; Maier et al., 2016) (see more in Chapter 1). In this thesis, examples of all three types of scenarios are applied (Chapters 3-4).

Examples from this thesis: In Chapter 3, the future baseline scenario is developed by the combination of SSP5-RCP8.5 and applied as an explorative framed scenario. Moreover, a backcasting approach is used for the identification of the required reductions in nutrient export by the top ten polluted rivers to avoid future coastal eutrophication in Europe under global change, which is an example of normative transforming scenarios. The backcasting approach includes quantification backwards from a desired end-point (e.g., target, which is ICEP at zero in this thesis) to the present mainly to determine the possible measures required to reach the desired point (Gomi et al., 2011; Li et al., 2019). On the other hand, in Chapter 4, six new scenarios are applied to predict "what will happen?", in particular to project the effects of BBFs and other nutrient management strategies on future nutrient emissions to the air, rivers and coastal waters of Europe, which is an example of predictive what-if scenarios. Applying these various scenario types demonstrates the capacity of the new MARINA-Nutrients model for future projections as well as the comprehensiveness of this PhD research.

5.4.2 Weaknesses

The three main weaknesses of this thesis include (1) a limited definition of hotspots towards impact (Chapter 2), (2) a steady-state approach and missing (local) sources (Chapters 2-4), and (3) missing other BBF types (Chapters 4, Figure 5.3).

o *Hotspots definition*

Definitions in other studies: Hotspot analyses help prioritize the areas of concern in mitigating air and/or water pollution as well as provide opportunities for targeting interventions. This is because there are various definitions of hotspots depending on the research question. Some studies used fixed thresholds, such as safe N levels within the planetary boundaries (Schulte-Uebbing et al., 2022). Other studies used impact-based thresholds depending on the impacts of water pollution on ecosystems and human health (Bouwman et al., 2024). In this case, if the simulated levels of nutrients exceed the threshold (non-effect) concentrations, hotspots are identified (Bouwman et al., 2024). Large-scale studies often applied a quantile-based approach and define hotspots as the areas with pollutant losses above the 75th percentile (the top 25%) (Wang et al., 2018; Micella et al., 2024a).

Definitions in this thesis: Hotspots are defined rather simply as basins with relatively higher pollution levels than surrounding basins following the approaches of Wang et al. (2018) and Li et al. (2022b). The aim was to identify hotspots for air pollution, surface water pollution, and both air and surface water pollution in Europe in a spatially explicit way (basins). Air pollution hotspots are defined as the top 20% of basins with the highest total N emission (the sum of NH_3, N_2O and NO_x) to the air. Water pollution hotspots are defined as the top 20% of basins with the highest inputs of total dissolved N (TDN) and/or total dissolved P (TDP) to rivers. Air and water pollution hotspots represent the top 20% of basins with the highest total N emission to the air and TDN and/or TDP inputs to rivers or all three. This hotspot definition has limitations. For instance, this approach does not account for pollution trends over time (Strokal et al., 2021; Micella et al., under review), nor for human health and/or ecological impacts (de Vries et al., 2021; Bouwman et al., 2024). Despite these limitations, I believe the approach of this thesis is suitable for the purpose, which focuses on hotspots of nutrient emissions to the air and water in Europe. Still, it should be interpreted with care and not for impact analyses of human and environmental health. The approach of this PhD thesis should be used as an indication of the identified highly polluted basins compared to other basins. This helps prioritize the focus of environmental policies for action today. Impact-associated pollution hotspots could be applied in future studies using the modeling tool that this PhD offers.

o *Steady-state approach and missing (local) sources*

Steady-state approach: To my knowledge, steady-state and dynamic models serve different purposes in air and water pollution research. Steady-state models such as GAINS and GREEN are often used for large-scale scenario analysis (Gomez-Sanabria et al., 2022; Grizzetti et al., 2025), while dynamic models such as Daisy can capture short-term variability (Abrahamsen & Hansen, 2000; Rashid et al., 2025). This PhD thesis focuses on large-scale assessments of basin-specific nutrient emissions to the air, and surface waters and scenarios. Therefore, the MARINA-Nutrients for Europe is developed as a steady-state model, suitable for the scale (601 basins), air and water pollutants studied, and the future scenario applications. However, there are two main limitations of the steady-state approach. First, the MARINA-Nutrients for Europe cannot consider legacy processes such as the accumulation of N and P in soils (biogeochemical legacy) (Beusen et al., 2022; Nguyen et al., 2022) and groundwater N storage (hydrological legacy) (Nguyen et al., 2022; Liu et al., 2024). These legacy processes influence the flows of nutrients from land to rivers over time (Nguyen et al., 2022). For instance, even when the application of fertilizers is reduced or stopped, legacy P could continue to leach from agricultural soils into rivers for years (Wang et al., 2020b). As a result, the simulated P inputs to rivers from agriculture may be overestimated in high-input areas or underestimated in low-input areas, and management strategies may show delayed effects. Nevertheless, this limitation is less critical in this PhD study as often sewage systems dominate inputs of DIP and river exports of DIP in European basins (Chapters 2-4). Second, the MARINA-Nutrients for Europe runs on an annual time scale and takes

a long-term perspective (up to 2050) while ignoring seasonal or intra-annual trends (e.g., temperature, rainfall) that could be important for nutrient emissions to the air (Ge et al., 2020) and surface waters (Bodirsky et al., 2014). A few attempts have been made to include seasonality into the MARINA models for river exports of N by an individual basin (Chen et al., 2019a) and lakes (Yang et al., 2021). The MARINA-Nutrients of this PhD research also includes N emissions to the air, and thus more time and effort would be needed to include seasonality. Future versions of the MARINA-Nutrients for Europe could build on this study and enhance intra-annual variability in air and surface water pollution.

Missing (local) sources of nitrogen emissions to the air: In this thesis, NH_3, N_2O and NO_x emissions to the air from agricultural sources such as animal housing, manure management systems, and agricultural soils are considered and simulated by the MARINA-Nutrients model. However, other sources also contribute to these emissions such as energy, industry, waste and wastewater handling, transport (EEA, 2024a; 2025a), and inland waters (lakes, rivers, reservoirs) (Zheng et al., 2022; Tian et al., 2024). The missing sources only account for a minor share of NH_3 emissions by the EU, as the agricultural sector accounts for 93% of total NH_3 emissions in 2022 (EEA, 2024b). On the other hand, some of the missing sources are major contributors to NO_x emissions (EEA, 2024a; 2025a) (see more in Chapter 1). Nevertheless, considering the agriculture-associated N emissions to the air serves the research objective focusing on the effects of using BBFs on reducing air and surface water pollution across the EU in combination with other nutrient management strategies (see Section 5.1 and Table S5.1). This all justifies my choice of the pollution sources. Future research can build on this thesis by adding more sources of N emissions to mitigate air pollution comprehensively.

Missing (local) sources of nutrient emissions to surface waters: The MARINA-Nutrients model includes the most important sources of nutrient inputs to rivers and nutrient exports by rivers to coastal waters, but ignores a few, which are often locally or regionally specific. For instance, the model does not account for nutrient inputs to rivers from industrial wastewater treatment systems and aquaculture, nor for direct atmospheric N deposition on waters. Vigiak et al. (2023) has quantified the loads of N and P to European seas from industrial and domestic wastewater treatment systems as 27% and 43% of the total loads in 1990-2018, respectively, indicating that industrial inputs contribute < 1% of total nutrient inputs to European freshwaters. Moreover, aquaculture contributes to nutrient inputs to freshwaters through the discharge of effluent including fertilizers and solid waste (e.g., fish feed and feces) (Ahmad et al., 2021). On the other hand, these missing sources account for a relatively minor share in surface water pollution compared to agriculture globally (Beusen et al., 2022) and in Europe (Vigiak et al., 2023), while they can be significant for some local situations (e.g., heavy industrial activities).

Missing sources of particulate nutrients in surface waters: The MARINA-Nutrients model does not quantify the river exports of particulate nutrient forms as the sources of surface

water pollution. Mayorga et al. (2010) simulated the river export of particulate N (PN) and particulate P (PP) as 20% of total N and 50% of total P, respectively, for Europe in 2000. This means that water pollution levels might likely be underestimated, especially for areas with a lot of erosion processes (e.g., Italy, Panagos et al. (2022)). Nevertheless, particulate nutrients are usually less bioavailable compared to dissolved forms. This PhD thesis focuses on reducing the risk of future coastal eutrophication (Chapter 3), which is largely influenced by bioavailable nutrients and silica (Billen & Garnier, 2007). Moreover, Chapter 4 of this thesis analyzes the effects of replacing synthetic fertilizers with BBFs, which contribute largely to bioavailable nutrients. Thus, it could be argued that missing (local) sources of nutrient emissions and ignoring particulate nutrients affect our results and conclusions to only a limited extent, and do not change the main messages. Yet, future studies may need to include particulate nutrient forms as the sources of water pollution for a comprehensive analysis of eutrophication issues.

- ***Missing other BBF types***

For N: This PhD study shows the environmental effects of implementing BBFs – not all, but it is the first step indicating that BBFs are promising options, but should be combined with other management strategies. However, this thesis is limited to two types of BBFs, namely ReNuRe and RPSS (Chapter 4). There are many other types of BBFs (Wester-Larsen et al., 2022; Müller et al., 2024; Jasiński et al., 2026). However, these two selected types of BBFs are produced through the processing of two large organic waste groups: animal manure (Velthof et al., 2024) and sewage sludge (see more in Chapter 1). In the EU, the livestock dense regions have issues with atmospheric N deposition in Natura 2000 domains (Vingerhoets et al., 2023b), and with manure surplus, which could turn this excess organic waste into an opportunity to produce, e.g., ReNuRe fertilizers, and thus reducing the dependence on external fertilizers (Sigurnjak et al., 2019; Huygens et al., 2020; Vingerhoets et al., 2024). ReNuRe products produced from manure processing are promising alternatives to synthetic N fertilizers (Ural-Janssen et al., 2025).

For P: Moreover, high P recovery from municipal wastewater treatment systems could be an opportunity to minimize the dependence of non-renewable external P sources to produce P fertilizers (Huygens et al., 2019; Vingerhoets et al., 2023b; Ural-Janssen et al., 2025). The results of Chapter 4 demonstrate that recovered P from treated sewage sludge (RPSS) could fulfill the P fertilizer need in Europe for 2050 under global change. Thus, the choices of BBFs in this thesis also present opportunities for the two environmental burdens of Europe and support decision-makers for the adoption of these products while contributing to reducing future air and surface water pollution associated with nutrient emissions.

5.4.3 Opportunities

The three main opportunities of this thesis for modeling and mitigation of nutrient pollution include (1) a soft-coupling design of the MARINA-Nutrients model to facilitate model

development (Chapters 2-4), (2) an uncalibrated approach to facilitate future analysis of nutrient pollution (Chapters 2-4), and (3) interactions to support nutrient pollution control (Chapters 2-4) (Figure 5.3).

o Soft-coupling design

Approach: In this thesis, the MITERRA-Europe model (Velthof et al., 2009; Duan et al., 2025) is soft-coupled to the MARINA-Nutrients model (Strokal et al., 2016; Wang et al., 2020a) by which two models interact through data exchange while running individually. The agriculture-associated model inputs and N emissions to the air are derived from the MITERRA-Europe for the recent past (2017) (Duan et al., 2021). Simply, this data from MITERRA-Europe are used into the MARINA-Nutrients after adjusting the spatial level of detail from regional (NUTS2) to the basin scale. This soft-coupling design provides three major opportunities.

Opportunity 1: soft coupling enhances efficiency in the quantification of spatial variabilities in sources by enabling detailed analysis of the major sources of nutrient emissions to air and surface waters in Europe (Chapters 2-4). The MITERRA-Europe model accounts for nutrient balances in crop and livestock production based on various animal, crop and soil types, manure management (e.g., stable type) and application practices (Duan et al., 2021; Duan et al., 2025). Thus, the integration of detailed model inputs and emissions of NH_3, N_2O and NO_x to the air strengthens the European version of MARINA-Nutrients to simulate N emissions to the air, inputs of nutrients to rivers and their exports by rivers to coastal waters from agriculture. Consequently, soft coupling makes this new MARINA-Nutrients suitable for this PhD research (see Section 5.1 and Table S5.1).

Opportunity 2: the MARINA-Nutrients model combines the biogeophysical (basin) and administrative (NUTS2) scales by coupling with MITERRA-Europe. The information from MITERRA-Europe is aggregated from NUTS2 to the basin scale via 0.5° x 0.5° grids (see more in Chapter 2 and S2). Therefore, the model provides quantitative information on the levels of nutrient emissions to the air and surface waters at the biogeophysical scale, while the major causes of the emissions are processed from the administrative scale where policy decisions could be made.

Opportunity 3: soft coupling provides flexibility and adaptability. This enables the MARINA-Nutrients to explore the effects of other BBFs and nutrient management strategies on future nutrient emissions to the air, rivers and coastal waters without recalibrating the entire system. Therefore, the model of this thesis offers further opportunities for future applications and scenario analysis.

o *Uncalibrated approach*

Approach: To my knowledge, calibrated and uncalibrated models often serve different purposes. Calibrated models are promising tools for local and regional analyses where observational data allow better representation of site-specific conditions, and offer valuable details on local processes (ten Hoeve et al., 2018; Rydgård et al., 2024), and better simulation of nutrient fluxes (Muntwyler et al., 2024; Rashid et al., 2025). Calibrated models are often used to increase an understanding of certain processes in nutrient pollution and require a lot of observational data for calibration. Such models are also often applied to the historical and present years. In contrast, uncalibrated models such as MARINA-Nutrients of this PhD thesis provide a transparent, process-based framework for large-scale assessments and often aim for future analyses (Strokal et al., 2016; Wang et al., 2020a; Ural-Janssen et al., 2023). Such uncalibrated models could be used to understand sources of environmental pollution at a larger scale (especially in data-poor areas), develop various scenarios and explore the effects of solutions. Building trust and validation are essential for uncalibrated models. Although the MARINA-Nutrients model of this PhD thesis is an uncalibrated model, similar to several other MARINA models, it has been carefully validated against both observational and independent studies, indicating a good performance for river export of nutrients to coastal waters (R_P^2 = 0.85, R_{NSE}^2 = 0.85 and R_{RSR}^2 = 0.38, see more in Chapter 2). I highlight two main opportunities of using an uncalibrated approach for nutrient pollution modeling.

Opportunity 1: The uncalibrated approach of MARINA models (existing versions and of this PhD thesis) enables consistent application across pollutants, regions, and scenarios (Li et al., 2024; Micella et al., 2024b; Wang et al., 2024; Ural-Janssen et al., 2025). Moreover, the uncalibrated design facilitates soft coupling with other models such as NUtrient flows in Food chains, Environment and Resources (NUFER) to improve modeling of nutrient flows on land (Wang et al., 2020a), and MAgPIE to assess future global clean-water scarcity under climate and socio-economic changes (Wang et al., 2024). In this thesis, the European version of MARINA-Nutrients is soft-coupled with MITERRA-Europe to improve modeling of nutrient emissions to the air and surface waters, mainly from agriculture.

Opportunity 2: The model of this PhD thesis can directly be applied for future years and scenario development. This allows to project future nutrient emissions to the air, rivers and coastal waters under changing climate and socio-economic conditions. Future analysis also allows to explore the effects of mitigation strategies without depending on the historical data and calibration need. Chapter 4 is an excellent example of such scenarios and future analyses.

o *Interactions to support nutrient pollution control*

Interaction types: This thesis offers opportunities for at least three types of interactions in nutrient pollution control: (1) interactions among sources of nutrient emissions to the air and surface waters (land-air and land-river-sea), (2) interactions among mitigation strategies (scenarios), and (3) policy interactions. The MARINA-Nutrients model explicitly addresses

(1) and (2), which are the core of this thesis. The latter is related to the contribution of mitigation strategies to EU policies and Sustainable Development Goals (SDGs).

Interaction type 1 (sources): The MARINA-Nutrients model focuses on land-air and land-river-sea interactions among sources of nutrient emissions (Chapter 2). For example, agricultural practices (e.g., application of synthetic fertilizers and manure, manure deposited on land during grazing) release N emissions to the air via denitrification and NH_3 volatilization (land-air), and inputs of N and P to rivers via surface runoff (land-river). Moreover, treated and untreated domestic sewage flows carry nutrients into rivers. These shared sources of nutrient emissions to the air and surface waters create opportunities for identifying synergistic strategies to reduce these emissions simultaneously (Chapter 4). On the other hand, through nutrient exports by rivers to coastal waters (land-river-sea), this PhD thesis demonstrates how much nutrients reach coastal waters from land activities and how much future river export of nutrients on land is required to be reduced to avoid future coastal eutrophication in Europe under global change (Chapter 3). This analysis helps define priority basins (i.e., the top ten polluted rivers) and contribute to developing effective strategies to avoid coastal eutrophication in Europe in the future (Chapter 3).

Interaction type 2 (scenarios): The scenario analysis in Chapter 4 provides examples of interactions among mitigation strategies. Interactions can be synergistic, antagonistic, and additive (Dunne, 2010). Synergy occurs, for example, when a mitigation strategy for one nutrient emission decreases another, while an antagonistic effect occurs if a strategy reduces one emission but increases another (e.g., pollution swapping (Velthof et al., 2024)). For instance, a scenario combining the use of ReNuRe with higher nitrogen use efficiencies in agricultural land (ReNuRe-NUE) is found to be synergistic in reducing both total N emission to the air (30%) and TDN inputs to rivers (23%) in 2050 compared to the future baseline (SSP5-RCP8.5). Another example is that improvement of the wastewater treatment shows an additive effect through reducing nutrient inputs to rivers and exports by rivers to coastal waters, as well as meeting P fertilizer demand with RPSS in Europe by 2050 relative to the future baseline (SSP5-RCP8.5). On the other hand, the use of ReNuRe alone is expected to reduce only N emissions to the air (22%). Thus, the MARINA-Nutrients model for Europe offers an opportunity to explore both synergetic and antagonistic effects of nutrient management strategies in mitigating nutrient emissions to the air, rivers and coastal waters in the future. The model could motivate decision-makers for proactive actions for the simultaneous reduction of nutrient-driven air and surface water pollution in Europe in the future.

Interaction type 3 (policies/SDGs): Further interactions of mitigation strategies considered in this thesis with existing EU policies and SDGs (Chapter 4). For example, the P-scenarios combining the use of RPSS with medium improvement of wastewater treatment (Pmed) or with advanced improvement of wastewater treatment (Padv), and their combinations with higher phosphorus use efficiencies in agricultural land (Pmed-PUE and Padv-PUE) help

achieve the EU Green Deal Farm-to-Fork target on reducing nutrient losses by at least 50% (EC, 2020). Moreover, more than 20% decline in N fertilizer use can be possible by 2050 under the ReNuRe-NUE scenario (EC, 2020). This analysis also supports the River Basin Management Plans (EC, 2000) by providing basin-specific opportunities for reducing air and water pollution simultaneously, and minimizing the risk of pollution swapping to help solve the N crisis in Europe. On the other hand, combining the use of ReNuRe and RPSS with increased nutrient use efficiencies in agriculture interacts with SDG 2 (zero hunger) SDG 6 (clean water and sanitation), SDG 12 (responsible consumption and production), SDG 13 (climate action), and SDG 14 (life below water) (UN, 2015) through increasing circularity, promoting sustainable food systems, and reducing environmental impacts of agriculture. Furthermore, improved wastewater treatments interact with SDG 6 (clean water and sanitation), SDG 11 (sustainable cities and communities), and SDG 14 (life below water) (UN, 2015; Wang et al., 2022a) while contributing to the circular economy.

5.4.4 Threats

Despite the opportunities, the PhD thesis identifies three main threats that might have contributed to the weaknesses of this thesis. These include (1) uncertainty along the modeling chain (Chapters 2-4), (2) potential trade-offs of modeling, scenarios and BBFs (Chapters 2-4), and (3) policy implications of implementing BBFs for nutrient pollution reductions (Chapter 4, Figure 5.3).

o Uncertainty along the modeling chain

Model chain: Uncertainties can be caused by model structure, and data (model inputs and parameters), (Wang et al., 2020a), and accumulates along the modeling chain (Bak et al., 2025). The model chain starts with the development of the model (model structure), continues with the preparation of data for model inputs, and ends with the model outputs. Thus, it is important to consider and mitigate threats of uncertainties in the model structure and data to build trust in model results.

Uncertainty in model structure: Uncertainties in the model structure are due to simplifications of the modeling approach to simulate nutrient flows from land to rivers and nutrient exports by rivers to coastal waters (see more in Chapters 2-4). A potential threat to model results is the ignorance of dynamics that may overestimate the effects of BBFs on reducing nutrient emissions. Another potential threat is associated with the fact that the application of the model might be limited to smaller scales (e.g., city, field) because the model is too coarse, eliminating heterogeneity within the basin through a lumped approach. Therefore, the MARINA-Nutrients for Europe cannot capture the effects of local interventions (e.g., precision fertilization). In such cases, high-resolution models are more appropriate (e.g., INTEGRATOR, Table 1.1). Nevertheless, the model of this PhD thesis is developed for large-scale analyses. The MARINA-Nutrients approach has been validated for many model

versions (Strokal et al., 2016; Wang et al., 2020a; Chen et al., 2022; Micella et al., 2024a) as well as in this thesis indicating a good performance (Ural-Janssen et al., 2023).

Uncertainty in data: Uncertainties exist in the available data to derive model inputs and parameters. For example, the use of long-term averages of natural discharges (Chapters 2-4) to reduce bias from short-term variability (e.g., climate extremes) (Strokal et al., 2021) masks interannual variability. This contributes to uncertainty since the differences in precipitation and temperature affect estimates of runoff and discharge, and in turn, nutrient inputs to rivers and exports to coastal waters. For instance, high flows can flush nutrients into rivers increasing loads, while low flows can increase their concentrations (van Vliet et al., 2023). On the other hand, in Chapter 4, several model parameters are derived based on expert knowledge and supported by literature. For instance, a low emission factor (<0.01) for NH_3 emission from the ammonia stripping-scrubbing process is considered (Vingerhoets et al., 2023a), and fertilizer replacement values of nutrients for ReNuRe and RPSS, respectively, are determined "1" as equivalent to synthetic fertilizers based on expert knowledge and literature (Huygens et al., 2020; Vingerhoets et al., 2024). Moreover, the back calculation and use of emission factors from 2017 to project N emissions to the air might overestimate N emissions in 2050, as future improvements in agricultural systems could lead to less emissions. All these threats associated with the data might result in over- or underestimation of nutrient emissions to the air, rivers and coastal waters in Europe, especially for basins with intensive agriculture. On the one hand, simplifications in data are a threat affecting model results. On the other hand, the data for model inputs such as emission factors and fertilizer replacement values for BBFs are often limited for studied 601 basins. Thus, the support and verification by experts, as was done in this thesis, are important to mitigate the threats associated with limited data and help build trust in model results.

o *Potential trade-offs*

Trade-offs among nutrients: An important threat that might affect model results is associated with the fact that the MARINA-Nutrients model does not account explicitly for biogeochemical interactions among nutrients. For example, the limitation of one nutrient can affect the other in water systems, showing a trade-off between them (Garnier et al., 2019; de Vries et al., 2024). In particular, reducing N inputs to rivers and coastal waters might shift the aquatic system from P-limited to N-limited or vice versa (Garnier et al., 2019), which can fail to control eutrophication. It is not easy to account for trade-offs among nutrients in terms of eutrophication control when modeling their flows by the MARINA-Nutrients model for Europe. Therefore, the results of this thesis should be interpreted with caution regarding biogeochemical interactions and seasonality effects.

Trade-offs between scenario assumptions and model results: Scenario analysis is a powerful tool to project future pollution levels. However, the assumptions made for scenario development can result in potential trade-offs for model results. An example of a trade-off is between

proposed higher nutrient use efficiencies and their implementation in reality. In this thesis, it was assumed that nutrient use efficiency is increased to 0.75 in all basins, following Schulte-Uebbing and de Vries (2021). It represents an optimistic yet policy-relevant target. However, achieving such nutrient use efficiency levels on agricultural land in practice might be constrained by various agronomic (e.g., farming systems) and socioeconomic (e.g., nutrient recycling systems) factors. These external factors, in turn, influence the feasibility of achieving the targeted nutrient use efficiencies for agricultural land. Another potential trade-off may exist between scenario assumptions and model outcomes. Scenario assumptions are often biased toward our opinion and are shaped based on our expertise and knowledge (Chapter 4). This means that the results of the MARINA-Nutrients model for Europe rely on these biased assumptions for 2050. Some studies overcome the threat of biased scenario assumptions on model results with transdisciplinary approaches in which scenarios are co-developed with relevant stakeholders from the start and scenarios are evaluated continually during the process (Pedde et al., 2025). This could be an approach for future work. Nevertheless, the best available knowledge in the BBFs-related scenarios is integrated in this thesis using the expertise of two host groups: Earth Systems and Global Change Group (ESC) in Wageningen University & Research and Lab for Bioresource Recovery (RE-SOURCE Lab) in Ghent University. ESC is strong in scenario development, while RE-SOURCE Lab is strong in the knowledge of nutrient recovery and recycling as well as BBFs. Combining these two knowledge hubs made the interpretation of the BBFs implementation into scenarios trustworthy.

Trade-offs between pollutants and BBFs: A potential threat of implementing BBFs could be associated with multiple pollutants (see more in Section 5.5.1). Some BBFs may include multiple pollutants (Schoumans et al., 2019; Albert & Bloem, 2023; Dong et al., 2023) (Table S5.3). This might create a potential trade-off between upcycling organic wastes (for applying as BBFs) and the release of multiple pollutants to rivers. However, measuring a wide range of organic contaminants in BBFs and their potential interaction with the soil matrix is difficult due to the pollutants' complex structure (Das et al., 2023). Standard protocols for detection, extraction and quantification of contaminants in BBFs (e.g., ecotoxicological tests) are required (Albert & Bloem, 2023) for sustainable production and application of BBFs. Therefore, policy-makers need to invest in overcoming these trade-offs for the promotion, adoption and implementation of BBFs in the EU.

o Policy associated barriers for implementing BBFs

Recovered P: Policy associated barriers could pose a threat for implementing P-recovered BBFs. This thesis indicates the projected reductions in P inputs to rivers and exports by rivers to coastal waters when RPSS -produced by 90% P recovery from treated sewage sludge- is considered to be used as BBF across the EU and the UK in 2050 (Chapter 4). However, recovery and recycling of P from human waste (e.g., municipal wastewater, sewage sludge) has not been implemented across the EU, yet. An example of policy barriers is the EU Fertilising Products Regulation (FPR). This FPR regulation allows the recycled P products recovered

from treated municipal wastewater and sewage sludge to be used as fertilizers. But this is possible only if phosphate is recovered from sewage sludge ash or precipitated phosphate salts (e.g., struvite) (EU, 2019). Another barrier of implementing BBFs is the regional differences in policies. Implementation of BBFs varies among the EU Member States: while some countries already have a national legislation for recovered phosphate fertilizers (e.g., the Netherlands (Overheid, 2015)); some will make recovery and recycling of P from sewage sludge or sewage sludge ash obligatory in the near future (e.g., Switzerland by 2026 (FOEN, 2024)). This thesis estimates that "Recovered P from treated sewage sludge can fulfill P fertilizer need in Europe in 2050" (Ural-Janssen et al., 2025). This highlights the potential that cannot be obtained unless the production and implementation of RPSS is recognized across the EU and the UK regulations (see Section 5.5). Therefore, this result showing the effects of BBFs in this PhD thesis should be interpreted with care. More research is needed to better understand the policy barriers to implementing P recovered BBFs across regions.

Recovered N: On the other hand, policy barriers seem to be less for N recovery from manure processing in the form of ammonium salts. The recently approved ReNuRe (REcovered Nitrogen from manURE) proposal (EC, 2025a) allows these manure-derived products to be used as mineral fertilizers even in Nitrate Vulnerable Zones (NVZs) (Huygens et al., 2020) in line with the Fertilising Products Regulation (EU, 2019). This BBF presents a promising opportunity for the integration of manure-derived products into the mineral fertilizer market (Vingerhoets et al., 2024). Therefore, this thesis offers a comprehensive analysis regarding the effects of the ReNuRe products on nutrient emissions to the air, rivers and coastal waters of Europe (see Section 5.5).

5.5 Reflection on perspectives

Section 5.5 discusses the main perspectives on the implementation of BBFs and their role in air and water pollution (reduction) by synthesizing the existing knowledge and the results of this thesis. In this PhD research, the effects of using BBFs in the EU are explored from an environmental point of view. However, there are other aspects requiring careful attention, as well. For example, agronomic efficiency, technological feasibility, and societal acceptability are important factors that could affect the adoption and implementation of BBFs. Although there are various studies on these aspects of BBFs, none have integrated these aspects in terms of mitigating air and water pollution.

Thus, I argue the importance of the five main perspectives towards the implementation of BBFs and their role in preventing and/or reducing air and water pollution (Figure 5.4):
(1) environmental effectiveness (Chapter 4 as an example; Section 5.5.1),
(2) agronomic efficiency (Section 5.5.2),
(3) technological feasibility (Section 5.5.3),
(4) policy and socioeconomic relevance (Section 5.5.4),
(5) integration of perspectives 1-4 for effective implementation across scales (Section 5.5.5) .

To this end, relevant literature is reviewed on BBFs for the last decade (2014-2025) to understand where the knowledge gaps are, and which areas are underrepresented and require further attention. Table S5.2 lists the references for each perspective derived from the literature review and Figure S5.1 for the reported frequency of each perspective.

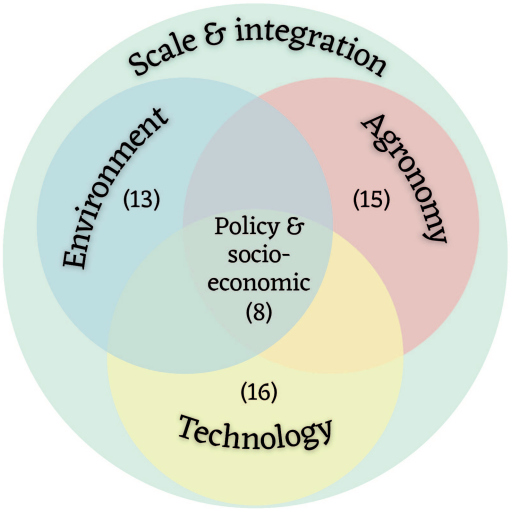


Figure 5.4 Illustration of the five main perspectives and their connections for the effective implementation of BBFs and their role in air and water pollution (reduction). Colors represent different perspectives and intersections represent the connections between the perspectives. The numbers in parentheses refer to papers that are part of the literature review for Section 5.5.

5.5.1 Environmental effectiveness

BBFs offer significant environmental advantages, including minimizing waste by enhanced nutrient recycling (Sniatala et al., 2023), reducing the emissions of GHG (Egene et al., 2022; Zilio et al., 2022; Hu et al., 2024) and NH_3 (Wester-Larsen et al., 2022; Zilio et al., 2022), and reducing NO_3^- leaching (Zilio et al., 2022) (Table S5.3). Besides, BBFs reduce our dependence on non-renewable materials with a high environmental burden during their extraction and transformation (Clagnan et al., 2023). However, the effectiveness and sustainability of BBFs in air and water pollution control depend on careful consideration of environmental conditions (e.g., soil types) and application techniques (Wester-Larsen et al., 2022), as well as understanding their nutrient release patterns (Agostini et al., 2024). Thus, environmental impacts are related to agronomic efficiencies and technologies for producing and applying BBFs (Figure 5.4).

On the other hand, BBFs might contain multiple pollutants (Albert & Bloem, 2023) such as heavy metals, polycyclic aromatic hydrocarbons (PAHs), emerging contaminants (e.g., microplastics), pathogens, antibiotics, pesticides, and pharmaceuticals (Schoumans et al., 2019; Sharma et al., 2019; Lehmann & Bloem, 2021; Dong et al., 2023; Estoppey et al., 2024) (Table S5.3). For example, Lehmann and Bloem (2021) reported that the frequency of antibiotic contamination in cattle and pig slurry was 60% and 89%, respectively, with sewage sludge exhibiting the highest frequency. Therefore, scientific evidence suggests the implementation of realistic limits for the majority of potential contaminants to promote

the adoption of BBFs as well as a combined chemical and ecotoxicological assessment to minimize their negative impacts, especially in long-term application (Albert & Bloem, 2023). Integrated methods to assess different types of BBFs and their environmental impacts facilitate the production of optimal BBFs according to regional needs (Vaneeckhaute, 2021).

Nevertheless, the upcycling of organic waste such as animal slurry/manure and sewage sludge after sufficient treatment to produce renewable BBFs can offer an environmental gain compared to non-renewable synthetic fertilizers. This thesis contributes to this environmental perspective by being a promising first step to explore the effectiveness of the two BBFs, namely ReNuRe and RPSS, on reducing nutrient emissions to the air, rivers, and coastal waters. The results indicate that using ReNuRe helps reducing N emissions to the air from agriculture in 2050 (by 22%), but not enough for reducing N pollution in waters relative to the future baseline scenario (SSP5-RCP8.5). On the other hand, the use of RPSS as a replacement for synthetic P fertilizers does not directly reduce P pollution in rivers as the total P input to land does not change relative to the future baseline scenario (Figure S4.5). Only the combination of using RPSS with upgrading wastewater treatment one level up (Pmed) or to tertiary treatment (Padv) could help reducing P inputs to rivers by 53-65% (Figure 4.3). For this reason, BBFs are suggested to be combined with other nutrient management strategies such as increasing nutrient use efficiency in agriculture and enhancing wastewater treatment systems (Ural-Janssen et al., 2025).

5.5.2 Agronomic efficiency

The agronomic efficiencies of BBFs could be similar to (Huygens & Saveyn, 2018; Shi et al., 2022; Saju et al., 2023; Müller et al., 2024) or higher than synthetic fertilizers (Ai et al., 2020; Clagnan et al., 2023). The literature on the agronomic assessment of BBFs often focuses on parameters to assess the plant availability of nutrients, the organic matter content, and its biodegradability (Schoumans et al., 2019). As main indicators, N and P fertilizer replacement values – indicating the rate of mineral fertilizer substitution – should be determined based on the origin and characteristics of BBFs (Brod et al., 2022). For example, rapid nutrient mineralization and overall biostimulant effects enhance the fertilizer value of BBFs (Clagnan et al., 2023). While mineral-like BBFs can balance N supply with plant uptake and reduce N losses, they do not contribute to long-term soil fertility (Rashid et al., 2025). Whereas, BBFs with higher organic N content like anaerobic digestate can enhance N supply capacity and soil fertility over time (Rashid et al., 2025). Moreover, C in some BBFs (e.g., struvite, biochar, ash) is retained longer in soil, providing a more stable and sustainable option to increase soil C stocks and sequestration (Hu et al., 2021). On the other hand, fertilization techniques could also alter their mineral fertilizer substitutions (Rydgård et al., 2024). Accurate prediction of nutrient mineralization dynamics of BBFs could therefore support farmers in the right timing and adequate application based on the crop demand (Agostini et al., 2024). Overall, BBFs can improve soil quality (Kurniawati et al., 2023b) and benefit plant productivity (Sharma et al., 2019), depending on their nutrient mineralization dynamics. As

the nutrient mineralization dynamics are influenced by application technologies and alter nutrient emissions of BBFs, this agronomy perspective interacts with the environment and technology perspectives in mitigating air and water pollution (Figure 5.4).

In this thesis, the effects of implementing the two BBFs, namely ReNuRe and RPSS, are explored on an annual basis. The effects of seasonal changes and application techniques are not considered explicitly. Whereas, it is assumed that the use of BBFs will not negatively affect the nutrient yields (defined as nutrient exported from agricultural land through crop harvesting and grazing) since the total effective nutrient input level remained the same (Ural-Janssen et al., 2025). On one hand, this thesis is just a first step for understanding the effects of these BBFs on reducing nutrient emissions to the air and surface waters (rivers and coastal waters) at the EU-wide scale. On the other hand, this thesis research has an opportunity to be improved in the future to better represent the agronomic value of BBFs. This would require integrating the agronomy better into the MARINA-Nutrients model for Europe. Therefore, future research can build on this thesis to better understand the implications and long-term benefits of BBFs based on local conditions and needs.

5.5.3 Technological feasibility

Technologies for producing BBFs through nutrient recovery from organic waste vary greatly from physicochemical to biological (Table S5.4). There is no single technology to recover nutrients effectively and to be used globally (Sniatala et al., 2023). The production technology should also improve nutrient use efficiency as well as minimize environmental impacts while considering local limitations (Melo et al., 2022). On the other hand, the cost, feasibility, and implications of the technology should also be taken into account (Sniatala et al., 2023). To determine the optimal technological feasibility for the production of BBFs, two approaches could be: (1) setting up a treatment system for a tailor-made end product with desired characteristics (Vaneeckhaute, 2021), and (2) designing an optimal treatment train based on the source (Vaneeckhaute et al., 2018). As an example of the former, ammonium sulfate and ammonium nitrate produced by NH_3 (stripping-)scrubbing technology are listed by the European Commission as high-priority products such as ReNuRe to replace synthetic N fertilizers (Huygens et al., 2020) (Table S5.4). The latter option is more suitable for regions with e.g., manure surplus, where they have a large amount of source to produce BBFs. On the other hand, high P recovery from treated municipal wastewater or sewage sludge-derived ash is a sustainable alternative for P while reducing the dependence on external fertilizers and contributing to the circular economy (Huygens et al., 2019; Liu et al., 2021; Fournie et al., 2022; Vingerhoets et al., 2023b; Zhu et al., 2023; Ural-Janssen et al., 2025). However, limitations such as heavy metal contamination (Husek et al., 2022) and regional surplus/deficiency (Panagos et al., 2022) should be considered while selecting processes.

In this thesis, therefore, ReNuRe and RPSS are assessed as BBFs based on nutrient emissions to the air, rivers and coastal waters of Europe from their production and application in

2050 (Ural-Janssen et al., 2025). For ReNuRe, N recovery from manure through anaerobic digestion followed by NH_3 (stripping-)scrubbing technology is considered. Combining both technologies produces ReNuRe fertilizer (Sigurnjak et al., 2019), and an organic matter-rich effluent. For RPSS, P recovery from treated sewage sludge is considered through recently proposed technologies (> 90% of P recovery, Huygens et al. (2019)). These BBFs are chosen due to being sustainable options with high organic waste amounts/potential (e.g., animal manure and sewage sludge) in Europe. Furthermore, other nutrient management strategies assessed in this thesis, namely increased nutrient use efficiencies in agricultural land and improving wastewater treatment technologies, demonstrate synergistic and additive effects in terms of environmental feasibility combined with BBFs (see more in Chapter 4). The model developed in this thesis can further be used to explore other feasibilities of nutrient management strategies such as economic (Vingerhoets et al., 2025). For example, the cost-optimization approach was developed for the Yangtze and Pearl Rivers to reduce future coastal eutrophication and water scarcity, respectively, in the cheapest way (Strokal et al., 2020; Baccour et al., 2024). These approaches are implemented in the MARINA-Nutrients models for China. They can also be implemented in the MARINA-Nutrients for Europe to support decision-making.

5.5.4 Policy and socioeconomic relevance

A number of EU policies that are directly/indirectly related to nutrient management and BBFs are summarized in Chapter 1 (see Section 1.1.6 and Table S1.2). For example, the Farm-to-Fork (F2F) strategy aims for sustainable food production by reducing food waste, enhancing circularity via waste recycling and improving nutrient use efficiency to mitigate nutrient losses by at least 50% without deteriorating soil fertility, claiming that this will reduce fertilizer use by at least 20% in 2030 (EC, 2020). By its ambitious targets, F2F focuses on the principles of circularity, efficiency and sufficiency (Spiller et al., 2024). By circularity, the aim is to close the loop of nutrient flows by recovery and recycling of nutrients in the agri-food chain by increasing nutrient use efficiency and reducing nutrient losses to the environment (Spiller et al., 2024). Improving nutrient use efficiency is commonly recommended to ensure sustainable agricultural production (Spiller et al., 2024). Sufficiency accounts for reforming our consumption patterns to sustain food security while minimizing environmental impacts (Spiller et al., 2024). By contributing to the circular economy through effective and sustainable use of resources by minimizing waste, increasing recycling and reducing the extraction of non-renewable materials (Le Pera et al., 2022), BBFs help close nutrient loops while improving nutrient use efficiency and mitigating nutrient emissions to the air and water systems. Consequently, BBFs help achieve SDG 2 (zero hunger), SDG 11 (sustainable cities and communities), SDG 12 (responsible consumption and production), SDG 13 (climate action), SDG 14 (life below water), and SDG 15 (life on land) (Clagnan et al., 2023), and the aforementioned Farm-to-Fork targets. Nevertheless, the implementation of BBFs requires ongoing efforts to address regulatory challenges in the EU (Kurniawati et al., 2023a), as some of them are subject to legal barriers preventing the effective use of signifi-

cant sources to produce them (Kvakkestad et al., 2023). Under the EU Fertilising Products Regulation (FPR), this category includes fertilizers containing organic carbon and nutrients of biological origin while specific inorganic components are still permitted (EU, 2019). For instance, the recovered P from treated sewage sludge is considered in the FPR only if phosphate is recovered from sewage sludge ash or precipitated phosphate salts (e.g., struvite) (EU, 2019). Hopefully, the recent limitation by the EU REACH regulation for intentionally added microplastics to cosmetics and household products could provide safer BBFs from treated sewage sludge in the future (Estoppey et al., 2024).

On the other hand, the costs of transportation and production, content variability, suitability for crops and policy framework challenge societal acceptance and adoption of BBFs by farmers (Kurniawati et al., 2023b; Garmendia-Lemus et al., 2024). For example, the contribution of BBFs to the maintenance of soil organic matter, which is recognized by farmers, has no clear economic value as there is no system or approach to determine the economic value of a certain soil quality. Thus, economic value assessment of BBFs can be based on 1) the value of nutrients in BBF products, 2) the cost of maintaining soil organic matter balance, and 3) the economic impact of improved soil organic matter content on crop yields (Schoumans et al., 2019). Another example is that farmers prefer similar properties as synthetic fertilizers with a certain N content, high organic matter, low volume, safety (e.g., hygiene) and easy application (Bonnichsen et al., 2020; Egan et al., 2022; Kvakkestad et al., 2023), but lower price than synthetic fertilizers (Tur-Cardona et al., 2018) to maximize the market share (Moshkin et al., 2023). Furthermore, cost effectiveness of the processing technology is significant for the adoption of BBFs (Lessmann et al., 2023). Therefore, challenges related to changing the perception and behavior of farmers need to be addressed by EU policies. Overcoming these challenges in the adoption of BBFs and introducing them to the market requires systemic and participatory approaches, collaborative and inclusive decision-making. Therefore, the policy and socioeconomic perspective is the core of the aforementioned perspectives in three aspects (Figure 5.4). First, policies can minimize the environmental impacts of BBFs by ensuring the production of safe and clean products, and limiting the excess application (e.g., precision fertilization). Second, policies can identify the characteristics of novel BBFs by setting criteria in terms of agronomic efficiency (e.g., fertilizer replacement value for nutrients). Third, policies can support the selection of processing technologies by providing incentives (e.g., subsidies).

In this thesis, the future baseline scenario (SSP5-RCP8.5) considers fossil fuel-based rapid economic development; high mitigation challenges; non-climate policies including environmental policies to protect local air, soil and water quality; high-tech increased agricultural production (O'Neill et al., 2014; O'Neill et al., 2017); high urbanization and low population growth (Strokal et al., 2021) in terms of socioeconomic development. Ultimately, the implementation of BBFs and other nutrient management strategies would be feasible under this global change scenario. Thus, in Chapter 4, the six alternative sce-

narios that consist of different combinations of the use of BBFs, increased nutrient use efficiencies in agricultural land, and improved wastewater treatment systems are built on the future baseline (SSP5-RCP8.5) from Chapter 3.

5.5.5 Scale and integration

This perspective takes a different context compared to the four perspectives above. This perspective has two essential parts: scale and integration. I argue to consider a *scale* when exploring options to implement BBFs. This is because our knowledge is still limited to laboratory (Egene et al., 2022; Wester-Larsen et al., 2022; Das et al., 2023; Shrivastava et al., 2023; Wester-Larsen et al., 2024) or field-scale (Zilio et al., 2022; Saju et al., 2023; Hu et al., 2024; Kebalo et al., 2024; Müller et al., 2024). Although these small scales or finer resolutions could yield better insights into the spatial dynamics of nutrient transport and its ecological implications, they do not give an overview of the effectiveness of BBFs on reducing nutrient losses to the air and waters at a larger scale. In addition, those studies cover only a short period of time. Long-term impacts of novel BBFs both on air and surface water pollution in the future are not well studied.

On the other hand, the existing studies for BBFs largely focus on agronomic and/or environmental impacts (see more in Sections 5.5.1 and 5.5.2). They often lack the assessment of technical/technological feasibility and socioeconomic relevance. Furthermore, scale is underrepresented in the literature. Because of these, an *integration* of knowledge from the four abovementioned perspectives is urgently needed, which is the second part of this perspective. Therefore, scale and integration provide a unifying perspective by showing how the other four are interconnected in practice and why their integration is needed for the adoption and upscaling of the implementation of BBFs (Figure 5.4). Modeling tools could be used for this integration. The MARINA-Nutrients model of this PhD thesis offers this opportunity and can be used for such integration. Perspective 1 (environmental effectiveness) has been explored in Chapter 4 and could be extended to more pollutants to provide a solid basis for policy development. Agronomic efficiency, technological feasibility, and policy/socioeconomic relevance can be added to the model via scenario development with participatory approaches in which farmers, technologists and policy-makers participate.

5.6 Future outlook

Section 5.6 starts with an outlook by summarizing the benefits of this PhD research in a general context (Section 5.6.1). Next, recommendations for future developments in science and policy for improving air and water quality in Europe are provided (Sections 5.6.2-5.6.3). Finally, concluding remarks are summarized (Section 5.6.4).

5.6.1 What benefits can be achieved in the general context

This PhD thesis presents three main benefits that support improving air and water quality in Europe.

Benefit 1 (a modeling tool): This thesis provides a new European version of the MARINA-Nutrients model that integrates air and water, including annual emissions of NH_3, N_2O and NO_x from crop and livestock production. This was never done before, as the existing MARINA models for river basins are limited to water pollution. The new MARINA-Nutrients for Europe is also applied for the first time to explore the effects of using BBFs alone and in combination with other nutrient management strategies on reducing nutrient emissions to the air, rivers and coastal waters in the future across the EU and the UK basins. By these and basin-scale assessments, the model of this thesis contributes to the River Basin Management Plans (EC, 2000).

Benefit 2 (scientific insights): The findings of this thesis indicate that nutrient pollution in surface waters of Europe is expected to increase in 2050 under socioeconomic developments and climate change (SSP5-RCP8.5). To avoid future coastal eutrophication in Europe, nutrient exports by rivers to coastal waters are required to be reduced. These findings inform where and how much nutrient pollution reductions are needed for improving water quality and minimizing the risk of coastal eutrophication, while supporting several of the EU policies (e.g., Water Framework Directive, Marine Strategy Framework Directive) and SDGs (e.g., SDG 14: life below water).

Benefit 3 (solutions): This thesis develops and applies a number of scenarios with strategies including synergistic and additive effects on nutrient emissions to the air, rivers and coastal waters in 2050. These scenarios are based on the use of BBFs (ReNuRe: REcovered Nitrogen from manURE and RPSS: Recovered Phosphorus from treated Sewage Sludge), increased nutrient use efficiencies in agriculture, and improved wastewater treatment efficiencies for nutrient removal. The results reveal that the RPSS can replace synthetic P fertilizers fully for the EU and the UK in 2050. The combinations of using BBFs with increased nutrient use efficiencies in agriculture and upgraded wastewater treatments are projected to reduce nutrient emissions to the air (by 30% for total N emissions), rivers (by 23% for TDN and up to 68% for TDP) and coastal waters (by 9% for TDN and up to 62% for TDP), simultaneously. A significant advantage of this thesis is the potential to build on for exploring the effects of other BBFs and nutrient management strategies for Europe as well as to learn for global applications.

5.6.2 Recommendations for science

Building on the MARINA-Nutrients model for Europe: To eliminate weaknesses and increase the accuracy of the results, process representation in the MARINA-Nutrients should be improved considering biogeochemical interactions, legacy effects, retention and seasonality

dynamics. A few attempts are made to include seasonality into the MARINA models for river exports of N by individual basins (Chen et al., 2019a) and lakes (Yang et al., 2021), while other efforts are ongoing. In addition, the model should account for more pollutants beyond nutrients to be able to assess the environmental and human health impacts of BBFs comprehensively. Quite some efforts have already been made in the family of the MARINA models for multiple pollutants. Examples are MARINA-Multi (Global 1.0) (Strokal et al., 2021), MARINA-Multi (Global 2.0) (Li et al., 2022b), MARINA-Multi (Global 3.0) (Zhang et al., 2024b), and MARINA-Multi (Global 4.0) (Micella et al., 2024a; Micella et al., 2024b; Bak et al., 2025; Micella et al., under review). Furthermore, the next step is to integrate PFAS (per- and polyfluoroalkyl substances). This all offers an opportunity to add multiple pollutants to the model of this PhD thesis.

Building on the backcasting approach for desirable futures: The backcasting approach used in this thesis can be developed further taking more pollutants and uncertainty ranges into account. This thesis used the Indicator for Coastal Eutrophication Potential (ICEP, Billen and Garnier (2007)) to set a target for safe levels of nutrient fluxes at the river mouth to avoid coastal eutrophication in Europe. Other researchers have used impact-based environmental thresholds for N to quantify critical N losses from and critical N inputs to European agriculture (de Vries et al., 2021). However, backcasting studies exploring options to reach a desirable future are scarce (Li et al., 2019; Ural-Janssen et al., 2024; Van Eynde et al., 2025). Therefore, future research should extend backcasting studies by broadening the technical (applying the approach for pollutants beyond nutrients) and scientific (adding uncertainty ranges) scope.

Building on the assessment of BBFs: A comprehensive assessment for the adoption and implementation of BBFs must include agronomic efficiency, feasibility of technologies to produce and apply, policy context and implications, and socioeconomic viability and acceptability. Therefore, an integration of knowledge from the four aforementioned perspectives at a large spatial scale is needed for the adoption and upscaling of the implementation of BBFs. This PhD thesis is a promising start for large-scale exploration of the effects of two BBFs, ReNuRe and RPSS, on nutrient emissions to the air, rivers and coastal waters. Future studies can learn from this research and build on it for further evaluations of different BBFs.

5.6.3 Recommendations for policy

Contribution to EU policies: The results of this thesis reveal that combining the use of RPSS with improved municipal wastewater treatments (one level up or to tertiary treatment), and both with increased P use efficiencies in agriculture could help achieve the Green Deal Farm-to-Fork target on reducing nutrient losses by at least 50% for P (EC, 2020). Moreover, more than 20% decline in N fertilizer use can be possible in 2050 due to the increased N use efficiencies in agriculture (EC, 2020). Another notable result of this thesis is RPSS being able to meet the P fertilizer need in the EU and the UK by 2050 under global change, thereby contributing to circular economy. In addition, the BBFs explored, namely ReNuRe

and RPSS, support the Fertilising Products Regulation (EU, 2019). This research also supports the River Basin Management Plans (EC, 2000) by basin-specific opportunities for reducing air and water pollution simultaneously, and minimizing the risk of pollution swapping to help solve the N crisis in Europe. Thus, all these are the recommendations provided by this PhD thesis.

Contribution to global sustainable development goals: By sustainable use of resources through minimizing waste and increasing recycling, and reducing the extraction of non-renewable materials (Le Pera et al., 2022), the use of BBFs contributes to several SDGs, as well. For example, combining the use of BBFs with increased nutrient use efficiencies in agriculture helps achieve SDG 2 (zero hunger), SDG 6 (clean water and sanitation), SDG 12 (responsible consumption and production), SDG 13 (climate action), and SDG 14 (life below water) (UN, 2015) through increasing circularity, promoting sustainable food systems, and reducing the environmental impacts of agriculture. Furthermore, enhanced municipal wastewater treatment systems contribute to SDG 6 (clean water and sanitation), SDG 11 (sustainable cities and communities), and SDG 14 (life below water) (UN, 2015; Wang et al., 2022a). Therefore, the strategies provided by this PhD thesis help achieve at least six SDGs.

5.6.4 Concluding remarks

While concluding this PhD thesis, I remember my initial curiosity and motivation, which guided my career path: how human activities contribute to environmental pollution and how to reduce this (Chapter 1). This thesis has shown the contribution of human activities, mainly agricultural practices and sewage systems, to air and surface water pollution associated with nutrients (Chapters 2-4). Nutrient pollution affects soil, air, and water systems. Therefore, integrated models are needed to assess nutrient emissions to both air and surface waters (Chapters 2 and 4). The MARINA-Nutrients model for Europe is a step in this direction (Chapters 2 and 4). The results highlight both the urgency of action (Chapter 3) and the potential of BBFs and other nutrient management strategies as synergetic or additive solutions (Chapter 4). Therewith, this thesis makes steps to improve our understanding of basin-specific nutrient emissions to the air and surface waters, and the role of BBFs and other nutrient management strategies on reducing future nutrient pollution under global change across the EU and the UK.

After five years of experience in the Ministry of Agriculture and Forestry in Türkiye, I started this PhD journey as an early-stage researcher. Knowing enough about the difficulties to take action on the policy side, I was quite enthusiastic about my PhD research and hoping to make a small contribution to society and the Earth. I believe my research has an impact through new scientific insights and all the dissemination and outreach opportunities during my PhD. I hope these efforts and my contributions to the existing knowledge with this research make a butterfly effect to reach an optimistic future with clean air and water resources for the next generations.

Appendix

Appendices contain supporting information for the following chapters of the PhD thesis:

S1: Supplementary materials for Chapter 1

S2: Supplementary materials for Chapter 2 (published as Ural-Janssen et al., 2023)

S3: Supplementary materials for Chapter 3 (published as Ural-Janssen et al., 2024)

S4: Supplementary materials for Chapter 4 (published as Ural-Janssen et al., 2025)

S5: Supplementary materials for Chapter 5

S1: Supplementary materials for Chapter 1

Text S1.1 Study area

The study area of this PhD covers 601 river basins (593 individual basins and 8 sub-basins of the Danube River) in the EU including the United Kingdom (Chapters 2-4, and Figure S1.1). The total area of the basins included is 5,768,449 km^2 with a range of 1303–288,123 km^2. River basins are delineated based on the global drainage direction map on 30-arcmin resolution (Döll & Lehner, 2002). The drainage area of the Danube (797,289 km^2) is divided into eight sub-basins by Strokal et al. (2019) to better indicate the spatial variation in river export of nutrients. These sub-basins include upstream (i.e. Upstream, Drava, Sava, and Tisza), middlestream (i.e. Siret, Prut and Middlestream) and downstream (i.e. Downstream delta) sub-basins. In the study area, 15 rivers discharge into the Arctic Ocean, 160 rivers discharge into the Atlantic Ocean, 147 rivers discharge into the Baltic Sea, 10 rivers discharge into the Black Sea (9 river basins and 8 sub-basins of the Danube River), 166 rivers discharge into the Mediterranean Sea and 96 rivers flow into the North Sea.

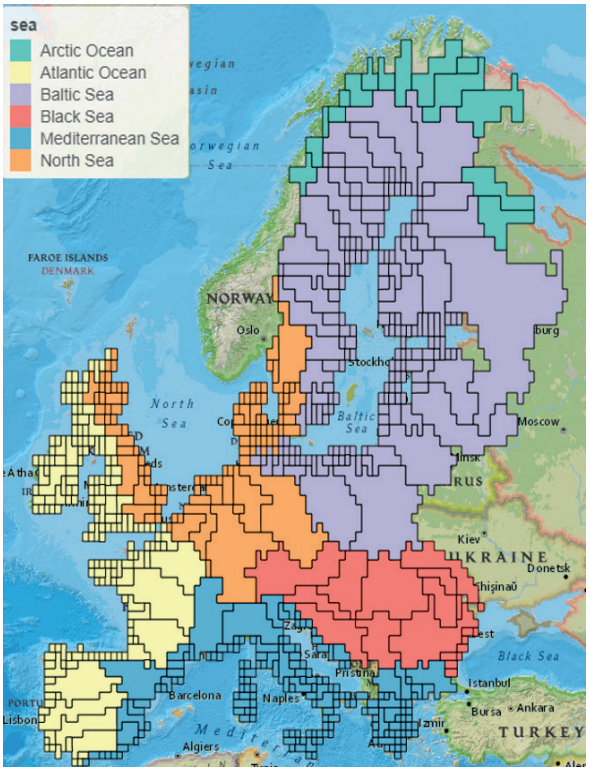


Figure S1.1 Study area considered in this PhD research covers 601 river basins in the European Union including the United Kingdom (593 individual basins and 8 sub-basins of the Danube River) (Strokal et al., 2025). The colors represent the basins draining to the seas/oceans. Source: The MARINA-Nutrients model for Europe. MARINA-Nutrients is short for Model to Assess River Inputs of pollutaNts to seAs for Nutrients.

Table S1.1 Impacts of emissions of nitrogen (N) forms to the air, and losses of N and phosphorus (P) to surface waters (rivers, seas) on human health and the environment. NH_3 is ammonia, NO_x is nitrogen oxides, N_2O is nitrous oxide, and GHG is greenhouse gas.

Form	Emission/loss to	Impact	Reference
NH_3, NO_x	Air	Increased aerosol/particulate matter formation, thereby influencing the radiation balance and cooling the atmosphere	(Wyer et al., 2022; de Vries et al., 2024)
NH_3	Air	Loss of biodiversity in terrestrial ecosystems	(de Vries et al., 2024)
NH_3, N_2O	Air	Climate change through altering GHG balance	(de Vries et al., 2024)
NH_3, NO_x	Air	Adverse effects on forest ecosystems by the acidification of forest soils	(de Vries & Du, 2024)
N_2O	Air	Depletion of stratospheric ozone, thereby increasing ultra-violet light exposure for humans (e.g., skin cancer) and plants (e.g., damage)	(Portmann et al., 2012; Barnes et al., 2019)
N_2O	Air	Increased risk of mortality from respiratory diseases	(de Vries et al., 2024; EEA, 2024d)
NO_x	Air	Increased levels of tropospheric ozone production, thereby leading to warming effect and reductions in crop yields and grassland productivity	(de Vries et al., 2024)
N, P	Surface waters	Freshwater and marine eutrophication, thereby leading to algal blooms, changes in aquatic vegetation, depletion of light, increased sedimentation and in turn, oxygen depletion and fish kills	(Glibert et al., 2018a; 2018b; Kudela et al., 2018; Njock et al., 2023; EEA, 2024c; Giani et al., 2024; EEA, 2025b)

Table S1.2 Summary of the European Union (EU) policies and action plans for mitigating air and surface water pollution associated with nutrient emissions, and implementing bio-based fertilizers. GHG is greenhouse gas, NO_x is nitrogen oxides, and WWTPs are wastewater treatment plants.

Name	Year	Aim	Scope	Source
Common Agricultural Policy	1962, 2023-27	To ensure a sustainable future for European farmers while contributing to climate change mitigation and adaptation (e.g., reducing GHG emissions), and efficient management of natural resources (e.g., reducing chemical dependency).	Air and water	(EC, 1962; 2023)
Sewage Sludge Directive (86/278/EEC)	1986	To protect soil against the contaminants in the sewage sludge that is used in agriculture, while recommending the recycling of sewage sludge to farmland as a source of nutrients and organic matter.	Water	(EC, 1986)
Nitrates Directive (91/676/EEC)	1991	To preserve ground- and surface water quality across Europe by limiting agricultural nitrate pollution through good farming practices.	Water	(EC, 1991a)
Urban Wastewater Treatment Directive (EU) 2024/3019	1991, 2024	To maintain the sufficient treatment of urban wastewaters to prevent nutrient pollution in surface waters and safeguard both the environment and public health, enhance circularity through water reuse and the recovery of valuable resources from wastewater, and mitigate GHG emissions from WWTPs.	Air and water	(EC, 1991b; EU, 2024b)
Habitats Directive (92/43/EEC)	1992	To ensure the long-term preservation of biodiversity by halting the decline of species and habitats and restoring them to a favorable conservation status within the EU.	Air and water	(EC, 1992)
Water Framework Directive (2000/60/EC)	2000	To achieve "good chemical and ecological status" in water bodies (i.e. ground- and inland, transitional and coastal surface waters) by 2027.	Water	(EC, 2000)
National Emission reduction Commitments Directive (2016/2284/EU)	2016	To reduce the effect of air pollution on human health by half in 2030 relative to 2005 through the emission reduction targets for the EU countries.	Air	(EU, 2016)
Marine Strategy Framework Directive (2008/56/EC)	2008	To protect marine ecosystems and achieve "good environmental status" of marine waters based on eleven qualitative descriptors.	Water	(EU, 2008a)
Waste Framework Directive (2008/98/EC)	2008	To lower production and consumption levels, re-use, recycle and recover the unpreventable waste to minimize the effects on the environment and human health.	Air and water	(EU, 2008b)
Paris Agreement (COP21)	2015	To reduce GHG and NO_x emissions with an overarching aim of limiting the global warming to well below 2°C compared to pre-industrial levels and follow practices to limit it to 1.5°C.	Air	(UNFCCC, 2015)
Fertilising Products Regulation (EU) 2019/1009	2019	To harmonize the quality, safety, and environmental requirements for all fertilizing products in the EU including novel organic and recycled fertilizers that are not covered previously, while providing a market opportunity for them.	Water	(EU, 2019)

Name	Year	Aim	Scope	Source
Green Deal (COM/2019/640) Farm-to-Fork Strategy (COM/2020/381)	2019 2020	To ensure fair, healthy and environmentally-friendly food systems while targeting a reduction of GHG emissions by at least 55% by 2030 compared to 1990 levels. To accelerate the transition to a sustainable food system while targeting a reduction of nutrient losses by at least 50% without deteriorating soil fertility and the use of fertilizers by at least 20% by 2030.	Air and water	(EC, 2019; 2020)
Zero Pollution Action Plan	2021	To reduce air, water and soil pollution to unharmful levels for health and natural ecosystems while targeting a reduction of the number of premature deaths caused by air pollution by 55%, the area of EU ecosystems where air pollution threatens biodiversity by 25%, and residual municipal waste by 50%.	Air and water	(EC, 2021a)
Commission Delegated Regulation (EU) 2023/1605	2023	To define end points in the manufacturing chain of certain organic fertilizers and soil improvers made from animal by-products that are allowed to be used after sufficient processing and no longer subject to the requirements of the Animal By-Products Regulation (EC) 1069/2009. These products fall under the Fertilising Products Regulation (EU) 2019/1009 once an endpoint is reached.	Air and water	(EU, 2023)
Industrial and Livestock Rearing Emissions Directive (EU) 2024/1785	2010, 2024	To limit pollution from large industrial facilities and intensive livestock farms and minimize the impact of pollution on health and environment. The revised version (IED 2.0) covers the largest and most polluting intensive pig and poultry farms – accounting for 30% of pig and poultry farms – in the EU to reduce N pollution in the air, water and soil.	Air and water	(EU, 2010; 2024a)

S2: Supplementary materials for Chapter 2

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Supplementary materials

Hotspots of nutrient losses to air and water: an integrated modeling approach for European river basins

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The Supplementary materials contain

Supplementary Text S2.1-S2.2

Tables S2.1-S2.13

Figure S2.1

Supplementary Text S2.1 Developments in the new MARINA-Nutrients model for Europe

The new MARINA-Nutrients model for Europe was built upon the existing approaches of MARINA (Strokal et al., 2016; Wang et al., 2020a) and MITERRA-Europe models (Velthof et al., 2009). Our improvements of the new MARINA-Nutrients model for Europe are listed below:

Addition of atmospheric P deposition over agricultural areas by using the MITERRA-Europe model outputs, previous MARINA-Nutrients model versions did not have P deposition;

- Accounting for losses of N to air and leaching from animal housing and storage systems based on the MITERRA-Europe model outputs. The N emissions to air (e.g., NH_3, N_2O) and leaching (i.e. NO_3, P_2O_5) are calculated as a function of emission and leaching fractions (Velthof et al. (2009) for further description);
- Accounting for direct (e.g., application of synthetic fertilizers and animal manure, grazing) and indirect (e.g., N_2O emissions from leaching and ammonia) losses of N to air from agricultural land;
- Including human waste that does not go to sewage systems as a diffuse source for the inputs of N and P to land and rivers.

Supplementary Text S2.2 Study area

We studied 601 basins (i.e. 593 river basins and 8 sub-basins) in the European Union including the United Kingdom (EU-28). The total area of these basins is 5,768,449 km^2 with a range of 1,303–288,123 km^2. In the study area, 15 rivers discharge into the Arctic Ocean, 160 rivers discharge into the Atlantic Ocean, 147 rivers discharge into the Baltic Sea, 10 rivers discharge into the Black Sea (9 river basins and 8 sub-basins of the Danube River), 166 rivers discharge into the Mediterranean Sea and 96 rivers flow into the North Sea.

River basins were delineated based on the global drainage direction map on 30-arcmin resolution (Döll & Lehner, 2002). The drainage area of the Danube (797,289 km^2) was divided into eight sub-basins following Strokal et al. (2016) to better indicate the spatial variation in river export of nutrients (Strokal et al., 2019). These sub-basins include upstream (i.e. Upstream, Drava, Sava, and Tisza), middlestream (i.e. Siret, Prut and Middlestream) and downstream (i.e. Downstream delta) sub-basins (Figure S2.1).

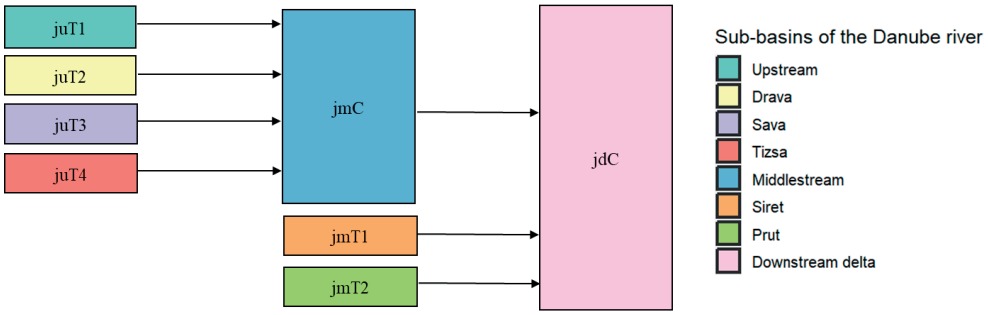


Figure S2.1 Upstream tributary (juT), middle stream tributary (jmT) and main channel (jmC), and downstream main channel (jdC) sub-basins of the Danube River (Strokal et al., 2019). The sub-basins were delineated by Strokal et al. (2019) based on the Drainage Direction Map (DDM30) (Döll & Lehner, 2002). The main channel was defined using the Strahler Order of the Drainage Direction Map (DDM30) following Strokal et al. (2016). The outlet of Downstream delta is the river mouth where nutrients are discharged to the sea. The fraction of the sub-basin area (0–1) draining directly to the main channels (i.e. ${}^{juT}A_{jmC}, {}^{juT}A_{jdC}, {}^{jmT}A_{jdC}, {}^{jmC}A_{jdC}$) was calculated as the fraction to the total area of these sub-basins (0–1) following Strokal et al. (2016) (Table S4 for explanations of the variables).

Table S2.1 Comparison of strengths and weaknesses of MITERRA-Europe (Velthof et al., 2009) and MARINA-Nutrients (Strokal et al., 2016) models.

Model name	Strengths	Weaknesses
MITERRA-Europe	+ Simplified processes for air and water emissions from agriculture + First model to assess synergistic and antagonistic effects of agricultural measures on the emissions + Includes agricultural policy scenario simulations and analysis (e.g., Nitrates Directive)	- No seasonal and annual dynamics - Only processes occur at soil surface considered - Not the whole food system considered (only crop and animal production) - For scenario analysis the model depends on other models (e.g., CAPRI) to provide changes in the activity data/driving factors
MARINA-Nutrients	+ Nutrient forms (i.e. DIN, DON, DIP, DOP) + Modeling of nutrient flows from land to coastal waters at basin/sub-basin scale + Source attribution for nutrients + Upstream, middlestream, downstream sub-basin contribution to pollution + Scenario analysis to reduce future water pollution	- No seasonality - Steady state model

Table S2.2 Comparison of inputs, processes and outputs of MITERRA-Europe (Velthof et al., 2009) and MARINA-Nutrients (Strokal et al., 2016) models.

Model name	Input data	Modeling processes	Output data
MITERRA-Europe	• Main inputs; o livestock numbers, o livestock distribution, o animal production, o feed, o crop areas, o crop yields, o fertilizer consumption	• Calculations are annual and on a deterministic basis; o GHG and NH_3 emissions o N leaching o The surplus of gross N balance o The soil N balance o The soil P balance	• Annual at country, NUTS2 and EU-27 levels o GHG emissions (CH_4, N_2O and NO_x), o NH_3 emissions, o N leaching, o N and P balance, o soil organic C stock changes
MARINA-Nutrients	• Point sources of nutrients: o Sewage § Wastewater treatment § Direct discharges o Manure (direct discharges) • Diffuse sources of nutrients: o Atmospheric N deposition o Biological N_2 fixation o Fertilizer application o Manure application • Human population • Meteorology • Hydrology • Land use	• Calculation of steady-state annual exports of nutrients at the outlets of sub-basins and at the river mouth o Mass-balance approach § Dissolved inorganic N and P § Dissolved organic N, P, and C • Calculation of wastewater treatment removal and river retention of nutrients • Scenario simulations (2050)	• Annual nutrient export at the river mouth o Dissolved inorganic N and P o Dissolved organic N and P • Source attribution of nutrient export at the river mouth o Sub-basin contributions to nutrient export at the river mouth

Table S2.3 Equations of the new version of the MARINA-Nutrients model for Europe to quantify inputs of N and P to land, rivers and to the seas by the European basins adjusted from Strokal et al. (2016).

Equation	Number
$RSdif_{F.y.j} = WSdif_{E.y.j} \times G_{F.j} \times FE_{ws.F.j}$	Main equation Eq.1
$RSdif_{DIN.fe.j} = WSdif_{N.fe.j} \times G_{DIN.j} \times FE_{ws.DIN.j}$	Eq.2
$RSdif_{DIN.ma.j} = WSdif_{N.ma.j} \times G_{DIN.j} \times FE_{ws.DIN.j}$	Eq.3
$RSdif_{DIN.dep.ant.j} = WSdif_{N.dep.ant.j} \times G_{DIN.j} \times FE_{ws.DIN.j}$	Eq.4
$RSdif_{DIN.fix.ant.j} = WSdif_{N.fix.ant.j} \times G_{DIN.j} \times FE_{ws.DIN.j}$	Eq.5
$RSdif_{DIN.hum.uncon.j} = WSdif_{N.hum.uncon.j} \times G_{DIN.j} \times FE_{ws.DIN.j}$	Eq.6
$RSdif_{DIN.dep.nat.j} = WSdif_{N.dep.nat.j} \times FE_{ws.DIN.j}$	Eq.7
$RSdif_{DIN.fix.nat.j} = WSdif_{N.fix.nat.j} \times FE_{ws.DIN.j}$	Eq.8
$G_{DIN.j} = 1 - \left(WSdif_{N.ex.j}/WSdif_{DIN.gross.j}\right)$	Eq.9
$FE_{ws.DIN.j} = CR_{DIN} \times Rnat_j$	Eq.10[a]
$WSdif_{DIN.gross.j} = WSdif_{N.fe.j} + WSdif_{N.ma.j} + WSdif_{N.dep.ant.j} + WSdif_{N.fix.ant.j} + WSdif_{N.hum.uncon.j}$	Eq.11
$RSdif_{DON.fe.j} = WSdif_{N.fe.j} \times G_{DON.j} \times FE_{ws.DON.j}$	Eq.12
$RSdif_{DON.ma.j} = WSdif_{N.ma.j} \times G_{DON.j} \times FE_{ws.DON.j}$	Eq.13
$RSdif_{DON.hum.uncon.j} = WSdif_{N.hum.uncon.j} \times G_{DON.j} \times FE_{ws.DON.j}$	Eq.14
$RSdif_{DON.lech.ant.j} = Area_j \times Agr_{fr.j} \times EC_{DON} \times f_{DON}\left(Rnat_j\right)$	Eq.15[b]
$RSdif_{DON.lech.nat.j} = Area_j \times \left(1 - Agr_{fr.j}\right) \times EC_{DON} \times f_{DON}\left(Rnat_j\right)$	Eq.16[b]
$G_{DON.j} = 1 - \left(WSdif_{N.ex.j}/WSdif_{DON.gross.j}\right)$	Eq.17
$FE_{ws.DON.j} = CR_{DON} \times Rnat_j$	Eq.18[c]
$WSdif_{DON.gross.j} = WSdif_{N.fe.j} + WSdif_{N.ma.j} + WSdif_{N.hum.uncon.j}$	Eq.19
$WSdif_{N.ma.j} = N_{ma} + N_{gr}$	Eq.20
$WSdif_{N.hum.uncon.j} = WSdif_{N.hum.uncon.urb.j} + WSdif_{N.hum.uncon.rur.j}$	Eq.21
$WSdif_{N.hum.uncon.urb.j} = Nexc_{hum.uncon.urb.j} \times (1 - fr_{NH3.hum})$	Eq.22[d]
$WSdif_{N.hum.uncon.rur.j} = Nexc_{hum.uncon.rur.j} \times (1 - fr_{NH3.hum})$	Eq.23[d]
$Nexc_{hum.uncon.urb.j} = E^N_{hum.j} \times Pop_{uncon.urb.j}$	Eq.24
$Nexc_{hum.uncon.rur.j} = E^N_{hum.j} \times Pop_{uncon.rur.j}$	Eq.25
$RSdif_{DIP.fe.j} = WSdif_{P.fe.j} \times G_{DIP.j} \times FE_{ws.DIP.j}$	Eq.26
$RSdif_{DIP.ma.j} = WSdif_{P.ma.j} \times G_{DIP.j} \times FE_{ws.DIP.j}$	Eq.27
$RSdif_{DIP.dep.ant.j} = WSdif_{P.dep.ant.j} \times G_{DIP.j} \times FE_{ws.DIP.j}$	Eq.28
$RSdif_{DIP.hum.uncon.j} = WSdif_{P.hum.uncon.j} \times G_{DIP.j} \times FE_{ws.DIP.j}$	Eq.29
$RSdif_{DIP.wth.ant.j} = Area_j \times Agr_{fr.j} \times EC_{DIP} \times f_{DIP}\left(Rnat_j\right)$	Eq.30[e]

Equation	Number
$RSdif_{DIP.wth.nat.j} = Area_j \times (1 - Agr_{fr.j}) \times EC_{DIP} \times f_{DIP}(Rnat_j)$	Eq.31[e]
$G_{DIP.j} = 1 - (WSdif_{P.ex.j}/WSdif_{DIP.gross.j})$	Eq.32
$FE_{ws.DIP.j} = CR_{DIP} \times Rnat_j$	Eq.33[f]
$WSdif_{DIP.gross.j} = WSdif_{P.fe.j} + WSdif_{P.ma.j} + WSdif_{P.dep.ant.j} + WSdif_{P.hum.uncon.j}$	Eq.34
$RSdif_{DOP.fe.j} = WSdif_{P.fe.j} \times G_{DOP.j} \times FE_{ws.DOP.j}$	Eq.35
$RSdif_{DOP.ma.j} = WSdif_{P.ma.j} \times G_{DOP.j} \times FE_{ws.DOP.j}$	Eq.36
$RSdif_{DOP.hum.uncon.j} = WSdif_{P.hum.uncon.j} \times G_{DOP.j} \times FE_{ws.DOP.j}$	Eq.37
$RSdif_{DOP.lech.ant.j} = Area_j \times Agr_{fr.j} \times EC_{DOP} \times f_{DOP}(Rnat_j)$	Eq.38[g]
$RSdif_{DOP.lech.nat.j} = Area_j \times (1 - Agr_{fr.j}) \times EC_{DOP} \times f_{DOP}(Rnat_j)$	Eq.39[g]
$G_{DOP.j} = 1 - (WSdif_{P.ex.j}/WSdif_{DOP.gross.j})$	Eq.40
$FE_{ws.DOP.j} = CR_{DOP} \times Rnat_j$	Eq.41[h]
$WSdif_{DOP.gross.j} = WSdif_{P.fe.j} + WSdif_{P.ma.j} + WSdif_{P.hum.uncon.j}$	Eq.42
$WSdif_{P.ma.j} = P_{ma} + P_{gr}$	Eq.43
$WSdif_{P.hum.uncon.j} = WSdif_{P.hum.uncon.urb.j} + WSdif_{P.hum.uncon.rur.j}$	Eq.44
$WSdif_{P.hum.uncon.urb.j} = Pexc_{hum.uncon.urb.j}$	Eq.45[i]
$WSdif_{P.hum.uncon.rur.j} = Pexc_{hum.uncon.rur.j}$	Eq.46[i]
$Pexc_{hum.uncon.urb.j} = E^P_{hum.j} \times Pop_{uncon.urb.j}$	Eq.47
$Pexc_{hum.uncon.rur.j} = E^P_{hum.j} \times Pop_{uncon.rur.j}$	Eq.48
$Rnat_j = Q_{nat.j}/Area_j$	Eq.49
$RSpnt_{F.y.j} = RSpnt_{E.hum.con.j} \times FEpnt_{F.hum.con.j}$	Main equation Eq.50 (Eq.2 in the paper)
$RSpnt_{DIN.hum.con.j} = RSpnt_{N.hum.con.j} \times FEpnt_{DIN.hum.con.j}$	Eq.51[j]
$RSpnt_{DON.hum.con.j} = RSpnt_{N.hum.con.j} \times FEpnt_{DON.hum.con.j}$	Eq.52[k]
$RSpnt_{N.hum.con.j} = RSpnt_{N.hum.con.urb.j} + RSpnt_{N.hum.con.rur.j}$	Eq.53
$RSpnt_{N.hum.con.urb.j} = Nexc_{hum.con.urb.j} \times (1 - hw_{frem.N.j})$	Eq.54[l]
$RSpnt_{N.hum.con.rur.j} = Nexc_{hum.con.rur.j} \times (1 - hw_{frem.N.j})$	Eq.55[l]
$Nexc_{hum.con.urb.j} = E^N_{hum.j} \times Pop_{con.urb.j}$	Eq.56
$Nexc_{hum.con.rur.j} = E^N_{hum.j} \times Pop_{con.rur.j}$	Eq.57
$E^N_{hum.j} = I^N_{hum.j} \times 0.365$	Eq.58
$I^N_{hum.j} = 4 + 17 \times [GDP_{ppp.j}/68{,}673]^{0.3}$	Eq.59
$FEpnt_{DIN.hum.con.j} = 0.485 + 0.225 \times (hw_{frem.N.j} \,/\, 0.88)$	Eq.60[l]
$RSpnt_{DIP.hum.con.j} = (RSpnt_{P.hum.con.j} + RSpnt_{P.det.con.j}) \times FEpnt_{DIP.hum.con.j}$	Eq.61[m]

Equation	Number
$RSpnt_{DOP.hum.con.j} = \left(RSpnt_{P.hum.con.j} + RSpnt_{P.det.con.j}\right) \times FEpnt_{DOP.hum.con.j}$	Eq.62[n]
$RSpnt_{P.hum.con.j} = RSpnt_{P.hum.con.urb.j} + RSpnt_{P.hum.con.rur.j}$	Eq.63
$RSpnt_{P.hum.con.urb.j} = Pexc_{hum.con.urb.j} \times (1 - hw_{frem.P.j})$	Eq.64[o]
$RSpnt_{P.hum.con.rur.j} = Pexc_{hum.con.rur.j} \times \left(1 - hw_{frem.P.j}\right)$	Eq.65[o]
$Pexc_{hum.con.urb.j} = E^{P}_{hum.j} \times Pop_{con.urb.j}$	Eq.66
$Pexc_{hum.con.rur.j} = E^{P}_{hum.j} \times Pop_{con.rur.j}$	Eq.67
$E^{P}_{hum.j} = E^{N}_{hum.j} \times \left(1/6\right)$	Eq.68[p]
$RSpnt_{P.det.con.j} = RSpnt_{P.det.con.urb.j} + RSpnt_{P.det.con.rur.j}$	Eq.69
$RSpnt_{P.det.con.urb.j} = \left(1 - hw_{frem.P.j}\right) \times WShw_{cap.P.det.j} \times Pop_{con.urb.j}$	Eq.70[o, r]
$RSpnt_{P.det.con.rur.j} = \left(1 - hw_{frem.P.j}\right) \times WShw_{cap.P.det.j} \times Pop_{con.rur.j}$	Eq.71[o,r]
$N_{total.y.j} = NH_{3.y.j} + N_2O_{y.j} + NO_{X.y.j}$	Main equation Eq.72 (Eq.3 in the paper)
$M_{F.y.j} = \left(RSdif_{F.y.j} + RSpnt_{F.y.j}\right) \times FE_{riv.F.outlet.j} \times FE_{riv.F.mouth.j}$	Main equation Eq.73 (Eq.4 in the paper)
$FE_{riv.DIN.outlet.j} = (1 - D_{DIN.j}) \times (1 - L_{DIN.j}) \times \left(1 - FQrem_j\right)$	Eq.74
$D_{DIN,j} = (1/Q_{act,j}) \times \Sigma_{i=1...n}(Q_{act,i} \times D_{DIN,i})$	Eq.75[s]
$L_{DIN,j} = 0.0605 \times \ln(Area_j) - 0.0443$	Eq.76[t]
$FE_{riv.DON.outlet.j} = \left(1 - FQrem_j\right)$	Eq.77
$FE_{riv.DIP.outlet.j} = (1 - D_{DIP.j}) \times (1 - L_{DIP.j}) \times \left(1 - FQrem_j\right)$	Eq.78[u]
$D_{DIP,j} = (1/Q_{act,j}) \times \Sigma_{i=1...n}(Q_{act,i} \times D_{DIP,i})$	Eq.79[v]
$FE_{riv.DOP.outlet.j} = \left(1 - FQrem_j\right)$	Eq.80
$FQrem_j = 1 - \left(Q_{act.j}/Q_{nat.j}\right)$	Eq.81
$FE_{riv.F.mouth.juT} = {}^{juT}FE_{riv.F.outlet.jmC} \times {}^{juT}FE_{riv.F.outlet.jdC}$	Eq.82
$FE_{riv.F.mouth.jmT} = {}^{jmT}FE_{riv.F.outlet.jdC}$	Eq.83
$FE_{riv.F.mouth.jmC} = {}^{jmC}FE_{riv.F.outlet.jdC}$	Eq.84
$FE_{riv.F.mouth.jdC} = 1$	Eq.85
${}^{juT}FE_{riv.F.outlet.jmC} = \left(1 - [D_{F.jmC} \times {}^{juT}A_{jmC}]\right) \times \left(1 - [L_{F.jmC} \times {}^{juT}A_{jmC}]\right) \times \left(1 - [FQrem_{jmC} \times {}^{juT}A_{jmC}]\right)$	Eq.86
${}^{juT}FE_{riv.F.outlet.jdC} = \left(1 - [D_{F.jdC} \times {}^{juT}A_{jdC}]\right) \times \left(1 - [L_{F.jdC} \times {}^{juT}A_{jdC}]\right) \times \left(1 - [FQrem_{jdC} \times {}^{juT}A_{jdC}]\right)$	Eq.87
${}^{jmT}FE_{riv.F.outlet.jdC} = \left(1 - [D_{F.jdC} \times {}^{jmT}A_{jdC}]\right) \times \left(1 - [L_{F.jdC} \times {}^{jmT}A_{jdC}]\right) \times \left(1 - [FQrem_{jdC} \times {}^{jmT}A_{jdC}]\right)$	Eq.88

Equation	Number
${}_{\mathrm{jmC}}FE_{riv.F.outlet.jdC} = \left(1 - [D_{F.jdC} \times {}_{\mathrm{jmC}}A_{jdC}]\right) \times \left(1 - [L_{F.jdC} \times {}_{\mathrm{jmC}}A_{jdC}]\right) \times \left(1 - [FQrem_{jdC} \times {}_{\mathrm{jmC}}A_{jdC}]\right)$	Eq.89

Note: Inputs of N and P to rivers and seas are quantified from point and diffuse sources. Point sources include effluents of treated wastewater from sewage systems to rivers. Diffuse sources include inputs of the pollutants from land via surface runoff. MARINA-Nutrients is short for the soft-linked model system. Abbreviations are explained in Table S4. [a] Uncalibrated approach was used to calculate the fraction of nutrients that is exported from land to rivers in basin j following Li et al. (2022b). In this approach, the European basins are clustered based on the climate zones (i.e. subtropic/tropic and other temperature zones). CR_{DIN} is the export coefficient for runoff exporting nutrients from land to surface waters in a form of DIN and derived from Li et al. (2022b) based on the subtropic/tropic (wet climate) and other temperature zones (dry climate). More nutrient export is expected from land to rivers with higher runoff. Thus, higher export coefficients are considered for basins with runoff ≥ 0.1 m·yr^{-1}. $CR_{DIN} = 0.9$ (unitless) for runoff ≥ 0.1 m·yr^{-1}, and $CR_{DIN} = 0.8$ (unitless) for runoff < 0.1 m·yr^{-1}. CR_{DIN} is relatively higher as the dissolved inorganic forms of N (e.g., NO_3^-) are more mobile in the environment having higher tendency to reach rivers by surface runoff than DON, DOP and DIP (Li et al., 2022b). [b] EC_{DON} is the coefficient for leaching of organic matter and derived from Mayorga et al. (2010). EC_{DON} = 280 kg·km^{-2}·yr^{-1}. $f_{DON}(Rnat_j) = Rnat_j{}^{aDON}$ where a_{DON} is a model parameter to quantify annual runoff (unitless). $a_{DON} = 0.95$ (Strokal et al., 2016). [c] CR_{DON} is the export coefficient for runoff exporting nutrients from land to surface waters in a form of DON and derived from Li et al. (2022b) (footnote a for further description). $CR_{DON} = 0.02$ (unitless) for runoff ≥ 0.1 m·yr^{-1}, and $CR_{DON} = 0.01$ (unitless) for runoff < 0.1 m·yr^{-1}. [d] We assumed no direct discharge of the untreated wastewater from population (urban and rural) not connected to sewage systems. Thus, we considered the unconnected population (urban and rural) wastewater as input to the land ($WSdif_{N.hum.uncon.j}$) and adjusted the equation from Strokal et al. (2016). $fr_{NH3.hum}$ is the fraction of N losses from human excretion to the air (0–1) and derived from Mayorga et al. (2010). $fr_{NH3.hum} = 0.24$. [e] EC_{DIP} is coefficient for P weathering and derived from Mayorga et al. (2010). EC_{DIP} = 26 kg·km^{-2}·yr^{-1}. $f_{DIP}(Rnat_j) = [1 + (Rnat_j/a_{DIP})^{-bDIP}]^{-1}$ where a_{DIP} and b_{DIP} are model parameters to quantify annual runoff (unitless). $a_{DIP} = 0.85$ and $b_{DIP} = 2$ (Strokal et al., 2016). [f] CR_{DIP} is the export coefficient for runoff exporting nutrients from land to surface waters in a form of DIP and derived from Li et al. (2022b) (footnote a for further description). $CR_{DIP} = 0.15$ (unitless) for runoff ≥ 0.1 m·yr^{-1}, and $CR_{DIP} = 0.1$ (unitless) for runoff < 0.1 m·yr^{-1}. [g] EC_{DOP} is coefficient for leaching of organic matter and derived from Mayorga et al. (2010). EC_{DOP} = 15 kg·km^{-2}·yr^{-1}. $f_{DOP}(Rnat_j) = Rnat_j{}^{aDOP}$ where a_{DOP} is a model parameter to quantify annual runoff (unitless). $a_{DOP} = 0.95$ (Strokal et al., 2016). [h] CR_{DOP} is the export coefficient for runoff exporting nutrients from land to surface waters in a form of DOP and derived from Li et al. (2022b) (footnote a for further description). $CR_{DOP} = 0.02$ (unitless) for runoff ≥ 0.1 m·yr^{-1}, and $CR_{DOP} = 0.01$ (unitless) for runoff < 0.1 m·yr^{-1}. [i] We assumed no direct discharge of the untreated wastewater from population (urban and rural) not connected to sewage systems. Thus, we considered the unconnected population (urban and rural) wastewater as input to the land ($WSdif_{P.hum.uncon.j}$) and adjusted the equation from Strokal et al. (2016). [j] $FEpnt_{DIN.hum.con}$ is calculated for each basin according to the Global *NEWS-2* as a linear empirical function of N removal during treatment (Dumont et al., 2005; van Drecht et al., 2009; Mayorga et al., 2010) as: $FEpnt_{DIN.hum.con}$ = 0.485 + 0.225 x ($hw_{fremN.j}$ / 0.8) where 0.485 is the fraction of total N (TN) in sewage effluents that is DIN without treatment; 0.225 is the maximum increase in DIN/TN ratio that can be achieved during treatment; $hw_{fremN.j}$ is N removal during treatment in sewage systems

of each basin; and 0.8 is the maximum observed value for N treatment (represents Finland) (Strokal et al., 2016). In this study, the maximum observed value for N treatment was around 0.88 and that is used instead of "0.8" in the equation. [k] $FEpnt_{DON.hum.con}$ = 0.14 derived from the Global *NEWS-2* model (Mayorga et al., 2010). [l] $hw_{frem.N.j}$ is the removal fraction of N during treatment in sewage systems in basin j and calculated following the approach of Strokal et al. (2021). $hw_{frem.N.j}$ (0–1) is the same for urban and rural treatment systems. [m] $FEpnt_{DIP.hum.con}$ = 1 derived from the Global *NEWS-2* model (Mayorga et al., 2010; Strokal et al., 2016). [n] $FEpnt_{DOP.hum.con}$ = 0.010 derived from the Global *NEWS-2* model (Mayorga et al., 2010). [o] $hw_{frem.P.j}$ is the removal fraction of P during treatment in sewage systems in basin j and calculated following the approach of Strokal et al. (2021). $hw_{frem.P.j}$ (0–1) is the same for urban and rural treatment systems. [p] 1/6 is the ratio between human P and human N excretion based on measurements of 27 waste water treatment plants in Austria (van Drecht et al., 2009). [r] $WShw_{cap.P.det.j}$ is the consumption rate of P detergents (from laundry and washing) in basin j and derived from Strokal et al. (2021) (kg cap^{-1} yr^{-1}). [s] Data on the characteristics of reservoirs and (regulated) lakes were derived from HydroLAKES (Messager et al., 2016). $Q_{act.i}$ is actual water discharge for reservoir i ($km^3 \cdot yr^{-1}$) and derived from Messager et al. (2016) using the approach of Strokal et al. (2016). $D_{DIN.i}$ is the fraction of DIN retained in reservoir i (0–1) and calculated by the equation: $D_{DIN.i} = 0.8845 \times (h_i / \boxtimes_{R.i})^{-0.3677}$ using the approach of Strokal et al. (2016). h_i is depth of reservoir i (meter) and derived from Messager et al. (2016) using the approach of Strokal et al. (2016). $\boxtimes_{R.i}$ is water residence time for reservoir i (year) calculated by the equation: $\boxtimes_{R.i} = V_i / Q_{act.i}$ (Strokal et al., 2016). V_i is volume of reservoir i (km^3 yr^{-1}) and derived from Messager et al. (2016) using the approach of Strokal et al. (2016). The maximum value of $D_{DIN.j}$ is set as 0.965 since this is the maximum value of the basins used to calibrate parameters (Strokal et al., 2016). [t] 0.0605 and 0.0443 in the equation are fitted coefficients and derived from Strokal et al. (2016). $Area_j$ is drainage area of basin j (km^2) and derived from Strokal et al. (2016). The maximum value of $L_{DIN.j}$ is set as 0.65 using the approach of Strokal et al. (2016). [u] The value of $L_{DIP.j}$ is set as 0.9 for dry rivers having runoff of less than 0.1 m yr^{-1} and as 0.5 for other rivers using the approach of Strokal et al. (2016). [v] $Q_{act.i}$ is actual water discharge for reservoir i (km^3 yr^{-1}) and derived from Messager et al. (2016) using the approach of Strokal et al. (2016). $D_{DIP.j}$ is the fraction of DIP retained in reservoir i (0–1) and calculated by the equation: $D_{DIP.i} = 0.85 \times [1 - \exp(-0.0807 \times 365 \times \boxtimes_{R.i})]$ using the approach of Strokal et al. (2016). The maximum value of $D_{DIP.j}$ is set as 0.85 using the approach of Strokal et al. (2016).

Table S2.4 Descriptions of the abbreviations in Table S2.3 adjusted from Strokal et al. (2016).

Parameter	Description	Unit
E	Nutrient element: o N o P	-
F	Nutrient form: o Dissolved inorganic N and P (DIN, DIP) o Dissolved organic N and P (DON, DOP)	-
y	Source	-
j	Basin (sub-basins for Danube)	-
i	Reservoir	-
$RSdif_{F.y.j}$	Inputs of nutrient form F (DIN, DON, DIP and DOP) to rivers from a diffuse source y in basin j	kg·yr^{-1} N or P
$RSdif_{F.fe.j}$	Inputs of nutrient form F (DIN, DON, DIP and DOP) to rivers from synthetic fertilizer application (fe) in basin j	kg·yr^{-1} N or P
$RSdif_{F.ma.j}$	Inputs of nutrient form F (DIN, DON, DIP and DOP) to rivers from livestock manure application (ma) in basin j	kg·yr^{-1} N or P
$RSdif_{F.dep.ant.j}$	Inputs of DIN and DIP to rivers from atmospheric deposition (dep) over agricultural land (ant) in basin j	kg·yr^{-1} N or P
$RSdif_{DIN.fix.ant.j}$	Inputs of DIN to rivers from biological N_2 fixation (fix) by agricultural crops (ant) in basin j	kg·yr^{-1} N
$RSdif_{F.hum.uncon.j}$	Inputs of nutrient form F (DIN, DON, DIP and DOP) to rivers from human excretion (hum) unconnected to sewage systems (uncon) in basin j	kg·yr^{-1} N or P
$RSdif_{DIN.dep.nat.j}$	Inputs of DIN to rivers from atmospheric deposition (dep) over natural land (nat) in basin j	kg·yr^{-1} N
$RSdif_{DIN.fix.nat.j}$	Inputs of DIN to rivers from biological N_2 fixation (fix) by natural vegetation (nat) in basin j	kg·yr^{-1} N
$RSdif_{F.lch.ant.j}$	Inputs of DON and DOP to rivers from organic matter leaching (lch) over agricultural areas (ant) in basin j	kg·yr^{-1} N or P
$RSdif_{F.lch.nat.j}$	Inputs of DON and DOP to rivers from organic matter leaching (lch) over natural areas (nat) in basin j	kg·yr^{-1} N or P
$RSdif_{DIP.wth.ant.j}$	Inputs of DIP to rivers from P weathering (wth) in agricultural areas (ant) in basin j	kg·yr^{-1} P
$RSdif_{DIP.wth.nat.j}$	Inputs of DIP to rivers from P weathering (wth) in natural areas (nat) in basin j	kg·yr^{-1} P
$WSdif_{E.fe.j}$	Inputs of nutrient element E (N and P) from synthetic fertilizers (fe) that are applied to agricultural land in basin j	kg·yr^{-1} N or P
$WSdif_{E.ma.j}$	Inputs of nutrient element E (N and P) from animal manure (ma) that is applied to agricultural land in basin j	kg·yr^{-1} N or P
$WSdif_{E.dep.ant.j}$	Inputs of nutrient element E (N and P) from atmospheric deposition (dep) over agricultural land (ant) in basin j	kg·yr^{-1} N or P
$WSdif_{N.fix.ant.j}$	Inputs of N from biological N_2 fixation (fix) by agricultural crops (ant) in basin j	kg·yr^{-1} N
$WSdif_{N.dep.nat.j}$	Inputs of N from atmospheric deposition (dep) over natural land (nat) in basin j	kg·yr^{-1} N

Parameter	Description	Unit
$WSdif_{N.fix.nat.j}$	Inputs of N from biological N_2 fixation (fix) by natural vegetation (nat) in basin j	kg·yr^{-1} N
$WSdif_{E.hum.uncon.j}$	Inputs of nutrient element E (N and P) from excretion of population (hum) not connected to sewage systems (uncon) in basin j	kg·yr^{-1} N or P
$WSdif_{E.hum.uncon.urb.j}$	Inputs of nutrient element E (N and P) from excretion of urban population (urb) not connected to sewage systems (uncon) in basin j	kg·yr^{-1} N or P
$WSdif_{E.hum.uncon.rur.j}$	Inputs of nutrient element E (N and P) from excretion of rural population (rur) not connected to sewage systems (uncon) in basin j	kg·yr^{-1} N or P
$WSdif_{E.ex.j}$	Export (ex) of nutrient element E (N and P) from crop harvesting and animal grazing in basins j	kg·yr^{-1} N or P
$G_{DIN.j}$	Fraction of DIN that is remained in soil after correcting for crop harvesting and livestock grazing in basin j	0–1
$G_{DON.j}$	Fraction of DON that is remained in soil after correcting for crop harvesting and livestock grazing in basin j	0–1
$G_{DIP.j}$	Fraction of DIP that is remained in soil after correcting for crop harvesting and livestock grazing in basin j	0–1
$G_{DOP.j}$	Fraction of DOP that is remained in soil after correcting for crop harvesting and livestock grazing in basin j	0–1
$FE_{ws.DIN.j}$	Fraction of DIN that is exported from land to rivers in basin j	0–1
$FE_{ws.DON.j}$	Fraction of DON that is exported from land to rivers in basin j	0–1
$FE_{ws.DIP.j}$	Fraction of DIP that is exported from land to rivers in basin j	0–1
$FE_{ws.DOP.j}$	Fraction of DOP that is exported from land to rivers in basin j	0–1
CR_{DIN}	The export coefficient for runoff exporting nutrients from land to rivers in a form of DIN	Unitless
CR_{DON}	The export coefficient for runoff exporting nutrients from land to rivers in a form of DON	Unitless
CR_{DIP}	The export coefficient for runoff exporting nutrients from land to rivers in a form of DIP	Unitless
CR_{DOP}	The export coefficient for runoff exporting nutrients from land to rivers in a form of DOP	Unitless
$Rnat_j$	Annual surface runoff from land to streams in basin j	m· yr^{-1}
$Q_{nat.j}$	Natural (nat) river discharge at the outlet of basin j, before water is taken for consumption	km^3· yr^{-1}
$Area_j$	The drainage area of basin j	km^2
N_{ma}	Inputs of N from animal manure that is applied to agricultural land by the MITERRA-Europe model	kg·yr^{-1} N
N_{gr}	Inputs of N from animal manure that is stayed on agricultural land during grazing by the MITERRA-Europe model	kg·yr^{-1} N
P_{ma}	Inputs of P from animal manure that is applied to agricultural land by the MITERRA-Europe model	kg·yr^{-1} P
P_{gr}	Inputs of P from animal manure that is stayed on agricultural land during grazing by the MITERRA-Europe model	kg·yr^{-1} P
$Nexc_{hum.uncon.urb.j}$	N in human excretion from urban population unconnected to sewage systems in basin j	kg·yr^{-1} N

Parameter	Description	Unit
$Nexc_{hum.uncon.rur.j}$	N in human excretion from rural population unconnected to sewage systems in basin j	kg·yr^{-1} N
$fr_{NH3.hum}$	Fraction of N losses from human excretion to the air	0–1
$E^{N}_{hum.j}$	Human N excretion in basin j	kg·person^{-1}·yr^{-1}
$I^{N}_{hum.j}$	Protein N intake for basin j	gr·person^{-1}·day^{-1}
$GDP_{ppp.j}$	Annual gross domestic product (purchasing power parity in 2010)	US$·person^{-1}·yr^{-1}
$Pop_{uncon.urb.j}$	Urban population without sewage connection in basin j	People
$Pop_{uncon.rur.j}$	Rural population without sewage connection in basin j	People
$Pexc_{hum.uncon.urb.j}$	P in human excretion from urban population unconnected to sewage systems in basin j	kg·yr^{-1} P
$Pexc_{hum.uncon.rur.j}$	P in human excretion from rural population unconnected to sewage systems in basin j	kg·yr^{-1} P
$E^{P}_{hum.j}$	Human P excretion in basin j	kg·person^{-1}·yr^{-1}
$Agr_{fr.j}$	Fraction of agricultural area in basin j	0–1
EC_{DON}	The coefficient for leaching of organic matter for DON	kg·km^{-2}·yr^{-1}
EC_{DIP}	The coefficient for P weathering	kg·km^{-2}·yr^{-1}
EC_{DOP}	The coefficient for leaching of organic matter for DOP	kg·km^{-2}·yr^{-1}
$f_{DON}(Rnat_j)$	a function of annual runoff for DON from land to streams in basin j	Unitless
$f_{DIP}(Rnat_j)$	a function of annual runoff for DIP from land to streams in basin j	Unitless
$f_{DOP}(Rnat_j)$	a function of annual runoff for DOP from land to streams in basin j	Unitless
$RSpnt_{F.y.j}$	Inputs of nutrient form F (DIN, DON, DIP and DOP) to rivers from a point source y in basin j	kg·yr^{-1} N or P
$RSpnt_{DIN.hum.con.j}$	Inputs of DIN to rivers from urban and rural sewage systems in basin j	kg·yr^{-1} N
$RSpnt_{DON.hum.con.j}$	Inputs of DON to rivers from urban and rural sewage systems in basin j	kg·yr^{-1} N
$RSpnt_{DIP.hum.con.j}$	Inputs of DIP to rivers from urban and rural sewage systems in basin j	kg·yr^{-1} P
$RSpnt_{DOP.hum.con.j}$	Inputs of DOP to rivers from urban and rural sewage systems in basin j	kg·yr^{-1} P
$RSpnt_{N.hum.con.urb.j}$	N in sewage effluents that enter rivers from urban population in basin j	kg·yr^{-1} N
$RSpnt_{N.hum.con.rur.j}$	N in sewage effluents that enter rivers from rural population in basin j	kg·yr^{-1} N
$RSpnt_{P.hum.con.urb.j}$	P in sewage effluents that enter rivers from urban population in basin j	kg·yr^{-1} P
$RSpnt_{P.hum.con.rur.j}$	P in sewage effluents that enter rivers from rural population in basin j	kg·yr^{-1} P
$RSpnt_{P.det.con.urb.j}$	P from detergent consumption that enter rivers from urban sewage systems in basin j	kg·yr^{-1} P

Parameter	Description	Unit
$RSpnt_{P.det.con.rur.j}$	P from detergent consumption that enter rivers from rural sewage systems in basin j	kg·yr^{-1} P
$Nexc_{hum.con.urb.j}$	N in human excretion from urban population connected to sewage systems in basin j	kg·yr^{-1} N
$Nexc_{hum.con.rur.j}$	N in human excretion from rural population connected to sewage systems in basin j	kg·yr^{-1} N
$Pexc_{hum.con.urb.j}$	P in human excretion from urban population connected to sewage systems in basin j	kg·yr^{-1} P
$Pexc_{hum.con.rur.j}$	P in human excretion from rural population connected to sewage systems in basin j	kg·yr^{-1} P
$hw_{frem.N.j}$	Removal fraction of N during treatment in sewage systems in basin j (the same for urban and rural)	0–1
$hw_{frem.P.j}$	Removal fraction of P during treatment in sewage systems in basin j (the same for urban and rural)	0–1
$WShw_{cap.P.det.j}$	Consumption rate of P detergents (laundry and washing) in basin j	kg·cap^{-1}· yr^{-1}
$Pop_{con.urb.j}$	Urban population with sewage connection in basin j	People
$Pop_{con.rur.j}$	Rural population with sewage connection in basin j	People
$FEpnt_{DIN.hum.con.j}$	Fraction of DIN entering rivers from urban and rural sewage systems in basin j	0–1
$N_{total.y.j}$	Total emission of N to air by agricultural source y and basin j	kg·yr^{-1} N
$NH_{3.y.j}$	Total emission of NH_3 to air by agricultural source y and basin j	kg·yr^{-1} N
$N_2O_{y.j}$	Total emission of N_2O to air by agricultural source y and basin j	kg·yr^{-1} N
$NO_{X.y.j}$	Total emission of NO_x to air by agricultural source y and basin j	kg·yr^{-1} N
$M_{F.y.j}$	River export of nutrient form F (DIN, DON, DIP and DOP) to sea from source y	kg·yr^{-1} N or P
$FE_{riv.DIN.outlet.j}$	Fraction of DIN that is exported to the outlet of basin j	0–1
$FE_{riv.DON.outlet.j}$	Fraction of DON that is exported to the outlet of basin j	0–1
$FE_{riv.DIP.outlet.j}$	Fraction of DIP that is exported to the outlet of basin j	0–1
$FE_{riv.DOP.outlet.j}$	Fraction of DOP that is exported to the outlet of basin j	0–1
$D_{DIN.j}$	Fraction of DIN retained in reservoirs and lakes in basin j	0–1
$D_{DIP.j}$	Fraction of DIP retained in reservoirs and lakes in basin j	0–1
$L_{DIN.j}$	Fraction of DIN retained in or/and lost from water systems (e.g., denitrification) in basin j	0–1
$L_{DIP.j}$	Fraction of DIP retained in or/and lost from water systems (e.g., sedimentation) in basin j	0–1
$FQrem_j$	Fraction of nutrient form F (generic for DIN, DON, DIP and DOP) removed from water systems in basin j by water consumption	0–1
$Q_{act.j}$	Actual (act) water discharge at the outlet of basin j after water consumption	km^3· yr^{-1}
$FE_{riv.F.mouth.juT}$	Fraction of nutrient form F (DIN, DON, DIP and DOP) exported from the outlet of upstream tributary (juT) to the river mouth	0–1

Parameter	Description	Unit
$FE_{riv.F.mouth.jmT}$	Fraction of nutrient form F (DIN, DON, DIP and DOP) exported from the outlet of middlestream tributary (jmT) to the river mouth	0–1
$FE_{riv.F.mouth.jmC}$	Fraction of nutrient form F (DIN, DON, DIP and DOP) exported from the outlet of middlestream main channel (jmC) to the river mouth	0–1
$FE_{riv.F.mouth.jdC}$	Fraction of nutrient form F (DIN, DON, DIP and DOP) exported from the outlet of downstream main channel (jdC) to the river mouth	0–1
${}^{juT}FE_{riv.F.outlet.jmC}$ ${}^{juT}FE_{riv.F.outlet.jdC}$	Fractions of nutrient form F (DIN, DON, DIP and DOP) exported from the outlet of upstream tributary (upper case: juT) to the outlets of the main channel in middlestream (lower case: jmC) and downstream (lower case: jdC) sub-basins	0–1
${}^{jmT}FE_{riv.F.outlet.jdC}$	Fraction of nutrient form F (DIN, DON, DIP and DOP) exported from the outlet of middlestream tributary (upper case: jmT) to the outlet of the main channel in downstream (lower case: jdC) sub-basins	0–1
${}^{jmC}FE_{riv.F.outlet.jdC}$	Fraction of nutrient form F (DIN, DON, DIP and DOP) exported from the outlet of middlestream subbasin with the main channel (upper case: jmC) to the outlet of the main channel in downstream (lower case: jdC) sub-basin (the outlet of the this downstream sub-basin is the river mouth)	0–1
$D_{F.jmC}$ $D_{F.jdC}$	Fractions of DIN and DIP retained in reservoirs of middlestream (jmC) and downstream (jdC) sub-basins with the main channel (C). These fractions were calculated for sub-basins using equations for $D_{DIN,j}$ and $D_{DIP,j}$ from Table S2.3.	0–1
$L_{F.jmC}$ $L_{F.jdC}$	Fractions of DIN and DIP that is lost from surface waters of middlestream (jmC) and downstream (jdC) sub-basins with the main channel (C). These fractions were calculated using the equation for $L_{DIN,j}$ and $L_{DIP,j}$ from Table S2.3.	0–1
$FQrem_{jmC}$ $FQrem_{jdC}$	Fractions of nutrient form F (generic for DIN, DON, DIP and DOP) that are lost from surface waters of middlestream (jmC) and downstream (jdC) sub-basins with the main channel (C) via water consumption. These fractions were calculated using equations for $FQrem_j$ from Table S2.3.	0–1
${}^{juT}A_{jmC}$ ${}^{juT}A_{jdC}$	Drainage area (A) of the main channel (C) in middlestream (lower case: jmC) and downstream (lower case: jdC) sub-basins that exports nutrients from the outlet of upstream tributary (upper case: juT). This drainage area was calculated as the fraction to the total sub-basin area.	0–1
${}^{jmT}A_{jdC}$	Drainage area (A) of the main channel (C) in downstream (lower case: jdC) sub-basin that exports nutrients from the outlet of middlestream tributary (upper case: jmT). This drainage area was calculated as the fraction to the total sub-basin area.	0–1
${}^{jmC}A_{jdC}$	Drainage area (A) of the main channel (C) in the downstream (lower case: jdC) subbasin that exports nutrients from the outlet of middlestream main channel (upper case: jmC). This drainage area was calculated as the fraction to the total sub-basin area.	0–1

Table S2.5 Description of how model inputs are processed to basins for the MARINA-Nutrients model of this study.

Equations from Table S3	Description of how model inputs are processed to basins
Eq. 2, 11, 12, 19, 26, 34, 35, 42	Synthetic fertilizer application to agricultural land was derived from the MITERRA-Europe model outputs (2017) which were calculated by the CAPRI model per crop type (Duan et al., 2021). In this study, the synthetic fertilizer application was converted from the NUTS2 regions to basins and sub-basins (for Danube). First, the synthetic fertilizer applied to each NUTS2 region (kg) was divided by the area of this region (km^2) to get the synthetic fertilizer application per NUTS2 region ($kg{\cdot}km^{-2}$ N or P). Second, the regional data was assigned to 0.5° resolution ($kg{\cdot}km^{-2}$ N or P per grid). Third, the gridded synthetic fertilizer application ($kg{\cdot}km^{-2}$ N or P) was multiplied by the grid area (km^2) to get synthetic fertilizer application per grid (kg N or P). Finally, the synthetic fertilizer application was summed for basins over corresponding grids to get the total amount of synthetic fertilizer applied per basin ($kg{\cdot}yr^{-1}$ N or P).
Eq. 3, 11, 13, 19, 27, 34, 36, 42	Animal manure application on agricultural land and manure deposited during grazing were derived from the MITERRA-Europe model outputs (2017) which were calculated as a function of excretion rates per animal type, and number of animals in housing and grazing systems (Duan et al., 2021). We accounted for losses of N and P during storage (e.g., denitrification of N, leaching of N and P) before manure application. In this study, the animal manure application was converted from the NUTS2 regions to basins and sub-basins (for Danube). First, the animal manure (sum of manure applied and stayed on agricultural land during grazing) for each NUTS2 region (kg) was divided by the area of this region (km^2) to get the animal manure application per NUTS2 region ($kg{\cdot}km^{-2}$). Second, the regional data was assigned to 0.5° resolution ($kg{\cdot}km^{-2}$ per grid). Third, the gridded animal manure application ($kg{\cdot}km^{-2}$) was multiplied by the grid area (km^2) to get animal manure application per grid (kg N or P). Finally, the animal manure application was summed for basins over corresponding grids to get the total amount of animal manure applied per basin ($kg{\cdot}yr^{-1}$ N or P).
Eq. 4, 11, 28, 34	Atmospheric deposition on agricultural land was derived from the MITERRA-Europe model outputs (Duan et al., 2021). In this study, the atmospheric deposition was converted from the NUTS2 regions to basins and sub-basins (for Danube). First, the atmospheric deposition for each NUTS2 region (kg) was divided by the area of this region (km^2) to get the atmospheric deposition per NUTS2 region ($kg{\cdot}km^{-2}$). Second, the regional data was assigned to 0.5° resolution ($kg{\cdot}km^{-2}$ per grid). Third, the gridded atmospheric deposition ($kg{\cdot}km^{-2}$) was multiplied by the grid area (km^2) to get atmospheric deposition per grid (kg N or P). Finally, the atmospheric deposition was summed for basins over corresponding grids to get the total amount of atmospheric deposition per basin ($kg{\cdot}yr^{-1}$ N or P).
Eq. 5, 11	Biological N_2 fixation by agricultural crops was derived from the MITERRA-Europe model outputs (Duan et al., 2021). In this study, the biological N_2 fixation was converted from the NUTS2 regions to basins and sub-basins (for Danube). First, the biological N_2 fixation for each NUTS2 region (kg) was divided by the area of this region (km^2) to get the biological N_2 fixation application per NUTS2 region ($kg{\cdot}km^{-2}$). Second, the regional data was assigned to 0.5° resolution ($kg{\cdot}km^{-2}$ per grid). Third, the gridded biological N_2 fixation ($kg{\cdot}km^{-2}$) was multiplied by the grid area (km^2) to get biological N_2 fixation per grid (kg N). Finally, the biological N_2 fixation application was summed for basins over corresponding grids to get the total amount of biological N_2 fixation per basin ($kg{\cdot}yr^{-1}$ N).

Equations from Table S3	Description of how model inputs are processed to basins
Eq. 7	Atmospheric deposition over non-agricultural land (kg·ha^{-1} N) was derived from the Integrated Model to Assess the Global Environment (IMAGE) model outputs (2010) which were in 0.5° resolution (Beusen et al., 2016). In this study, the atmospheric deposition was aggregated from 0.5°grids to basins and sub-basins (for Danube). First, the atmospheric deposition (kg·ha^{-1} N per grid) was multiplied by grid area (km^2) and by fraction of non-agricultural area per grid (0–1) to get total atmospheric deposition per grid (kg N). Then, the atmospheric deposition was summed for basins over corresponding grids to get the total amount of atmospheric deposition per basin (kg·yr^{-1} N).
Eq. 8	Biological N_2 fixation by natural vegetation (kg·ha^{-1} N) was derived from the IMAGE model outputs (2010) which were in 0.5° resolution (Beusen et al., 2016). In this study, the biological N_2 fixation was aggregated from 0.5°grids to basins and sub-basins (for Danube). First, the gridded biological N_2 fixation (kg·ha^{-1} N) was multiplied by grid area (km^2) and by fraction of non-agricultural area per grid (0–1) to get biological N_2 fixation per grid (kg N). Then, the biological N_2 fixation was summed for basins over corresponding grids to get the total amount of biological N_2 fixation per basin (kg·yr^{-1} N).
Eq. 15, 16, 38, 39	Leaching of organic matter from agricultural and non-agricultural soils was calculated by using model parameters from Mayorga et al. (2010).
Eq. 30, 31	Weathering of P-contained minerals on agricultural and natural land was calculated by using model parameters from Mayorga et al. (2010).
Eq. 9, 17, 32, 40	Exports of N and P from agricultural land by crop harvesting and animal grazing were derived from the MITERRA-Europe model outputs (2017) (Duan et al., 2021). The MITERRA-Europe model calculates N and P in harvested grassland and cropland (Velthof et al., 2009). In this study, the nutrient exports by crop harvesting and animal grazing were converted from the NUTS2 regions to basins and sub-basins (for Danube). First, the nutrient exports for each NUTS2 region (kg) were divided by the area of this region (km^2) to get the nutrient exports per NUTS2 region (kg·km^{-2}). Second, the regional data was assigned to 0.5° resolution (kg·km^{-2} per grid). Third, the gridded nutrient exports (kg·km^{-2}) were multiplied by the grid area (km^2) to get nutrient exports per grid (kg N or P). Finally, the nutrient exports were summed for basins over corresponding grids to get the total amount of nutrient exports per basin (kg·yr^{-1} N or P).
Eq. 49, 81	Natural water discharges were derived from Variable Infiltration Capacity (VIC) macroscale hydrological model (van Vliet et al., 2016). The water discharges were 30-year average data (1981-2010) for 2010 to avoid the influence of climate extremes, and aggregated from the scale of 0.5° grids to the basins. The water consumption (water use) rates were based on the gridded data of the Global *NEWS-2* model (Fekete et al., 2010; Mayorga et al., 2010). Information on the characteristics of reservoirs and (regulated) lakes (e.g., volume) were derived from HydroLAKES (Messager et al., 2016). Actual water discharges (after water consumption) were calculated by using the water consumption rates by (Strokal et al., 2019).
Eq. 24, 25, 47, 48, 56, 57, 66, 67, 70, 71	Total, urban and rural populations were derived from the NCAR database in 0.125° resolution (Gao, 2017). Population data was projected to 2020 based on SSP2 scenario (van Puijenbroek et al., 2023). First, the total, urban and rural populations were aggregated from 0.125° to 0.5° grids. Then, the total, urban and rural populations were summed for basins over corresponding grids to get the total, urban and rural populations per basin (people·yr^{-1}).

Equations from Table S3	Description of how model inputs are processed to basins
Eq. 56, 57, 66, 67, 70, 71	Inputs of pollutants from sewage systems were quantified as a function of the population that is connected to sewage systems (0–1), the excretion rates of pollutants per person per year (kg·person^{-1}·yr^{-1}) and removal efficiencies of pollutants during treatment (0–1). The data from country scale was converted to 0.5×0.5° gridded format and aggregated to basins and sub-basins (for Danube) following the approach of Strokal et al. (2021). Fractions of urban and rural population connected to sewage systems were derived from the literature (van Puijenbroek et al., 2023). The country scale projections for 2020 were used based on the SSP2-Moderate (SSP2-M) scenario (van Puijenbroek et al., 2023). This dataset was consistent in terms of urban connection rates with the Joint Monitoring Program (JMP) dataset for 2020 based on national reports (WHO/UNICEF, 2021). However, the dataset of van Puijenbroek et al. (2023) had some inconsistencies in rural connection rates of some countries (0% values). Therefore, we considered the JMP data for those countries (e.g., Croatia, Cyprus, Finland, Ireland, Portugal, Sweden) prior to modeling (WHO/UNICEF, 2021). For rural connection rates of Austria, Bulgaria, Czechia, Estonia, Hungary, Italy, Latvia, Lithuania, Slovakia, UK; JMP data was higher than the SSP2-M 2020 projections. Thus, the JMP data was used for those countries (WHO/UNICEF, 2021). Nearly 43% of the rural data was taken from van Puijenbroek et al. (2023) projections for 2020 based SSP2-M scenario, and the rest (57%) was from the JMP data (WHO/UNICEF, 2021). First, the country scale sewage system connection (SC) rates for urban and rural population were assigned to 0.5° resolution (SC rates per grid). Second, the gridded fractions were multiplied by the total population (urban and rural) per grid to get the urban and rural populations with SC per grid. Third, the gridded urban and rural populations with SC were summed for basins over corresponding grids to get the urban and rural populations with SC per basin. Forth, the urban and rural populations with SC were divided by the urban and rural populations per basin. The outcome was the faction of urban population with SC per basin (0–1) and the faction of rural population with SC per basin (0–1).
Eq. 59	Data on gross domestic product at purchasing power parity (GDPppp) was derived from the literature (van Puijenbroek et al., 2023). The country scale projection for 2020 was used based on the SSP2-M scenario (van Puijenbroek et al., 2023). First, the country scale GDPppp was assigned to 0.5° resolution (US$·capita^{-1}·yr^{-1}per grid). Second, the gridded GDPppp was multiplied by the total number of people per grid (US$·yr^{-1}). Third, the GDPppp was summed for basins over corresponding grids to get the GDPppp per basin (US$·yr^{-1}). Finally, the basin GDPppp values were divided by the total population per basin to get the GDPppp per capita per basin (US$·capita^{-1}·yr^{-1}). The unit of GDPppp was adjusted to US$2010 (US$2010·capita^{-1}·yr^{-1}) (Strokal et al., 2019).
Eq. 24, 25, 47, 48, 56, 57, 58, 66, 67, 68, 70, 71	Excretion or consumption rates of N and P per capita were calculated as a function of the GDPppp following the approach of van Drecht et al. (2009). The consumption rate of P in detergents (laundry and washing) was taken from van Drecht et al. (2009) for 2000. First, this consumption rate was assigned to 0.5° resolution and used for 2010. Second, the gridded consumption rates of P in detergents was multiplied by the total number of people per grid (kg·yr^{-1} P). Third, the P from detergents consumed by total population per grid was summed for basins over corresponding grids to get the P from detergent consumption by total population per basin (kg·yr^{-1} P). Finally, the P from detergents consumption by total population per basin (kg·yr^{-1} P) was divided by the total population per basins to get the P from detergent consumption per capita per basin (kg·capita^{-1}·yr^{-1} P).

Equations from Table S3	Description of how model inputs are processed to basins
Eq. 54, 55, 60, 64, 65, 70, 71	Fractions of N and P removal in wastewater treatment systems (the same for both urban and rural) were derived from the literature (van Puijenbroek et al., 2023). The country scale fractions of N and P removal in wastewater treatment systems were calculated by using removal efficiencies (pollutant dependent) and fractions of people with treatment types per country from the updated version of van Puijenbroek et al. (2023). First, the country scale fractions of N and P removal in wastewater treatment systems were assigned to 0.5° resolution (fractions of N and P removal per grid). Second, the gridded per capita human excretion ($kg{\cdot}capita^{-1}{\cdot}yr^{-1}$ N or P) was multiplied by the population (urban and rural) with SC per grid (people) to get the N and P entering sewage systems per grid ($kg{\cdot}yr^{-1}$ N or P). Third, the N and P entering sewage systems were summed for basins over corresponding grids ($kg{\cdot}yr^{-1}$ N or P per basin). Fourth, the N and P entering sewage systems per basin was multiplied by removal fractions per grid to get the N and P removed during treatment per grid ($kg{\cdot}yr^{-1}$ N or P). Fifth, the N and P removed during treatment were summed for basins over corresponding grids ($kg{\cdot}yr^{-1}$ N or P per basin). Finally, the N and P removed during treatment per basin were divided by the amount of N and P entering sewage systems per basin to get the removal fractions of N and P in wastewater treatment systems (hw_{fremN} and hw_{fremP}) per basin (0–1).
Eq.72	Reactive N emissions to air (i.e. NH_3, N_2O, NO_x) from animal housing and storage systems were derived from the MITERRA-Europe model outputs (2017) which were calculated by using emissions factors (Duan et al., 2021). Reactive N emissions from animal housing and storage systems included NH_3, N_2O and NO_x emissions. In this study, each N emission to air was converted from the NUTS2 regions to basins and sub-basins (for Danube). First, the N emissions for each NUTS2 region (kg) were divided by the area of this region (km^2) to get the N emissions to air per NUTS2 region ($kg{\cdot}km^{-2}$ N). Second, the regional was assigned data to 0.5° resolution ($kg{\cdot}km^{-2}$ N per grid). Third, the gridded N emissions ($kg{\cdot}km^{-2}$ N) were multiplied by the grid area (km^2) to get N emissions to air per grid (kg N). Finally, the N emissions were summed for basins over corresponding grids to get the total amount of N emissions to air per basin ($kg{\cdot}yr^{-1}$ N). The N emissions to air from animal housing and storage systems were considered in total N emissions to air from agriculture per basin ($kg{\cdot}yr^{-1}$ N). Reactive N emissions to air from agricultural soils (i.e. NH_3, N_2O, NO_x) were derived from the MITERRA-Europe model outputs (2017) which were calculated by using emissions factors (Duan et al., 2021). Reactive N emissions from agricultural soil contained NH_3 emissions from applications of synthetic fertilizer and animal manure, grazing; N_2O emissions from applications of synthetic fertilizer and animal manure, grazing, biological fixation, cultivated peatland and indirect N_2O emissions (i.e. from N leaching and ammonia); and NO_x emissions from applications of synthetic fertilizer and animal manure, grazing. In this study, each N emission to air was converted from the NUTS2 regions to basins and sub-basins (for Danube). First, the N emissions for each NUTS2 region (kg) were divided by the area of this region (km^2) to get the N emissions to air per NUTS2 region ($kg{\cdot}km^{-2}$ N). Second, the regional data was assigned to 0.5° resolution ($kg{\cdot}km^{-2}$ N per grid). Third, the gridded N emissions ($kg{\cdot}km^{-2}$ N) were multiplied by the grid area (km^2) to get N emissions to air per grid (kg N). Finally, the N emissions were summed for basins over corresponding grids to get the total amount of N emissions to air per basin ($kg{\cdot}yr^{-1}$ N). To present the total reactive N emissions from agriculture, we summed the N emissions from animal housing and storage systems and from agricultural soil.

Note: The model equations and data sources are described in Table S2.3, Table S2.4, and Table S2.6.

Table S2.6 Sources of the MITERRA-Europe model inputs.

Input data description	Years	Source
Fertilizer use data at country level in the EU28	2016–2018	(FAOSTAT, 2022)
Natural grassland data at NUTS2 level	2016–2018	(Eurostat, 2019a)
Animal numbers, crop areas and crop yields[a]	2015	CAPRI model based on Eurostat (Britz & Witzke, 2014)
Soil data on coarse fragments, texture, pH, organic C content, bulk density and data on perennial grass cover		ESDB and LUCAS (Tóth et al., 2013)
N excretion of animals and CH_4 emissions from manure management and enteric fermentation	2017	Common Reporting formats (CRFs) database (UNFCCC, 2019)
Arable farm size, farming system, crop rotation, livestock units, and areas with organic farming, irrigation, crop cover, and grass cover	2016–2018	(Eurostat, 2019b)
N deposition	2010	(EMEP, 2010)
NH_3 emission factors and the implementation of NH_3 mitigation measures on national level		Greenhouse Gas - Air Pollution Interactions and Synergies (GAINS) model (Amann et al., 2011; IIASA, 2018)
N_2O emission factors and global warming potentials		Reports of the Intergovernmental Panel on Climate Change (IPCC) for National Greenhouse Gas Inventories (IPCC, 2006)
NOx emission factors		(Velthof et al., 2014)
N surface runoff fraction: Fraction of soil N input by inorganic and organic fertilizers, calculated as a function of slope class, land use, precipitation surplus, soil type and depth to rock		(Velthof et al., 2009)
N leaching fraction: Fraction of soil N surplus, calculated as a function of soil type, land use, SOC content, precipitation surplus, temperature and rooting depth		(Velthof et al., 2009)

Note: a Most of the data are derived from Eurostat, but a gap filling procedure and consistency check is applied (Duan et al., 2021).

Table S2.7 Description of the data used for model validation.

Parameters	Type	Number of basins	Number of observations	Years	Explanation	Reference
DIN, DON, TDN, DIP, TDP	Concentration	45	148	2000–2017	We calculated yearly average concentrations ($mg \cdot L^{-1}$) out of weekly/monthly measurements prior to comparing the annual model results. We considered only the stations close to or at the river mouth for the study area. Measured loads ($kg \cdot yr^{-1}$) were calculated from concentrations multiplying water discharges. Measured yields ($kg \cdot km^{-2} \cdot yr^{-1}$) were calculated from concentrations dividing basin areas and multiplying water discharges.	(UNEP, 2022)
DIN, DON, DIP, DOP	Yield	19	34	1990–2000	We compared our annual river export yields ($kg \cdot km^{-2} \cdot yr^{-1}$) by the observed yields of DIN, DON, DIP and DOP ($kg \cdot km^{-2} \cdot yr^{-1}$) that were used to validate a nutrient model including Europe (Global *NEWS-2* model) for the period of 1990–2000. Measured loads ($kg \cdot yr^{-1}$) were calculated from yields multiplying basin areas. Measured concentrations ($mg \cdot L^{-1}$) were calculated from yields multiplying by basin areas and dividing water discharges.	(Mayorga et al., 2010)
DIN, TN, DIP, TP	Load	4	16	1990–2012	We compared total N (TN) and total P (TP) loads ($kg \cdot yr^{-1}$) with the loads of total dissolved N (TDN) and total dissolved P (TDP) from the MARINA-Nutrients model. Our model did not calculate the particulate forms of N (PN) and P (PP) (TN is the sum of DIN, DON and PN; TP is the sum of DIP, DOP and PP). Measured yields ($kg \cdot km^{-2} \cdot yr^{-1}$) were calculated from loads dividing basin areas. Measured concentrations ($mg \cdot L^{-1}$) were calculated from loads dividing water discharges.	(Cozzi et al., 2018)
DIN, DON, TN, DIP, TP	Load	1	9	1980–1993, 2001	See the explanation above for TN and TP, and for the calculations of yield and concentration from load.	(Ylöstalo et al., 2016)
TN, TP	Load	1	4	1997–2003, 2010–2014	See the explanation above for TN and TP, and for the calculations of yield and concentration from load.	(Friedland et al., 2019)
DIN, DIP	Concentration	1	2	2001–2010	Measured loads ($kg \cdot yr^{-1}$) were calculated from concentrations multiplying water discharges. Measured yields ($kg \cdot km^{-2} \cdot yr^{-1}$) were calculated from concentrations dividing basin areas and multiplying water discharges.	(Hesse & Krysanova, 2016)

Parameters	Type	Number of basins	Number of observations	Years	Explanation	Reference
TN, TP	Load	5	10	1995	See the explanation above for TN and TP, and for the calculations of yield and concentration from load.	(Räike et al., 2020)
DIN, DON, TN, DIP, DOP, TP	Load	1	30	2012–2016	See the explanation above for TN and TP, and for the calculations of yield and concentration from load.	(Vybernaite-Lubiene et al., 2018)
TN, TP	Load	5	10	1995–2014	See the explanation above for TN and TP, and for the calculations of yield and concentration from load.	(Räike et al., 2018)

Note: Number of basins indicates the river basins where observational data are available. Number of observations indicates total number of observations per data source (observation per parameter per year). Year indicates the period of the measurements or observed data (i.e. load, yield). Explanation describes our methods to process the data for model validation.

Table S2.8 Inputs of N and P to land, and their export from agricultural land by crop harvesting and animal grazing.

Inputs to land and export from agricultural land	**N ($Tg \cdot yr^{-1}$)**	**P ($Tg \cdot yr^{-1}$)**
fertilizer application	14	0.63
manure	10.2	0.96
excretion of population not connected to sewage systems	0.29	0.064
biological fixation by crops	1	-
atmospheric deposition over agricultural areas	3.7	0.096
biological fixation by natural vegetation	1.6	-
atmospheric deposition over non-agricultural areas	3.6	-
nutrient export from agricultural land by crop harvesting and animal grazing	18	1.2

Note: Source: The MITERRA-Europe model (Duan et al., 2021) for agricultural land and the IMAGE model (Beusen et al., 2016) for non-agricultural land.

Table S2.9 Comparison of NH_3, N_2O and NO_x emissions with other modeling studies for Europe ($Gg \cdot yr^{-1}$ N).

		This study	Other studies			
N source	**Emission source**	**EU28 (2017)**	**EU25 (2002)[a]**	**EU27 (2000)[b]**	**EU27 (2000)[c]**	**EU27 (2000)[d]**
NH_3	Housing and storage systems	1487	1428	1189	1279	1048
	Applied fertilizer	677	678	1413	540	798
	Applied manure	959	759		823	683
	Grazing	277	201	271	231	319
	Total	**3401**	**3066**	**2873**	**2873**	**2848**
N_2O	Housing and storage systems	22	48	55	54	52
	N applied	217[e]	316[f]	242[f]	208[f]	289[f]
	Grazing	67	67	124	66	92
	Indirect emissions[g]	67	80	43	51	76
	Total	**374**	**511**	**464**	**379**	**510**
NO_x	Housing and storage systems	25	32	20	20	0
	N applied	130[h]	16	123	28	23
	Grazing	-	59	63	29	196
	Total	**155**	**107**	**206**	**77**	**219**

Note: [a] Results are estimated by IDEAg model in the EU-25 for the year 2002 (Leip et al., 2009). [b] Results are estimated by INTEGRATOR model in the EU-27 for the year 2000 (de Vries et al., 2011). NH_3 emissions from applied fertilizer include emissions through fertilizer and manure applications (de Vries et al., 2011). [c] Results are estimated by MITERRA model in the EU-27 for the year 2000 (de Vries et al., 2010). [d] Results are estimated by IMAGE model in the EU-27 for the year 2000 (Bouwman et al., 2006). [e] In this study, N applied includes N_2O emissions from synthetic fertilizer and manure applications. [f] N applied includes N_2O emissions through synthetic fertilizer and manure applications, deposition, mineralization, fixation and crop residues (de Vries et al., 2011). [g] Indirect N_2O emissions include N_2O emissions from N leaching and ammonia. [h] In this study, NO_x emissions from the soil (e.g., synthetic fertilizer and manure applications) are indicated as N applied.

Table S2.10 Share of diffuse and point sources in river export of N and P in Europe ($Gg \cdot yr^{-1}$ N or P).

River export to seas		Arctic Ocean	Atlantic Ocean	Baltic Sea	Black Sea	Mediterranean Sea	North Sea
TDN	Total export	35	759	456	286	595	576
	Diffuse sources	34.5	672	412	241	481	487
	Point sources	0.6	87	44	45	114	89
TDP	Total export	1.6	29	19	11	31	21
	Diffuse sources	1.5	14	11	6	10	8
	Point sources	0.1	15	8	5	21	13

Note: Source: The new MARINA-Nutrients model for Europe.

Table S2.11 Comparison of N budget of agricultural land in the EU including N emissions from housing systems and soil (NH_3, N_2O, NO_x and N_2) ($kg \cdot ha^{-1} \cdot yr^{-1}$ N).

Source	MITERRA-**Europe** (2017)	INTEGRATOR (2010)
Fertilizer	63.5	72
Excretion	57	54
Deposition	17	10
Biological N fixation	4.5	7
Total N input	142	145[a]
Crop uptake	82	92
Emissions to air (NH_3, N_2O, NO_x)	18	19
Losses to surface waters by runoff	9	8
Denitrification (N_2)	26	17
Leaching to groundwater	7	9
Total N output	142	145

Note: Source: The MITERRA-Europe model. [a] Results are estimated using INTEGRATOR model for total agricultural land (cropland and grassland) in the EU (including the UK) for the year 2010 (de Vries et al., 2021). Total N input includes biosolids ($1 kg \cdot ha^{-1} \cdot yr^{-1}$) and mineralization ($1 kg \cdot ha^{-1} \cdot yr^{-1}$).

Table S2.12 Comparison of annual total dissolved N (TDN) export to European seas by the source with other modeling studies for Europe ($Gg \cdot yr^{-1}$ N).

Pollution source	**This study**	**MARINA-Nutrients (Global, version 1.0 with IMAGE data)**	**MARINA-Nutrients (Global, version 1.0 with MAgPIE data)**	**GREEN**	**IMAGE-GNM**	**Global *NEWS-2***	**Global *NEWS-2***
	2017-2020	2010	2010	2012	2000	2000	1995
Sewage systems	379	444	444	914			
Population unconnected to sewage systems	12	6	6	84			
Agriculture	1435	1243	1407	1900			
Atmospheric deposition (natural)	374	374	346	964			
Biological fixation (natural)	158	158	131				
Organic matter leaching (natural)	343	338	326				
Total	**2701**[a]	**2563**[b]	**2660**[c]	**3862**[d]	**3038**[e]	**2518**[f]	**2300**[g]

Note: [a] Results are estimated by MARINA-Nutrients model as the river export of total dissolved N (TDN) for the period 2017–2020. TDN is the sum of dissolved inorganic N (DIN) and dissolved organic N (DON). [b] Results are estimated by MARINA-Global-IMAGE model as the river export of total dissolved N (TDN) for the year 2010 (Beusen et al., 2016). [c] Results are estimated by MARINA-Global-MAgPIE model as the river export of total dissolved N (TDN) for the year 2010. [d] Results are estimated by GREEN model as the river export of total N (TN) to the European seas for the year 2012 (Grizzetti et al., 2021). Atmospheric deposition is the sum of anthropogenic and natural, and sewage systems include industrial wastewater as well. [e] Results are estimated by IMAGE-GNM model as the river export of total N (TN) for the year 2000 (Beusen et al., 2015). [f] Result is estimated by Global NEWS-2 model as the river export of total dissolved N (TDN) for the year 2000 (Mayorga et al., 2010). [g] Result is estimated by Global NEWS-2 model as the river export of total dissolved inorganic N (DIN) for the year 1995 (Dumont et al., 2005).

Table S2.13 Comparison of annual total dissolved P (TDP) export to European seas by the source with other modeling studies for Europe ($Gg \cdot yr^{-1}$ P).

Pollution source	**This study**	**MARINA-Nutrients (Global, version 1.0 with IMAGE data)**	**GREEN**	**GREEN**	**IMAGE-GNM**	**Global *NEWS***
	2017-2020	2010	2012	2005	2000	2000
Sewage systems	64	81	142			
Population unconnected to sewage systems	0.5	1	17			
Agriculture	27	23	88			
P weathering (natural)	4	4	44			
Organic matter leaching (natural)	18	18				
Total	**114**[a]	**127**[b]	**290**[c]	**282**[d]	**328**[e]	**235**[f]

Note: a Results are estimated by MARINA-Nutrients model as the river export of total dissolved P (TDP) for the period 2017-2020. TDP is the sum of dissolved inorganic P (DIP) and dissolved organic P (DOP). [b] Results are estimated by MARINA-Global-IMAGE model as the river export of total dissolved P (TDP) for the year 2010 (Beusen et al., 2016). [c] Results are estimated by GREEN model as the river export of total P (TP) to the European seas for the year 2012 (Grizzetti et al., 2021). Sewage systems include industrial wastewater as well. [d] Result is estimated by GREEN model as the river export of total P (TP) to the European seas (i.e. Atlantic Sea, Baltic Sea, Black Sea, Mediterranean Sea, North Sea) for the year 2005 (Grizzetti et al., 2012). Black Sea includes discharges from Turkey. [e] Results are estimated by IMAGE-GNM model as the river export of total P (TP) for the year 2000 (Beusen et al., 2015). [f] Result is estimated by Global *NEWS-2* model as the river export of total dissolved P (TDP) for the year 2000 (Mayorga et al., 2010).

S3: Supplementary materials for Chapter 3

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Supplementary information

Large reductions in nutrient losses needed to avoid future coastal eutrophication across Europe

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The Supplementary information contains

Supplementary Text S3.1

Boxes S3.1-S3.2

Tables S3.1-S3.2

Figure S3.1

Supplementary Text S3.1 Study area

We studied 594 rivers (593 basins and 8 sub-basins of the Danube River) in the European Union including the United Kingdom (EU-28) (see details in Ural-Janssen et al. (2023)). These rivers discharge into six seas/oceans namely, the Arctic Ocean, Atlantic Ocean, Baltic Sea, Black Sea, Mediterranean Sea and North Sea. The Danube River includes eight sub-basins (i.e. Upstream, Drava, Sava, Tisza, Siret, Prut, Middlestream and Downstream delta) and covers 14% (797,289 km^2) of the study area (5,768,449 km^2) (Ural-Janssen et al., 2023).

Box S3.1 Summarized equations of the MARINA-Nutrients model for Europe to quantify river export of nitrogen (N) and phosphorus (P) to European seas from diffuse and point sources (see details in Ural-Janssen et al. (2023)). Parameters are described in Box S3.2.

Equation	Number
$RSdif_{F.lech.ant.j} = Area_j \times Agr_{fr.j} \times EC_F \times CL_F Rnat_j$ for DON and DOP, used in Eq.1 in the paper	Eq.S1
$RSdif_{F.lech.nat.j} = Area_j \times (1 - Agr_{fr.j}) \times EC_F \times CL_F Rnat_j$ for DON and DOP, used in Eq.1 in the paper	Eq.S2
$RSdif_{DIP.wth.ant.j} = Area_j \times Agr_{fr.j} \times EC_{DIP} \times CW_{DIP} Rnat_j$ for DIP, used in Eq.1 in the paper	Eq.S3
$RSdif_{DIP.wth.nat.j} = Area_j \times (1 - Agr_{fr.j}) \times EC_{DIP} \times CW_{DIP} Rnat_j$ for DIP, used in Eq.1 in the paper	Eq.S4
$Rnat_j = Q_{nat.j} / Area_j$ used in Eq.5 in the paper	Eq.S5
$WSdif_{F.gross.j} = WSdif_{E.fe.j+} WSdif_{E.ma.j+} WSdif_{E.dep.ant.j+} WSdif_{E.fix.ant.j} + WSdif_{E.hum.uncon.j}$ for DIN, used in Eq.4 in the paper	Eq.S6
$WSdif_{F.gross.j} = WSdif_{E.fe.j+} WSdif_{E.ma.j+} WSdif_{E.dep.ant.j+} WSdif_{E.hum.uncon.j}$ for DIP, used in Eq.4 in the paper	Eq.S7
$WSdif_{F.gross.j} = WSdif_{E.fe.j+} WSdif_{E.ma.j+} WSdif_{E.hum.uncon.j}$ for DON and DOP, used in Eq.4 in the paper	Eq.S8
$WSdif_{E.hum.uncon.j} = WSdif_{E.hum.uncon.urb.j} + WSdif_{E.hum.uncon.rur.j}$	Eq.S9
$WSdif_{N.hum.uncon.urb.j} = Nexc_{hum.uncon.urb.j} \times (1 - fr_{NH3.hum})$	Eq.S10
$WSdif_{N.hum.uncon.rur.j} = Nexc_{hum.uncon.rur.j} \times (1 - fr_{NH3.hum})$	Eq.S11
$WSdif_{P.hum.uncon.urb.j} = Pexc_{hum.uncon.urb.j}$	Eq.S12
$WSdif_{P.hum.uncon.rur.j} = Pexc_{hum.uncon.rur.j}$	Eq.S13
$Nexc_{hum.uncon.urb.j} = E^N_{hum.j} \times Pop_{uncon.urb.j}$	Eq.S14
$Nexc_{hum.uncon.rur.j} = E^N_{hum.j} \times Pop_{uncon.rur.j}$	Eq.S15
$E^N_{hum.j} = I^N_{hum.j} \times 0.365$	Eq.S16
$I^N_{hum.j} = 4 + 17 \times [GDP_{ppp.j} / 68673]^{0.3}$	Eq.S17
$Pexc_{hum.uncon.urb.j} = E^P_{hum.j} \times Pop_{uncon.urb.j}$	Eq.S18
$Pexc_{hum.uncon.rur.j} = E^P_{hum.j} \times Pop_{uncon.rur.j}$	Eq.S19
$E^P_{hum.j} = E^N_{hum.j} \times (1/6)$	Eq.S20

Equation	Number
$Nexc_{hum.con.urb.j} = E^{N}_{hum.j} \times Pop_{con.urb.j}$ used in Eq.7 in the paper	Eq.S21
$Nexc_{hum.con.rur.j} = E^{N}_{hum.j} \times Pop_{con.rur.j}$ used in Eq.8 in the paper	Eq.S22
$Pexc_{hum.con.urb.j} = E^{P}_{hum.j} \times Pop_{con.urb.j}$ used in Eq.7 in the paper	Eq.S23
$Pexc_{hum.con.rur.j} = E^{P}_{hum.j} \times Pop_{con.rur.j}$ used in Eq.8 in the paper	Eq.S24
$D_{F,j} = (1/Q_{act,j}) \times \Sigma_{i=1...n}(Q_{act,i} \times D_{F,i})$ for DIN and DIP, used in Eq.10 in the paper	Eq.S25
$D_{DIN,i} = 0.8845 \times (h_i / \tau_{R,i})^{-0.3677}$	Eq.S26
$D_{DIP,i} = 0.85 \times [1 - \exp(-0.0807 \times 365 \times \tau_{R,i})]$	Eq.S27
$\tau_{R,i} = V_i / Q_{act,i}$	Eq.S28
$L_{DIN,j} = 0.0605 \times \ln(Area_j) - 0.0443$ for DIN and constants for DIP, used in Eq.10 in the paper	Eq.S29
$FQrem_j = 1 - (Q_{act.j}/Q_{nat.j})$ used in Eqs.10-11 in the paper	Eq.S30
${}_{juT}FE_{riv.F.outlet.jmC} = \left(1 - [D_{F.jmC} \times {}_{juT}A_{jmC}]\right) \times \left(1 - [L_{F.jmC} \times {}_{juT}A_{jmC}]\right) \times \left(1 - [FQrem_{jmC} \times {}_{juT}A_{jmC}]\right)$ used in Eq.12 in the paper	Eq.S31
${}_{juT}FE_{riv.F.outlet.jdC} = \left(1 - [D_{F.jdC} \times {}_{juT}A_{jdC}]\right) \times \left(1 - [L_{F.jdC} \times {}_{juT}A_{jdC}]\right) \times \left(1 - [FQrem_{jdC} \times {}_{juT}A_{jdC}]\right)$ used in Eq.12 in the paper	Eq.S32
${}_{jmT}FE_{riv.F.outlet.jdC} = \left(1 - [D_{F.jdC} \times {}_{jmT}A_{jdC}]\right) \times \left(1 - [L_{F.jdC} \times {}_{jmT}A_{jdC}]\right) \times \left(1 - [FQrem_{jdC} \times {}_{jmT}A_{jdC}]\right)$ used in Eq.13 in the paper	Eq.S33
${}_{jmC}FE_{riv.F.outlet.jdC} = \left(1 - [D_{F.jdC} \times {}_{jmC}A_{jdC}]\right) \times \left(1 - [L_{F.jdC} \times {}_{jmC}A_{jdC}]\right) \times \left(1 - [FQrem_{jdC} \times {}_{jmC}A_{jdC}]\right)$ used in Eq.14 in the paper	Eq.S34

Box S3.2 Descriptions of the abbreviations in Box S3.1 adjusted from Ural-Janssen et al. (2023).

Parameter	Description	Unit
y	Source	-
j	Basin (sub-basins for the Danube River)	-
i	Reservoir	-
$RSdif_{F.lch.ant.j}$	Inputs of nutrient form F (DON and DOP) to rivers from organic matter leaching (lch) over agricultural areas (ant) in basin j	kg N or P yr^{-1}
$RSdif_{F.lch.nat.j}$	Inputs of nutrient form F (DON and DOP) to rivers from organic matter leaching (lch) over non-agricultural areas (nat) in basin j	kg N or P yr^{-1}
$RSdif_{DIP.wth.ant.j}$	Inputs of DIP to rivers from P weathering (wth) in agricultural areas (ant) in basin j	kg P yr^{-1}
$RSdif_{DIP.wth.nat.j}$	Inputs of DIP to rivers from P weathering (wth) in non-agricultural areas (nat) in basin j	kg P yr^{-1}
$Area_j$	The drainage area of basin j	km^2
$Agr_{fr.j}$	Fraction of agricultural area in basin j	0-1
EC_F	Watershed export rate for nutrient form F (DON and DOP) from leaching of organic matter	kg km^{-2} yr^{-1}
EC_{DIP}	Watershed export rate for DIP from P weathering	kg km^{-2} yr^{-1}
CW_F	Uncalibrated export coefficient for leaching of nutrient form F (DON and DOP) from land to surface waters	-
CW_{DIP}	Uncalibrated export coefficient for weathering of DIP from land to surface waters	-
$Rnat_j$	Annual surface runoff from land to streams in basin j	m yr^{-1}
$Q_{nat.j}$	Natural (nat) river discharge at the outlet of basin j, before water is taken for consumption	km^3 yr^{-1}
$WSdif_{F.gross.j}$	The sum of all the inputs of nutrient form F (DIN, DON, DIP or DOP) to agricultural land in basin j	kg N or P yr^{-1}
$WSdif_{E.fe.j}$	Inputs of nutrient element E (N and P) from synthetic fertilizers (fe) that are applied to agricultural land in basin j	kg N or P yr^{-1}
$WSdif_{E.ma.j}$	Inputs of nutrient element E (N and P) from animal manure (ma) that is applied to agricultural land in basin j	kg N or P yr^{-1}
$WSdif_{E.dep.ant.j}$	Inputs of nutrient element E (N and P) from atmospheric deposition (dep) over agricultural land (ant) in basin j	kg N or P yr^{-1}
$WSdif_{N.fix.ant.j}$	Inputs of N from biological N_2 fixation (fix) by agricultural crops (ant) in basin j	kg N yr^{-1}
$WSdif_{E.hum.uncon.j}$	Inputs of nutrient element E (N and P) from excretion of population (hum) not connected to sewage systems (uncon) in basin j	kg N or P yr^{-1}
$WSdif_{E.hum.uncon.urb.j}$	Inputs of nutrient element E (N and P) from excretion of urban population (urb) not connected to sewage systems (uncon) in basin j	kg N or P yr^{-1}
$WSdif_{E.hum.uncon.rur.j}$	Inputs of nutrient element E (N and P) from excretion of rural population (rur) not connected to sewage systems (uncon) in basin j	kg N or P yr^{-1}
$Nexc_{hum.uncon.urb.j}$	N in human excretion from urban population unconnected to sewage systems in basin j	kg N yr^{-1}
$Nexc_{hum.uncon.rur.j}$	N in human excretion from rural population unconnected to sewage systems in basin j	kg N yr^{-1}

Parameter	Description	Unit
$fr_{NH3.hum}$	Fraction of N losses from human excretion to the air	0-1
$Pexc_{hum.uncon.urb.j}$	P in human excretion from urban population unconnected to sewage systems in basin j	kg P yr^{-1}
$Pexc_{hum.uncon.rur.j}$	P in human excretion from rural population unconnected to sewage systems in basin j	kg P yr^{-1}
$E^{N}_{hum.j}$	Human N excretion rate in basin j	kg $person^{-1}$ yr^{-1}
$I^{N}_{hum.j}$	Protein N intake for basin j	g N $person^{-1}$ day^{-1}
$GDP_{ppp.j}$	Annual gross domestic product at purchasing power parity	US$ 2005 $person^{-1}$ yr^{-1}
$E^{P}_{hum.j}$	Human P excretion rate in basin j	kg $person^{-1}$ yr^{-1}
$Pop_{uncon.urb.j}$	Urban population without sewage connection in basin j	People
$Pop_{uncon.rur.j}$	Rural population without sewage connection in basin j	People
$Nexc_{hum.con.urb.j}$	N in human excretion from urban population connected to sewage systems in basin j	kg N yr^{-1}
$Nexc_{hum.con.rur.j}$	N in human excretion from rural population connected to sewage systems in basin j	kg N yr^{-1}
$Pexc_{hum.con.urb.j}$	P in human excretion from urban population connected to sewage systems in basin j	kg P yr^{-1}
$Pexc_{hum.con.rur.j}$	P in human excretion from rural population connected to sewage systems in basin j	kg P yr^{-1}
$Pop_{con.urb.j}$	Urban population with sewage connection in basin j	People
$Pop_{con.rur.j}$	Rural population with sewage connection in basin j	People
$D_{F.j}$	Fraction of nutrient form F (DIN and DIP) retained in reservoirs and (regulated) lakes in basin j	0-1
$L_{F.j}$	Fraction of nutrient form F (DIN and DIP) retained in or/and lost from water systems (e.g., denitrification) in basin j	0-1
$Q_{act.j}$	Actual (act) water discharge at the outlet of basin j after water consumption	km^3 yr^{-1}
${}^{juT}FE_{riv.F.outlet.jmC}$ ${}^{juT}FE_{riv.F.outlet.jdC}$	Fractions of nutrient form F (DIN, DON, DIP and DOP) exported from the outlet of upstream tributary (upper case: juT) to the outlets of the main channel in middlestream (lower case: jmC) and downstream (lower case: jdC) sub-basins	0-1
${}^{jmT}FE_{riv.F.outlet.jdC}$ ${}^{jmC}FE_{riv.F.outlet.jdC}$	Fraction of nutrient form F (DIN, DON, DIP and DOP) exported from the outlet of middlestream subbasin with the main channel (upper case: jmC) to the outlet of the main channel in downstream (lower case: jdC) sub-basin (the outlet of the this downstream subbasin is the river mouth)	0-1
$D_{F.jmC}$ $D_{F.jdC}$	Fractions of nutrient form F (DIN and DIP) retained in reservoirs of middlestream (jmC) and downstream (jdC) sub-basins with the main channel (C). These fractions were calculated for sub-basins using equations for $D_{DIN,j}$ and $D_{DIP,j}$ from Table S1	0-1

Parameter	Description	Unit
$L_{F.jmC}$ $L_{F.jdC}$	Fractions of nutrient form F (DIN and DIP) that is lost from surface waters of middlestream (jmC) and downstream (jdC) sub-basins with the main channel (C). These fractions were calculated using the equation for $L_{DIN,j}$ and $L_{DIP,j}$ from Table S1	0-1
$FQrem_{jmC}$ $FQrem_{jdC}$	Fractions of nutrient form F (generic for DIN, DON, DIP and DOP) that are lost from surface waters of middlestream (jmC) and downstream (jdC) sub-basins with the main channel (C) via water consumption. These fractions were calculated using equations for $FQrem_j$ from Box S3.1.	0-1
${}^{juT}A_{jmC}$ ${}^{juT}A_{jdC}$	Drainage area (A) of the main channel (C) in middlestream (lower case: jmC) and downstream (lower case: jdC) sub-basins that exports nutrients from the outlet of upstream tributary (upper case: juT). This drainage area was calculated as the fraction to the total sub-basin area.	0-1
${}^{jmT}A_{jdC}$	Drainage area (A) of the main channel (C) in downstream (lower case: jdC) sub-basin that exports nitrogen from the outlet of middlestream tributary (upper case: jmT). This drainage area was calculated as the fraction to the total sub-basin area.	0-1
${}^{jmC}A_{jdC}$	Drainage area (A) of the main channel (C) in the downstream (lower case: jdC) subbasin that exports nitrogen from the outlet of middlestream main channel (upper case: jmC). This drainage area was calculated as the fraction to the total sub-basin area.	0-1

Table S3.1 Main characteristics and approaches of the baseline (SSP5-RCP8.5) scenario for 2050, and changes relative to current situation (2017-2020) in Europe.

Characteristics	2017-2020	2050 based on SSP5-RCP8.5	Change	Approaches
Population (person)	547,997,738	647,278,070	+18%	Urban, rural and total populations per basin for 2020 are derived from Ural-Janssen et al. (2023) and for 2050 are derived from Strokal et al. (2021) based on the SSP5 scenario. Strokal et al. (2021) assumes high urbanization (e.g., increasing number of people living in cities and people with sewage connections) and moderate improvement in wastewater treatment plants (e.g., shifting from secondary to tertiary treatment).
urban population	419,695,198	580,757,907	+38%	
rural population	128,302,540	66,520,163	-48%	
Sewage connection rates (0-1)		high		
Wastewater treatment rates (0-1)		moderate improvement		
GDP (US$ capita^{-1} yr^{-1})	27,098	69,843	+158%	Average GDP per basin is calculated as a function of the GDP and population per basin. The GDP data are derived from Strokal et al. (2021) for 2010 representing the current and for 2050 based on the SSP5 scenario.
Agricultural area (cropland, grassland, area set aside and fallow) (km^2)	2,205,561	2,242,472	~ +2%	Data on agricultural area is derived from the MITERRA-Europe model for 2017 at NUTS2 scale (Duan et al., 2021), and projected for 2050 per basin as a function of the percent changes between 2015-2050 in the IMAGE model based on the SSP5-RCP8.5 scenario (Beusen et al., 2022). Beusen et al. (2022) considers the same trajectories for land use between 2015-2020 based on SSP2 for all SSPs, and the scenarios start to differ from 2021 onwards.
N inputs to agricultural land	29.2 Tg N yr^{-1} 132 kg N ha^{-1}	31.2 Tg N yr^{-1} 139 kg N ha^{-1}	+7%	Data on N inputs to agricultural land is derived from the MITERRA-Europe model for 2017 at NUTS2 scale (Duan et al., 2021), and projected for 2050 per basin as a function of the percent changes between 2015-2050 in the IMAGE model based on the SSP5-RCP8.5 scenario (Beusen et al., 2022).
N inputs to non-agricultural land	5.1 Tg N yr^{-1}	5.5 Tg N yr^{-1}	+6%	Data on N inputs to non-agricultural land is derived from the IMAGE model for 2015 at 0.5° grids, and projected for 2050 per basin as a function of the percent changes between 2015-2050 in the IMAGE model based on the SSP5-RCP8.5 scenario (Beusen et al., 2022).
P inputs to agricultural land	1.7 Tg P yr^{-1} 7.9 kg P ha^{-1}	1.8 Tg P yr^{-1} 8 kg P ha^{-1}	+3%	Data on P inputs to agricultural land is derived from the MITERRA-Europe model for 2017 at NUTS2 scale (Duan et al., 2021), and projected for 2050 per basin as a function of the percent changes between 2015-2050 in the IMAGE model based on the SSP5-RCP8.5 scenario (Beusen et al., 2022).

Table S3.2 Description of how model inputs are processed to basins for the MARINA-Nutrients model for Europe. Numbers indicate the equation numbers in Box S3.1.

Numbers	Description of how model inputs are processed for 2050
Eqs.S1-S2 and Eq.1 in the paper	Organic matter (e.g., litter, decomposition products, organic residues) can bring dissolved organic nutrients via precipitation and runoff to surface waters (Gmach et al., 2020). Inputs of N (DON) and P (DOP) to rivers from the leaching of organic matter from agricultural and non-agricultural soils (kg N or P yr^{-1} per basin) were calculated for 2050 as a function of agricultural/non-agricultural area (km^2) in basin, watershed export rates for leaching of organic matter (EC_{DON} and EC_{DOP}, kg km^{-2} yr^{-1}), uncalibrated export coefficients for leaching of nutrients from land to waters and annual surface runoff from land to streams ($CL_F x Rnat_j$, calculated using runoff values, unitless). Basin area was derived from Strokal et al. (2016). The fraction of agricultural area in basin ($Agr_{fr.j}$, 0-1) was calculated for 2050 by dividing the agricultural area estimated for 2050 by basin area. The coefficients for organic matter leaching: EC_{DON} = 280 kg km^{-2} yr^{-1} and EC_{DOP} = 15 kg km^{-2} yr^{-1} were derived from Mayorga et al. (2010). For $Rnat_j \geq 0.1$ m, uncalibrated export coefficient CL_F for DON and DOP was considered as 1.0, and for $Rnat_j < 0.1$ m, 0.9, as these coefficients gave globally comparable results for DON and DOP leaching to surface waters from agricultural areas. $Rnat_j$ was calculated for 2050 by dividing the natural discharge with the basin area.
Eqs.S3-S4 and Eq.1 in the paper	Inputs of P to rivers from the weathering of P-contained minerals from agricultural and non-agricultural areas (kg N or P yr^{-1} per basin) were calculated for 2050 as a function of agricultural/non-agricultural area (km^2) in basin, watershed export rates for DIP weathering (EC_{DIP}, kg km^{-2} yr^{-1}), uncalibrated export coefficient for weathering of DIP from land to waters and annual surface runoff from land to streams ($CL_{DIP} x Rnat_j$, calculated using runoff values, unitless). EC_{DIP} = 26 kg km^{-2} yr^{-1} was derived from Mayorga et al. (2010). For $Rnat_j \geq 0.1$ m, CL_{DIP} was considered as 0.4, and for $Rnat_j < 0.1$ m, 0.2, as these coefficients gave globally comparable results for DIP weathering from agricultural areas.
Eqs.S6-S8 and Eq.2 in the paper	Inputs of N and P to land from synthetic fertilizer application to agricultural land (kg N or P yr^{-1} per basin) were calculated for 2050 as a function of the current (2017) synthetic fertilizer application derived from the MITERRA-Europe (Duan et al., 2021) and the percent change in the synthetic fertilizer application to agricultural land between 2015-2050 based on the SSP5 scenario derived from the IMAGE model (Beusen et al., 2022).
Eqs.S6-S8 and Eq.2 in the paper	Inputs of N and P to land from animal manure (applied to agricultural land and deposited on land during grazing) (kg N or P yr^{-1} per basin) were calculated for 2050 as a function of the current (2017) animal manure derived from the MITERRA-Europe (Duan et al., 2021) and the percent change in the animal manure recycled to agricultural land between 2015-2050 based on the SSP5 scenario derived from the IMAGE model (Beusen et al., 2022).
Eqs.S6-S7 and Eq.2 in the paper	Inputs of N and P to land from atmospheric deposition on agricultural land (kg N or P yr^{-1} per basin) were calculated for 2050 as a function of the current (2017) atmospheric deposition derived from the MITERRA-Europe (Duan et al., 2021) and the percent change in the atmospheric deposition on agricultural land between 2015-2050 based on the SSP5 scenario derived from the IMAGE model (Beusen et al., 2022).
Eq.S6 and Eq.2 in the paper	Inputs of N to land from biological N_2 fixation by agricultural crops (kg N yr^{-1} per basin) were calculated for 2050 as a function of the current (2017) biological N_2 fixation derived from the MITERRA-Europe (Duan et al., 2021) and the percent change in the biological N_2 fixation by agricultural crops between 2015-2050 based on the SSP5 scenario derived from the IMAGE model (Beusen et al., 2022).

Numbers	Description of how model inputs are processed for 2050
Eqs.S6-S13 and Eq.2 in the paper	Inputs of N and P to land from the excretion of population unconnected to sewer systems (kg N or P yr^{-1} per basin) were calculated for 2050 as a function of unconnected population (people) and the rates of human excretion for N and P (kg cap^{-1} yr^{-1}) in basins based on the SSP5 scenario. Total, urban and rural population unconnected to sewage systems were derived from Strokal et al. (2021) for 2050 based on the SSP5 scenario (people per basin). We used the approach of Ural-Janssen et al. (2023) by considering there is no direct discharge of the untreated wastewater from population (urban and rural) not connected to sewage systems in Europe, and the unconnected human waste is lost to rivers through land. Therefore, we considered the untreated human wastewater as a diffuse source for the nutrient inputs to rivers through land (i.e. $WSdif_{N.hum.uncon.j}$ and $WSdif_{P.hum.uncon.j}$). The fraction of N loss to air from human excretion not connected to sewage systems $fr_{NH3.hum}$ = 0.24 was derived from Ma et al. (2010).
Eq.2 in the paper	Inputs of N to land from atmospheric deposition on non-agricultural land (kg N yr^{-1} per basin) were derived from the IMAGE model for 2015 (current) and for 2050 based on the SSP5 scenario (Beusen et al., 2022).
Eq.2 in the paper	Inputs of N to land from biological N_2 fixation by natural vegetation (kg N yr^{-1} per basin) were derived from the IMAGE model for 2015 (current) and for 2050 based on the SSP5 scenario (Beusen et al., 2022).
Eq.4 in the paper	Exports of N and P from agricultural land by crop harvesting and animal grazing (kg N or P yr^{-1} per basin) were calculated for 2050 as a function of the current (2017) exports of N and P derived from the MITERRA-Europe (Duan et al., 2021) and the percent change in the exports of N and P from agricultural land by crop harvesting and animal grazing between 2015-2050 based on the SSP5 scenario derived from the IMAGE model (Beusen et al., 2022).
Eqs.S5, S25 and S30	Natural water discharges were derived from Variable Infiltration Capacity (VIC) macroscale hydrological model for 2010 as current (by averaging the data over 30 years of the period 1980-2010 to avoid the influence of climate extremes) and for 2050 (van Vliet et al., 2016) and used to calculate inputs of nutrients to rivers. Actual water discharges were calculated for 2010 as current (see details in (Ural-Janssen et al., 2023)) and for 2050 by using the water consumption projections for 2050 based on SSP5-RCP8.5 scenario from Khan et al. (2022). First, the annual water consumption data for 2050 was derived from Khan et al. (2022) at 0.5° resolution (km^3 yr^{-1} per grid cell). Total water consumption (Q_{rem}) was the sum of six sectors, namely irrigation, livestock, domestic, electricity, manufacturing and mining (km^3 yr^{-1}) (Khan et al., 2022). Second, the gridded Q_{rem} data was aggregated for basin over the corresponding grids (km^3 yr^{-1} per basin). Third, the actual discharges (Q_{act}) were quantified by subtracting Q_{rem} from the natural discharge Q_{nat} for 2050 (km^3 yr^{-1} per basin). The $Q_{act.j} \leq 0$ was considered as "0.036 km^3 yr^{-1}", as this was the lowest $Q_{act.j}$ value per basin among the current actual water discharges (2010).
Eqs.S1-S5 and Eq.5 in the paper	Surface runoff (m yr^{-1} per basin) was calculated by dividing the natural discharge for 2050 (Q_{nat}, km^3 yr^{-1} per basin) with the area of basin (km^2) following the approach of Strokal et al. (2016).

Numbers	Description of how model inputs are processed for 2050
Eqs.2, 4-5 in the paper	Uncalibrated approach was used to calculate the export fraction of nutrients entering rivers of basin ($FE_{ws.F,j}$, 0-1) and the fraction of nutrients applied to agricultural land that remained in soils of basin ($G_{F,j}$, 0-1) following Li et al. (2022b). In this approach, the world basins were clustered based on the climate zones from FAO (2007) to calculate the fractions of nutrients retained in soils after crop harvesting and animal grazing ($G_{F,j}$). Among the European basins (593 basins and 8 sub-basins of the Danube River), 223 basins were in subtropic/tropic zones and assumed to have a rather wet climate, and 378 basins were in other temperature zones and assumed to have a rather dry climate (Li et al., 2022b). Based on the uncalibrated approach of Li et al. (2022b): o If $R_{nat,j} \geq 0.1$ m yr^{-1}, $FE_{ws.DIN,j} = 0.9 \times R_{nat,j}$ and if $R_{nat,j} < 0.1$ m yr^{-1}, $FE_{ws.DIN,j} = 0.8 \times R_{nat,j}$ o If $R_{nat,j} \geq 0.1$ m yr^{-1}, $FE_{ws.DON,j} = 0.02 \times R_{nat,j}$ and if $R_{nat,j} < 0.1$ m yr^{-1}, $FE_{ws.DON,j} = 0.01 \times R_{nat,j}$ o If $R_{nat,j} \geq 0.1$ m yr^{-1}, $FE_{ws.DIP,j} = 0.15 \times R_{nat,j}$ and if $R_{nat,j} < 0.1$ m yr^{-1}, $FE_{ws.DIP,j} = 0.1 \times R_{nat,j}$ o If $R_{nat,j} \geq 0.1$ m yr^{-1}, $FE_{ws.DOP,j} = 0.02 \times R_{nat,j}$ and if $R_{nat,j} < 0.1$ m yr^{-1}, $FE_{ws.DOP,j} = 0.01 \times R_{nat,j}$ o Any of $FE_{ws.F,j} > 0.9$ was assumed that equal to 0.9. o If $G_{F,j} < 0$, $G_{F,j}$ was validated as 0.05 for the basins in subtropic/tropic zones, and as 0.1 for the basins in other temperature zones.
Eqs. S14-S15, S18-S19, S21-S24 and Eqs.3, 6-9 in the paper	Total, urban and rural population projections (people per basin) and sewer connection rates (0-1 per basin) were derived from Strokal et al. (2021) for 2050 based on the SSP5 scenario.
Eq.S17	Data on gross domestic product at purchasing power parity (GDPppp, US$2005 $person^{-1}$ yr^{-1} per basin) was derived from Strokal et al. (2021) for 2050 based on the SSP5 scenario.
Eqs.S16 and S20	Excretion or consumption rates of N and P per person (kg $person^{-1}$ yr^{-1} per basin) were calculated for 2050 as a function of the GDPppp following the approach of van Drecht et al. (2009). The consumption rate of P in detergents (i.e. laundry and washing) (kg P $person^{-1}$ yr^{-1} per basin) was derived from Strokal et al. (2021) for 2050 based on the SSP5 scenario.
Eqs.S21-24 and Eqs.7-8 in the paper	Fractions of N and P removal in wastewater treatment systems (the same for both urban and rural, 0-1 per basin) were derived from Strokal et al. (2021) for 2050 based on the SSP5 scenario.

Numbers	Description of how model inputs are processed for 2050
Eq.9 in the paper	Fractions of nutrient form F (DIN, DON, DIP or DOP) entering rivers from point source ($FEpnt_{F.hum.con.j}$, 0-1 per basin) were derived from Mayorga et al. (2010) as 0.14, 1 and 0.01 for DON, DIP and DOP, respectively. For DIN, the Eq. 9 in the paper is used: $FEpnt_{DIN.hum.con.j} = 0.485 + 0.225 \times (hw_{frem.N.j} / 0.86)$ where 0.485 is the fraction of total N (TN) in sewage effluents that is DIN without treatment, 0.225 is the maximum increase in DIN/TN ratio that can be achieved during treatment, hw_{fremN} is N removal in wastewater treatment systems of each basin, and 0.86 is the maximum observed value for N treatment for Europe for 2050 based on the SSP5 scenario.
Eqs.S25-S29 and Eq.10 in the paper	Data on the characteristics of reservoirs and (regulated) lakes was derived from HydroLAKES Project to calculate current nutrient retentions in rivers (Messager et al., 2016). $Q_{act.i}$ is actual water discharge for reservoir i ($km^3\ yr^{-1}$) and derived from Messager et al. (2016). No change in the characteristics of reservoirs and (regulated) lakes was assumed for 2050. The fraction of DIN retained in reservoir i ($D_{DIN.i}$, 0-1) was calculated by following the approach of Strokal et al. (2016). H_i is depth of reservoir I (m) and derived from Messager et al. (2016). $\tau_{R.i}$ is water residence time for reservoir i (year) (Strokal et al., 2016). V_i is volume of reservoir i ($km^3\ yr^{-1}$) and derived from Messager et al. (2016). The nutrient retentions of individual reservoirs were averaged for basins using the actual water discharges at the outlets of the basins (Strokal et al., 2016). The maximum value of $D_{DIN.j}$ was set as 0.965 as this was the maximum value for the basins used to calibrate parameters (Strokal et al., 2016). The fraction of DIN retained in reservoir i (D_{DIPi}, 0-1) was calculated by following the approach of Strokal et al. (2016).The maximum value of D_{DIPj} was set as 0.85 following the approach of Strokal et al. (2016). The fraction of DIN retained in and/or lost from water systems (e.g., denitrification) in basin j ($L_{DIN.j}$, 0-1) was calculated as a function of basin area (Strokal et al., 2016). The fitted coefficients 0.0605 and 0.0443 in the equation were derived from Strokal et al. (2016). The maximum value of $L_{DIN.j}$ was set as 0.65 following the approach of Strokal et al. (2016). The fraction of DIP retained in and/or lost from water systems (e.g., sedimentation) in basin j (L_{DIPj}, 0-1) was set as 0.9 for dry rivers having runoff of less than 0.1 m yr^{-1} and as 0.5 for the rest of the rivers following the approach of Strokal et al. (2016).
Eqs.15-18 in the paper	Dissolved Silica loads (Mg yr^{-1}) per basin were derived from the Global *NEWS2* model for 2050 based on the Global Orchestration scenario (Seitzinger et al., 2010) as this scenario was the closest option to our baseline scenario. First, the DSi loads (Mg yr^{-1}) per basin were selected by using the spatial data in two steps: (1) matching the river mouths by coordinates (i.e. longitude and latitude of the river mouths from the Global *NEWS2* and MARINA-Nutrients models) and (2) matching the remaining river mouths based on the shortest distance between the river mouths from the Global *NEWS2* and MARINA-Nutrients models. Second, the DSi loads were converted from Mg yr^{-1} to kg km^{-2} day^{-1} by dividing the basin area (km^2) and 365 (day yr^{-1}) for unit conversion. Third, the limiting nutrient was determined based on the N:P ratio (when TN:TP ratio > 16, P is the limiting nutrient, and when TN:TP ratio < 16, N is limiting) (Billen & Garnier, 2007). Fourth, the maximum allowable river exports of nutrients were calculated by following the back-casting approach of (Li et al., 2019) for 2050 based on the SSP5-RCP8.5 scenario (kg C-eq. km^{-2} day^{-1}). Based on the limiting nutrient (N or P), the N-ICEP and P-ICEP equations were used for back-casting the maximum allowable river export of N and P for 2050 based on the ICEP = 0 (with an uncertainty range of -1 and +1) (kg C-eq. km^{-2} day^{-1}).

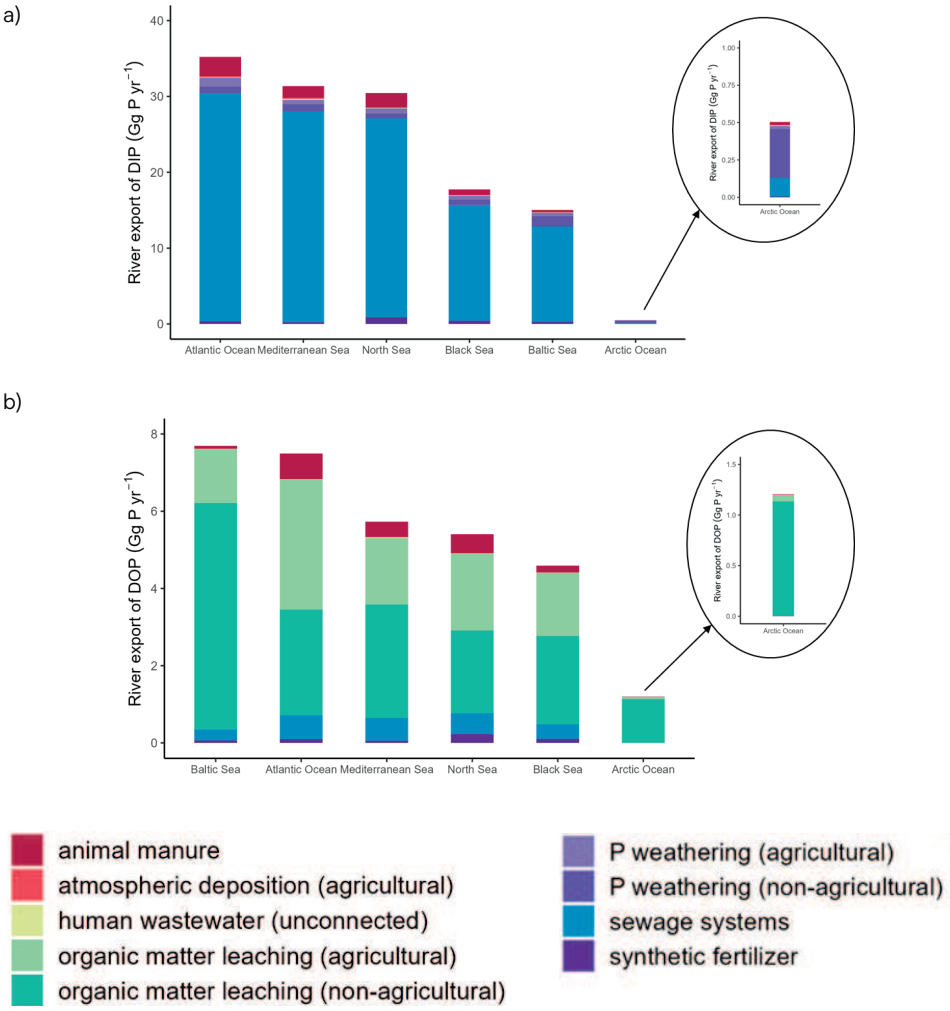


Figure S3.1 Future (2050) river export of phosphorus by source to the European seas based on the SSP5-RCP8.5 scenario. (a) River export of dissolved inorganic phosphorus (DIP, Gg P yr^{-1}). (b) River export of dissolved organic phosphorus (DOP, Gg P yr^{-1}). SSP5 is short for Shared Socioeconomic Pathway 5 (fossil fueled development) and RCP8.5 is short for Representative Concentration Pathways (high-end climate change). Source: MARINA-Nutrients model for Europe (Section 3.2.1).

S4: Supplementary materials for Chapter 4

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Supplementary information

Mitigating nutrient losses in Europe: Synergistic solutions for air and water pollution by 2050

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The Supplementary information contains

Supplementary Text S4.1-S4.2

Tables S4.1-S4.4

Figures S4.1-S4.9

Supplementary Text S4.1 Description of baseline scenario for 2050

Our baseline scenario follows the Shared Socioeconomic Pathway 5 (SSP5, O'Neill et al. (2014)) and the Representative Concentration Pathway 8.5 (RCP8.5, van Vuuren et al. (2011)) to project changes in future air and water pollution as a result of socioeconomic and climatic changes. We incorporated the existing storylines of how agriculture might be developed in European basins (e.g., increased crop and livestock production) by 2050 following Beusen et al. (2022). We also incorporated the existing storylines on urbanization (e.g., increased migration from rural areas to cities) and sewage systems (e.g., moderate improvement) in European basins for 2050 following Strokal et al. (2021). We mention our baseline scenario (SSP5-RCP8.5) as "future baseline" or "2050 baseline" throughout the manuscript and supplementary information.

Table S4.1 Main equations of the MARINA-Nutrients model to inputs of nitrogen (N) and phosphorus (P) to rivers, and their river exports to the coastal waters of Europe from diffuse and point sources (Ural-Janssen et al., 2023). Source: The MARINA-Nutrients model for Europe.

Equation	No
for calculating crop P demand	
$P_{cropdemand.j} = WSdif_{P.ex.j} \times P_{index.j}$	S1
for calculating inputs of nutrients to land from production and application of ReNuRe producst and effluent from ammonia stripping-scrubbing	
$Nslurry_{a.h.j} = Nexcretion_{a.h.j} - \left(\sum Nemission_{f.a.y.j}\right)$	S2
$Ndigestate_{a.j} = Nslurry_{a.h.j}$	S3 [a]
$NH_{3.a.y.j} = Ndigestate_{a.j} \times EF_{NH3.y}$	S4
$NReNuRe_{a.j} = \left(Ndigestate_{a.j} - NH_{3.a.y.j}\right) \times C_a$	S5
$Neffluent_{a.j} = Ndigestate_{a.j} - NH_{3.a.y.j} - NReNuRe_{a.j}$	S6
$WSdif_{N.ReNuRe.a.j} = NReNuRe_{a.j} \times NFRV_{ReNuRe}$	S7
$WSdif_{N.effluent.a.j} = Neffluent_{a.j} \times NFRV_{effluent.a}$	S8
$WSdif_{P.effluent.a.j} = Pexcretion_{a.j} \times PFRV_{effluent.a}$	S9
for calculating inputs of nutrients to rivers	
$RSdif_{f.y.ag.j} = Area_j \times Agr_{fr.j} \times EC_f \times CL_f \times Rnat_j$	S10
$RSdif_{f.y.nonag.j} = Area_j \times \left(1 - Agr_{fr.j}\right) \times EC_f \times CL_f \times Rnat_j$	S11
$G_{f.j} = 1 - \left(WSdif_{e.ex.j} / \sum WSdif_{f.y.j}\right)$	S12 [b]
$FE_{ws.f.j} = CR_f \times Rnat_j$	S13
$Rnat_j = Q_{nat.j} / Area_j$	S14

Equation	No
$WScap_{N.j} = I^{N}_{hum.j} \times 0.365$ for N	S15
$WScap_{P.j} = I^{N}_{hum.j} \times 0.365 \times (1/6)$ for P	S16
$I^{N}_{hum.j} = 4 + 17 \times [GDP_{ppp.j}/68673]^{0.3}$	S17
$FEpnt_{DIN.j} = 0.485 + 0.225 \times (hw_{frem.N.j}/0.86)$, constants for DON, DIP and DOP	S18 [c]
for calculating production and application of RPSS	
$Psludge_{t.j} = WScap_{P.j} \times Pop_j \times fr_{pop.sew.j} \times hw_{frem.P.j}$	S19
$Precovered_{t.j} = Psludge_{t.j} \times 0.90$	S20
$WSdif_{P.RPSS.j} = Precovered_{t.j} \times PFRV_{RPSS}$	S21
for calculating river exports of nutrients to coastal waters	
$FE_{riv.f.outlet.j} = (1-\mathrm{D_{f.j}}) \times (1-\mathrm{L_{f.j}}) \times (1 - FQrem_j)$ for DIN and DIP	S22
$FE_{riv.f.outlet.j} = (1 - FQrem_j)$ for DON and DOP	S23
$\mathrm{D_{f.j}} = (1/\mathrm{Q_{act.j}}) \times \Sigma_{i=1\ldots n}(\mathrm{Q_{act.i}} \times \mathrm{D_{f.i}})$	S24
$\mathrm{D_{DIN.i}} = 0.8845 \times (\mathrm{h_i}/\tau_{\mathrm{R.i}})^{-0.3677}$	S25
$\mathrm{D_{DIP.i}} = 0.85 \times [1 - \exp(-0.0807 \times 365 \times \tau_{\mathrm{R.i}})]$	S26
$\tau_{\mathrm{R.i}} = \mathrm{V_i}/\mathrm{Q_{act.i}}$	S27
$\mathrm{L_{DIN.j}} = 0.0605 \times ln(Area_j) - 0.0443$, constant for DIP	S28
$FQrem_j = 1 - (Q_{act.j}/Q_{nat.j})$	S29
$FE_{riv.f.mouth.juT} = {}^{\mathrm{juT}}FE_{riv.f.outlet.jmC} \times {}^{\mathrm{juT}}FE_{riv.f.outlet.jdC}$	S30
$FE_{riv.f.mouth.jmT} = {}^{\mathrm{jmT}}FE_{riv.f.outlet.jdC}$	S31
$FE_{riv.f.mouth.jmC} = {}^{\mathrm{jmC}}FE_{riv.f.outlet.jdC}$	S32
$FE_{riv.f.mouth.jdC}$ = 1 as the outlets of down-stream sub-basins discharge directly to seas	S33
${}_{\mathrm{juT}}FE_{riv.f.outlet.jmC} = \left(1 - [D_{f.jmC} \times {}_{\mathrm{juT}}A_{jmC}]\right) \times \left(1 - [L_{f.jmC} \times {}_{\mathrm{juT}}A_{jmC}]\right) \times \left(1 - [FQrem_{jmC} \times {}_{\mathrm{juT}}A_{jmC}]\right)$	S34
${}_{\mathrm{juT}}FE_{riv.f.outlet.jdC} = \left(1 - [D_{f.jdC} \times {}_{\mathrm{juT}}A_{jdC}]\right) \times \left(1 - [L_{f.jdC} \times {}_{\mathrm{juT}}A_{jdC}]\right) \times \left(1 - [FQrem_{jdC} \times {}_{\mathrm{juT}}A_{jdC}]\right)$	S35
${}_{\mathrm{jmT}}FE_{riv.f.outlet.jdC} = \left(1 - [D_{f.jdC} \times {}_{\mathrm{jmT}}A_{jdC}]\right) \times \left(1 - [L_{f.jdC} \times {}_{\mathrm{jmT}}A_{jdC}]\right) \times \left(1 - [FQrem_{jdC} \times {}_{\mathrm{jmT}}A_{jdC}]\right)$	S36

Equation	No
${}_{jmC}FE_{riv.F.outlet.jdC} = \left(1 - [D_{f.jdC} \times {}_{jmC}A_{jdC}]\right) \times \left(1 - [L_{f.jdC} \times {}_{jmC}A_{jdC}]\right) \times \left(1 - [FQrem_{jdC} \times {}_{jmC}A_{jdC}]\right)$	S37

[a] **The** N content of digested slurry (digestate) is considered the same as the N content of animal slurry as a result of assuming no N loss during anaerobic digestion.

[b] **The** sum of nutrient inputs to agricultural land differs based on the nutrient form f (Ural-Janssen et al., 2023):

$\sum WSdif_{DIN.y.j} = WSdif_{N.fe.j} + WSdif_{N.ma.j} + WSdif_{N.gr.j} + WSdif_{N.dep.ag.j} + WSdif_{N.fix.ag.j} + WSdif_{N.hum.uncon.j}$

$\sum WSdif_{DIP.y.j} = WSdif_{P.fe.j} + WSdif_{P.ma.j} + WSdif_{P.gr.j} + WSdif_{P.dep.ag.j} + WSdif_{P.hum.uncon.j}$

$\sum WSdif_{DON.y.j} = WSdif_{N.fe.j} + WSdif_{N.ma.j} + WSdif_{N.gr.j} + WSdif_{N.hum.uncon.j}$

$\sum WSdif_{DOP.y.j} = WSdif_{P.fe.j} + WSdif_{P.ma.j} + WSdif_{P.gr.j} + WSdif_{P.hum.uncon.j}$

[c] **0.86** is the maximum observed value for N removal from the wastewater treatment systems in Europe for 2050 under the future baseline (Ural-Janssen et al., 2024). The maximum value for N removal is 0.88 for 2050 under the Pmed and Padv scenarios.

Table S4.2 Descriptions of the abbreviations from the equations of the MARINA-Nutrients model (Table S4.1), adjusted from Ural-Janssen et al. (2023). Source: The MARINA-Nutrients model for Europe.

Equation	Parameter	Description	Unit
S1	$P_{cropdemand.j}$	Crop P demand by basin j	kg P yr^{-1}
S1	$P_{index.j}$	Recommended P rate relative to crop demand by basin j	-
1, S2	$Nemission_{f.a.y.j}$	N emissions to the air from animal housing and manure storage by form f, animal type a, source y in basin j	kg N yr^{-1}
1, S2, S9	$Nexcretion_{a.y.j}$, $Pexcretion_{a.y.j}$	N and P excretions produced by animals per animal type a, source y in basin j	kg N or P yr^{-1}
1	$EF_{f.a.y.j}$	Emission factors per N form f, animal type a, source y in basin j	0-1
S2	$Nslurry_{a.h.j}$	N content of animal slurry per animal type a, animal housing h in basin j	kg N yr^{-1}
S4-S6	$NH_{3.a.y.j}$	NH_3 emission to the air from manure processing by animal type a, source y in basin j	kg N yr^{-1}
S3-S6	$Ndigestate_{a.j}$	N content of digested slurry by animal type a in basin j	kg N yr^{-1}
S4	$EF_{NH3.y}$	Emission factor for NH_3 emission from ammonia stripping-scrubbing process	0-1
2	$Nemission_{f.y.j}$	N emissions to the air from agricultural soils by form f, by source y in basin j	kg N yr^{-1}
2, S7-S9	$WSdif_{N.y.j}$, $WSdif_{P.y.j}$	Input of N and P to agricultural or non-agricultural land by source y (the application of synthetic fertilizer, untreated manure, ReNuRe products/RPSS and effluent, manure deposited on land during grazing, human waste from population unconnected to sewage systems, atmospheric deposition and biological N_2 fixation over agricultural and non-agricultural areas) in basin j	kg N or P yr^{-1}
2	$EF_{f.y.j}$	Emission factors per N form f, source y in basin j	0-1
S5-S8	$NReNuRe_{a.j}$	N content of ReNuRe product per animal type a in basin j	kg N yr^{-1}
S5	C_a	Coefficient to calculate the portion of ReNuRe product produced per animal type a	0-1
S6, S8	$Neffluent_{a.j}$	N content of effluent from ammonia stripping-scrubbing process per animal type a in basin j	kg N yr^{-1}
S7	$WSdif_{N.ReNuRe.a.j}$	N input to agricultural land from the application of ReNuRe products by animal type a in basin j	kg N yr^{-1}
S7	$NFRV_{ReNuRe}$	Nitrogen fertilizer replacement value for ReNuRe products	0-1
S8	$WSdif_{N.effluent.a.j}$	N input to agricultural land from the application of effluent from ammonia stripping-scrubbing process by animal type a in basin j	kg N yr^{-1}

Equation	Parameter	Description	Unit
S8	$NFRV_{effluent.a}$	Nitrogen fertilizer replacement value for effluent from ammonia stripping-scrubbing process by animal type a	0-1
S9	$WSdif_{P.effluent.a.j}$	P input to agricultural land from the application of effluent from ammonia stripping-scrubbing process by animal type a in basin j (P is considered not to be affected by anaerobic digestion and ammonia stripping-scrubbing)	kg N yr^{-1}
S9	$PFRV_{effluent.a}$	Phosphorus fertilizer replacement value for effluent from manure processing by animal type a	0-1
3, 5, S10-S11	$RSdif_{f.y.j}$	Annual inputs of nutrients to rivers by form f, diffuse source y in basin j ($RSdif_{f.y.j} = RSdif_{f.y.ag.j} + RSdif_{f.y.nonag.j}$)	kg N or P yr^{-1}
S10	$RSdif_{f.y.ag.j}$	Annual inputs of nutrients to rivers by form f, diffuse agricultural source y in basin j	kg N or P yr^{-1}
S11	$RSdif_{f.y.nonag.j}$	Annual inputs of nutrients to rivers by form f, diffuse non-agricultural source y in basin j	kg N or P yr^{-1}
S10-S11, S14, S28	$Area_j$	The drainage area of basin j	km^2
S10-S11	$Agr_{fr.j}$	Fraction of agricultural area in basin j	0-1
S10-S11	EC_f	Watershed export coefficient for nutrient form f from organic matter leaching and P weathering	kg km^{-2} yr^{-1}
S10-S11	CL_f, CW_f	Uncalibrated export coefficient for nutrient form f from land to surface waters by organic matter leaching and P weathering	-
3, S12	$G_{f.j}$	Fraction of nutrient form f that is remained in soils of basin j after nutrient export from agricultural land by crop harvesting and animal grazing	0-1
S12	$WSdif_{e.ex.j}$	Export of nutrient element e from agricultural land via crop harvesting and animal grazing	kg N or P yr^{-1}
3, S13	$FE_{ws.f.j}$	Watershed export fraction of nutrient form f entering rivers of basin j as a function of runoff	0-1
S13	CR_f	Uncalibrated export coefficient of nutrient form f for runoff exporting nutrients from land to rivers	-
S10-S11, S13-S14	$Rnat_j$	Annual surface runoff from land to streams in basin j	m yr^{-1}
S14, S29	$Q_{nat.j}$	Natural river discharge at the outlet of basin j, before water is taken for consumption	km^3 yr^{-1}
4-5	$RSpnt_{f.y.j}$	Annual inputs of nutrients to rivers by form f, point source y in basin j	kg N or P yr^{-1}
4	Pop_j	Population in basin j	person

Equation	Parameter	Description	Unit
4	$fr_{pop.sew.j}$	Fraction of population that is connected to sewage systems basin j	0-1
4, S15-S16, S19	$WScap_{N.j}$, $WScap_{P.j}$	Human N and P excretion/consumption rates in domestic wastewater in basin j	kg N or P person^{-1} yr^{-1}
S15-S17	$I^{N}_{hum.j}$	Protein N intake for basin j	g N person^{-1} day^{-1}
S17	$GDP_{ppp.j}$	Annual gross domestic product at purchasing power parity in basin j	US$ 2005 person^{-1} yr^{-1}
4, S18	$hw_{frem.e.j}$	N and P removal rates during wastewater treatment (same for urban and rural treatment systems)	0-1
4, S18	$FEpnt_{f.j}$	Fraction of nutrient form f entering rivers of basin j by the effluent from wastewater treatment systems	0-1
S19	$Psludge_{t.j}$	P content of treated sewage sludge by treatment type t (e.g., secondary, tertiary) in basin j	kg P yr^{-1}
S20	$Precovered_{t.j}$	Recovered P from treated sewage sludge through 90% P recovery by treatment type t in basin j	kg P yr^{-1}
S21	$WSdif_{P.RPSS.j}$	P input to agricultural land from the application of recovered P from treated sewage sludge (RPSS) per basin j	kg P yr^{-1}
S21	$PFRV_{RPSS}$	Phosphorus fertilizer replacement value for recovered P from treated sewage sludge	0-1
5	$M_{f.y.j}$	Annual river export of nutrient by form f, source y in basin j	kg N yr^{-1}
5, S22-S23	$FE_{riv.f.outlet.j}$	Fraction of nutrient form f exported to the outlet of basin j	0-1
5	$FE_{riv.f.mouth.j}$	Fraction of nutrient form f exported from the basin j outlet to the river mouth	0-1
S24	$D_{f,j}$	Fraction of nutrient form f retained in reservoirs and (regulated) lakes in basin j	0-1
S24, S27	$Q_{act.i}$	Actual water discharge for reservoir i	km^3 yr^{-1}
S24-S26	$D_{f,i}$	Fraction of nutrient form f retained in reservoir i	0-1
S25	h_i	Depth of reservoir i	meter
S25-S27	$\tau_{R.i}$	Water residence time for reservoir i	year
S27	V_i	Volume of reservoir i	
S28	$L_{f,j}$	Fraction of nutrient form f retained in or/and lost from water systems (e.g., denitrification) in basin j	0-1

Equation	Parameter	Description	Unit
S22-S23, S29	$FQrem_j$	Fraction of nutrient form f removed from water systems in basin j by water consumption	0-1
S24, S29	$Q_{act.j}$	Actual water discharge at the outlet of basin j after water consumption	$km^3\ yr^{-1}$
S30	$FE_{riv.f.mouth.juT}$	Fraction of nutrient form f exported from the outlet of upstream tributary (juT) to the river mouth	0-1
S30, S34	$^{juT}FE_{riv.f.outlet.jmC}$	Fraction of nutrient form f exported from the outlet of upstream tributary (upper case: juT) to the outlet of the main channel in middlestream (lower case: jmC) sub-basins	0-1
S30, S35	$^{juT}FE_{riv.f.outlet.jdC}$	Fraction of nutrient form f exported from the outlet of upstream tributary (upper case: juT) to the outlet of the main channel in downstream (lower case: jdC) sub-basins	0-1
S31	$FE_{riv.f.mouth.jmT}$	Fraction of nutrient form f exported from the outlet of middlestream tributary (jmT) to the river mouth	0-1
S31, S36	$^{jmT}FE_{riv.f.outlet.jdC}$	Fraction of nutrient form f exported from the outlet of middlestream subbasin with the tributary (upper case: jmT) to the outlet of the main channel in downstream (lower case: jdC) sub-basin (the outlet of the this downstream subbasin is the river mouth)	0-1
S32	$FE_{riv.f.mouth.jmC}$	Fraction of nutrient form f exported from the outlet of middlestream main channel (jmC) to the river mouth	0-1
S32, S37	$^{jmC}FE_{riv.f.outlet.jdC}$	Fraction of nutrient form f exported from the outlet of middlestream subbasin with the main channel (upper case: jmC) to the outlet of the main channel in downstream (lower case: jdC) sub-basin (the outlet of the this downstream subbasin is the river mouth)	0-1
S33	$FE_{riv.f.mouth.jdC}$	Fraction of nutrient form f exported from the outlet of down-stream main channel (lower case: jdC), which is the river mouth	0-1
S34-S37	$D_{f.jmC}$, $D_{f.jdC}$	Fractions of nutrient form f retained in reservoirs of middlestream (jmC) and downstream (jdC) sub-basins with the main channel. These fractions were quantified for sub-basins using equations for $D_{DIN,j}$ and $D_{DIP,j}$ in Table S1.	0-1
S34-S37	$L_{f.jmC}$, $L_{f.jdC}$	Fractions of nutrient form f that is lost from surface waters of middlestream (jmC) and downstream (jdC) sub-basins with the main channel. These fractions were quantified using the equation for $L_{DIN,j}$ and $L_{DIP,j}$ in Table S1.	0-1
S34-S37	$FQrem_{jmC}$, $FQrem_{jdC}$	Fractions of nutrient form f that are lost from surface waters of middlestream (jmC) and downstream (jdC) sub-basins with the main channel via water consumption. These fractions were quantified using the equation for $FQrem_j$ in Table S1.	0-1
S34	$^{juT}A_{jmC}$	Drainage area (A) of the main channel in middlestream (lower case: jmC) sub-basins that exports nutrients from the outlet of upstream tributary (upper case: juT). This drainage area was calculated as the fraction to the total sub-basin area.	0-1

Equation	Parameter	Description	Unit
S35	${}^{juT}A_{jdC}$	Drainage area (A) of the main channel in downstream (lower case: jdC) sub-basins that exports nutrients from the outlet of upstream tributary (upper case: juT). This drainage area was calculated as the fraction to the total sub-basin area.	0-1
S36	${}^{jmT}A_{jdC}$	Drainage area (A) of the main channel (C) in downstream (lower case: jdC) sub-basin that exports nitrogen from the outlet of middlestream tributary (upper case: jmT). This drainage area was calculated as the fraction to the total sub-basin area.	0-1
S37	${}^{jmC}A_{jdC}$	Drainage area (A) of the main channel (C) in the downstream (lower case: jdC) subbasin that exports nitrogen from the outlet of middlestream main channel (upper case: jmC). This drainage area was calculated as the fraction to the total sub-basin area.	0-1

Table S4.3 Overview of the data sources and calculations for the alternative scenarios of this study for 2050 (see Ural-Janssen et al. (2024) for the overview of the data sources and calculations for the 2050 baseline). The scenarios were applied to the MARINA-Nutrients model for Europe to quantify nitrogen (N) emissions to the air from agriculture (including manure processing), inputs of N and phosphorus (P) to rivers, and their river exports to the coastal waters of Europe from diffuse and point sources. Source: The MARINA-Nutrients model for Europe.

Equation	Description
1-2	We quantified NH_3, N_2O and NO_x emissions from crop and livestock production for 2050 by European basins under the six alternative scenarios using the emissions factors (EFs) from the MITERRA-Europe model based on the data from 2017 (Duan et al., 2021). Emission factors (EFs) for ammonia (NH_3), nitrous oxide (N_2O) and nitrogen oxides (NO_x) emissions from animal housing, manure storage and application, and grazing were based on the MITERRA-Europe model for the EU-27 considering the practices of the Member States (based on manure type, stable type and application technique) and IPCC guidelines (Velthof et al., 2009; Lesschen et al., 2011). We calculated EF per basin for the current period (2017) based on the basin data. For example, we back-calculated the basin EFs (0-1) for N emissions to the air from animal housing systems by dividing the N emission to the air (kg N yr^{-1}) per basin by the N excretion per animal type (kg N yr^{-1}) per basin (e.g., $EF_{NH3h} = NH_{3h} / Nexc_h$). Similarly, we calculated the basin EFs (0-1) for N emissions to the air from agricultural soils by dividing the N emission to the air (kg N yr^{-1}) per basin by each N input to soil (kg N yr^{-1}) per basin. These EFs ($EF_{f.a.y.j}$, $EF_{f.y.j}$) were calculated at the basin scale for 2017 based on N emissions to the air from the MITERRA-Europe model (Duan et al., 2021). We used these calculated EFs to quantify N emissions to the air per basin by 2050, thereby accounting for spatially explicit drivers affecting nutrient losses to air and water (Table S2).
1	NH_3 emissions from animal housing (kg N yr^{-1}) were quantified per animal type (dairy cattle, other cattle, pig, poultry, other animals) as a function of animal N excretion (kg N yr^{-1}) and EF (0-1) per animal type.
1	NH_3 emissions from manure storage (kg N yr^{-1}) were quantified per animal type (dairy cattle, other cattle, pig, poultry, other animals) as a function of animal N excretion (kg N yr^{-1}) and EF (0-1) per animal type.
1	N_2O emissions from animal housing and manure storage (kg N yr^{-1}) were quantified per animal type (dairy cattle, other cattle, pig, poultry, other animals) as a function of animal N excretion (kg N yr^{-1}) and EF (0-1) per animal type.
1	NO_x emissions from animal housing and manure storage (kg N yr^{-1}) were quantified as for all animal types as a function of animal N excretion (kg N yr^{-1}) and EF (0-1).

Equation	Description
1, S2, S9	The N and P excretions by dairy cattle, other cattle and pigs (kg N or P yr^{-1}) were considered to be processed via anaerobic digestion (AD) and ammonia stripping-scrubbing (AS) after animal housing by 2050 under the ReNuRe and ReNuRe-NUE scenarios. The N excretions produced by the three animal types were 54% of total N excretion under the future baseline (Figure S4.2). We quantified the animal slurry to be sent to manure processing after animal housing as follows: $WSdif_{N.dairycattle.untreated.j} = Nexc_{dairycattle.h.j} - NH_{3dairycattle.h.j} - NH_{3dairycattle.s.j} - N_2O_{dairycattle.hs.j} - NO_{Xdairycattle.hs.j} - N_{2dairycattle.hs.j}$ (example for dairy cattle) where $WSdif_{N.dairycattle.untreated.j}$ is N excretion produced by dairy cattle in animal housing that is corrected for N emissions to the air from animal housing in basin j. This is also N input to agricultural land from the application of untreated manure that is corrected for N emissions to the air from animal housing in basin j under the future baseline (kg N yr^{-1}). $Nexc_{dairycattle.h.j}$ is N excretion produced by dairy cattle in animal housing in basin j (kg N yr^{-1}). $NH_{3dairycattle.h.j}$ is NH_3 emission produced by dairy cattle in animal housing in basin j (kg N yr^{-1}). $NH_{3dairycattle.s.j}$ is NH_3 emission produced by dairy cattle in manure storage in basin j (kg N yr^{-1}). $N_2O_{dairycattle.hs.j}$ is N_2O emission produced by dairy cattle in animal housing and manure storage in basin j (kg N yr^{-1}). $NO_{Xdairycattle.hs.j}$ is NO_x emission produced by dairy cattle in animal housing and manure storage in basin j (kg N yr^{-1}). $N_{2dairycattle.hs.j}$ is N_2 emission produced by dairy cattle in animal housing and manure storage in basin j (kg N yr^{-1}). We considered no N leaching in animal housing and manure storage systems. We assumed that the P excretions are not affected by the manure processing and left unchanged in the effluent (kg P yr^{-1}) from the ammonia stripping-scrubbing process. Therefore, P from this effluent fraction was indicated as "P from untreated manure application" in Figure 4.5, Figure S4.5, and Figure S4.6.
S3	No N losses were assumed during the AD process. Thus, the N content of the digested slurry was considered the same as the N content of the slurry per animal type.
S4	Low EF (0-1) was used for NH_3 emission from the ammonia stripping-scrubbing process due to e.g., NH_3 loss during aeration (Vingerhoets et al., 2023a). EF (0.00000009) for NH_3 emission from the ammonia stripping-scrubbing was derived from Vingerhoets et al. (2023a). NH_3 emission to the air the ammonia stripping-scrubbing process was quantified based on the N content of the digested slurry (kg N yr^{-1}) by animal type a in basin j, and $EF_{NH3.y}$ (0-1) for NH_3 emission from ammonia stripping-scrubbing.
S5	The N content of the ReNuRe product (kg N yr^{-1}) was quantified per animal type by subtracting the NH_3 (kg N yr^{-1}) emission from ammonia stripping-scrubbing from the N content of the digestate (kg N yr^{-1}), and multiplying the result with a coefficient (0-1). The coefficient was derived from literature to calculate how much of the digestate was converted to ReNuRe product per animal type during the ammonia stripping-scrubbing process. The coefficients per animal were $C_{dairycattle}$ = 0.39 kg N/kg N, $C_{othercattle}$ = 0.39 kg N/kg N and C_{pig} = 0.46 kg N/kg N (Vingerhoets et al., 2024). We considered the N input to agricultural land from the application of ReNuRe product as follows: $WSdif_{N.dairycattle.ReNuRe.j} = NReNuRe_{a.j}$ (Eqn. S5 for the calculation of $NReNuRe_{a.j}$) where $WSdif_{N.dairycattle.ReNuRe.j}$ is N input to agricultural land from the application of ReNuRe product that is corrected for N emissions to the air from animal housing and manure processing in basin j (kg N yr^{-1}).

Equation	Description
S6	The N content of the effluent (kg N yr^{-1}) from the ammonia stripping-scrubbing process was quantified per animal type by subtracting the NH_3 emission (kg N yr^{-1}) from ammonia stripping-scrubbing and the N content of the ReNuRe product (kg N yr^{-1}) from the N content of the digestate (kg N yr^{-1}). The ReNuRe products have a higher mineral N concentration than the untreated manure (Huygens et al., 2020). This mineral N content is more readily available for crop uptake and less prone to N emissions to the air, and hence ReNuRe products have a higher NUE (Huygens et al., 2020). As the ammonium (NH^+_4) in ReNuRe products is stabilized and more efficiently incorporated into the soil, this reduces NH_3 volatilization (Huygens et al., 2020). Also, separating the organic matter-rich fraction (effluent) in the ammonia stripping-scrubbing process reduces the risk of GHG emissions e.g., N_2O which are unavoidable from untreated manure due to the uncontrolled microbial activity during manure storage and application (Huygens et al., 2020).
2	NH_3 emissions from agricultural soils (kg N yr^{-1}) were quantified as a function of N inputs to agricultural land (kg N yr^{-1}) and EFs (0-1). N inputs to agricultural land were from the application of synthetic fertilizers, untreated manure, ReNuRe products and effluent from ammonia stripping-scrubbing, and manure deposited on land during grazing.
2	N_2O emissions from agricultural soils (kg N yr^{-1}) were quantified as a function of N inputs to agricultural land (kg N yr^{-1}) and EFs (0-1). N inputs to agricultural land were from the application of synthetic fertilizers, untreated manure, ReNuRe products and effluent from ammonia stripping-scrubbing, manure deposited on land during grazing, and atmospheric N deposition.
2	NO_x emissions from agricultural soils (kg N yr^{-1}) were quantified as a function of N inputs to agricultural land (kg N yr^{-1}) and EFs (0-1). N inputs to agricultural land were from the application of synthetic fertilizers and untreated manure, and manure deposited on land during grazing.
2	EFs (0-1) to calculate the N emissions to the air from the application of the ReNuRe products and the stripping-scrubbing effluent were derived from literature (Foqué & Demeyer, 2009; Ravi et al., 2023). EFs for the application of the ReNuRe products were 0.02 and 0.0038 for NH_3 and N_2O, respectively, being lower than the EFs for the application of untreated manure. EFs for NH_3 emissions from the application of the stripping-scrubbing effluent were 0.096 and 0.129 for cattle and pig, respectively. EFs for N_2O emissions from the application of the stripping-scrubbing effluent were 0.01 for both cattle and pig. No NO_x emissions from the application of the ReNuRe products and the stripping-scrubbing effluent were considered.
S7	We quantified how much ReNuRe products can substitute the synthetic N fertilizers by 2050 under the ReNuRe scenario by using mineral N fertilizer replacement value (NFRV, 0-1). We considered $NFRV_{ReNuRe}$=1 based on the characteristics of the ReNuRe products (ammonium salts) that can be regarded as a substitute for mineral N (Huygens et al., 2020; Vingerhoets et al., 2024). Then, we subtracted the amount of the ReNuRe products produced per basin (kg N yr^{-1}) from the synthetic N fertilizer application per basin (kg N yr^{-1}) as in the future baseline. We considered the difference as the synthetic N fertilizer needs for that basin (kg N yr^{-1}) by 2050 under the ReNuRe scenario to maintain the same amount of reactive N inputs to agricultural land as in the future baseline (Figure S4.5).
S8	The effluent from ammonia stripping-scrubbing (kg N yr^{-1}) was considered to be applied to agricultural land, as well. $NFRV_{effluent.a}$ was derived from literature as 0.60 for both cattle and pig (VLM, 2023).

Equation	Description
S9	We quantified the P input to agricultural land (kg P yr^{-1}) from the application of the ammonia stripping-scrubbing effluent as follows: $WSdif_{P,effluent.a,j} = Pexcretion_{a.j} \times PFRV_{effluent.a}$ P was considered not to be affected by anaerobic digestion and ammonia stripping-scrubbing. Thus, the P content of the effluent was the same as the P excretion per animal. $PFRV_{effluent.a}$ was derived from literature as 0.80 and 1 for cattle and pig, respectively (Meers et al., 2020).
3-4, S12	Inputs of N and P to land (kg N or P yr^{-1}) were quantified for the future baseline scenario in our previous work (Ural-Janssen et al., 2024). Inputs of N and P to agricultural land (the application of synthetic fertilizers and untreated manure, manure deposited on land during grazing, atmospheric deposition, and biological N_2 fixation by crops) were derived from the MITERRA-Europe model for 2017 (Duan et al., 2021). N and P yields from crop harvesting and animal grazing were also from the MITERRA-Europe model for 2017 (Duan et al., 2021). We projected nutrient inputs to agricultural land and exported from agricultural land for 2050 based on the changes in the future baseline scenario (see Ural-Janssen et al. (2024) for details). Inputs of N to non-agricultural land (atmospheric N deposition and biological N_2 fixation by natural vegetation) were derived from the IMAGE model for 2050 based on the SSP5 regarding our future baseline scenario (SSP5-RCP8.5). For the ReNuRe, Pmed and Padv scenarios, we kept the overall N and P use efficiencies on agricultural land (NUE, PUE) as the same as in the future baseline. Hence, we considered the total nutrient inputs to land by 2050 as in the future baseline for these alternative scenarios. We only changed the synthetic fertilizer use by substituting with the BBFs (ReNuRe products under the ReNuRe scenario, and RPSS under the Pmed and Padv scenarios). For the ReNuRe-NUE, Pmed-PUE and Padv-PUE scenarios, we considered higher NUE (N harvested/total N input to agricultural land) and PUE (P harvested/total P input to agricultural land) together with the substitution of synthetic fertilizers by BBFs. By increasing the overall nutrient use efficiencies, the total inputs of nutrients to land were reduced. In this case, we reduced the synthetic fertilizer use as a function of the reduction in total nutrient input to land. See Table S4 for the details of the alternative scenarios.
3, S12	Uncalibrated approach Li et al. (2022b) was used to calculate the fraction of nutrients applied to agricultural land that remained in soils of basin. In this approach, the world basins were clustered based on the climate zones from FAO (2007) to calculate the fractions of nutrients retained in soils after crop harvesting and animal grazing. Among the European basins (593 basins and 8 sub-basins of the Danube River), 223 basins were in subtropic/tropic zones and assumed to have a rather wet climate, and 378 basins were in other temperature zones and assumed to have a rather dry climate (Li et al., 2022b). Following this approach: o If $G_{f.j} < 0$, $G_{f.j}$ was validated as 0.05 for the basins in subtropic/tropic zones, and as 0.1 for the basins in other temperature zones. Exports of N and P from agricultural land by crop harvesting and animal grazing ($WSdif_{e.ex.j}$, kg N or P yr^{-1}) were quantified for the future baseline scenario in our previous work (Ural-Janssen et al., 2024).

Equation	Description
3-4, S13	Uncalibrated approach of Li et al. (2022b) was used to calculate the export fraction of nutrients entering rivers of basin (0-1). Following this approach, CR_f differs based on $Rnat_j$ as follows: o If $Rnat_j \geq 0.1$ m yr^{-1}, CR_{DIN} = 0.9 and if $Rnat_j < 0.1$ m yr^{-1}, CR_{DIN}= 0.8 o If $Rnat_j \geq 0.1$ m yr^{-1}, CR_{DON}= 0.02 and if $Rnat_j < 0.1$ m yr^{-1}, CR_{DON}= 0.01 o If $Rnat_j \geq 0.1$ m yr^{-1}, CR_{DIP}= 0.15 and if $Rnat_j < 0.1$ m yr^{-1}, CR_{DIP}= 0.1 o If $Rnat_j \geq 0.1$ m yr^{-1}, CR_{DOP}= 0.02 and if $Rnat_j < 0.1$ m yr^{-1}, CR_{DOP}= 0.01 o Any of $FE_{ws.f.j} > 0.9$ was assumed that equal to 0.9.
S10-S11	Nutrient inputs to rivers (kg N or P yr^{-1}) through leaching of organic matter and weathering of P-contained minerals from agricultural and non-agricultural areas are parameterized processes, estimated following the approach of Strokal et al. (2016). Parameterized nutrient inputs to rivers were quantified in our previous work for the future baseline scenario (Ural-Janssen et al., 2024). In this study, we kept them as in the future baseline for the alternative scenarios. Nutrient inputs to rivers through leaching of organic matter from agricultural areas were quantified as a function of basin area, agricultural fraction of the basin area, the watershed export rate for nutrient form, the uncalibrated export coefficient for nutrient form and annual surface runoff from land to streams. Nutrient inputs to rivers through weathering of P-contained minerals from agricultural areas were quantified in the same way. Nutrient inputs to rivers through leaching of organic matter and weathering of P-contained minerals from non-agricultural areas were quantified based on the fraction of non-agricultural area per basin. Basin areas (km^2) were derived from Strokal et al. (2016). The fraction of agricultural area per basin (0-1) was quantified by dividing the agricultural area projected for 2050 (Ural-Janssen et al., 2024) by basin area. The watershed export rates (kg km^{-2} yr^{-1}) for organic matter leaching and P weathering were derived from Mayorga et al. (2010): o EC_{DON} = 280 kg km^{-2} yr^{-1} and EC_{DOP} = 15 kg km^{-2} yr^{-1} for leaching of organic matter o EC_{DIP} = 26 kg km^{-2} yr^{-1} for P weathering The uncalibrated export coefficients (unitless) were derived from Li et al. (2022b) to calculate the inputs of DON and DOP to rivers from organic matter leaching, and inputs of DIP to rivers from P weathering: o If $Rnat_j \geq 0.1$ m yr^{-1}, CL_{DON} = 1.0 and if $Rnat_j < 0.1$ m yr^{-1}, CL_{DON}= 0.9 for leaching of organic matter o If $Rnat_j \geq 0.1$ m yr^{-1}, CL_{DOP}= 1.0 and if $Rnat_j < 0.1$ m yr^{-1}, CL_{DOP}= 0.9 for leaching of organic matter o If $Rnat_j \geq 0.1$ m yr^{-1}, CW_{DIP}= 0.40 and if $Rnat_j < 0.1$ m yr^{-1}, CW_{DIP}= 0.20 for P weathering
S14-S17	All these parameters were quantified in our previous work for the future baseline scenario (Ural-Janssen et al., 2024), and used for the alternative scenarios in this study. Natural river discharges per basin (km^3 yr^{-1}) were derived from van Vliet et al. (2016) for the future baseline (by averaging the data over 30 years of the period 2020-2050 to avoid the influence of climate extremes), and also used for the alternative scenarios. Per capita excretion or consumption rates of nutrients (kg N or P person^{-1} yr^{-1}) in domestic wastewater per basin were derived from Strokal et al. (2021) for the future baseline and used for the alternative scenarios in this study.

Equation	Description
5, S18	We assumed that the urbanization-related inputs per basin will follow the storyline of SSP5. Projections on total, urban and rural population (person) and sewer connection rates (%) were derived from Strokal et al. (2021) per basin for the future baseline scenario. Fractions of N and P removal in wastewater treatment systems per basin ($hw_{frem.e.j}$, 0-1, the same for both urban and rural) were derived from Strokal et al. (2021) for the future baseline. The maximum N removal rate was 0.88 for 2050 under the Pmed and Padv scenarios. Fractions of nutrient forms entering rivers from point source ($FEpnt_{f.j}$, 0-1) were constants derived from Mayorga et al. (2010) for DON, DIP and DOP as 0.14, 1 and 0.01, respectively, for the future baseline. For DIN, we quantified this fraction in our previous work for the future baseline scenario (Ural-Janssen et al., 2024), and used for the alternative scenarios in this study.
S19-S21	We quantified the P removal from the wastewater treatment plant (WWTP) per basin as a function of per capita P excretion/consumption rate (kg P person^{-1} yr^{-1}), population connected to sewage systems (person) and P removal rate in wastewater treatment systems (0-1). By considering all the treated P stays in sewage sludge (> 90% of all P ends up concentrated in sewage sludge according to Fang et al. (2021)), we quantified the recovered P from treated sewage sludge (RPSS, kg P yr^{-1}). To this end, we quantified the RPSS by 90% P recovery by 2050 under the P-focused alternative scenarios (Eqn. S20). We quantified the P input to agricultural land (kg P yr^{-1}) by the application of RPSS as a function of PFRV (Eqn. S21). We considered $PFRV_{RPSS}$=1 as equivalent to synthetic P fertilizers based on expert opinion. Then, we subtracted the amount of the RPSS produced per basin from the synthetic P fertilizer use per basin as in the future baseline scenario. We considered the difference as the synthetic P fertilizer need for that basin (kg P yr^{-1}) by 2050 under the Pmed and Padv scenarios to maintain the same amount of P inputs to agricultural land as in the future baseline (Figure S4.5). For most of the European basins, the RPSS ≥ the synthetic P fertilizer as in the future baseline.
S22-S37	All these parameters were quantified in our previous work for the future baseline scenario (Ural-Janssen et al., 2024), and we used them also for the alternative scenarios.

Table S4.4 Overview of the six alternative scenarios and their assumptions to quantify nutrient inputs to agricultural areas, nitrogen (N) emissions to the air from agriculture, inputs of N and phosphorus (P) to rivers, and their river exports to the coastal waters of Europe from diffuse and point sources. Scenario names are short for REcovered Nitrogen from manURE (ReNuRe), ReNuRe + N use efficiency (ReNuRe-NUE), P medium (Pmed), P advanced (Padv), P medium + P use efficiency (Pmed-PUE) and P advanced + PUE (Padv-PUE), respectively. RPSS is short for recovered P from treated sewage sludge and WWTPs is short for wastewater treatment plants. References were used to make some assumptions. Source: Scenario development.

Scenario name	Targeted nutrient	Focus	Description and underlying assumptions for 2050	Reference
ReNuRe	N	ReNuRe products	o ReNuRe products will be produced through the anaerobic digestion and ammonia stripping-scrubbing of the animal slurry from dairy cattle, other cattle, and pigs (54% and 50% of total N and P excretions, respectively, Figure S4.2). o Synthetic N fertilizers will be replaced by ReNuRe products by considering the same amount of reactive N inputs to agricultural land as in the future baseline scenario (Table S3). o In 2050, atmospheric N deposition on agricultural areas that is in the future baseline will be reduced. 50% of atmospheric N deposition consists of NH_3 emissions based on Kelleghan et al. (2021) assuming that emission and deposition occur within the same basin. As a result, we assumed that reductions in N deposition are related to the reductions in NH_3 emissions from agriculture. First, we estimated the percent reduction in NH_3 emissions from agriculture per basin as a result of the ReNuRe products. Then, we considered that 50% of these reductions will be applied to the reduction of atmospheric N deposition on agricultural areas. o NUE (N harvested/total N input to agricultural land) will be as in the future baseline.	(Huygens et al., 2020; Kelleghan et al., 2021)
ReNuRe-NUE	N	ReNuRe products+increased NUE	o All assumptions of the ReNuRe scenario will be applied. o NUE will be increased to 0.75 for the basins with NUE < 0.75 (Figure S4.3). o Synthetic N fertilizer application that is in the future baseline will be reduced based on the increased NUE per basin.	(Schulte-Uebbing & de Vries, 2021)

Pmed	P	RPSS+medium upgrade of WWTPs	o o o o	All basins will upgrade their nutrient removal technologies in the WWTPs to one level up. For example, basins with primary treatment will upgrade the treatment technology to secondary treatment (overall 42% of N and 51% of P removal). Basins with secondary treatment will upgrade the treatment technology to tertiary treatment (overall 88% of N and 95% of P removal). RPSS will be produced through 90% of P recovery from treated sewage sludge from the WWTPs based on Huygens et al. (2019). Synthetic P fertilizers will be replaced by RPSS relative to the future baseline. PUE (P harvested/total P input to agricultural land) will be as in the future baseline.	(Huygens et al., 2019)
Padv	P	RPSS+advanced upgrade of WWTPs	o o o o	All basins will upgrade their nutrient removal technologies to tertiary treatment (overall 88% of N and 95% of P removal). RPSS will be produced through 90% of P recovery from treated sewage sludge from the WWTPs based on Huygens et al. (2019) as in the Pmed scenario. Synthetic P fertilizers will be replaced by RPSS relative to the future baseline as in the Pmed scenario. PUE will be as in the future baseline.	(Huygens et al., 2019)
Pmed-PUE	P	RPSS+medium upgrade of WWTPs+increased PUE	o o o	All assumptions of the Pmed scenario will be applied. PUE will be increased to 0.75 for the basins with PUE < 0.75 (Figure S4.3). Synthetic P fertilizer application that is in the future baseline will be reduced based on the increased PUE per basin.	(Schulte-Uebbing & de Vries, 2021)
Padv-PUE	P	RPSS+advanced upgrade of WWTPs+increased PUE	o o o	All assumptions of the Padv scenario will be applied. PUE will be increased to 0.75 for the basins with PUE < 0.75 (Figure S4.3). Synthetic P fertilizer application that is in the future baseline will be reduced based on the increased PUE per basin.	(Schulte-Uebbing & de Vries, 2021)

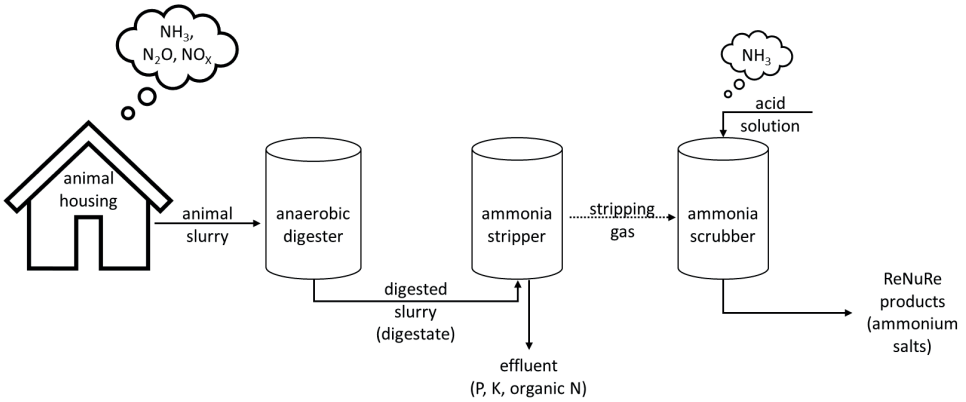


Figure S4.1 Schematic overview of the production of ReNuRe products and effluent through animal housing, anaerobic digestion and ammonia stripping-scrubbing. Arrows indicate liquid (solid line) and gas (dashed line) forms. Ammonia (NH_3), nitrous oxide (N_2O) and nitrogen oxides (NO_x) are emissions to the air from animal housing and manure processing. Source: Scenario development.

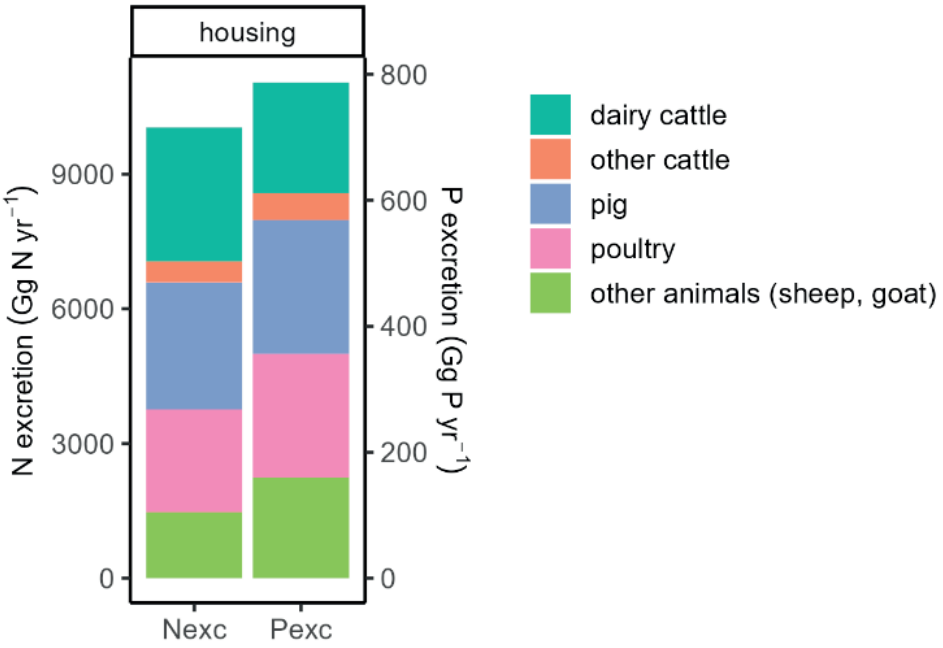


Figure S4.2 Nitrogen (N) and phosphorus (P) excretions (exc) produced in housing systems in 2050 under the baseline scenario (Gg N or P yr^{-1}). The baseline is based on the combination of SSP5-RCP8.5 (Shared Socioeconomic Pathway 5 and Representative Concentration Pathway 8.5). Source: The MARINA-Nutrients model for Europe.

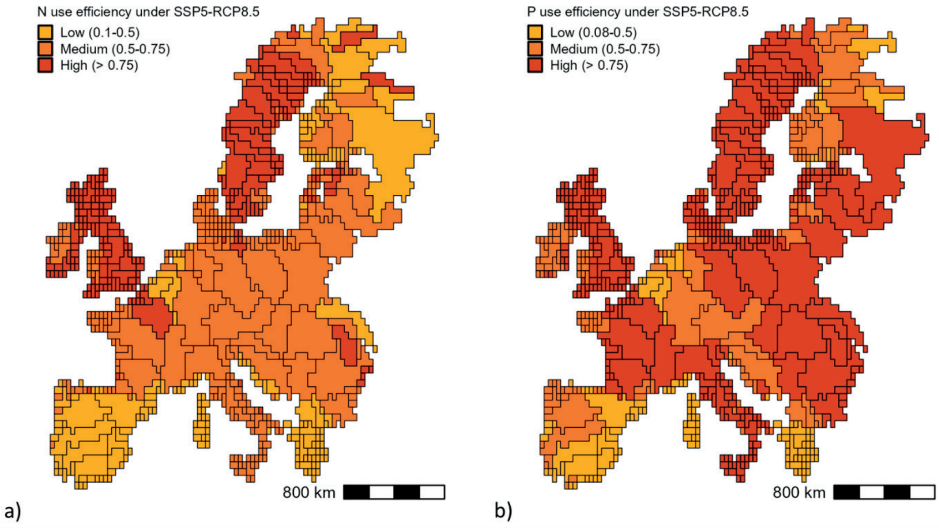


Figure S4.3 Overall nutrient use efficiencies under the future baseline scenario (unitless). a) Nitrogen use efficiency (NUE). b) Phosphorus use efficiency (PUE). The baseline is based on the combination of SSP5-RCP8.5 (Shared Socioeconomic Pathway 5 and Representative Concentration Pathway 8.5). The classification (low, medium and high) is arbitrary. Source: The MARINA-Nutrients model for Europe.

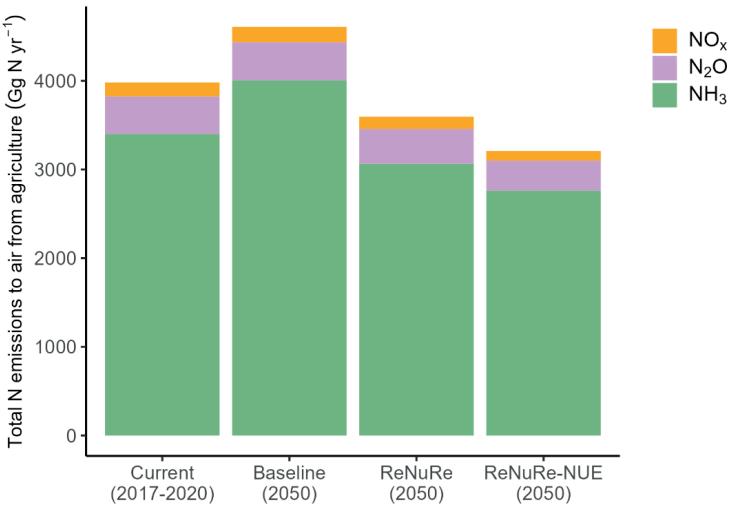


Figure S4.4 Total nitrogen (N) emission to the air from agriculture for the current (2017-2020), future baseline, and two alternative scenarios in 2050 (Gg N yr^{-1}). N emissions include ammonia (NH_3), nitrous oxide (N_2O) and nitrogen oxides (NO_x). The baseline is based on the combination of SSP5-RCP8.5 (Shared Socioeconomic Pathway 5 and Representative Concentration Pathway 8.5). The two alternative scenarios are ReNuRe (ReNuRe products) and ReNuRe-NUE (ReNuRe products + increased NUE). ReNuRe stands for REcovered Nitrogen form manURE and NUE is short for nitrogen use efficiency. Source: The MARINA-Nutrients model for Europe.

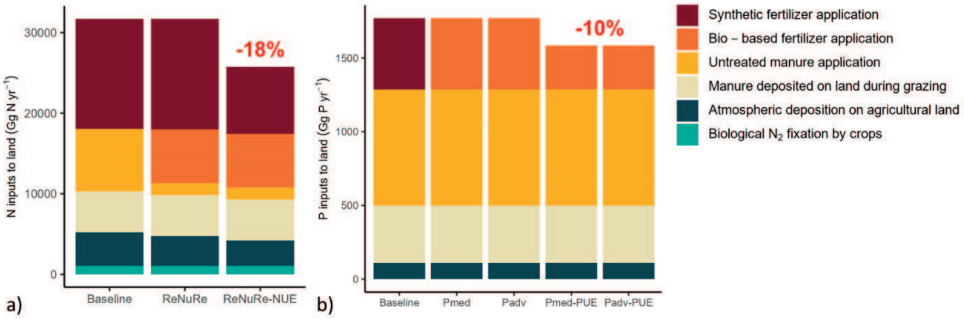


Figure S4.5 Inputs of nutrients to agricultural areas under the baseline and six alternative scenarios in 2050 (Gg N or P yr^{-1}). a) Nitrogen (N) and b) phosphorus (P). The baseline is based on the combination of SSP5-RCP8.5 (Shared Socioeconomic Pathway 5 and Representative Concentration Pathway 8.5). The six alternative scenarios are ReNuRe (ReNuRe products), ReNuRe-NUE (ReNuRe products + increased NUE), Pmed (RPSS + medium upgrade of WWTPs), Pmed-PUE (RPSS + medium upgrade of WWTPs + increased PUE), Padv (RPSS + advanced upgrade of WWTPs) and Padv-PUE (RPSS + advanced upgrade of WWTPs + increased PUE). ReNuRe stands for REcovered Nitrogen form maNURE, NUE is short for nitrogen use efficiency, RPSS is short for recovered phosphorus from treated sewage sludge, WWTPs is short for wastewater treatment plants, and PUE is short for phosphorus use efficiency. Source: The MARINA-Nutrients model for Europe.

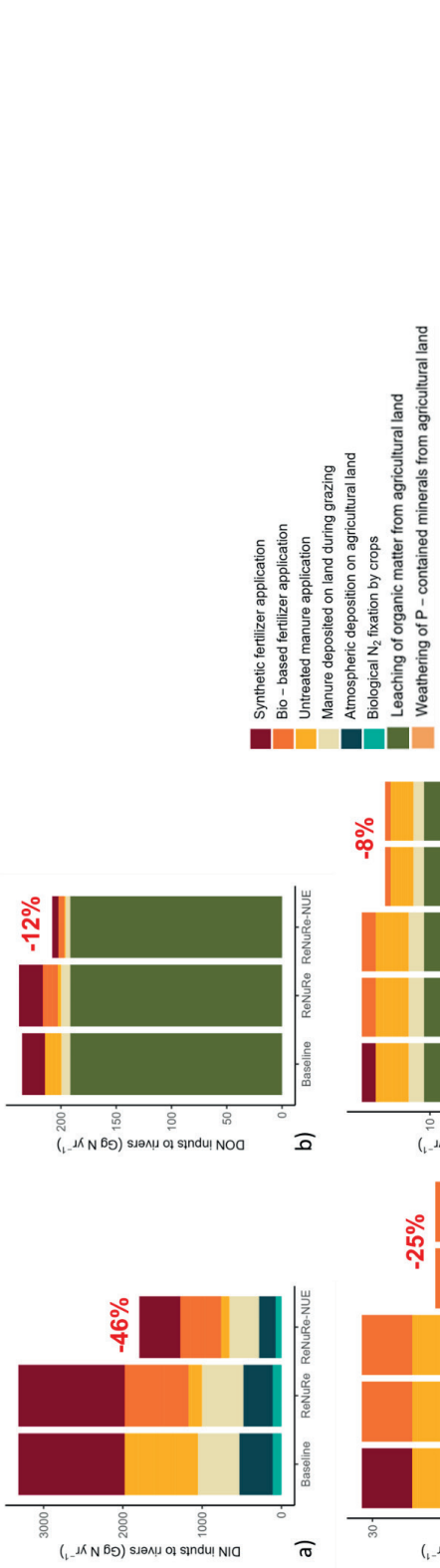


Figure S4.6 Inputs of nutrients to rivers from agricultural sources under the baseline and six alternative scenarios in 2050 (Gg N or P yr^{-1}). a) Dissolved inorganic nitrogen (DIN). b) Dissolved organic nitrogen (DON). c) Dissolved inorganic phosphorus (DIP). d) Dissolved organic phosphorus (DOP). The baseline is based on the combination of SSP5-RCP8.5 (Shared Socioeconomic Pathway 5 and Representative Concentration Pathway 8.5). The six alternative scenarios are ReNuRe (ReNuRe products), ReNuRe-NUE (ReNuRe products + increased NUE), Pmed (RPSS + medium upgrade of WWTPs), Pmed-PUE (RPSS + medium upgrade of WWTPs + increased PUE), Padv (RPSS + advanced upgrade of WWTPs) and Padv-PUE (RPSS + advanced upgrade of WWTPs + increased PUE). ReNuRe stands for REcovered Nitrogen form manURE, NUE is short for nitrogen use efficiency, RPSS is short for recovered phosphorus from treated sewage sludge, WWTPs is short for wastewater treatment plants, and PUE is short for phosphorus use efficiency. Source: The MARINA-Nutrients model for Europe.

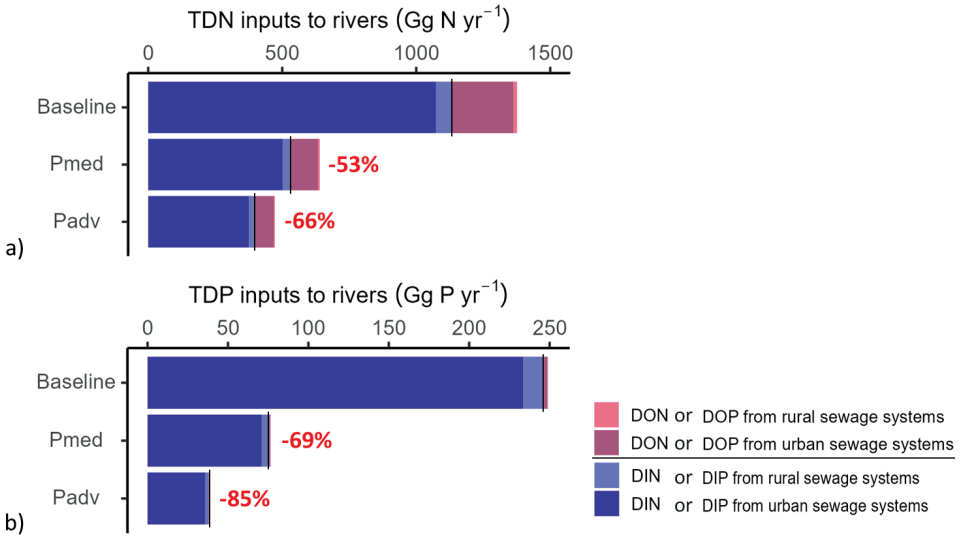


Figure S4.7 Nutrient inputs to rivers from point sources under the baseline and two alternative scenarios by 2050 (Gg N or P yr^{-1}). a) Total dissolved nitrogen (TDN) including dissolved inorganic (DIN) and dissolved organic nitrogen (DON) from rural and urban wastewater treatment systems. b) Total dissolved phosphorus (TDP) including dissolved inorganic (DIP) and dissolved organic phosphorus (DOP) from rural and urban wastewater treatment systems. The percentages are estimated relative to the baseline. The baseline is based on the combination of SSP5-RCP8.5 (Shared Socioeconomic Pathway 5 and Representative Concentration Pathway 8.5). The two alternative scenarios are Pmed (RPSS + medium upgrade of WWTPs) and Padv (RPSS + advanced upgrade of WWTPs). RPSS is short for recovered phosphorus from treated sewage sludge and WWTPs is short for wastewater treatment plants. Source: The MARINA-Nutrients model for Europe.

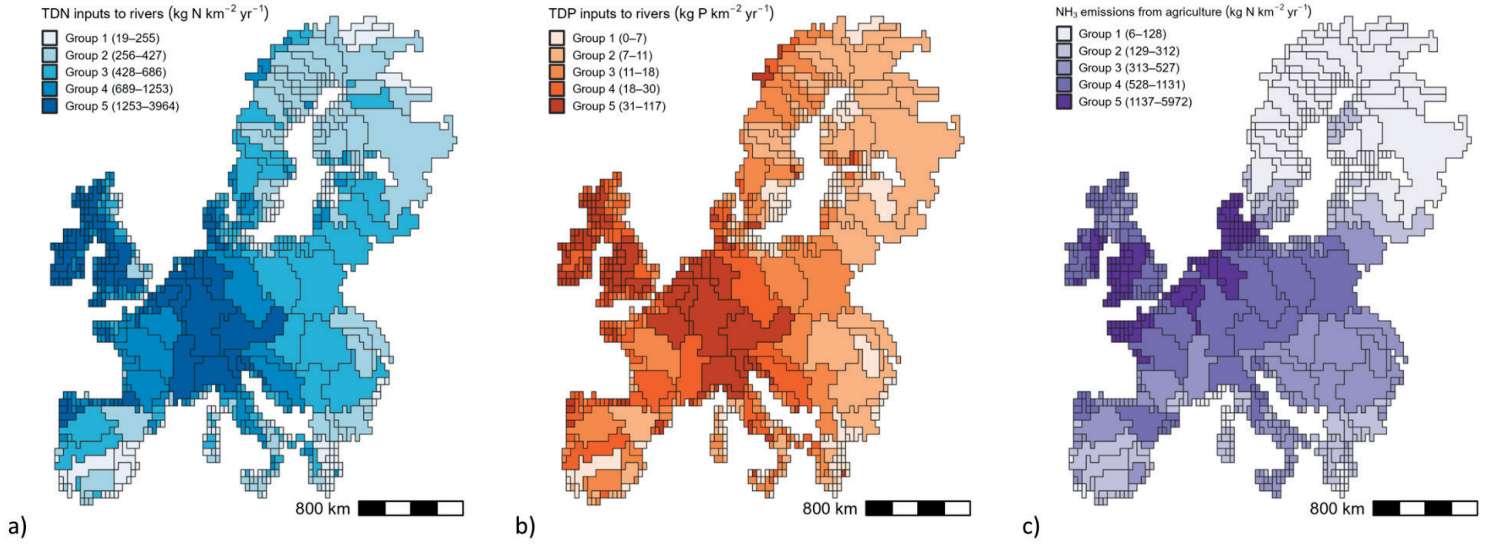


Figure S4.8 a) Total dissolved N (TDN) inputs to rivers from agricultural, non-agricultural and urbanization-related sources under the ReNuRe-NUE (ReNuRe products + increased NUE) scenario (kg N km^{-2} yr^{-1}). b) Total dissolved P (TDP) inputs to rivers from agricultural, non-agricultural and urbanization-related sources under the Padv-PUE (RPSS + advanced upgrade of WWTPs + increased PUE) scenario (kg P km^{-2} yr^{-1}). c) Total NH_3 emissions to the air from agricultural sources under the ReNuRe-NUE (ReNuRe products + increased NUE) scenario (kg N km^{-2} yr^{-1}). Groups 1-5 are defined per pollution type, and each group has 20% of basins from lowest to highest pollution. ReNuRe stands for REcovered Nitrogen from manURE, NUE is short for nitrogen use efficiency, RPSS is short for recovered phosphorus from treated sewage sludge, WWTPs is short for wastewater treatment plants, and PUE is short for phosphorus use efficiency. Source: The MARINA-Nutrients model for Europe.

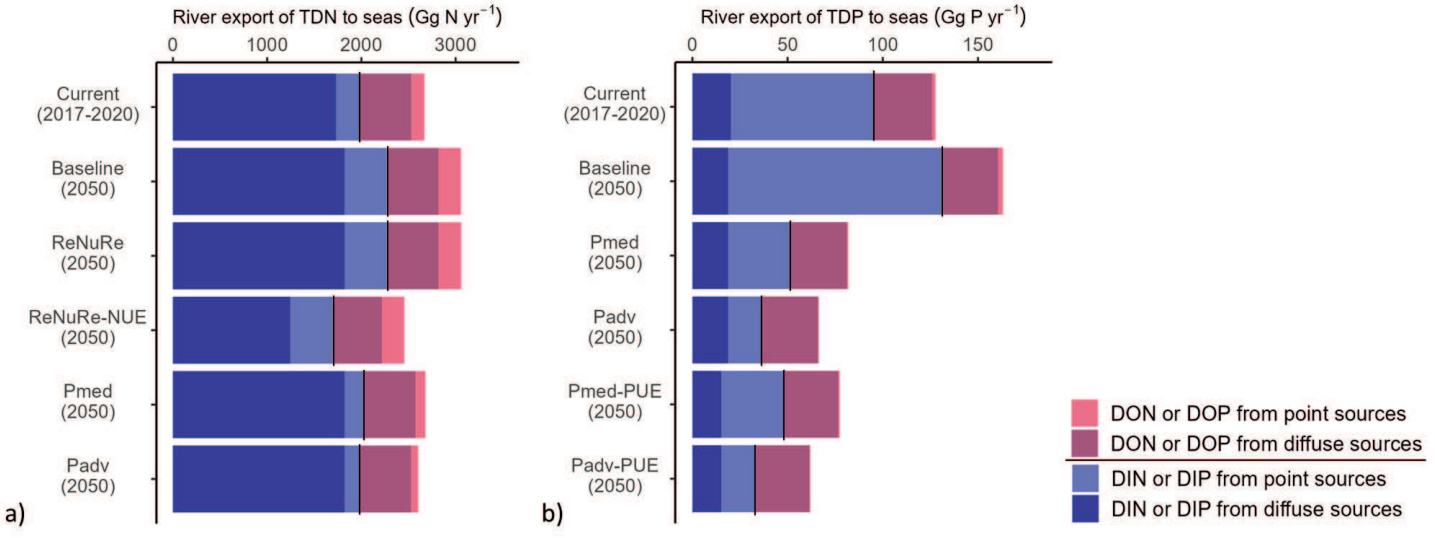


Figure S4.9 River exports of nutrients to seas from diffuse (agricultural, non-agricultural) and point (sewage systems) sources for the current (2017-2020), baseline and six alternative scenarios by 2050 (Gg N or P yr^{-1}). a) Total dissolved nitrogen (TDN) consisting of dissolved inorganic and organic forms. b) Total dissolved phosphorus (TDP) consisting of dissolved inorganic and organic forms. The baseline is based on the combination of SSP5-RCP8.5 (Shared Socio-economic Pathway 5 and Representative Concentration Pathway 8.5). The six alternative scenarios are ReNuRe (ReNuRe products), ReNuRe-NUE (ReNuRe products + increased NUE), Pmed (RPSS + medium upgrade of WWTPs), Pmed-PUE (RPSS + medium upgrade of WWTPs + increased PUE), Padv (RPSS + advanced upgrade of WWTPs) and Padv-PUE (RPSS + advanced upgrade of WWTPs + increased PUE). ReNuRe stands for REcovered Nitrogen form manURE, NUE is short for nitrogen use efficiency, RPSS is short for recovered phosphorus from treated sewage sludge, WWTPs is short for wastewater treatment plants, and PUE is short for phosphorus use efficiency. Source: The MARINA-Nutrients model for Europe.

Supplementary Text S4.2 Uncertainties and limitations

Our estimates have uncertainties and limitations associated with two main aspects: (1) model approaches, inputs and parameters, and (2) scenario assumptions.

Model approaches, inputs and parameters

MARINA-Nutrients is a part of the MARINA model family (The MARINA models - WUR), which has a lot of scientific trust in model development and evaluation for nutrients at different scales built over the past 10 years (Strokal et al., 2016; Chen et al., 2019b; Wang et al., 2020a; Ural-Janssen et al., 2023; Micella et al., 2024c). "Building trust circle" has been developed to address uncertainties in model inputs, approaches, and outputs by covering the entire model development chain (Strokal et al., 2024). Results showed that the model outputs are rather sensitive to variabilities in hydrology (e.g., water discharges) as well as to human activities on land (e.g., synthetic fertilizer application) (Strokal et al., 2016; Wang et al., 2020a). The MARINA-Nutrients model for Europe has been validated for the river exports of N and P based on long term observations (Ural-Janssen et al., 2023). Here, we applied the model for future analysis. MARINA-Nutrients is a process-based, steady-state model that takes a lumped approach to quantify the nutrient losses by river basins. The model does not account for soil dynamics, particulate nutrients, and losses via soil erosion. As a consequence, total nutrient losses to European rivers and seas could be underestimated especially in areas that suffer from erosion. On the other hand, the contribution of agricultural inputs to P losses might be overestimated in basins where crop P uptake exceeds the actual P inputs (Muntwyler et al., 2024). The contribution of soil P erosion could be within the range of estimated P losses based on the PUE values used in our study. This could imply that positive and negative P surpluses across basins cancel out or actual P surpluses on agricultural land in 2050 remain above zero in most EU countries (Muntwyler et al., 2024).

Moreover, we made several assumptions in our projections. Examples are reducing atmospheric N deposition on agricultural areas as a result of reduced NH_3 emissions from agriculture, replacing manure storage with manure processing systems, and increasing nutrient use efficiencies. Our assumptions may lead to over-/underestimation of our results for future N emissions to the air and N inputs to rivers and seas by European basins. Nevertheless, our assumptions are supported by literature and expert knowledge. Here we modeled the impact of production and application of BBFs from manure and sewage sludge treatments, increased nutrient use efficiency and improved sewage treatments on future nutrient losses from agricultural, non-agricultural and urbanization-related sources given a future baseline scenario. These sources mainly contribute to dissolved nutrient losses and when validated with the measured nutrient exports by rivers, the model performed reasonably well for the period of 2017-2020 (e.g., Pearson's determination coefficient = 0.85, Nash-Sutcliffe efficiency = 0.85) (Ural-Janssen et al., 2023). We recommend future research to look into finer resolutions for

in-depth analysis of the effects of BBFs on reducing air and water pollution at a field scale, and to investigate the implications of abovementioned limitations. A finer resolution could yield more reliable insights into the spatial dynamics of nutrient transport and its ecological implications.

Furthermore, the uncertainty is associated with data and parameters, especially the EFs to quantify the NH_3, N_2O and NO_x emissions from livestock and manure treatment systems, and field application. For example, Hou et al. (2017) found that the uncertainty of the model output (> 70%) mainly arises from the EFs for calculating NH_3 emissions from animal housing and manure storage, and for calculating N_2O emissions from manure application. In our study, the EFs calculated per basin for 2017 were used to project future N emissions to the air from animal housing, manure storage, the application of synthetic fertilizers and untreated manure, and manure deposited on land during grazing. By this approach we might overestimate N emissions by 2050, as future improvements in agricultural systems could lead to reduced emissions. On the other hand, the literature and expert-knowledge based NFRV and PFRV show substantial variation across individual sites, contributing to uncertainty on the more regional estimated effects of BBFs on nutrient losses to the environment. Thus, we acknowledge the uncertainty in the data and EFs used in our study.

Scenario assumptions

Our future baseline scenario accounts for a pessimistic future under the combination of SSP5 (fossil fuel development) and RCP8.5 (high-end climate change) pathways for population growth, urbanization and intensive agriculture. We made a number of assumptions in our alternative scenarios. For example, we considered the RPSS to be used as BBFs under the P-focused scenarios. However, P recycling of sewage sludge is still not allowed in several EU countries (e.g., the Netherlands). We identified the pollution reductions in rivers and coastal waters if this would be possible for the EU-28 (including the UK) by 2050. The assumption of increasing nutrient use efficiencies to 0.75 in basins below this value following Schulte-Uebbing and de Vries (2021), represents an optimistic yet policy-relevant target. However, achieving such nutrient use efficiency levels on agricultural land in practice might be constrained by various agronomic (e.g., farming systems) and socioeconomic (e.g., nutrient recycling systems) factors. Moreover, variability in land use intensity, soil and crop types, and hydrological conditions within basin could lead to over- or underestimation of nutrient retention and losses in certain areas. This in turn influences the feasibility of achieving the targeted nutrient use efficiency.

S5: Supplementary materials for Chapter 5

Table S5.1 Summary of the key findings of this PhD thesis across chapters in relation to the knowledge gaps and research sub-objectives (SO) presented in Chapter 1. EU is the European Union, MARINA-Nutrients is the Model to Assess River Inputs of pollutaNts to seAs for Nutrients, N is nitrogen, P is phosphorus, and ReNuRe stands for REcovered Nitrogen from manURE.

Chapter	Knowledge gap	Research sub-objective	Highlights
2	(1) Lack of an integrated model that quantifies the contribution of agriculture to nutrient emissions to the air, and surface waters (rivers and coastal waters) in a spatially explicit way across the EU and the UK basins	SO1: To quantify the contribution of agricultural activities to air, river and coastal water pollution simultaneously with a focus on nutrients in European basins	• A new European version of the MARINA-Nutrients model was developed to calculate nutrient emissions to air, rivers and coastal waters by river basins. • In the period of 2017-2020, agriculture was responsible for 55% of N and sewage for 67% of P in rivers. • Almost two-fifths of reactive N emissions to air were from animal housing and storage. • Nearly a third of the European basin areas were pollution hotspots producing over half of N emissions to air and nutrients in rivers. • Over 25% of river export of N ended up in the Atlantic Ocean and P in the Mediterranean Sea.
3	(2) Limited knowledge of basin-specific targets and required reductions in river exports of N and P to avoid coastal eutrophication in Europe in the future under global change	SO2: To assess the future river exports of N and P, and identify required reductions to avoid coastal eutrophication in Europe under global change	• Nutrient export to European seas is expected to increase by 13-28% under global change. • Manure and fertilizers together contribute to river export of N by 35% in 2050. • Sewage systems are responsible for 70% of future P export by rivers. • By 2050, the top ten polluted rivers for N and P host 42% of the European population. • Avoiding future coastal eutrophication requires over 47% less N and up to 77% less P exports by these polluted rivers.
4	(3) Lack of EU-wide assessment of the effects of bio-based fertilizers that are produced through recovered nutrients on reducing future nutrient emissions to the air, rivers and coastal waters	SO3: To quantify the effects of using recovered N from manure processing and recovered P from treated sewage sludge, increasing N and P use efficiencies in agriculture, and improving sewage treatments on reducing future nutrient emissions to the air, and losses to rivers and coastal waters of Europe	• We explored synergistic strategies to reduce future air and water pollution associated with nutrients in Europe. • In 2050, 30% less N is expected in the air via ReNuRe products and higher use efficiency. • Rivers are projected to receive 23-68% less nutrients relative to the future baseline. • Rivers are projected to export up to two-thirds less P relative to the future baseline. • Recovered P from treated sewage sludge can fulfill P fertilizer need in Europe in 2050.

Table S5.2 References derived from the literature review (2014-2025) for five perspectives on the implementation of bio-based fertilizers and their role in air and water pollution (reduction): (1) environment, (2) agronomy, (3) technology, (4) policy and socioeconomic, and (5) scale and integration. Some references used for several perspectives are not repeated here. For the perspective on scale and integration, assessment is done based on the scales of existing studies mainly related to environment and agronomy perspectives (see more in Section 5.5).

Perspective	Reference
Environment	1. Albert, S. & Bloem, E. Ecotoxicological methods to evaluate the toxicity of bio-based fertilizer application to agricultural soils - A review. *Sci Total Environ.* **879**, 163076 (2023). 2. Das, S. et al. Organic contaminants in bio-based fertilizer treated soil: Target and suspect screening approaches. *Chem.* **337**, 139261 (2023). 3. Dong, Y. et al. Simultaneous detection of pesticides and pharmaceuticals in three types of bio-based fertilizers by an improved QuEChERS method coupled with UHPLC-q-ToF-MS/MS. *J. Hazard. Mater.* **458**, 131992 (2023). 4. Egene, C.E. et al. Greenhouse gas emissions from a sandy loam soil amended with digestate-derived biobased fertilisers – A microcosm study. *App. Soil Eco.* **178**, 104577 (2022). 5. Estoppey, N. et al. Exposure assessment of plastics, phthalate plasticizers and their transformation products in diverse bio-based fertilizers. *Sci. Total Environ.* **918**, 170501 (2024). 6. Hu, Y. et al. Hydrochar from dairy sludge as phosphorus fertiliser affects greenhouse gas emissions and maize yield. *Acta Agr. Scan., Sec. B* **74**, 2310276 (2024). 7. Lehmann, L. & Bloem, E. Antibiotic residues in substrates and output materials from biogas plants - Implications for agriculture. *Chem.* **278**, 130425 (2021). 8. Schoumans, O.F. et al. *Development of a conceptual framework to evaluate organic fertilisers: Assessment on soil quality and agronomic, environmental and economic aspects.* Wageningen, Wageningen Environmental Research, Report 2964 (2019). 9. Sharma, B. et al. Recycling of Organic Wastes in Agriculture: An Environmental Perspective. *Int. J. Environ. Res.* **13**, 409-429 (2019). 10. Sniatala, B. et al. Macro-nutrients recovery from liquid waste as a sustainable resource for production of recovered mineral fertilizer: Uncovering alternative options to sustain global food security cost-effectively. *Sci. Total Environ.* **856**, 159283 (2023). 11. Wester-Larsen, L. et al. Potential ammonia volatilization from 39 different novel biobased fertilizers on the European market - A laboratory study using 5 European soils. *J. Environ. Manag.* **323**, 116249 (2022). 12. Wester-Larsen, L. et al. Effects of biobased fertilisers on soil physical, chemical and biological indicators – a one-year incubation study. *Soil Res.* **62**, SR23213 (2024). 13. Zilio, M. et al. Using highly stabilized digestate and digestate-derived ammonium sulphate to replace synthetic fertilizers: The effects on soil, environment, and crop production. *Sci. Total Environ.* **815**, 152919 (2022).

Perspective	Reference
Agronomy	1. Agostini, L. et al. Prediction of nitrogen mineralization from novel bio-based fertilizers using chemical extractions. *Environ. Tech. Innov.* **36**, 107781 (2024). 2. Ai, P. et al. Effect of application of different biogas fertilizer on eggplant production: Analysis of fertilizer value and risk assessment. *Environ. Tech. Innov.* **19**, 101019 (2020). 3. Brod, E. et al. Considering inorganic P binding in bio-based products improves prediction of their P fertiliser value. *Sci. Total Environ.* **836**, 155590 (2022). 4. Clagnan, E. et al. Effects of the application of microbiologically activated bio-based fertilizers derived from manures on tomato plants and their rhizospheric communities. *Sci. Rep.* **13**, 22478 (2023). 5. Hansen, V. et al. Long-term soil health effects of human urine and other bio-based fertilizers: A comprehensive field study. *Agri. Ecosys. Environ.* **398**, 110108 (2026). 6. Hu, Y. et al. Systematic Review of Dairy Processing Sludge and Secondary STRUBIAS Products Used in Agriculture. *Front. Sus. Food Sys.* **5**, 763020 (2021). 7. Huygens, D. & Saveyn, H.G.M. Agronomic efficiency of selected phosphorus fertilisers derived from secondary raw materials for European agriculture. A meta-analysis. *Agro. Sus. Dev.* **38**, 1-14 (2018). 8. Kebalo, L.F. et al. Using bio-based fertilizer derived from peri-urban wastes affects soil properties and lettuce yield and quality. *Scientia Horticultur.* **324**, 112599 (2024). 9. Kurniawati, A. et al. Opportunities and challenges of bio-based fertilizers utilization for improving soil health. *Org. Agri.* **13**, 335-350 (2023). 10. Müller, B. et al. Agronomic performance of novel, nitrogen-rich biobased fertilizers across European field trial sites. *Field Crops Res.* **316**, 109486 (2024). 11. Rashid, M.A. et al. Evaluating the performance of biobased, recovered nitrogen fertilizers in European cropping systems using modelling. *Farm. Sys.* 3, 100141 (2025). 12. Rydgård, M. et al. Regionalised modelling of recycled fertiliser P in agricultural fields: Development of the life cycle inventory model PLCI 2.0. *J. Clean. Prod.* **443**, 141088 (2024). 13. Saju, A. et al. Exploring the short-term in-field performance of Recovered Nitrogen from Manure (RENURE) materials to substitute synthetic nitrogen fertilisers. *Clean. Circ. Bioeco.* 5, 100043 (2023). 14. Shi, W. et al. Mineral fertiliser equivalent value of dairy processing sludge and derived biochar using ryegrass (Lolium perenne L.) and spring wheat (Triticumaestivum). *J. Environ. Manag.* **321**, 116012 (2022). 15. Shrivastava, V. et al. Ammonia water as a biobased fertiliser: Evaluating agronomic and environmental performance for Lactuca sativa compared to synthetic fertilisers. *Biocatal. Agri. Biotech.* **54**, 102907 (2023).

Perspective	Reference
Technology	1. Cano-Larrotta, A. et al. Optimizing the final rest period of sludge treatment wetlands: Assessment of biosolids quality. *J. Environ. Manag.* **370**, 122716 (2024). 2. Chojnacka, K. et al. Management of biological sewage sludge: Fertilizer nitrogen recovery as the solution to fertilizer crisis. *J. Environ. Manag.* **326**, 116602 (2023). 3. Fournie, T. et al. Phosphorus recovery and reuse potential from smouldered sewage sludge ash. *Waste Manag.* **137**, 241-252 (2022). 4. González, I. et al. Wastewater nutrient recovery using twin-layer microalgae technology for biofertilizer production. *IWA Wat. Sci. Tech.* **82**, 1044-1061 (2020). 5. González-García, I. et al. Two-phase nutrient recovery from livestock wastewaters combining novel membrane technologies. *Biomass Conv. Bioref.* **12**, 4563-4574 (2022). 6. Huygens, D. et al. *Technical proposals for selected new fertilising materials under the Fertilising Products Regulation (Regulation (EU) 2019/1009) – Process and quality criteria, and assessment of environmental and market impacts for precipitated phosphate salts & derivates, thermal oxidation materials & derivates and pyrolysis & gasification materials.* Luxembourg: Publications Office of the European Union, EUR 29841 EN (2019). 7. Jasiński, D. et al. The environmental sustainability of bio-based fertilisers produced via integrated nutrient recycling technologies. *Biomass Bioener.* **207**, 108705 (2026). 8. Liu, H. et al. Phosphorus recovery from municipal sludge-derived ash and hydrochar through wet-chemical technology: A review towards sustainable waste management. *Chem. Eng. J.* **417**, 129300 (2021). 9. Melo, L.C.A. et al. Biochar-based fertilizer effects on crop productivity: a meta-analysis. *Plant Soil* **472**, 45-58 (2022). 10. Sigurnjak, I. et al. Production and performance of bio-based mineral fertilizers from agricultural waste using ammonia (stripping-)scrubbing technology. *Waste Manag.* **89**, 265-274 (2019). 11. Vaneeckhaute, C. et al. Nutrient recovery from digested waste: Towards a generic roadmap for setting up an optimal treatment train. *Waste Manag.* **78**, 385-392 (2018). 12. Vaneeckhaute, C. Integrating resource recovery process and watershed modelling to facilitate decision-making regarding bio-fertilizer production and application. *npj Clean Water* **4**, 1-3 (2021). 13. Vingerhoets, R. et al. Detailed nitrogen and phosphorus flow analysis, nutrient use efficiency and circularity in the agri-food system of a livestock-intensive region. *J. Clean. Prod.* **410**, 137278 (2023). 14. Vingerhoets, R. et al. Ammonia stripping and scrubbing followed by nitrification and denitrification saves costs for manure treatment based on a calibrated model approach. *Chem. Eng. J.* **477**, 146984 (2023). 15. Vingerhoets, R. et al. Economic potential for nutrient recovery from manure in the European union. *Resour. Conserv. Recycl.* **215**, 108079 (2025). 16. Zhu, F. et al. Phosphorus recovery for circular Economy: Application potential of feasible resources and engineering processes in Europe. *Chem. Eng. J.* **454**, 140153 (2023).

Perspective	Reference
Policy and socioeconomic	1. Bonnichsen, O. et al. *Danish farmers' preferences for bio-based fertilisers - a choice experiment.* IFRO Working Paper, No. 2020/15, University of Copenhagen, Department of Food and Resource Economics (IFRO), Copenhagen (2020). 2. Egan, A. et al. What are the desired properties of recycling-derived fertilisers from an end-user perspective? *Clean. Respon. Consump.* **5**, 100057 (2022). 3. Kurniawati, A. et al. Understanding the future of bio-based fertilisers: The EU's policy and implementation. *Sustain. Chem. Clim. Act.* **3**, 100033 (2023). 4. Kvakkestad, V. et al. Circulation of nutrients through bio-based fertilizer products: perspectives from farmers, suppliers, and civil society. *Front. Sus. Food Sys.* **7**, 1-10 (2023). 5. Le Pera, A. et al. Environmental sustainability of an integrate anaerobic digestion-composting treatment of food waste: Analysis of an Italian plant in the circular bioeconomy strategy. *Waste Manag.* **139**, 341-351 (2022). 6. Lessmann, M. et al. Maximizing agricultural reuse of recycled nutrients: A spatially explicit assessment of environmental consequences and costs. *J Environ Manag.* **332**, 117378 (2023). 7. Spiller, M. et al. Beyond circularity! Integration of circularity, efficiency, and sufficiency for nutrient management in agri-food systems. *Nutr. Cycl. Agroecosyst.* **129**, 287-297 (2024). 8. Tur-Cardona, J. et al. Farmers' reasons to accept bio-based fertilizers: A choice experiment in seven different European countries. *J. Clean. Prod.* **197**, 406-416 (2018).
Scale and integration	-

Table S5.3 Summary of the positive (+) and negative (-) impacts of different bio-based fertilizers (BBFs) on emissions to the air and water systems, and other pollutants based on the literature review for Section 5.5.1. Positive impact is considered when BBFs reduce emissions to the air and losses to water systems relative to synthetic/mineral fertilizers. Negative impact represents the contribution of BBFs to air and/or water pollution with multiple pollutants. N_2O is nitrous oxide, NH_3 is ammonia, GHG is greenhouse gas, and NO_3^- is nitrate.

Pollutant	**Impact**	**Description**	**Reference**
Organic contaminants (e.g., pharmaceuticals)	No	Most organic contaminants detected in BBF-applied soil might come from non-BBFs sources	(Das et al., 2023)
Pesticides and pharmaceuticals	-	15 BBFs with various origins are found to contain at least one pesticide or pharmaceutical (often ibuprofen)	(Dong et al., 2023)
N_2O	+	Comparable or lower N_2O emissions from 18 digestate-derived BBFs relative to synthetic fertilizers	(Egene et al., 2022)
Microplastics	-	Microplastics, phthalate plasticizers and their transformation products are detected in BBFs from three types of organic wastes, and based on the detected amounts biowaste-BBFs are found not to be safe	(Estoppey et al., 2024)
N_2O	-	Not significantly different N_2O emissions from two hydrochars produced from Danish and Irish dairy sludge compared to mineral P fertilizer	(Hu et al., 2024)
Antibiotics	-	Antibiotics are detected in digestates (with frequency of 50-100%) from the anaerobic digestion of animal manure and sewage sludge that can be used as BBFs	(Lehmann & Bloem, 2021)
Microorganisms, medical drugs, other chemicals	-	Potential list of unwanted substances that organic fertilizers might contain (in this study, organic fertilizers are described as BBFs)	(Schoumans et al., 2019)
NH_3		Potential NH_3 emissions of 39 novel BBFs are assessed among each other – digestates are found with the highest accumulated NH_3 volatilization potential, and composts and struvites are with the lowest	(Wester-Larsen et al., 2022)
GHG, NH_3, NO_3^-	+	No higher emissions of NH_3 and GHGs, and no risks of NO_3^- leaching in the soil	(Zilio et al., 2022)

Table S5.4 Overview of common nutrient recovery technologies for processing of manure and sewage sludge based on the literature review for Section 5.5.3. BBF is bio-based fertilizer, N is nitrogen, LF is liquid fraction, P is phosphorus, $(NH_4)_2SO_4$ is ammonium sulfate, LCA is life cycle assessment, NH_3 is ammonia, NH_4NO_3 is ammonium nitrate, SF is solid fraction, and ReNuRe stands for REcovered Nitrogen from manURE. Plus sign (+) between technologies indicate consecutive treatments.

Technology/mechanism	Focus	Final product	Reference
Sludge treatment wetland	BBF production	Biosolids	(Cano-Larrotta et al., 2024)
Anaerobic digestion; Hydrothermal carbonization	N recovery for fertilizer production	Anaerobically digested sewage sludge; LF and hydrochar	(Chojnacka et al., 2023)
Smouldering sewage sludge with sand + acidic extraction; Co-smouldering sludge with woodchips + acidic extraction	P recovery for potential reuse	Ash	(Fournie et al., 2022)
Twin-layer microalgae	Nutrient recovery for BBF production	Solid BBF	(González et al., 2020)
Novel gas-permeable membrane + electro dialytic process	Nutrient recovery	$(NH_4)_2SO_4$ solution, struvite (slow-release P fertilizer)	(González-García et al., 2022)
Carboxylic acid technology; Microbial cultivation; Insect farming; Pyrolysis; Composting; Anaerobic digestion	LCA of valorizing fruit and vegetable wastes and residues to recover valuable components for producing novel BBFs	Various BBF blends including building blocks such as microbial biomass, compost, and biochar	(Jasiński et al., 2026)
NH_3 stripping-scrubbing	Mineral fertilizer production	NH_4NO_3; $(NH_4)_2SO_4$	(Sigurnjak et al., 2019)
NH_3 stripping-scrubbing + nitrification/denitrification	Cost minimization of manure treatment by optimizing its operational system parameters	NH_4NO_3; $(NH_4)_2SO_4$	(Vingerhoets et al., 2023a)

Table S5.4 Continued.

Technology/mechanism	Focus	Final product	Reference
Centrifuge + LF: NH_3 stripping-scrubbing; Centrifuge + LF: NH_3 stripping-scrubbing + nitrification/denitrification; Centrifuge + LF: NH_3 stripping-scrubbing + nitrification/denitrification + SF: Composting Acidification + Centrifuge + LF: Precipitation + nitrification/denitrification + SF: Composting	Economic and environmental assessment of N recovery from manure to produce ReNuRe products	Stripped LF, $(NH_4)_2SO_4$, SF; Effluent, $(NH_4)_2SO_4$, SF; Effluent, $(NH_4)_2SO_4$, dried SF; Effluent, struvite, dried SF	(Vingerhoets et al., 2025)

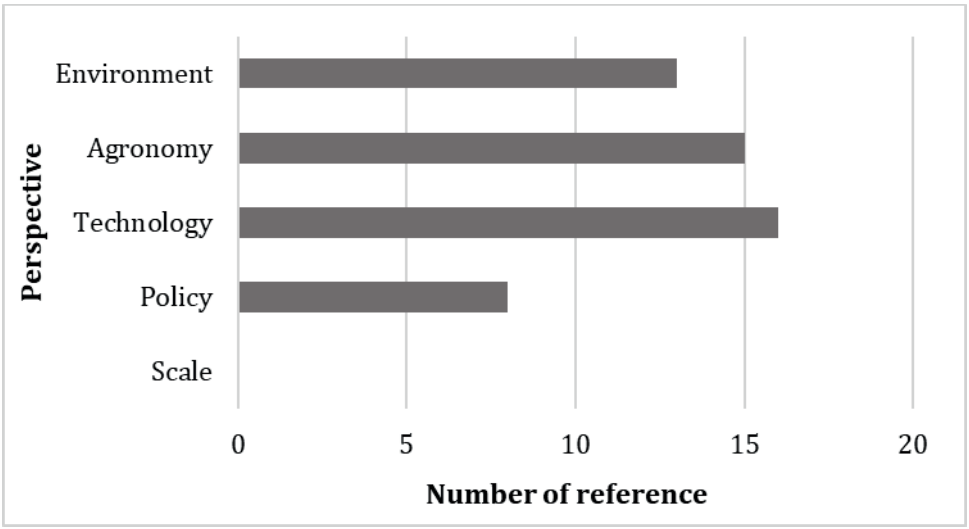


Figure S5.1 Bar chart showing reported frequency of each perspective based on the literature review (2014-2025) for Section 5.5.

Summary (English)

Intensive agriculture and urbanization are important sources of nutrient emissions to the air and surface waters (rivers and coastal waters) in Europe. Elevated levels of nitrogen (N) and phosphorus (P) emissions can cause impacts on human health and the environment. Integrated nutrient assessments of both air and surface waters are, so far, limited, especially on the basin scale for the European Union (EU) and the United Kingdom (UK). This hinders a better understanding of air and water pollution hotspots, their causes and future trends. As a result, it is not easy to identify basin-specific required reductions to avoid future pollution. Integrated models are needed to fulfill these knowledge gaps, but hardly exist for both air and surface water pollution assessments for the EU basins and the UK.

Despite the progress of the EU policies, nutrient management strategies to reduce both air and water pollution simultaneously are needed. However, their effects have not been explored for European basins and future years while considering socioeconomic developments and climate change (global change drivers). Increasing population and demand for agricultural productivity urge the renewable alternatives such as bio-based fertilizers (BBFs) to replace nonrenewable synthetic fertilizers. BBFs could potentially reduce nutrient pollution through nutrient recovery from animal manure and municipal sewage sludge. However, the environmental effects of BBFs are often analyzed at small scales and for either air or water pollution. Additionally, the combination of BBFs with other nutrient management strategies to reduce air and water pollution is poorly understood at the EU level under global change. Consequently, there is a lack of a comprehensive policy for the adoption and implementation of these BBF products in the EU and the UK.

The main objective of this PhD thesis is, therefore, to enhance our understanding of basin-specific nutrient emissions to the air and surface waters, and explore the role of BBFs and other nutrient management strategies to reduce future nutrient pollution under global change across the EU and the UK.

To this end, three research sub-objectives (SO) are achieved:

- **SO1:** To quantify the contribution of agricultural activities to air, river and coastal water pollution simultaneously with a focus on nutrients in European basins (**Chapter 2**);
- **SO2:** To assess the future river exports of N and P, and identify required reductions to avoid coastal eutrophication in Europe under global change (**Chapter 3**);
- **SO3:** To quantify the effects of using recovered N from manure processing and recovered P from treated sewage sludge, increasing N and P use efficiencies in agriculture, and improving sewage treatments on reducing future nutrient emissions to the air, and losses to rivers and coastal waters of Europe (**Chapter 4**).

This PhD thesis contains five chapters. **Chapter 1** justifies the research objectives. **Chapter 2** provides model development for the recent past (SO1). **Chapters 3** and **4** focus on model applications for future under socioeconomic developments and climate change (SO2) and under different scenarios including BBFs and other nutrient management strategies (SO3). **Chapter 5** provides the synthesis of research outcomes and their discussion, followed by a future outlook. Below, I summarize the research carried out for Chapters 2-4.

Chapter 2 (SO1) presents the newly developed MARINA-Nutrients model for Europe, which is short for Models to Assess River Inputs of pollutaNts to seAs for Nutrients. The model quantifies the annual N emissions to the air from agricultural activities, inputs of N and P to rivers, and their exports to coastal waters from point and diffuse sources. Diffuse sources include the application of synthetic fertilizers and animal manure on agricultural land, biological N_2 fixation by vegetation and atmospheric N deposition on land, leaching of organic matter and weathering of P-contained minerals. Point sources are sewage systems. The model is applied for 601 (sub)basins in 2017-2020. New modeling insights reveal that almost two-fifths of reactive N emissions to the air from agriculture are from animal housing and storage. Agriculture –mainly the application of synthetic fertilizers and manure– and sewage systems are responsible for 55% of N and 67% of P in rivers, respectively, at the EU level. Nearly a third of the European basin area is pollution hotspots (air, water, or both), producing over half of N emissions to the air and nutrient pollution in rivers. Moreover, 59% of the total population in the study area lives in those hotspots. Such hotspots are basins where total N emission to the air and nutrient inputs to rivers exceed the 80^{th} percentile (the top 20%).

Chapter 3 (SO2) provides the required reductions for future river exports of nutrients to avoid coastal eutrophication in 2050. To this end, a future baseline scenario is developed and a backcasting approach is applied. A future baseline scenario combines the storylines of the existing Shared Socio-economic Pathway 5 (fossil-fuel-based) and Representative Concentrative Pathways 8.6 (high-end climate change) and is executed by the MARINA-Nutrients model from Chapter 2. A backcasting approach establishes a target for low eutrophication using an Indicator for Coastal Eutrophication Potential (ICEP). New insights reveal that nutrient export to European seas is expected to increase by 13-28% in 2050 relative to the period of 2017-2020. Manure and fertilizers are projected to contribute to future N export by rivers by 35%, while the remainder is mainly from sewage systems (22%), atmospheric N deposition over agricultural areas (14%), and leaching of organic matter (10%) from agricultural areas. Sewage systems are projected to be responsible for 70% of future P export by rivers. Avoiding future coastal eutrophication requires over 47% less N and up to 77% less P exports individually by the top ten polluted rivers in 2050 relative to the period of 2017-2020. The basins of these polluted rivers are expected to host 42% of the population in 2050, which potentially might be affected by coastal eutrophication.

Chapter 4 (SO3) provides novel insights into the effects of BBFs on reducing future nutrient emissions to the air and surface waters across the EU and the UK in combination with other nutrient management strategies. The BBFs considered are REcovered Nitrogen from manURE (ReNuRe) and Recovered P from treated Sewage Sludge (RPSS). Six scenarios are developed for 2050 and include the use of BBFs alone and in combination with increased nutrient use efficiencies in agriculture and improved sewage treatments. These new scenarios are executed by the updated MARINA-Nutrients for Europe for the year 2050. New insights reveal that adopting ReNuRe products with increased N use efficiency is found to be a synergistic option to reduce N emissions to the air (30%), and N inputs to rivers (23%) and N export by rivers to coastal waters (20%) of Europe in 2050 compared to the future baseline scenario (taken from Chapter 3). By combining the use of RPSS, increased P use efficiency and enhanced sewage treatment, 68% less P inputs to rivers and 62% less P export by rivers to coastal waters are projected relative to the future baseline in 2050. Results show that the RPSS alone can fulfill the P fertilizer needs of the EU and the UK by 2050.

Chapter 5 provides the synthesis of the research outcomes and discusses them in terms of the strengths (S), weaknesses (W), opportunities (O) and threats (T) using the SWOT approach. Strengths include a novel integrated nutrient model for both air and surface waters, building trust approaches, and different scenario types (predictive, explorative, and normative). The main weaknesses of this research are related to the hotspot definition, a steady-state modeling approach and missing (local) sources, and missing other BBF types. Despite these weaknesses, the thesis offers opportunities for more in-depth analysis of future trends in air and water pollution. These opportunities lie in a soft-coupling design of the MARINA-Nutrients to facilitate model development, its uncalibrated approach that offers the flexibility for scenario development, and interactions between BBFs and other nutrient management strategies to support future pollution control. The thesis has threats that are related to uncertainty along the modeling chain, potential trade-offs, and policy associated barriers for implementing BBFs. Building on these reflections, the main perspectives on BBFs and their role in air and water pollution (reduction) are discussed by synthesizing the existing knowledge and the results of this thesis. These perspectives are associated with the environmental, agronomic, technological, policy and socioeconomic aspects. Additionally, effective implementation and adoption of BBFs across scales requires the integration of the aforementioned aspects. Thus, a future outlook should build on the modeling tool, scientific insights, and solutions of this thesis to further explore not only BBFs but also other strategies to reduce air and water pollution simultaneously.

To conclude, this PhD thesis shows that nutrient pollution in the EU and the UK basins is expected to increase in the future. As a result, considerable amounts of N and P emissions should be reduced. However, synergetic solutions exist for reducing nutrient emissions to both air and surface waters and avoiding future coastal eutrophication in EU seas. BBFs offer a promising strategy and their reduction potential is often undervalued. The thesis highlights

that BBFs are effective in pollution reduction when they are combined with higher nutrient use efficiencies and improved wastewater treatment. This all opens opportunities to support the EU environmental ambitions as well as the sustainable development goals for good air quality and clean water.

Samenvatting (Nederlands)

In Europa zijn intensieve landbouw en verstedelijking belangrijke bronnen van nutriëntenemissies naar de lucht en het oppervlaktewater (rivieren en kustwateren). Verhoogde emissies van stikstof (N) en fosfor (P) kunnen schadelijke effecten hebben op de menselijke gezondheid en het milieu. Geïntegreerde nutriëntenanalyses die zowel lucht als oppervlaktewater omvatten, zijn tot op heden beperkt beschikbaar, met name op stroomgebiedschaal voor de Europese Unie (EU) en het Verenigd Koninkrijk (VK). Dit belemmert een beter begrip van hotspots van lucht- en waterverontreiniging, hun oorzaken en toekomstige trends. Daardoor is het lastig om stroomgebiedsspecifieke reducties te identificeren die nodig zijn om toekomstige verontreiniging te voorkomen. Geïntegreerde modellen zijn noodzakelijk om deze kennislacunes te vullen, maar bestaan nauwelijks voor gecombineerde beoordelingen van lucht- en oppervlaktewaterverontreiniging voor EU-stroomgebieden, inclusief het VK.

Ondanks de vooruitgang van het EU-beleid zijn nutriëntenbeheer strategieën nodig die gelijktijdig lucht- en waterverontreiniging verminderen. De effecten van dergelijke strategieën zijn echter nog nauwelijks onderzocht voor Europese stroomgebieden en toekomstige perioden, waarbij rekening wordt gehouden met sociaaleconomische ontwikkelingen en klimaatverandering (drijvende krachten achter mondiale verandering). De groeiende bevolking en de toenemende vraag naar landbouwproductiviteit vergroten de noodzaak van hernieuwbare alternatieven, zoals bio-based meststoffen (BBFs), ter vervanging van niet-hernieuwbare kunstmest. BBFs kunnen potentieel bijdragen aan het verminderen van nutriëntenverontreiniging door het terugwinnen van nutriënten uit dierlijke mest en zuiveringsslib. De milieueffecten van BBFs worden echter vaak geanalyseerd op kleine schaal en doorgaans specifiek voor lucht- of waterverontreiniging. Bovendien is het gecombineerde effect van BBFs en andere nutriëntenbeheer strategieën op lucht- en waterverontreiniging onder aardsysteemveranderingen op EU-niveau onvoldoende onderzocht. Hierdoor ontbreekt een samenhangend beleid voor de adoptie en implementatie van BBF-producten in de EU en het VK.

Het hoofddoel van dit proefschrift is daarom het vergroten van inzicht in stroomgebiedsspecifieke patronen van nutriëntenemissies naar de lucht en het oppervlaktewater, en het verkennen van de rol van BBFs en andere nutriëntenbeheer strategieën bij het terugdringen van toekomstige nutriëntenverontreiniging onder mondiale veranderingen in de EU en het VK.

Daartoe worden drie subdoelen (S) gerealiseerd:

S1: Het kwantificeren van de bijdrage van landbouwactiviteiten aan lucht-, rivier- en kustwaterverontreiniging, met de nadruk op nutriënten in Europese stroomgebieden (**Hoofdstuk 2**);

S2: Het beoordelen van toekomstige riviertransporten van N en P en het identificeren van benodigde reducties om kust-eutrofiëring in Europa te voorkomen onder mondiale verandering (**Hoofdstuk 3**);

S3: Het kwantificeren van de effecten van het gebruik van teruggewonnen N uit mestverwerking en teruggewonnen P uit zuiveringsslib, het verhogen van N- en P-gebruikefficiënties in de landbouw en het verbeteren van rioolwaterzuiveringinstallaties om toekomstige nutriëntenemissies naar de lucht, de nutriëntentoevoer naar rivieren en het nutriëntentransport naar kustwateren in Europa te reduceren (**Hoofdstuk 4**).

Dit proefschrift bestaat uit vijf hoofdstukken. **Hoofdstuk 1** onderbouwt de onderzoeksdoelstellingen. **Hoofdstuk 2** beschrijft de modelontwikkeling voor het recente verleden (S1). **Hoofdstukken 3** en **4** richten zich op modeltoepassingen voor de toekomst onder respectievelijk sociaaleconomische en klimatologische ontwikkelingen (S2), en voor verschillende scenario's inclusief BBFs en andere nutriëntenbeheer strategieën (S3). **Hoofdstuk 5** bevat de synthese en discussie van de onderzoeksresultaten, gevolgd door een toekomstperspectief. Hieronder wordt het onderzoek uit Hoofdstukken 2–4 samengevat.

Hoofdstuk 2 (S1) presenteert het nieuw ontwikkelde MARINA-Nutrients-model voor Europa (Models to Assess River Inputs of pollutaNts to seAs for Nutrients). Het model kwantificeert jaarlijkse N-emissies naar de lucht vanuit landbouwactiviteiten, de toevoer van N en P naar rivieren en hun transport naar kustwateren vanuit punt- en diffuse bronnen. Diffuse bronnen omvatten het gebruik van kunstmest en dierlijke mest op landbouwgrond, biologische N_2-fixatie door vegetatie, atmosferische N-depositie op land, uitspoeling van organische stof en verwering van P-houdende mineralen. Puntbronnen bestaan uit rioolstelsels. Het model is toegepast op 601 (deel)stroomgebieden voor de periode 2017–2020. Nieuwe modelinzichten laten zien dat bijna twee vijfde van de reactieve N-emissies naar de lucht vanuit de landbouw afkomstig is van dierlijke huisvesting en opslag. Landbouw – met name het gebruik van kunstmeststoffen en dierlijke mest – en rioolstelsels zijn op EU-niveau verantwoordelijk voor 55% van de N- en 67% van de P-belasting van rivieren. Bijna een derde van het Europese stroomgebiedsoppervlak bestaat uit verontreinigingshotspots (lucht, water of beide), die samen meer dan de helft van de N-emissies naar de lucht en nutriëntenverontreiniging in rivieren veroorzaken. Bovendien woont 59% van de totale bevolking in het studiegebied in deze hotspots. Hotspots zijn gedefinieerd als stroomgebieden waar de totale N-emissie naar de lucht en nutriënteninput naar rivieren hoger is dan het 80e percentiel.

Hoofdstuk 3 (S2) beschrijft de benodigde reducties van toekomstige nutriëntentransporten via rivieren om kust-eutrofiëring in 2050 te voorkomen. Hiervoor is een toekomstig basisscenario ontwikkeld en een backcasting-benadering toegepast. Het basisscenario combineert de verhaallijnen van Shared Socio-economic Pathway 5 (fossiele brandstof-gedreven ontwikkeling) en Representative Concentration Pathway 8.6 (extreme klimaatverandering)

en is doorgerekend met het MARINA-Nutrients-model. Met behulp van de Indicator for Coastal Eutrophication Potential (ICEP) wordt een doelstelling voor lage eutrofiëring vastgesteld. De resultaten tonen aan dat de nutriëntentransport naar Europese zeeën in 2050 naar verwachting met 13–28% zal toenemen ten opzichte van 2017–2020. Dierlijke mest en kunstmest dragen naar verwachting voor 35% bij aan de toekomstige N-export via rivieren, terwijl de resterende bijdragen voornamelijk afkomstig zijn van rioolstelsels (22%), atmosferische N-depositie op landbouwgrond (14%) en uitspoeling van organische stof (10%). Rioleringssystemen zijn naar verwachting verantwoordelijk voor 70% van de toekomstige P-export. Het voorkomen van kust-eutrofiëring vereist voor de tien meest vervuilde rivieren een reductie van meer dan 47% voor N en tot 77% voor P in 2050 ten opzichte van 2017–2020. In deze stroomgebieden zal naar verwachting 42% van de bevolking wonen, die mogelijk wordt getroffen door kust-eutrofiëring.

Hoofdstuk 4 (S3) biedt nieuwe inzichten in de effecten van BBFs op het verminderen van toekomstige nutriëntenemissies naar de lucht en nutriëntentoevoer naar oppervlaktewateren in de EU en het VK, in combinatie met andere nutriëntenbeheer strategieën. De onderzochte BBFs zijn teruggewonnen stikstof uit mestverwerking (ReNuRe) en teruggewonnen fosfor uit behandeld zuiveringsslib (RPSS). Zes scenario's voor 2050 zijn ontwikkeld, waaronder het gebruik van BBFs alleen en in combinatie met verhoogde nutriëntengebruiksefficiënties in de landbouw en verbeterde rioolwaterzuivering. Deze scenario's zijn doorgerekend met het geactualiseerde MARINA-Nutrients-model voor 2050. De resultaten laten zien dat de inzet van ReNuRe -producten in combinatie met een verhoogde N-gebruiksefficiëntie een synergetische optie is, resulterend in reducties van 30% in N-emissies naar de lucht, 23% in N-toevoer naar rivieren en 20% in N-transport naar kustwateren ten opzichte van het toekomstig basisscenario. Door het combineren van RPSS, verhoogde P-gebruiksefficiëntie en verbeterde rioolwaterzuivering worden reducties van 68% in P-input naar rivieren en 62% in P-export naar kustwateren verwacht. De resultaten tonen aan dat RPSS alleen al kan voorzien in de fosfor mestbehoefte van de EU en het VK in 2050.

Hoofdstuk 5 synthetiseert de onderzoeksresultaten en behandelt deze aan de hand van een SWOT-analyse (Strengths, Weaknesses, Opportunities, Threats). De sterke punten omvatten een nieuw geïntegreerd nutriëntenmodel voor lucht en oppervlaktewater, vertrouwensopbouwende benaderingen en het gebruik van verschillende typen scenario's (predictief, exploratief en normatief). De zwakke punten zijn onder meer de hotspotdefinitie, de stationaire modelbenadering, ontbrekende (lokale) bronnen en het ontbreken van andere BBF-typen. Kansen zijn er voor diepgaandere analyses van toekomstige trends in lucht- en waterverontreiniging, onder andere door de flexibele soft-coupling-opzet van het MARINA-Nutrients-model en de interacties tussen BBFs en andere nutriëntenbeheer strategieën. Bedreigingen hebben betrekking op onzekerheden in de modelleringsketen, mogelijke afwegingen en beleidsmatige barrières voor de implementatie van BBFs. Op basis hiervan worden perspectieven op BBFs

en hun rol in het verminderen van lucht- en waterverontreiniging besproken vanuit milieu, agronomische, technologische, beleidsmatige en sociaaleconomische invalshoeken.

Concluderend laat dit proefschrift zien dat nutriëntenverontreiniging in stroomgebieden van de EU en het VK naar verwachting in de toekomst zal toenemen, waardoor aanzienlijke reducties van N- en P-emissies noodzakelijk zijn. Tegelijkertijd bestaan er synergetische oplossingen om nutriëntenemissies naar zowel de lucht als het oppervlaktewater te verminderen en toekomstige kust-eutrofiëring te voorkomen. BBFs vormen een veelbelovende strategie waarvan het reductiepotentieel vaak wordt onderschat. Het proefschrift benadrukt dat BBFs vooral effectief zijn wanneer zij worden gecombineerd met hogere nutriëntengebruiksefficiënties en verbeterde rioolwaterzuivering. Dit biedt belangrijke kansen om de milieudoelstellingen van de EU en de duurzame ontwikkelingsdoelen voor schone lucht en schoon water te ondersteunen.

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"The greatest danger to our future is apathy."

Dr Jane Goodall, in a public address, "Reasons for Hope" at Universiti Sains Malaysia, Penang, November 2019

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Authorship statement

PhD candidate's name: Aslıhan Ural-Janssen
Promotors: Maryna Strokal and Erik Meers
Title of PhD thesis: Integrated modeling of nutrient emissions to air and water: Synergetic solutions for the European future
Date of public defence: 1 June 2026

Chapter 1 Introduction. I wrote the first draft and revised three times based on feedback from my promotors.

Chapter 2 Hotspots of nutrient losses to air and water: an integrated modeling approach for European river basins (published). I designed the outline of the paper, collected data for the period of 2017-2020, and processed it for 601 European basins under supervision. I soft-linked the existing models to develop the new MARINA-Nutrients model for Europe, and validated it under supervision. I wrote the draft of the manuscript, made the figures supporting the story, and revised the manuscript several times in response to comments from my supervisors and co-authors. I submitted the paper, and handled the revision phases.

Chapter 3 Large reductions in nutrient losses needed to avoid future coastal eutrophication across Europe (published). I proposed the initiation and design to my supervisors, and we discussed the content together. I used the existing pathways to develop a future baseline scenario, collected and processed data for 2050. I ran the MARINA-Nutrients model for Europe with the future baseline scenario and used a backcasting approach based on an environmental target. I wrote the draft of the manuscript, made the figures supporting the story, and revised the manuscript several times in response to comments from my supervisors. I submitted the paper and led the revision phase.

Chapter 4 Mitigating nutrient losses in Europe: Synergistic solutions for air and water pollution by 2050 (published). The initiation and design, as well as data collection, were largely led by me with support from my supervisors and co-authors. I developed six alternative future scenarios, ran the MARINA-Nutrients model for Europe, formulated the aim, and drafted the manuscript with supporting figures. I used ChatGPT Free to help speed up coding in R for data visualization. I revised the manuscript several times in response to comments from my supervisors and co-authors. I submitted the paper and led the revision phase.

Chapter 5 Synthesis. I wrote the first draft and revised three times based on feedback from my promotors.

Date: 12/01/2026
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About the author



Aslıhan Ural-Janssen was born in Istanbul. Her interest in environmental protection led her to pursue a BSc degree in Environmental Engineering at Marmara University in 2009. Then, she received a master's degree in Environmental Biotechnology in Istanbul Technical University with a thesis about antibiotic removal in municipal wastewater treatment systems. Soon in 2012, she received a scholarship from the Ministry of National Education in Türkiye to do a MSc abroad. She applied to Wageningen University & Research (WUR) to pursue her second MSc in Aquatic Ecology and Water Quality Management. She did a master thesis at the Aquatic Ecology and Water Quality Management and Environmental Systems Analysis groups where she focused on modeling nitrogen delivery to rivers and coastal waters of the EU and management consequences. After that, she moved to Ankara and worked at the Ministry of Agriculture and Forestry, General Directorate of Water Management for five years on harmonization (e.g., the Nitrates Directive) and research projects (e.g., identification of nitrate vulnerable zones and sensitive areas in river basins). Besides, she improved her communication and presentation skills by giving lots of presentations to local authorities. Motivated by a growing interest in teaching, she decided to pursue a PhD. Since 2020, she has been a PhD candidate at WUR, Earth Systems and Global Change group, as part of the *Marie Skłodowska-Curie Innovative Training Network FertiCycle (2020-2023),* and at Ghent University (UGent), Lab for Bioresource Recovery, within the *Horizon Europe NutriBudget Project (2023-2024).* Her doctoral research is about developing and applying the integrated MARINA-Nutrients model for Europe to quantify nutrient emissions to air and water with a focus on synergetic solutions in the future. As part of her training activities within *FertiCycle*, she spent research periods at the University of Copenhagen (three months), UGent (one year) and Nutrient Management Institute (two weeks). In these visits, she learned about Life Cycle Assessment and expanded her knowledge in integrated modeling of bio-based fertilizers. During her PhD, she helped teaching, supervising students, and actively involved in workshops for the *Inter-Sectoral Impact Model Intercomparison Project (ISIMIP) Water Quality* sector. She also organized an event at UGent where water quality modelers from WUR met the RE-SOURCE Lab team to facilitate knowledge exchange and interaction between researchers and PhD candidates. She currently works as a researcher at WUR, Earth Systems and Global Change group (2 days per week), and spends the rest of her time with her 11-month-old daughter. Her commitment to contributing to an optimistic future with clean air and water resources for the next generations will continue to guide her after completing her PhD.

List of publications

Ural-Janssen, A., Kroeze, C., Lesschen, J. P., Meers, E., van Puijenbroek, P. J. T. M., & Strokal, M. (2023). Hotspots of nutrient losses to air and water: an integrated modeling approach for European river basins. *Frontiers of Agricultural Science and Engineering*, *10*(4), 579–592. https://doi.org/10.15302/J-FASE-2023526. **(This thesis, Chapter 2)**

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Conference proceedings

Ural-Janssen, A., Kroeze, C., and Strokal, M. (2023). Reducing future coastal eutrophication under global change in Europe, EGU General Assembly 2023, Vienna, Austria, 23–28 Apr 2023, EGU23-6212, https://doi.org/10.5194/egusphere-egu23-6212, 2023.

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Ural-Janssen, A., Meers, E., Ros, G. H., Strokal V., & Strokal, M. (2025). Bio-based strategies in agriculture: perspectives for environmental pollution reductions, RAMIRAN 2025, Wageningen, Netherlands, 15-17 Oct 2025, Book of Abstracts.

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- Life Cycle Assessment within biological production systems, University of Copenhagen (2022)
- Waste processing and up-cycling concepts, University of Ghent (2021)
- Nutrient cycling and environmental and agronomic impacts - Sustainability of upcycled bio based fertiliser production and use, University of Copenhagen, Bangor University (2021 - 2022)
- Implementation of waste and nutrient management in governmental and EU policies on bio-based fertilisers, Wageningen University & Research (2023)
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- *Future nutrient pollution and coastal eutrophication in Europe under global change*. RAMIRAN Conference, 12-14 September 2023, Cambridge, UK
- *Bio-based strategies in agriculture: perspectives for environmental pollution reductions*. RAMIRAN Conference, 15-17 October 2025, Wageningen, Netherlands

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