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RESEARCH ARTICLE

HOTSPOTS OF NUTRIENT LOSSES TO AIR AND WATER: AN INTEGRATED MODELING APPROACH FOR EUROPEAN RIVER BASINS

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KEYWORDS

agriculture, air-water modeling, European rivers, nutrient pollution, sewage systems, source attribution

HIGHLIGHTS

- A new MARINA-Nutrients model was developed to assess air and water pollution in Europe.
- Agriculture is responsible for 55% of N and sewage for 67% of P in rivers.
- Almost two-fifths of reactive N emissions to air are from animal housing and storage.
- Nearly a third of the basin area produces over half of N emissions to air and nutrients in rivers.
- Over 25% of river export of N ends up in the Atlantic Ocean and P in the Mediterranean Sea.

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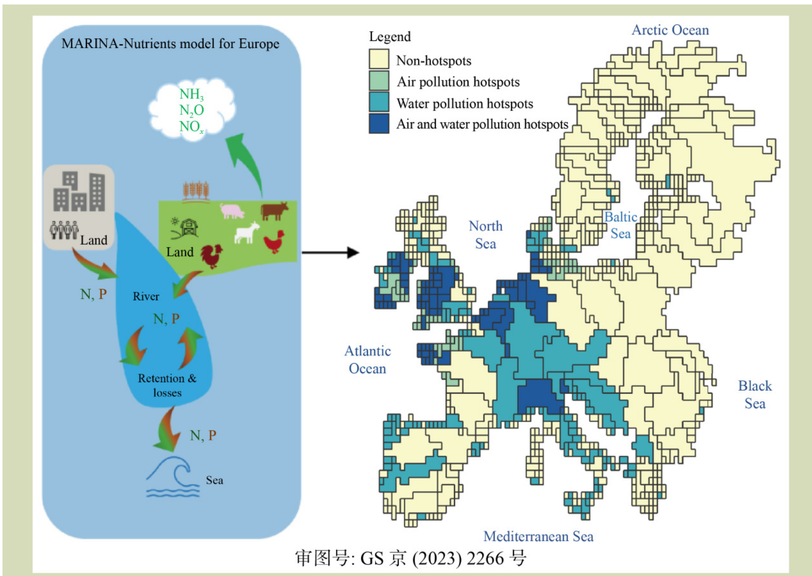
GRAPHICAL ABSTRACT



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·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·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.

1 INTRODUCTION

Intensive agriculture and high population density are often the main causes of air and water pollution with nutrients in Europe[1,2]. 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[3]. Leip et al.[4] 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[5]. Losses of N and P to the environment have substantially affected air[6–8] and water quality[9–11]. Giannakis et al.[6] 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[11,12].

Several studies have quantified N losses to air, often at the administrative scales[4,13,14]. Other studies focused on N and P losses to rivers and coastal waters, often at the grid and/or basin scales[12,15,16]. 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[17]) whereas water policies often cover basin scale (e.g., River Basin Management Plans required by the Directive 2000/60/EC[18]). 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 S1). In contrast, modeling studies generally focus either on a particular pollution source or a receiving body (e.g., surface or ground waters) (Table S1). The MARINA model (Model to Assess River Inputs of pollutaNts to seAs) is an example of this[19]. Several versions of MARINA exist. Some focus on river exports of nutrients[20–22] and some on other pollutants[23,24]. 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[19,25]. 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). However, such a model does not consider N emissions to air from agricultural activities in Europe (Table S2). 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 (Tables S1 and S2)[26]. 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)[16]. 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)[19,25]. 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)[26,27]. A detailed assessment of N losses to air, N and P losses to water from agricultural production at a basin scale can help to 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 MATERIALS AND METHODS

2.1 MARINA-Nutrients model for Europe

A new version of the MARINA-Nutrients model for European basins (Supplementary Text 1) was developed from the existing MARINA approaches[19,25] 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 (Fig. 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

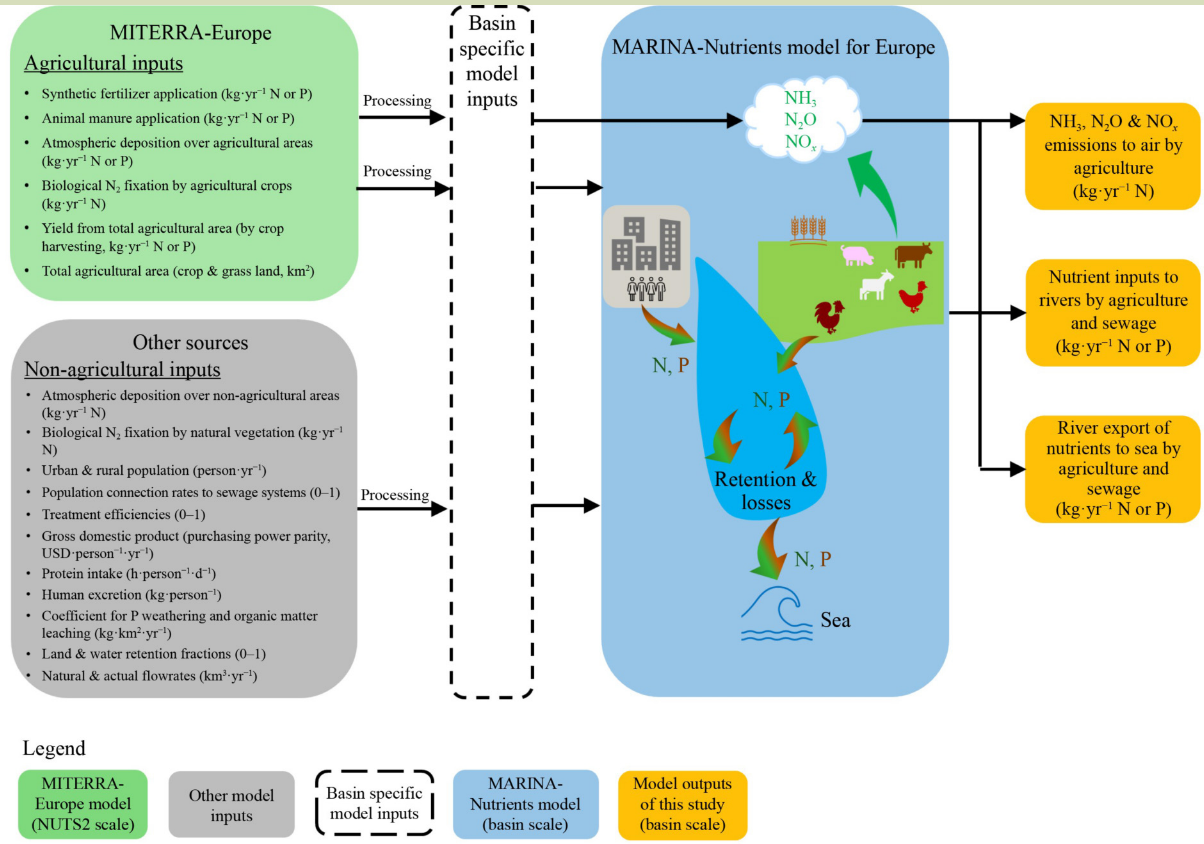


Fig. 1 Framework of the new MARINA-Nutrients model for Europe. NUTS2: Nomenclature of Territorial Units for Statistics level 2[28]. N emissions to air are ammonia, nitrous oxide and nitrogen oxides (NO_x).

sub-basin (only for the Danube River) scale (study area description in Supplementary Text 2 and Fig. S1). 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)[13,26]. 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 data sets (e.g., FAOSTAT)[29].

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 1 for details). More details on the input data and model calculations are provided in Tables S3–S5 and Sections 2.1.1–2.1.4.

2.1.1 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 (Fig. 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 (Fig. 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[14,26]. N inputs to non-agricultural land include atmospheric 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 (Fig. 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 S3). N inputs to non-agricultural land were derived from the IMAGE model (i.e., atmospheric deposition and biological N_2 fixation) (Table S5)[30].

2.1.2 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.[19] and Li et al.[23]. Nutrient inputs to rivers from agricultural and non-agricultural areas were quantified following Strokal et al.[19] and uncalibrated modeling approach of Li et al.[23].

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[19,23]:

$$RSdif_{F.y.j} = WSdif_{E.y.j} \times G_{F.j} \times FE_{ws.F.j} \tag{1}$$

$$RSpnt_{F.y.j} = RSpnt_{E.y.j} \times FEpnt_{F.y} \tag{2}$$

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 ($kg \cdot yr^{-1}$), $WSdif_{E.y.j}$ is the total input of nutrient element E to agricultural or non-agricultural land in basin j from source y ($kg \cdot yr^{-1}$), $G_{F.j}$ 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 ($kg \cdot yr^{-1}$), $RSpnt_{E.y.j}$ is the input of nutrient element E from point source y to rivers in basin j ($kg \cdot yr^{-1}$), and $FEpnt_{F.y}$ is the fraction of nutrient form F entering rivers from point source y in basin j (0–1) (Tables S3–S5 provide more details).

2.1.3 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 S6)[29]. We took the emissions from MITERRA-Europe in NUTS2 scale and aggregated them to the basin scale (Table S5). 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) (Tables S3–S5 provide more detail).

2.1.4 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[19]:

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

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 Eqs. (74–89) in Table S3 based on the data derived from HydroLAKES[31] (Tables S3–S5 provide more details).

2.1.5 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.[32] and Li et al.[23]. 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 (Fig. 2). Water pollution hotspots represent the basins in Group V based on only TDN, only TDP or both TDN and TDP inputs to rivers (Fig. 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 (Fig. 2). The ranges of the groups are described in Section 3.

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 (Fig. 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[33]. 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[16]. We also used the data from literature on concentrations (mg·L^{-1})[34] and loads (kg·yr^{-1})[35–40] 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 and 2016. All observed values for 58 rivers were normalized to the same unit (kg·yr^{-1}) for model validation. Details of the data are given in Table S7 and the validation results are indicated in Section 3.

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.

3.1 Nutrients on land from agriculture

Under the current practice, the new MARINA-Nutrients model estimated the total inputs of 34.4 Tg·yr^{-1} N and 1.8 Tg·yr^{-1} P to land in the EU-28. Agriculture was responsible for 84% of N and 96% of P on land (Table S8). 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·km^{-2}·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·km^{-2}·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.

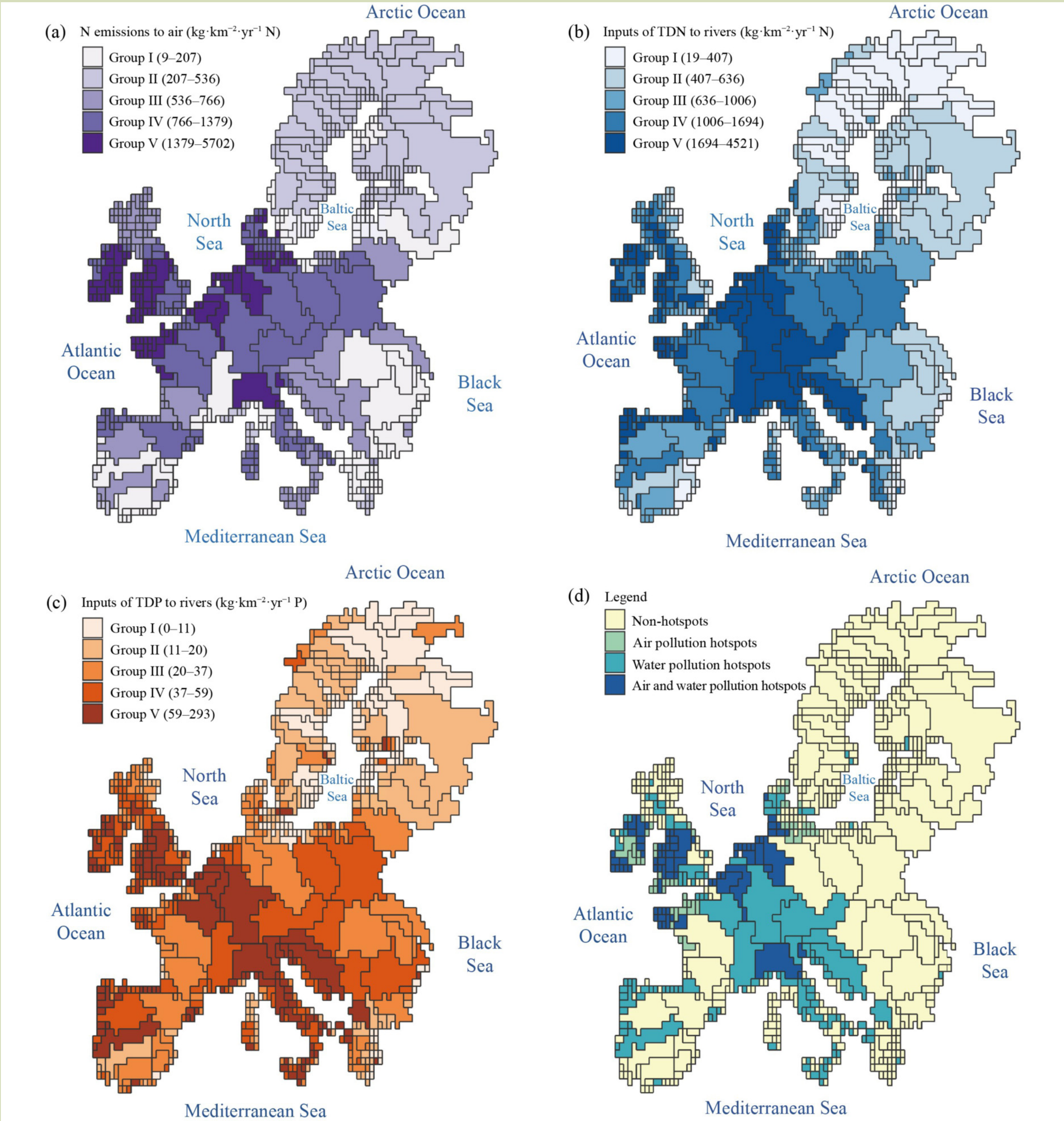


Fig. 2 Nutrient losses to air and waters by the European basins (审图号: GS 京 (2023) 2266 号). (a) Reactive N emissions to air by basin (kg·km^{-2}·yr^{-1} N). (b) Inputs of total dissolved N (TDN) to rivers by basin (kg·km^{-2}·yr^{-1} N). (c) Inputs of total dissolved P (TDP) to rivers by basin (kg·km^{-2}·yr^{-1} P). Groups I–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).

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 and 2020 (Fig. 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 and 4521 kg·km^{-2}·yr^{-1} N and 0–293 kg·km^{-2}·yr^{-1} P, respectively (Fig. 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 (Fig. 4). From agriculture, synthetic fertilizer application was the largest source of the

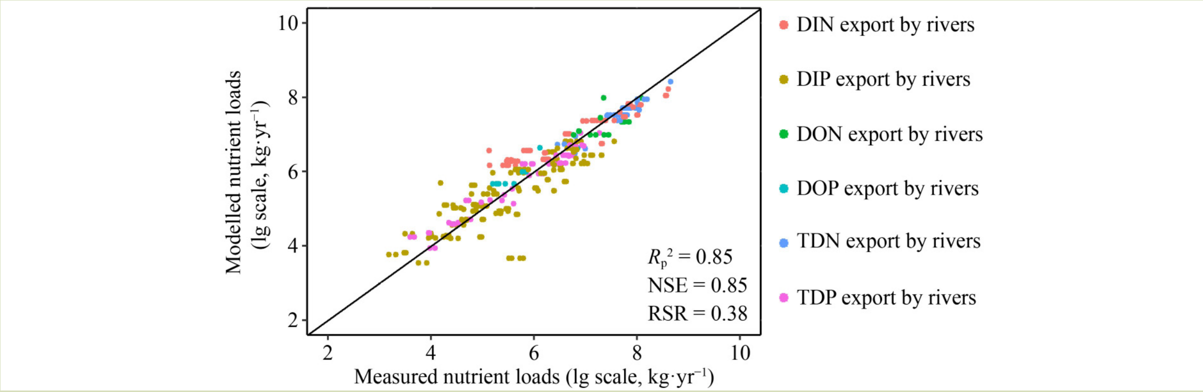


Fig. 3 Measured versus modeled river exports of dissolved inorganic N (DIN), dissolved organic N (DON), total dissolved N (TDN), dissolved inorganic P (DIP), dissolved organic P (DOP) and total dissolved P (TDP) to the European seas (lg scale; kg·yr^{-1}). Each dot represents an individual river mouth for 58 rivers. Measured loads indicate all the observed data (average per year) at or close to the river mouths in various years in 1990–2017 (Table S7 for details). Modeled nutrient loads are annual river exports of nutrients at the river mouths in 2017–2020.

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 (Fig. 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 (Fig. 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 (Fig. 4).

3.3 Nitrogen emissions to air from agriculture

Reactive N emissions to air were 4.0 Tg·yr^{-1} N between 2017 and 2020 including the emissions from agricultural soil (2.5 Tg·yr^{-1} N) and animal housing and storage systems (1.5 Tg·yr^{-1} N) (Fig. 2). Animal housing and storage systems were an important contributor by almost two-fifths (38%) of

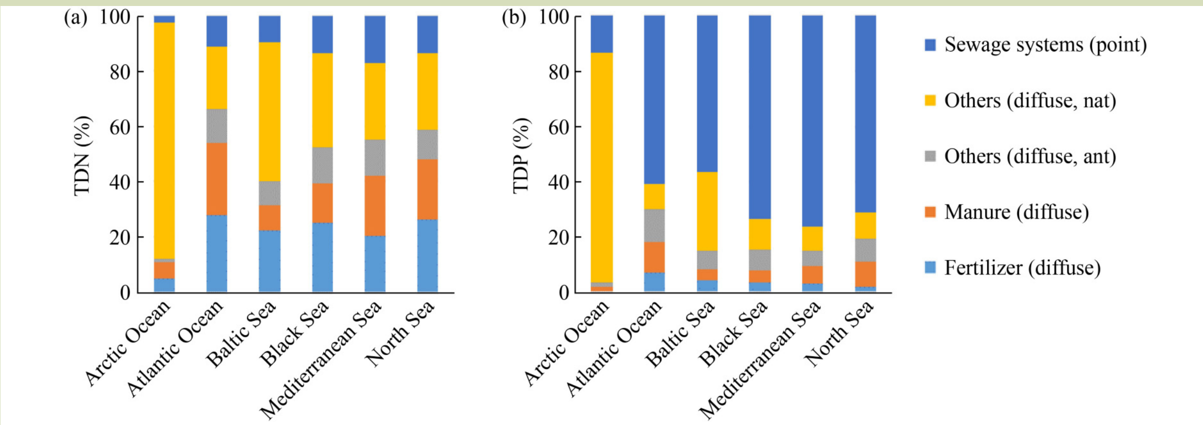


Fig. 4 (a) Relative contribution of sources to inputs of total dissolved N (TDN) to rivers per discharge sea (%). (b) Relative contribution of sources to inputs of total dissolved P (TDP) to rivers per discharge sea (%). In legend, fertilizer means synthetic fertilizers applied; manure means that applied 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.

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 and 5702 $kg \cdot km^{-2} \cdot yr^{-1}$ N (Fig. 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 S9). N_2O and NO_x emissions contributed to 12% and 4% of the reactive N emissions, respectively.

3.4 Air and water pollution hotspots

Air and river pollution was concentrated in the basins of western, central and southern Europe (Fig. 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 (Fig. 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 (Fig. 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 (Fig. 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 and 2020 (Fig. 2). In Europe, 7% of the total basin area having 18% of the population was polluted by nutrient losses to air and water simultaneously (Fig. 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 S10, Fig. 5 and Fig. 6). For river exports of TDN and TDP, five groups have ranges changing between 12 and 2746 $kg \cdot km^{-2} \cdot yr^{-1}$ N and 0–150 $kg \cdot km^{-2} \cdot yr^{-1}$ P, respectively (Fig. 5 and Fig. 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 (Fig. 5 and Fig. 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 (Fig. 5 and Fig. 6).

Source contribution to the river exports of nutrients varied to a large extent among the discharge seas (Fig. 5 and Fig. 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) (Fig. 5). Sewage systems accounted for two-thirds of the TDP export and were the major source of the DIP export (Fig. 6). Except for the Arctic Ocean, organic matter leaching over non-agricultural areas was the main source of the DOP export by rivers (Fig. 6).

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 to 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[41]. 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[42]. 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

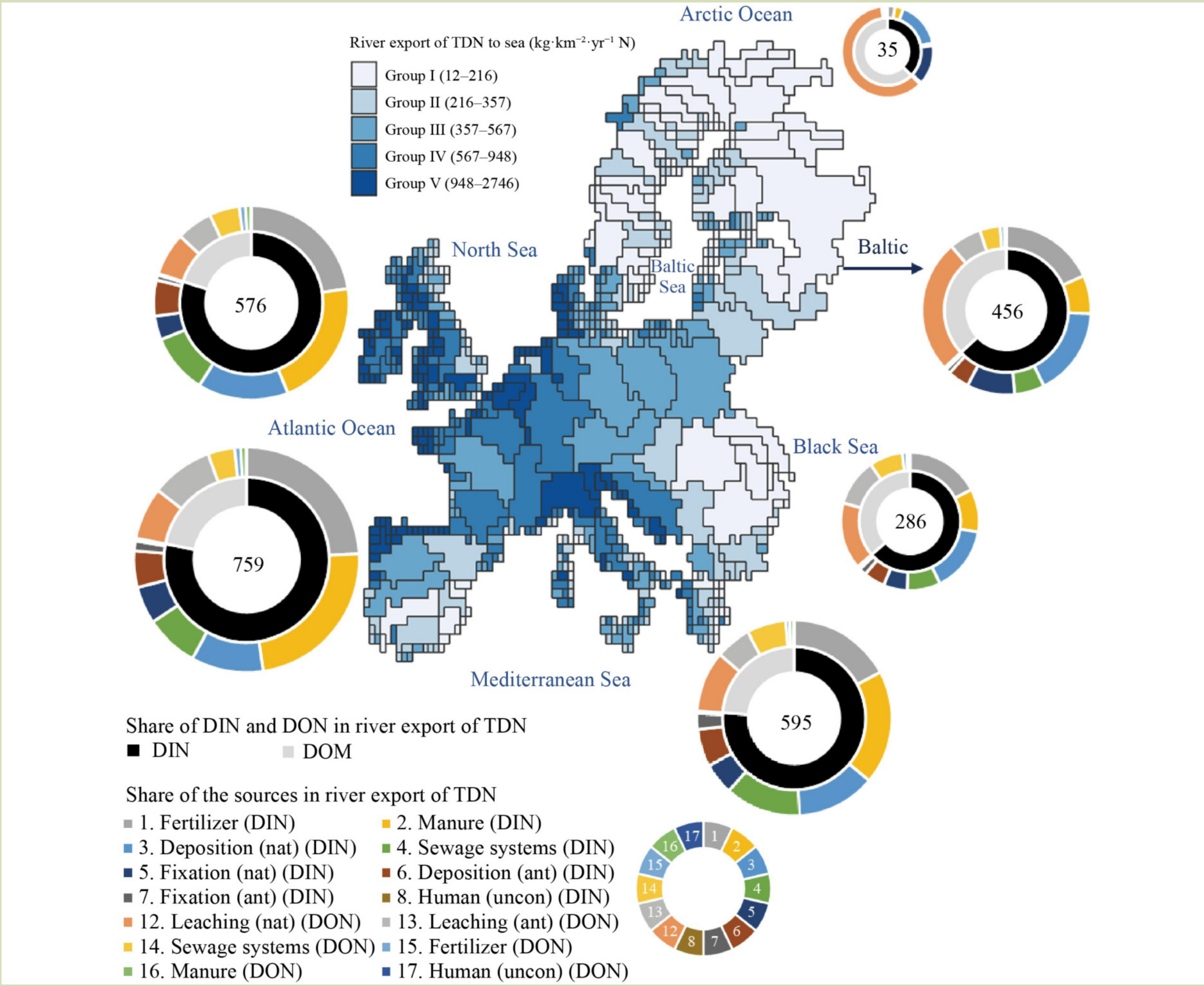


Fig. 5 River export of total dissolved N (TDN) per basin (kg·km^{-2}·yr^{-1} N) (审图号: GS 京 (2023) 2266 号). Groups I to V were defined based on the pollution levels (20% of basins for each group) for TDN export by basins. Doughnut pie charts indicate the river exports of dissolved inorganic N (DIN) and 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.

climate changes (footnotes of Table S3).

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)[43] as well as surface waters[44,45] 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 data sets 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.

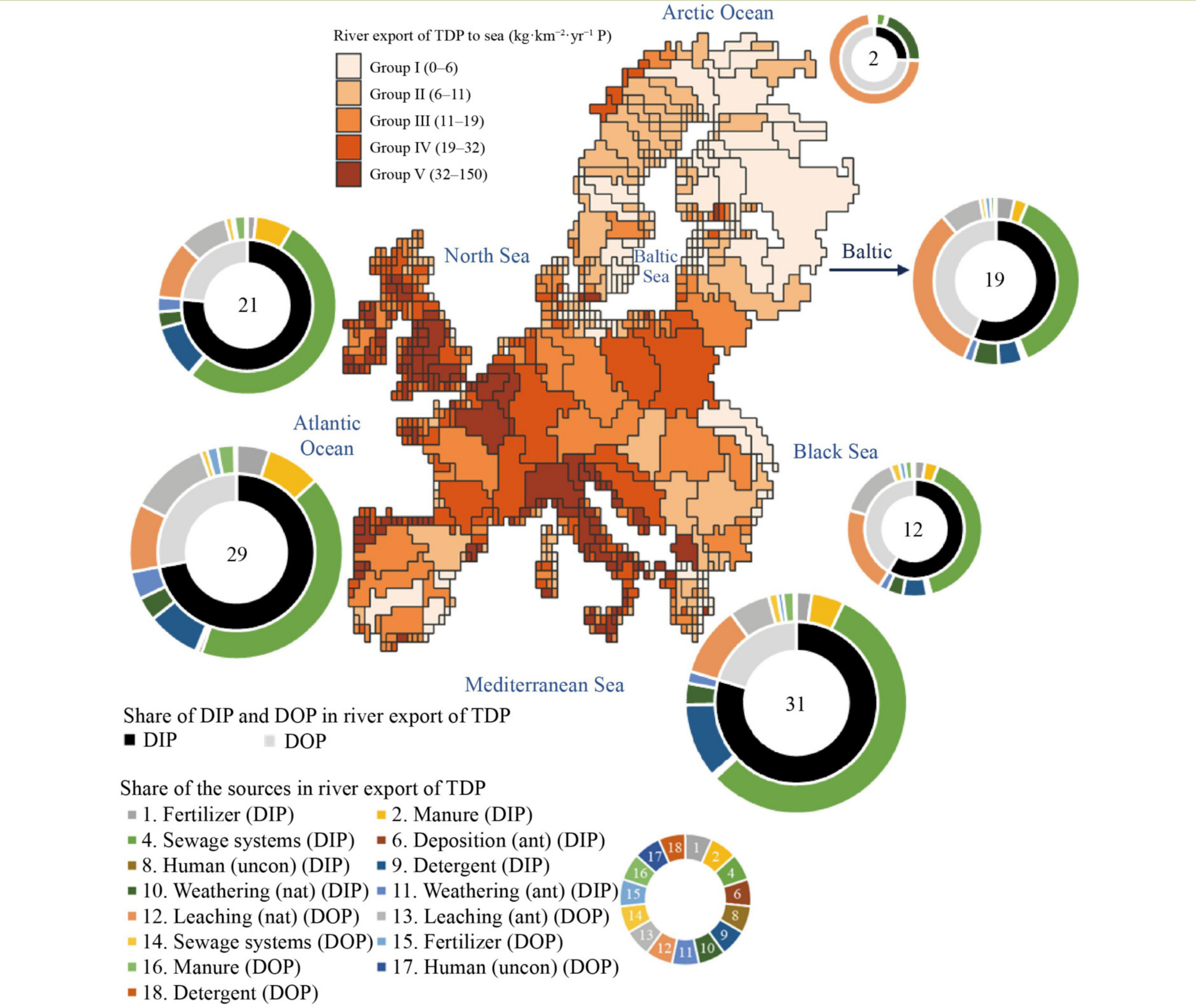


Fig. 6 River export of total dissolved P (TDP) per basin (kg·km^{-2}·yr^{-1} P) (审图号: GS 京 (2023) 2266 号). Groups I to V were defined based on the pollution levels (20% of basins for each group) for river export of TDP by basins. Doughnut pie charts indicate the dissolved inorganic P (DIP) and dissolved 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 Fig. 5 for details of abbreviations.

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[23,32]. 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.

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; $-\infty$ to 1), and the ratio of root mean square error (RMSE) and standard deviation of observations (RSR; 0 to $+\infty$) according to Moriasi et al.[46]. R^2 indicates the degree of collinearity between modeled and observed data[47]. NSE shows how well the modeled and observed data fits the 1:1 line[47]. 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[47]. Therefore, Moriasi et al.[46] 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[46,47] (Fig. 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[19]. 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.[48] for 2010 (Table S11). 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[23,48].

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[13,49]. 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 S9)[13,50,51]. Our modeled NH_3 (3401 Gg·yr^{-1} N) is higher than the other estimates (2848–3066 Gg·yr^{-1} N), N_2O (374 Gg·yr^{-1} N) is slightly lower (379–511 Gg·yr^{-1} N) and NO_x (155 Gg·yr^{-1} N) is within the range of other studies (77–219 Gg·yr^{-1} N) (Table S9). 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 S12)[15,16]. However, the modeled TDP is lower than other modeling studies (Table S13)[15,16]. This could be due to different scope of some other studies (footnotes in Table S13). TDN and TDP were compared with TN and TP in some cases (footnotes in Tables S12 and S13). This may also explain the differences between our results and other studies[12,42,52].

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.

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 and 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.

Supplementary materials and data availability

The online version of this article at https://doi.org/10.15302/J-FASE-2023526 contains supplementary materials (Texts 1–2; Fig. S1; Tables S1–S13). In addition, the main model results supporting Figs. 2–6 generated in this study have been deposited in the DANS Easy Repository under the Digital Object Identifier: 10.17026/dans-zg6-7wz4.

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Compliance with ethics guidelines

Aslıhan Ural-Janssen, Carolien Kroeze, Jan Peter Lesschen, Erik Meers, Peter J.T.M. van Puijenbroek, and Maryna Strokal declare that they have no conflicts of interest or financial conflicts to disclose. This article does not contain any studies with human or animal subjects performed by any of the authors.

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