Global anthropogenic and natural nutrient fluxes: from local to planetary assessments

Anna Malagò and Fayçal Bouraoui
European Commission, Joint Research Centre (JRC), Ispra, Italy
E-mail: anna.malago@ec.europa.eu

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Abstract
Nutrients are recognized as one of the nine planetary boundaries, which could increase risks of unacceptable global environmental changes. In this study we developed a recent and complete high-resolution nutrient flows compilation that can be used for assessing global nutrient annual fluxes from land to sea. It consists of annual nitrogen (N) and phosphorus (P) fluxes with spatial resolution of five arc-minutes (about 10 × 10 km at the equator) centered on year 2005, assessing potential nutrient delivery to rivers, lakes and oceans. The dataset includes: nutrient inputs in agricultural areas (mineral and organic fertilization, nitrogen fixation), crop/fodder/grass harvest, nutrient inputs by domestic and industrial activities (i.e. wastewater treatment plants, industries, and phosphorus from detergents), nutrients from built-areas, nitrogen atmospheric deposition, N and P transported via erosion, and phosphorus release by weathering. The dataset was compared with other studies, was analyzed at different spatial scales showing the main environmental hotspots, and finally a qualitative uncertainty analysis was performed. The results showed that nitrogen surplus was the largest contributor to the potential losses on all continents, while for phosphorus the major contributors included the surplus, erosion and inputs from human wastewater. Hotspots were identified mainly in China and India. Rates exceeding 100 kg ha⁻¹ of N were observed locally in Europe, Egypt and North America coinciding with intensive agriculture practices. We also showed that N and P transported via erosion, domestic and industrial nutrient emissions, as well as manure resulted in the most uncertain fluxes.

1. Introduction
Both nitrogen (N) and phosphorus (P) are essential, life-supporting elements, but their biogeochemical cycles have been greatly perturbed by human activities (Fowler et al 2013). Global N and P cycles were recognized as critical Earth-system processes and were identified as one of the nine ‘planetary boundaries’ (Rockström et al 2009), which could increase risks of unacceptable global environmental change. Many research groups have investigated N and P sources and transport from global terrestrial ecosystems to coastal marine ecosystems developing spatially resolved explicit soil budgets for N and P. For instance, Harrison et al (2005) provided P sources at spatial resolution of 0.5° (about 50 × 50 km at the equator) for year 1995 and Bouwman et al (2013) developed spatially explicit N and P inputs for agricultural and natural areas with a resolution of 0.5 by 0.5° considering the historical period of years 1900, 1950 and 2000. More recently Bouwman et al (2017) updated the input dataset creating long time series from 1970 to 2010, while Beusen et al (2016) analyzed global long-term changes in the delivery and retention of N and P from land to sea based on a grid model approach at 0.5° resolution. However to our knowledge, the highest resolution global N and P dataset was that of Liu et al (2010) (about 10 × 10 km at the equator) that considers also the distribution and management of 20 crops for the year 2000. Concerning non-agricultural sources, Van Drecht et al (2003) provided N and P inputs from wastewater treatment plants and industries (point sources) at a 0.5° resolution using data of population and connection rates at country level for year 2000, and more recently Van Puijenbroek et al (2019)
estimated global N and P emissions from sewage for the period 1970–2050. These studies are generally in good agreement when comparing total estimates, but there is high uncertainty on the spatial allocation of each component. To the best of our knowledge few studies have reported a detailed analysis of the uncertainty of nutrient fluxes. For instance Kros et al. (2012) analyzed the uncertainties in model inputs and their propagation to model outputs, De Vries et al. (2003) performed an uncertainty analysis using a Monte Carlo technique of all major nitrogen fluxes in the Netherlands, Van Drechtl et al. (2005) showed that the most uncertain aspects of the nutrient balance are related to biological N fixation and atmospheric N deposition, while other researchers have focused mainly on uncertainties in nutrient gaseous emissions at different spatial scales (e.g. Del Grosso et al. 2010, Nol et al. 2010).

Another limitation of many previous studies is that they focus only on cropland, thus excluding large portions of grassland and natural areas of the world.

In this study we aim at overcoming these issues, creating a harmonized dataset of N and P fluxes able to drive models for assessing riverine export of nutrients at different spatial scales. In particular, we developed and assessed a global spatially explicit five arc-minute resolution dataset of N and P fluxes, from anthropogenic and natural sources, based on the most recent available global spatial data centered on year 2005. The dataset provides a current understanding of the global nutrient fluxes identifying major gaps where priorities for control measures and future research should focus. Specifically, we presented and analyzed the potential N and P flux delivery to oceans and their components, we analyzed the fluxes at different spatial scales, and finally we provide a qualitative estimation of their uncertainty.

2. Material and method

2.1. The grid cell global nutrients delivered to rivers, lakes and oceans

Nitrogen (N) and phosphorus (P) are usually not transported conservatively through the environment but are subject to a variety of mostly biologically mediated losses and transformations in both terrestrial and aquatic settings (Ator et al. 2011). In this study we estimated total N and P fluxes potentially delivered to aquatic ecosystems as follows:

\[
N_{\text{pot}} = N_{\text{surplus CROP, FODG, GRAS}} + N_{\text{human}} + N_{\text{ero}} + N_{\text{dep,nat}} + N_{\text{dep,other}}
\]

\[
P_{\text{pot}} = P_{\text{surplus CROP, FODG, GRAS}} + P_{\text{surplus GRAS}} + P_{\text{human}} + P_{\text{ero}} + P_{\text{prel}} + P_{\text{man,nat}}
\]

where \(N_{\text{pot}}\) and \(P_{\text{pot}}\) are the N and P potential total fluxes (tons); \(N/P_{\text{surplus CROP, FODG, GRAS}}\) are the surplus on croplands, fodder and grassland, respectively, calculated as the positive budget between \(N/P_{\text{in}}\) (net mineral fertilizers and manure; nitrogen fixation and nitrogen atmospheric deposition) and \(N/P_{\text{out}}\) (harvested N and P content); \(N_{\text{human}}\) and \(P_{\text{human}}\) are the contribution of N and P from wastewater treatment plants, industries, scattered dwellings and detergents for P; \(N_{\text{ero}}\) and \(P_{\text{ero}}\) are the nutrient losses transported with erosion; \(P_{\text{prel}}\) is the phosphorus P release from weathering; \(N_{\text{dep,nat}}\) and \(N_{\text{dep,other}}\) are the nutrient inputs on natural areas and other areas respectively; \(P_{\text{man,nat}}\) is the phosphorus inputs on natural areas. These fluxes are described in detail in the supplementary information S1 (appendix A) (available online at stacks.iop.org/ERL/16/054074/mmedia). The fluxes are defined on a global grid with a spatial resolution of five arc-minutes, covering a total area of \(130 \times 10^6 \text{ km}^2\) resulting in \(2 \times 10^6\) pixels. For each grid cell we defined ten classes of landcover and within the cropland class, CROP, we distinguish 42 crops with four management systems (irrigated, high–input/commercial rainfed, low-input rainfed and subsistence) provided by the SPAM model (You et al. 2014). It is important to highlight that the dataset is centered on year 2005 that means that we collected all the data with reference to this year when it is possible (e.g. landuse, population, national statistics etc), but we extended to more recent years when not available.

The fluxes are calculated in tons applying a downscaling approach from national to grid-cell level and using the most recent high spatial resolution geodatabases available at the global scale during our study (S1, appendix A). Depending on its nature, each flux is associated with a corresponding landuse class and thus for instance the N and P fluxes of crops are spatialized (scaled) on different crops, while N and P fluxes on fodder refer to the fodder area. In the rest of the analysis forest, shrub and bare are considered ‘natural areas’, and in ‘other areas’ we included all the remaining landcover (water, sea and snow).

2.2. The spatial scale effects

The grid-cell structure used in this study allowed the analysis of the effects of the estimations of N and P flux estimations at continental, country, regional, river basin and grid-cell scales. In particular, the administrative spatial allocation is based on the GADM dataset (GADM 2020), while the river basins were defined using the procedure explained in Malagó et al. (2019a). In order to understand the effect of the spatial scale, only for this specific analysis (section 3.7) the nutrient budgets and potential deliveries were calculated after the aggregation of inputs and outputs. Instead, in tables 1 and 2 we report the positive aggregation of surplus from the results at grid cell level. This is done to avoid that a negative surplus will cancel a positive surplus resulting in a nil surplus while the true pressure is that of the positive surplus.
Table 1. Aggregated nitrogen fluxes by continent and developed and developing countries.

| Pressures                      | Short name       | Africa | Asia | Europe | North America | Oceania | South America | World | Developed country | Developing country |
|--------------------------------|------------------|--------|------|--------|---------------|---------|---------------|-------|------------------|--------------------|
| Total area $10^6$ (km$^2$)    |                  | 29.7   | 46.5 | 6.3    | 21.7          | 8.3     | 17.6          | 130.2 | 50.1             | 80.1               |
| Harvest area $10^6$ (km$^2$)  |                  | 2.1    | 5.9  | 1.4    | 1.5           | 0.2     | 1.1           | 12.2  | 3.47             | 8.75               |
| Mineral fertilizers (Tg)      | $N_{\text{min,CROP}}$ | 2.13   | 42.27| 9.80   | 11.84         | 0.86    | 0.86          | 3.26  | 70.2             | 47.95              |
| Manure (Tg)                   | $N_{\text{man,CROP}}$ | 1.43   | 10.30| 2.28   | 2.10          | 0.11    | 1.82          | 18.0  | 4.27             | 13.77              |
| Fixation (Tg)                 | $N_{\text{fix,CROP}}$ | 2.81   | 9.28 | 0.88   | 3.30          | 0.20    | 3.79          | 20.3  | 4.53             | 15.72              |
| Atmospheric deposition (Tg)   | $N_{\text{dep,CROP}}$ | 1.06   | 7.51 | 1.60   | 1.31          | 0.05    | 0.68          | 12.2  | 3.22             | 8.98               |
| Uptake (Tg)                   | $N_{\text{OUT,CROP}}$ | 4.09   | 25.12| 9.84   | 12.82         | 0.83    | 8.52          | 61.2  | 25.05            | 36.18              |
| Surplus (Tg)$^a$              | $N_{\text{SURPLUS,CROP}}$ | 3.47   | 45.13| 5.01   | 5.82          | 0.38    | 2.56          | 62.38 | 10.19            | 52.19              |
| Area fodder $10^6$ (km$^2$)   |                  | 0.95   | 4.22 | 0.89   | 0.24          | 0.46    | 1.44          | 8.2   | 2.77             | 5.42               |
| Mineral fertilizers (Tg)      | $N_{\text{min,FODG}}$ | 0.00   | 0.16 | 0.37   | 0.01          | 0.01    | 0.04          | 0.6   | 0.49             | 0.10               |
| Manure (Tg)                   | $N_{\text{man,FODG}}$ | 1.72   | 9.19 | 2.22   | 2.22          | 0.67    | 1.11          | 4.48  | 19.4             | 14.87              |
| Fixation (Tg)                 | $N_{\text{fix,FODG}}$ | 6.15   | 27.40| 5.81   | 1.57          | 2.97    | 9.34          | 53.2  | 18.02            | 35.21              |
| Atmospheric deposition (Tg)   | $N_{\text{dep,FODG}}$ | 0.46   | 3.92 | 1.00   | 0.16          | 0.10    | 0.82          | 6.4   | 2.06             | 4.39               |
| Uptake (Tg)                   | $N_{\text{OUT,FODG}}$ | 5.06   | 20.45| 6.21   | 1.58          | 2.45    | 8.82          | 44.6  | 14.88            | 29.69              |
| Surplus (Tg)$^a$              | $N_{\text{SURPLUS,FODG}}$ | 3.26   | 20.23| 3.27   | 0.82          | 1.73    | 5.86          | 35.18 | 10.28            | 24.90              |
| Area of grassland $10^6$ (km$^2$) |                  | 6.6    | 5.9  | 1.5    | 1.8           | 4.5     | 2.9           | 23.1  | 7.71             | 14.44              |
| Mineral fertilizers (Tg)      | $N_{\text{min,GRAS}}$ | 0.08   | 2.53 | 2.14   | 1.31          | 0.24    | 0.19          | 6.5   | 3.74             | 2.75               |
| Manure (Tg)                   | $N_{\text{man,GRAS}}$ | 6.42   | 12.71| 4.28   | 4.03          | 1.19    | 6.11          | 34.7  | 7.99             | 26.74              |
| Fixation (Tg)                 | $N_{\text{fix,GRAS}}$ | 2.63   | 2.37 | 0.59   | 0.70          | 1.81    | 1.16          | 9.3   | 3.08             | 6.17               |
| Atmospheric deposition (Tg)   | $N_{\text{dep,GRAS}}$ | 2.43   | 3.04 | 1.35   | 1.07          | 0.60    | 1.40          | 9.9   | 2.94             | 6.95               |
| Uptake (Tg)                   | $N_{\text{OUT,GRAS}}$ | 6.93   | 12.39| 5.01   | 4.27          | 2.30    | 5.32          | 36.2  | 10.65            | 25.57              |
| Surplus (Tg)$^a$              | $N_{\text{SURPLUS,GRAS}}$ | 4.62   | 8.26 | 3.34   | 2.85          | 1.53    | 3.55          | 24.15 | 7.10             | 17.05              |

(Continued)
| Pressures                              | Short name           | Africa | Asia  | Europe | North America | Oceania | South America | World | Developed country | Developing country |
|----------------------------------------|----------------------|--------|-------|--------|--------------|---------|---------------|-------|-------------------|-------------------|
| Natural areas                          |                      |        |       |        |              |         |               |       |                   |                   |
| Forest, Shrub and Bare area \(10^6\) (km\(^2\)) |                      |        |       |        |              |         |               |       |                   |                   |
| Manure (Tg)                            | N\(_{\text{man,nat}}\) | 2.84   | 3.51  | 0.35   | 1.90         | 0.19    | 1.69          | 10.5  | 2.45              | 8.02              |
| Atmospheric deposition (Tg)            | N\(_{\text{dep,nat}}\) | 3.1    | 5.6   | 1.0    | 2.7          | 0.2     | 2.2           | 14.9  | 4.92              | 9.94              |
| Other \(10^6\) (km\(^2\))             |                      |        |       |        |              |         |               |       |                   |                   |
| Atmospheric deposition (Tg)            | N\(_{\text{dep,other}}\) | 0.2    | 0.7   | 0.2    | 0.5          | 0.0     | 0.1           | 1.7   | 0.87              | 0.81              |
| Erosion                                | N\(_{\text{ero}}\)   | 3.88   | 18.94 | 0.87   | 2.82         | 1.90    | 9.30          | 37.7  | 3.35              | 34.35             |
| Human                                  |                      |        |       |        |              |         |               |       |                   |                   |
| Wastewater emissions (Tg)              | N\(_{\text{PS,dep}}\) | 0.28   | 2.63  | 0.77   | 0.57         | 0.03    | 0.40          | 4.7   | 1.54              | 3.16              |
| Industries (Tg)                        | N\(_{\text{PS,ind}}\) | 0.04   | 0.28  | 0.09   | 0.08         | 0.00    | 0.06          | 0.6   | 0.20              | 0.36              |
| Scattered dwellings (Tg)               | N\(_{\text{SD,imp,other}}\) | 0.89  | 2.76  | 0.20   | 0.30         | 0.01    | 0.26          | 4.4   | 0.51              | 3.91              |
| Open defection (Tg)                    | N\(_{\text{SD,open}}\) | 0.34   | 1.02  | 0.00   | 0.01         | 0.00    | 0.03          | 1.4   | 0.00              | 1.39              |
| Built area wash-off (Tg)               | WASHOFF\(_{\text{N}}\) | 0.13   | 0.67  | 0.17   | 0.15         | 0.01    | 0.12          | 1.3   | 0.37              | 0.89              |
| Potential nitrogen delivery (Tg)       | N\(_{\text{pot}}\)   | 23.05  | 109.76| 15.26  | 18.54        | 6.06    | 26.08         | 198.8 | 41.79             | 156.96            |

\(a\) The N surplus was calculated aggregated the values at grid cell level.
Table 2. Aggregated phosphorus fluxes by continent and developed and developing countries.

| Drivers          | Short name | North America | South America | Europe | Asia | Africa | Oceania | Worldwide | Developed country | Developing country |
|------------------|------------|---------------|---------------|--------|------|--------|---------|-----------|-------------------|-------------------|
| Total area (10^6 km²) |            |               |               |        |      |        |         |           |                   |                   |
| Harvest area (10^6 km²) |            |               |               |        |      |        |         |           |                   |                   |
| Mineral fertilizers (Tg) | P\textsubscript{min}\_CROP |               |               |        |      |        |         |           |                   |                   |
| Manure (Tg) | P\textsubscript{man}\_CROP |               |               |        |      |        |         |           |                   |                   |
| Uptake (Tg) | P\textsubscript{OUT}\_CROP |               |               |        |      |        |         |           |                   |                   |
| Surplus (Tg) | P\textsubscript{SURPLUS}\_CROP |               |               |        |      |        |         |           |                   |                   |
| Mineral fertilizers (Tg) | P\textsubscript{min}\_FODG |               |               |        |      |        |         |           |                   |                   |
| Manure (Tg) | P\textsubscript{man}\_FODG |               |               |        |      |        |         |           |                   |                   |
| Uptake (Tg) | P\textsubscript{OUT}\_FODG |               |               |        |      |        |         |           |                   |                   |
| Surplus (Tg) | P\textsubscript{SURPLUS}\_FODG |               |               |        |      |        |         |           |                   |                   |
| Mineral fertilizers (Tg) | P\textsubscript{min}\_GRAS |               |               |        |      |        |         |           |                   |                   |
| Manure (Tg) | P\textsubscript{man}\_GRAS |               |               |        |      |        |         |           |                   |                   |
| Uptake (Tg) | P\textsubscript{OUT}\_GRAS |               |               |        |      |        |         |           |                   |                   |
| Surplus (Tg) | P\textsubscript{SURPLUS}\_GRAS |               |               |        |      |        |         |           |                   |                   |

(Continued)
Table 2. (Continued.)

| Drivers                        | Short name | Africa | Asia | Europe | North America | Oceania | South America | World | Developed country | Developing country |
|--------------------------------|------------|--------|------|--------|---------------|---------|---------------|-------|-------------------|-------------------|
| Natural areas                  |            |        |      |        |               |         |               |       |                   |                   |
| Forest, shrub and bare area $10^6$ (km$^2$) |            | 20.0   | 29.7 | 2.3    | 16.5          | 2.9     | 11.9          | 83.5  | 33.53             | 49.95             |
| Manure (Tg)                    | $P_{\text{man_nat}}$ | 0.17   | 0.26 | 0.05   | 0.28          | 0.02    | 0.21          | 1.0   | 0.35              | 0.65              |
| Weathering                     | $P_{\text{rel}}$ | 0.23   | 0.43 | 0.06   | 0.21          | 0.06    | 0.15          | 1.1   | 0.44              | 0.70              |
| Erosion                        | $P_{\text{ero}}$ | 1.42   | 7.67 | 0.32   | 1.08          | 0.60    | 3.76          | 14.8  | 1.22              | 13.62             |
| Human                           |            |        |      |        |               |         |               |       |                   |                   |
| Wastewater emissions           | $P_{\text{PS_dep}}$ | 0.04   | 0.31 | 0.08   | 0.06          | 0.00    | 0.05          | 0.5   | 0.16              | 0.39              |
| Industries (Tg)                | $P_{\text{PS_ind}}$ | 0.01   | 0.04 | 0.01   | 0.01          | 0.00    | 0.01          | 0.1   | 0.02              | 0.05              |
| Detergents (Tg)                | $P_{\text{PS_det}}$ | 0.01   | 0.08 | 0.03   | 0.03          | 0.00    | 0.03          | 0.2   | 0.08              | 0.09              |
| Scattered dwellings (Tg)       | $P_{\text{SD_impr_other}}$ | 0.17   | 0.48 | 0.03   | 0.04          | 0.00    | 0.04          | 0.8   | 0.07              | 0.70              |
| Open defecation (Tg)           | $P_{\text{SD_open}}$ | 0.07   | 0.21 | 0.00   | 0.00          | 0.00    | 0.00          | 0.3   | 0.00              | 0.28              |
| Built area wash-off (Tg)       | WASHOFF$P$ | 0.03   | 0.15 | 0.04   | 0.03          | 0.00    | 0.03          | 0.3   | 0.08              | 0.20              |
| Potential phosphorus delivery (Tg) | $P_{\text{pot}}$ | 3.07   | 19.82| 2.02   | 3.16          | 1.23    | 5.97          | 35.3  | 5.66              | 29.62             |

a The $P$ surplus was calculated aggregated the values at grid cell level.
2.3. The estimation of uncertainty

To the best of our knowledge, until now a systematic uncertainty assessment of all nutrient fluxes has not been done, and uncertainties reported in literature vary widely (Del Grosso et al 2010, Nol et al 2010, Kros et al 2012, De Vries et al 2003). In addition, the analysis of the uncertainty of nutrient fluxes and budgets is complex due to the large number of inputs, spatial correlations and scale-dependency of uncertainties (Heuvelink and Pebesma 1999). Despite these difficulties and although the scale of the work does not allow a detailed analysis, we estimated the uncertainties of nutrient fluxes in a qualitative way using a score approach similar to that proposed in Kros and Oenema (2014). We identified the main factors of uncertainty with regards to the estimation and the allocation (SI, tables S7 and S8). The scores range from 0 to 2: the value 2 indicates a high uncertainty in the case of factors that are highly variable and maybe strongly influence by the assumptions made during calculation; a score of 1 has been assigned to factors which are moderately variable; 0 was assigned for small uncertainty, to those factors which have the lowest variability or effect on the definition of the flux. The sum of these scores by each flux provided an indication of the uncertainty of the flux.

3. Results and discussion

3.1. The potential nitrogen and phosphorus delivery

We have estimated that the potential nitrogen and phosphorus delivery to rivers, lakes and oceans was around 199 TgN yr$^{-1}$ and 35 TgP yr$^{-1}$, respectively. The main contributions were from Asia (55% for N, 56% for P), South America (13% for N, 16% for P) and Africa (12% for N, 9% for P).

Developed countries and developing countries accounted respectively for 21% and 79% for N potential delivery, and 16% and 84% for P potential delivery (tables 1 and 2). In particular, the countries with the highest N pollution are China, India, United States of America, Brazil and Indonesia, and Colombia and Peru for P. At the regional scale, hot-spot regions are Sichuan, Shandong, Henan, Xizang, Yunnan, Hebei in China and Uttar Pradesh in India, while the river basins most under pressures are Chang Jiang, Ganges, Amazon, Indus, Parana, Magdalena, Mississippi and the Nile. Figure 1 shows the spatial distribution of the specific nitrogen and phosphorus that can be potentially delivered to earth and aquatic ecosystem (kg ha$^{-1}$) at the grid-cell level. Hotspots for high potential losses of nitrogen and phosphorus occur in India and eastern China.

Globally, nitrogen surplus (from crop, fodder and grass) was the largest contributor to the potential losses on all continents (figure 2). For phosphorus, we observed that the surplus, erosion and inputs from human wastewater contributed the most to the potential losses to the oceans. However, it is noteworthy that these potential contributors have different pathways from the land surface to the aquatic environment as well as different main retention processes (soil, river and lake retention).

3.2. The nutrient surpluses and their components

Globally, the nitrogen surplus is about 1.6 times the amount of applied mineral fertilizer in cropland, fodder and grassland, while the phosphorus surplus was estimated to be half the amount of applied mineral fertilizer in the same areas (tables 1 and 2). The highest global contribution of surplus comes from Asia, Europe and North America both for nitrogen and phosphorus. In particular, Henan, Shandong, Hebei, and Sichuan in China as well as Uttar Pradesh in India, correspond to hotspots of net mineral fertilizers and manure on cropland, as well as atmospheric deposition for N and surplus (SI, appendix B, tables S5 and S6). The river basins Chang Jiang, which is the longest river in China and the third in the world, after the Ganges and Mississippi rivers are also affected by the same nutrient sources.

Spatially, rates exceeding 100 kg ha$^{-1}$ of N surplus in cropland were observed in India, in the north and eastern part of China, in the eastern part of North America (in the Mississippi Basin), in Egypt across the Nile Basin, as well as in Europe coinciding with intensive agriculture practices. Similar spatial trends were observed for phosphorus across the globe with the highest values exceeding 25 kg ha$^{-1}$ of surplus (figure 3).
3.3. Nitrogen and phosphorus losses via erosion

Our findings show that soil erosion contributes significantly to the potential nitrogen and phosphorus losses, with the same order of magnitude of that of domestic and industrial losses (tables 1 and 2). Globally N and P transported via erosion were estimated around 38 Tg and 14.8 Tg, respectively. These fluxes of N and P associated with erosion processes are comparable with those provided by Quinton et al (2010) who estimated ranges of 23–42 and 12.5–22.5 Tg for N and P, respectively (SI, appendix B). The spatial distribution of N and P transported via erosion is shown in figure 3. High N and P sediment-bound losses are predicted to occur in South America and Africa in areas characterized by basic volcanic rock lithology. Higher losses occur in India and Nepal where morphological rocks are dominant, mostly along the Himalaya Mountains. High losses are estimated in China across carbonate sedimentary rocks, and in regions of heavy rainfall, areas of steep slopes and high-relief topography (i.e. Andes and Tibetan Plateau). In particular, hotspot regions were the Papua region (the largest and easternmost province of Indonesia) with 1.3 Tg of N and 0.3 Tg of P. The river basins with the highest values of N and P transported via erosion were the Ganges and Amazon (around 2.4 Tg of N and 0.9 Tg of P for both), followed by the Chang Jiang (1.4 Tg of N and 0.7 of P) and the Magdalena in Colombia (1.3 Tg of N and 0.6 of P). Significant losses were also estimated for the Irrawaddy in Myanmar and the Nile.

3.4. Contributions of nitrogen and phosphorus from natural areas

Concerning nitrogen fluxes in natural areas (forest, shrub and bare soils), manure accounted for 5.3% (10.5 Tg) and atmospheric deposition for about 7.5% (14.9 Tg) (table 1) of total potential nutrient delivery. The atmospheric deposition in natural areas is about one half of that of agricultural areas while manure in natural areas is about one-seventh of that deposited in agricultural areas (crop, fodder, grassland). The continental partitioning of these fluxes reveals that Asia and Africa received the major contribution from manure in natural areas. Similarly, atmospheric deposition in natural areas is an important nitrogen pressure in Asia, Africa and North America. Regions with high atmospheric deposition were Para (0.23 Tg) and the Amazonas (0.2 Tg) in Brazil, while in Texas both atmospheric and P manure in natural
areas were high (around 0.2 Tg of N and 0.02 Tg of P respectively). In Europe, the rate of atmospheric deposition of nitrogen in natural areas was higher the other continents with a median value of around 4.2 kg ha\(^{-1}\) (SI, appendix B and figure S4).

The median of specific N and P manure in natural areas were similar in each continent, although in South America we observed the highest median value for manure of around 0.54 kg ha\(^{-1}\) (SI, appendix B and figure S5).

3.5. Phosphorus release from weathering
Chemical weathering fluxes of P amounted globally to 1.14 Tg (table 2). The highest contributions were from Asia, Africa and North America with 0.43 Tg, 0.23 Tg and 0.21 Tg of P respectively. Krasnoyarsk is
a region within Krasnoyarsk Krai in Russia with the highest value of P release from weathering (0.03 Tg) followed by Sakha in Asia (0.026 Tg) and Western Australia (0.018 Tg).

The median of P weathering release was around 0.06 kg ha$^{-1}$ in each continent (SI, appendix B and figure S5), and generally areas with abundant carbonates and active volcanism contributed considerably above the median as shown in figure 3.

3.6. Domestic and industrial nutrient inputs

In total, domestic and industrial inputs amounted to 12.3 Tg and 2.13 Tg of nitrogen and phosphorus, respectively, and our estimations of N and P from wastewater treatment plants and industries are very similar to those reported by Morée et al (2013) (SI, appendix B and table S5).

The hotspots were the regions of Henan, Guangdong and Jiangsu in China and Uttar Pradesh in India with values around 0.1 Tg for N and 0.13 Tg for P. N and P industrial inputs are largest in the same regions, but also in England with around 0.065 Tg of N. The spatial variations of N and P from human sources at the grid cell level are shown in figure 3. Open defecation accounted for about 10% of total domestic and industrial inputs (table 1), concentrated mainly in Africa and Asia where India has the largest number of people practicing open defecation in the world, around 600 million (JMP 2014). The main regions with high rates of open defecation are Uttar Pradesh, Bihar, West Bengal, Maharashtra, as well as for the scattered dwellings. Nutrient transported via runoff in built up areas accounted for 10% of total domestic and industrial inputs reaching higher values in Uttar Pradesh, Guangdong and Jiangsu regions due to the large urban populations. Similar findings were observed for phosphorus (table 2). It was calculated that the contribution of phosphorus-based detergents amounted to 0.18 Tg of phosphorus, corresponding to 8% of the global human inputs for P.

3.7. Spatial scale effects

There are important differences at continental, regional, country and river basin scales for all inputs and outputs in our datasets. Although the sample size has an effect on the distribution of values in the different spatial categories, the heterogeneity of the N and P potential fluxes is reduced with increasing spatial aggregation (figures 4–5). The same was observed for all other fluxes as shown in the supplementary materials (figures S9 and S10). The interquartile range increased from grid-cell to country scale and the outliers decreased. However, the violin plots that show the kernel density distribution, become less compact and more elongated from grid-cell cell to country scale denoting a lower density and less homogeneity around the median value.

It is important to note the large number of outliers characterizing the cells and basin potential losses. This is probably explained by the large number of cells and river basins (figures 4 and 5). The number of outliers is reduced at the regional level probably due to the effect of aggregation but also because many data used for the crop distribution is usually collected at regional level. Then the uncertainty decreases from regional to country and continental scale because all these spatial unit are characterized by a one to one relationship (one region belongs to only one country that belongs to only one continent). This is not the case for river basins and region as a basin can belong to multiple regions, or one region can include more than one basin.

Similar findings were observed analyzing the N and P budgets in crop land for each continent (figures S11 and S12).

It is noteworthy that the river basin scale has a similar distribution to the grid-cell. This means that also at this scale it is possible to identify hotspot zones where generally accurate data are often not available because much of the statistical data used in the budget is collected according to geographic and municipal
boundary rather than watershed boundaries (Shober et al 2011). At the country level where both intensive and extensive agriculture systems are present, N and P budgets are affected by averaging between intensive and extensive regions, as well as both the success and problems in N/P management, obscuring the local situations.

This highlights the need of selecting the appropriate spatial scale based on the specific purpose of the study or analysis. For instance, at the level of the European member states, the surplus calculated at country level is reported regularly by (Eurostat 2020) and used in the four-yearly report of the Nitrates Directive to assess the status of implementation of measures. At regional level (i.e. NUTS 2 administrative territorial units) nitrogen budgets are calculated for instance to assess the regional nitrate leaching (Poisvert et al 2017) and at the farm level they are used to investigate farm performance and as a tool for farm advice (Mulier et al 2003, Schröder et al 2007).

In this context, a harmonized grid-cell approach is extremely useful and can help to overcome the issue of comparison of the different budgets between countries since, as pointed out by Eurostat, currently the European budgets are not comparable due to the difference in the definitions, methodologies and data sources used (Eurostat 2013).

3.8. Uncertainties of nutrient fluxes
Analyzing the main factors that can affect the degree of uncertainty (figure 6 and tables S7 and S8 in SI), we estimated that the main uncertain inputs are nutrient transport via erosion, manure, and domestic nutrient emissions, followed by atmospheric deposition, nitrogen fixation and nutrient plant uptake.

Nutrient transport via erosion and human nutrient emissions resulted in more uncertain fluxes because their estimation involves several steps, each adding more uncertainty (figures S2 and S3, appendix A). Similarly, manure is less certain than mineral fertilizers and its spatial allocation between stable and meadow manure is an additional cause of uncertainty as also explained in Bouwman et al (1999). In addition, the quantification of nutrients in manure is affected by several factors that can vary across years, regions, weather, animal type, as well as the calculation method (Zhang et al 2020).

Atmospheric deposition follows manure in the rank of the uncertainty because it is generally obtained from coarse resolution chemistry-transport models that may not be consistent with the land cover and land use data used to compile the dataset (Van Drecht et al 2005). Bouwman et al (1997) pointed out that there is also the lack of measurements of deposition rates in many parts of the
world (Africa and South America) and the validation of models with point measurements involve serious scaling problems.

In agreement with Van Drecht et al (2005), N and P mineral fertilizers were more certain than other fluxes. However, Zhang et al (2020) pointed out the national scale disagreement from different sources of information. As a matter of fact, fertilizer input information is generally available at the country level from FAO and International Fertilizer Association (IFA) and although most countries have reported similar total synthetic fertilizer sources at the country level, a few countries (e.g. China) reported different values with a discrepancy of up to 30%. In addition, the fraction of synthetic fertilizers used for cropland versus fodder and grassland can vary significantly across countries (Lassaletta et al 2014) and there is no robust information about that.

However, with respect to other studies that assumed a constant rate of application for all crops and states (Nishina et al 2017, Houlton et al 2019, Zhang et al 2020), our downscaling approach considered the heterogeneity among regions and crops types, decreasing the uncertainties related to their spatial allocation.

Although N fixation is also affected by the same uncertainty of mineral fertilizers and plant uptake, it is considered in literature to be one of the most uncertain terms of the N budget (Zhang et al 2020) due to the difficulty in measuring it (Vitousek et al 2002).

Similar results were obtained for the estimation and allocation of nutrient plant uptake which depends on several factors including the crop variety used, climate soil, fertility, crop yields and management (Van Drecht et al 2005, Zhang et al 2020). In addition, major uncertainties arise from insufficient data, for instance, on crops that are not marketed and on the use of crop residues, as well as about the trade of crops.

The uncertainties related to N emissions are discussed in detail by Bouwman et al (1997). Kros et al (2012) showed smaller uncertainties for N emissions and that uncertainties can vary considerably across countries. In particular, they reported a relatively large contribution of N uptake (10%–30%) with respect to the N contribution of emissions (0%–20%).

The other fluxes (phosphorus nutrients from detergents, phosphorus release from chemical weathering and nutrients via wash-off) have less uncertainty with respect other fluxes due to the relatively few steps and factors involved in their calculations. However, it must be noted that very few data are available concerning the concentration of P in detergents at the national scale, and consequently the extrapolation of the information introduces uncertainty.

In addition, the analysis of uncertainty should also consider the spatial scale. It was observed that uncertainties at the continent scale were much smaller compared to uncertainty at the country, region or basin level because errors were partly cancelled out by spatial aggregation (Kros et al 2012). In addition, the fluxes can be correlated each other, which affects the degree of uncertainty (Heuvelink and Pebesma 1999).

4. Conclusion

Our study provides a consistent global dataset of nitrogen and phosphorus fluxes, highlighting areas of potential nutrient delivery to rivers, lakes and oceans.

Figure 6. Uncertainties of main nutrient fluxes considering both estimation and allocation. The numbers are the sum of scores reported in tables S7 and S8 in SI.
Currently these fluxes describe the nutrient budget around 2005, but the procedure is easily reproducible for updating and scenario analysis. We show that large nitrogen and phosphorus surpluses occur throughout the world, indicating that the additional supply of N and P fertilizers to meet the expected increasing agricultural demand by 2050 (Bruinsma 2009) can increase the risk of serious environmental problems. In this context, our dataset plays a strategic role providing high resolution spatial information of nutrient fluxes that are very difficult to retrieve at large scales (Bouraoui and Grizzetti 2011), or are only available at administrative geographical boundaries. We also showed the importance of the scale and the effects of the aggregation of fluxes at different spatial scales, highlighting the valuable resource of the proposed datasets. In particular, these are extremely useful for large scale water quality modeling. Malagó et al (2019a, 2019b) used these datasets for the prediction of annual and monthly nutrient loads, analysis of source apportionment and investigations of river and lake retentions from local, regional, country to transcontinental scale.

However, some limitations exist. First, the nutrient fluxes are affected by uncertainties that are difficult to assess analytically. However, in this study we proposed a qualitative methodology and we showed that nutrient transport via erosion, human nutrient emissions and manure resulted in more uncertain fluxes because their estimation involves different factors in several steps, each adding more uncertainty.

Second, in the current version we do not consider P in atmospheric deposition that, although its contribution is less that N, it was recently demonstrated to have an important role in the trophic status of alpine lakes and streams in Europe and North America (Brahney et al 2015, Stoddard et al 2016). In addition, we did not include also the amount of N release from chemical weathering due to the limited information available when this work started, although it has been demonstrated to be important in montane ecosystems and where high biological N fixation rates are temperature-limited (Houlton et al 2018, Sabo et al 2019).

In conclusion, our dataset provides key indicators of agricultural sustainability, including nitrogen use efficiency (SI, appendices A and B) and nitrogen surplus as proposed by Gil et al (2019) to evaluate the implementation of the global Sustainable Development Goals, and in particular SDG-2 ‘End hunger, achieve food security and improved nutrition and promote sustainable agriculture’.

Data availability statements

Our dataset (spatial data in raster format and csv) will be made available according to JRC Data policy (https://data.jrc.ec.europa.eu/). However, all data discussed in the paper will be made available to readers also upon request to authors.

The data that support the findings of this study are available upon reasonable request from the authors.

Author contributions

Anna Malagó: conceptualization, methodology, software, validation, formal analysis, investigation, resources, data curation, writing—original draft preparation, writing—review and editing, visualization.

Fayçal Bouraoui: conceptualization, methodology, software, validation, formal analysis, investigation, resources, writing—review and editing, project administration.

ORCID iD

Anna Malagó https://orcid.org/0000-0003-1301-6151

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