ENVIRONMENTAL RESEARCH
COMMUNICATIONS

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To cite this article: Uwe Häußermann *et al* 2021 *Environ. Res. Commun.* **3** 095004

IOP Publishing *Environ. Res. Commun.* **3** (2021) 095004 https://doi.org/10.1088/2515-7620/ac23e5

Environmental Research Communications

CrossMark

OPEN ACCESS

RECEIVED
28 April 2021

REVISED
18 June 2021

ACCEPTED FOR PUBLICATION
6 September 2021

PUBLISHED
16 September 2021

PAPER

National nitrogen budget for Germany

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Keywords: agriculture, energy, Germany, industry, national nitrogen budget, reactive nitrogen

Supplementary material for this article is available online

Abstract

Emissions of reactive nitrogen (N_r) give rise to a wide range of environmental problems. Nitrogen budgets for various systems and on different scales are an established tool to quantify the sources and fate of N_r. The national nitrogen budget (NNB) for Germany calculates the nitrogen flows for eight pools: Atmosphere, Energy and Fuels, Material and Products in Industry, Humans and Settlements, Agriculture, Forest and Semi-natural Vegetation, Waste, and Hydrosphere, as well as for the transboundary N-flows. In Germany, in total 6,275 kt N_r a^{-1} has been introduced into the nitrogen cycle annually (mean 2010 to 2014), of which 43% stem from ammonia synthesis. Domestic extraction and import of nitrogenous fossil fuels (lignite, coal, crude oil) releases another 2,335 kt N_r a^{-1}. Import of food, feed and materials contributes 745 kt N_r a^{-1}, while biological N fixation converts 308 kt N_r a^{-1} into organically bound nitrogen. In terms of N_r sinks, the combustion and denoxing of fuels and the refining of crude oil converts 2,594 kt N_r a^{-1} to N_2. In waters, soils, and wastewater treatment plants, denitrification leads to the release of 1,107 kt N_r a^{-1} as N_2. Via the atmosphere and hydrosphere, Germany exports 755 kt N_r a^{-1} to neighbouring countries and into coastal waters. On balance, Germany releases 1,627 kt N_r a^{-1} annually to the environment. However, the NNB as a whole and the individual pool balances involve substantial uncertainties, which have to be considered when interpreting the results.

1. Introduction

Since the development of the Haber-Bosch process for large-scale ammonia synthesis a century ago, humans have intervened in the nitrogen cycle more than in any other geochemical cycle (Galloway *et al* 2008). The total world ammonia production reached around 150 Mt N in 2019 (USGS 2020), by far the largest part of which is used as N fertilizer in agriculture (estimated 79% in 2013/14, calculated after Heffer and Prud'homme (2016). The planetary boundary for industrial and intentional biological fixation of nitrogen were quantified by Steffen *et al* (2015) to 63 Mt N per year, which is exceeded by a factor of more than two. The excessive release into the environment of reactive nitrogen (N_r; defined as all N forms other than N_2) causes numerous problems, including the loss of aquatic and terrestrial biodiversity, the formation of greenhouse gases, air pollution, and increased nitrate levels in groundwater and marine ecosystems. A nitrogen budget (NB) quantifies the N_r emission from the various sources, the circulation of N_r compounds through the biosphere and technosphere, and the final sinks of N_r, termed the eco-systemic nitrogen cascade by Galloway *et al* (2003). The NB has been introduced as an efficient instrument for determining the N_r flows, which helps to raise awareness of their potential impacts. Furthermore, the NB provides policymakers with information for identifying intervention points and developing efficient emission reduction measures (UNECE 2013).

Several studies on NBs have been published across a range of scales, various system boundaries of N flows, and different regional entities. On the global scale, Smil (1999) estimated nitrogen flows in crop production, while Fowler *et al* (2013) described the processing and fluxes of N_r in terrestrial and marine systems and the atmosphere. Quite a number of studies focus on agriculture and the food sector, e.g. Pierer *et al* (2015) assessed the consumer-related N flows with food and material use in Austria, and Lassaletta *et al* (2014) balanced the so-called hydrologic agro-food system in Spain. Agricultural nitrogen emissions to the atmosphere and the hydrosphere were calculated for Canada by Janzen *et al* (2003) and for New Zealand by Parfitt *et al* (2008). Olsthoorn and Fong (1998) focused on nitrogen losses from anthropogenic N inputs in the Netherlands. Based on these data, Kroeze *et al* (2003) illustrated the uncertainties and knowledge gaps in the fate of nitrogen in natural and terrestrial systems. The studies by Domene and Ayres (2001) and Saikku *et al* (2007) give examples of national nitrogen flow analysis in the industry and energy sector.

A national nitrogen budget (NNB) covers the relevant N inflows and outflows for all economic sectors within a nation. The US NNB is based on a total nitrogen turnover of 34,900 kt N in 2002 (Doering III *et al* 2011). Houlton *et al* (2013) interpreted the turnover of 37,000 kt N in 2002 as the total N fixation and assessed the intentional N fixation as five times higher than the unintentional N fixation for the US in 2007. Three NNBs have been calculated for China (Cui *et al* 2013, Gu *et al* 2013, Luo *et al* 2018), varying in the number of subsystems and N flows considered. Gu *et al* (2013) calculate 22,500 kt N a^{-1} as N accumulation in soil, biomass, products and inland water, while 2010 Cui *et al* (2013) reports 31,000 kt N accumulation for the same year, and Luo *et al* (2018) quantify the N loss and accumulation only in the food sector to 47,200 t N a^{-1} in 2014. A nitrogen flow analysis for Switzerland concluded that an N_r emission reduction by more than 70% is required to meet the national environmental targets Heldstab *et al* (2014). Projecting the temporal trend of the N budget surplus 1990 to 2012 for the United Kingdom, Worrall *et al* (2016) predict that the UK will become a net sink of total N in 2031. While all studies mentioned above rely mostly on statistical databases, the European Union nitrogen budget (Leip *et al* 2011) was almost completely model-based and illustrated the wide range of N_r emissions within the EU with high spatial resolution.

The above listed NNB applications differ considerably in the number of subsystems and N flows, the methodology to determine them, and the consideration of stock changes. The results are therefore only comparable with each other to a very limited extent. To overcome this problem, an international agreement under the revised 1999 'Gothenburg Protocol to the Convention on Long-Range Transboundary Air Pollution' (CLTRAP) established a NNB reporting scheme. With the 'Guidance document on national nitrogen budgets' (UNECE 2013) the Expert Panel on Nitrogen Budgets (EPNB) of the Task Force on Reactive Nitrogen (TFRN) presented guidelines on NNB calculation, mainly addressed to the bodies of the 'Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe' (EMEP). To the best of our knowledge, a NNB based on the NNB Guidance Document has not yet been carried out. We calculated the NNB for Germany adopting the UNECE (2013) methodology and focused on two questions: (i) What are the sources, quantities and species of N_r emissions in Germany and what are the final N_r sinks? (ii) What is the uncertainty of the NNB? Are the N inflows and outflows for Germany in balance, or are there significant gaps in the sources, fate and/or sinks of N_r?

2. Material and methods

We applied the NNB scheme of the 'Guidance document on national nitrogen budgets' (UNECE 2013) and calculated the N_r flows between eight pools for Germany: *Atmosphere*, *Energy and Fuels*, *Material and Products in Industry*, *Humans and Settlements*, *Agriculture*, *Forest and Semi-natural Vegetation*, *Waste,* and *Hydrosphere.* Additionally, the transboundary nitrogen flows with the *Rest of the World* are assessed. With the exception of *Atmosphere,* pools are subdivided into two to four sub-pools, based on the sector structuring used for the national greenhouse gas emissions inventory (IPCC 2006, EEA and EMEP 2013). In total, we determined N inflow and outflow for 20 sub-pools (table 1). For each of the eight major pools, the EPNB has developed an annex, which explains the methodology for the computation of the relevant pool's N flows (to date six annexes are available online).

The N flow calculation is based on different types of data. For the majority of pools, the N inflow and outflow involve nitrogen that is bound in biogenic or technical materials. These N flows are mainly calculated from the transported material flow multiplied by its mean nitrogen content, using data taken from official statistics and data bases. The emission of greenhouse gases (N_2O, NO_x) and ammonia is taken directly from the National Inventory Reports. The German Ministry of Agriculture reports the agricultural NB in tonnes N per year (BMEL 2020). Atmospheric transport models are applied to assess the atmospheric N deposition (model

Table 1. Pools and sub-pools of the national nitrogen budget for Germany.

Pool	Sub-pool
Atmosphere	—
Energy and Fuels	Energy Conversion
	Manufacturing Industries and Construction
	Transport
	Other Energy and Fuels
Materials and Products in Industry	Food and Feed Processing
	Nitrogen Chemistry
	Other Producing Industry
Humans and Settlements	Human Body
	Material World
Agriculture	Animal Husbandry
	Soil Management
	Biogas Production
Forest and Semi-natural Vegetation	Forest
	Other land
	Wetland
Waste	Solid Waste
	Wastewater
Hydrosphere	Groundwater
	Surface Water
	Coastal Water

LOTOS-EUROS, Schaap *et al* 2018) and the import and export of NH_y and NO_x (model MSC-W, Norwegian Meteorological Institute 2017). The N flows in the *Hydrosphere* pool are assessed using the MoRE model (Fuchs *et al* 2017). The statistics and data bases used, approaches to calculate the individual N flows, and results are explained in detail in the Supplement. The criterion for NNB inclusion was an N flow $\geqslant$1 kt N per year. For reasons of clarity, in the following all figures are rounded to full digits, outflows are indicated by a minus sign.

3. Results

3.1. National nitrogen flow analysis

In total, we quantified some 150 individual nitrogen flows for Germany. Figure 1 shows the (partly aggregated) annual N flows for the eight pools (mean 2010–2014). Summarizing the flows and allocating emissions from aggregated anthropogenic sources to air and surface waters (table 2), the values show that agriculture accounts for two-thirds of all reactive nitrogen released in Germany, it remains by far the most important source of N_r emissions into the air and into surface waters. Furthermore, it shows that two thirds of the overall anthropogenic nitrogen emissions are released to air and one third to the surface waters.

The annual N turnover for the eight NNB pools totals 22,760 kt N a^{-1} (here N denotes N_r and N_2). Ammonia synthesis, import and domestic extraction of fuels, import and export of chemical products, food and feed are the largest N_r flows in Germany's NNB. With regard to the primary sources and final sinks of N_r in Germany, two main domains can be distinguished (table 2). Energy production, domestic extraction and import of fossil fuels as well as the formation of thermal NO_x in combination release −2,527 kt N a^{-1}. This corresponds very closely to the N amount of 2,594 kt N a^{-1}, which is converted into N_2 by fuel combustion, flue gas denoxing and crude oil refining. Thus, power generation (including traffic) is obviously a sector with a large N turnover. Due to our assumptions this is largely closed, however it contributes to a relevant extent to the overall NO_x emissions (table 3).

The second domain includes all other N conversions. The most important input is the ammonia synthesis of 2,695 kt N a^{-1}, of which 1,664 kt N a^{-1} is used as nitrogen fertilizer. There are net imports of 745 kt N a^{-1} as constituents of food, feed, and chemicals and manufactured non-food products. With 308 kt N a^{-1}, biological N fixation plays only a minor role in Germany, and the major part of this is by legume cropping. This gives a total of 3,748 kt N a^{-1} for which the final sinks are only partially known. The denitrification in soils, groundwater, surface water and wastewater treatment plant is estimated at −1,107 kt N a^{-1}. With the transport of N_r species via the atmosphere und in rivers, a net total of −744 kt N a^{-1} leaves Germany. The disposal of wastes and an increase in timber storage is calculated to lead to only a very small N stock change in the German NNB. Thus,

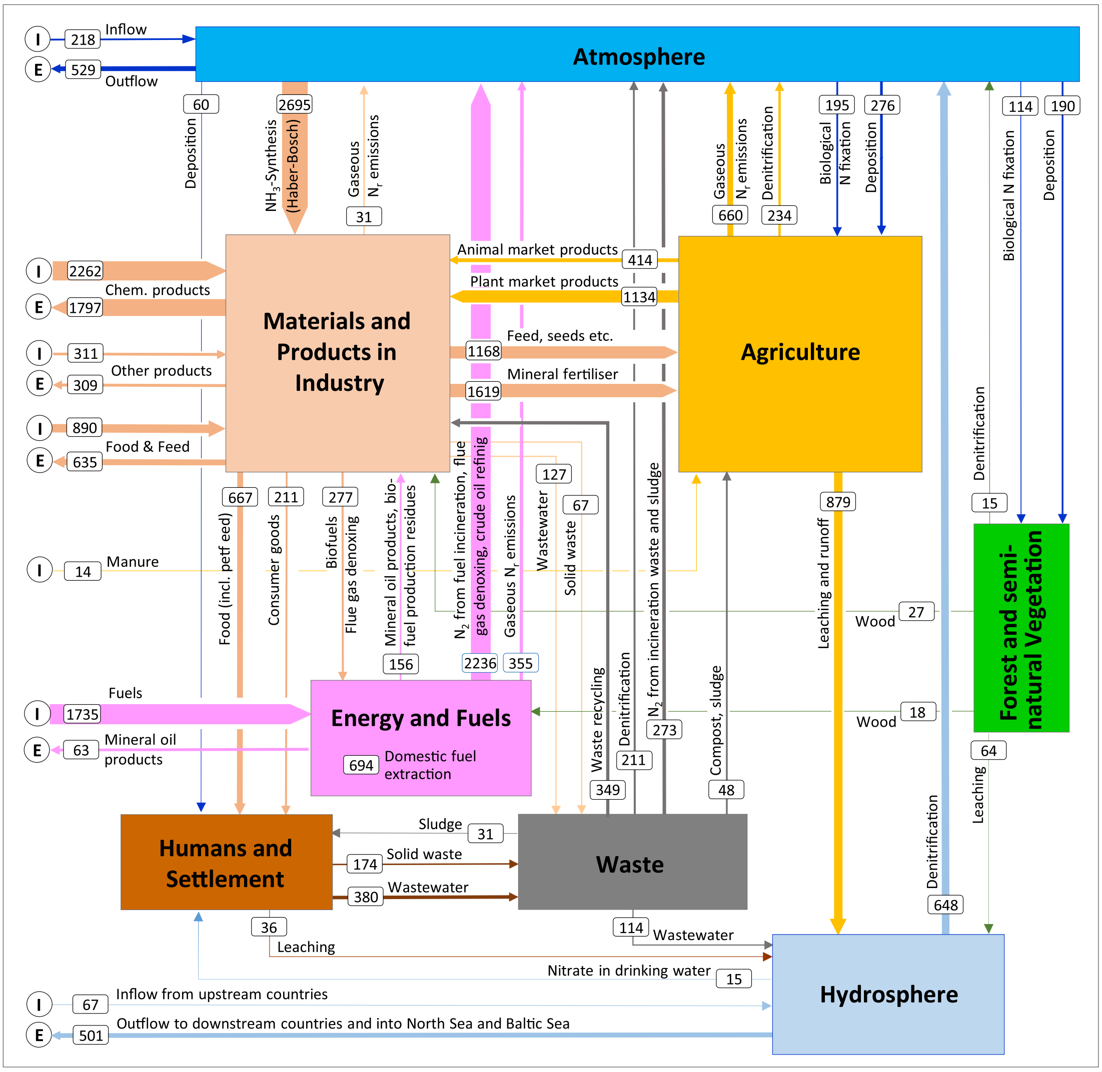

Figure 1. Nitrogen pools and N flows (kt N a^{-1}) of the National Nitrogen Budget for Germany (mean 2010–2014).[6]

Table 2. Anthropogenic sources and emissions of reactive nitrogen into air and surface waters in Germany (mean 2010–2014).

Source	NO_x-N kt N a^{-1}	NH_3-N kt N a^{-1}	N_2O-N kt N a^{-1}	NO_3-N kt N a^{-1}	Totals kt N a^{-1}
Agriculture	36.0	558.0	65.4	381.9	1041.3
Transport	159.6	11.5	3.0	0.0	174.1
Industry/Energy Conversion	184.2	16.6	11.7	29.9	242.4
Households/wastewater treatment plants/urban areas	0.1	2.9	2.1	84.4	89.5
Totals	379.9	589.0	82.2	496.2	1547.3

there is a gap of 1,804 kt N a^{-1} between the N_r quantities in the primary sources and final sinks, which corresponds to ~29% of the N_r sources.

Theoretically, the sums of the N inflows and outflows should be nearly equal. However, table 4 demonstrates that this is not the case for several pools, nor for the overall German NNB. A surplus of 2,126 kt N a^{-1}, corresponding to ~9% of the total N inflow, indicates the magnitude of the uncertainties in the NNB

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[6] Trans-boundary N flows are shown as import (I) and export (E). Only N flows at or above 10 kt N a^{-1} are displayed, N flows are partly aggregated, and change in N stock is not indicated.

Table 3. Sources and final sinks of reactive nitrogen in Germany (mean 2010–2014).

Process	Nitrogen species	N-flow kt N a^{-1}
Ammonia synthesis	NH_3	2,695
Domestic extraction and net import of fossil fuels	N(org)	2,335
Formation of thermal NO_x	NO_x	192
Biological N fixation in soils (agriculture and natural vegetation)	N(org)	308
Net import with food, feed and materials (without fuels)	N(org)	745
Sum of sources		6,275
Conversion of N_r to N_2 with combustion and denoxing	N_2	−1,706
Nitrogen losses with refining of crude oil	N_2	−818
Denitrification total, of which	N_2	−1,107
- Soils (agricultural crops and natural vegetation)	N_2	−248
- Waters (groundwater, surface waters)	N_2	−648
- Wastewater treatment plants	N_2	−211
Waste disposal (landfills)	N(org)	−85
Net export via atmosphere	NH_3, N_2O, NO_x	−312
Net export with rivers	NO_3, N(org)	−433
Sum of sinks		−4,471
Difference		1,804

Table 4. Nitrogen inflow and outflow in the pools in German NNB (mean 2010–2014).

Pool	Inflow kt N a^{-1}	Outflow kt N a^{-1}	Difference kt N a^{-1}	Difference % of inflow
Atmosphere	1,271	−1,062	209	16%
Energy and Fuels	2,662	−2,632	30	1%
Material and Products in Industry	8, 245	−6,841	1, 404	17%
Humans and Settlements	958	−590	368	38%
Agriculture	3, 320	−3,320	0	0%
Forest and Semi-natural Vegetation	598	−140	458	77%
Waste	763	−1110	−347	−45%
Hydrosphere	1, 167	−1, 164	3	0%
Trans-boundary N flows	3, 776	−3, 775	1	0%
Totals	22,856	−20, 732	2,124	9%

Table 5. Nitrogen inflow and outflow in *Atmosphere* pool (mean 2010–2014).

Nitrogen flow	NO_x kt N a^{-1}	N_2O kt N a^{-1}	NH_y kt N a^{-1}	Total kt N a^{-1}
Emissions in Germany	380	83	590	1,053
Inflow to Germany	115	n.r.	104	218
Total inflow	495	83	694	1,271
Deposition	−178	0	−354	−532
Outflow from Germany	−281	n.r.	−249	−530
Total outflow	−458	0	−603	−1,062
Net inflow	37	83	91	210

calculation. In the following section, we explain in more detail the most important N flows and the possible reasons for the differences between inflow and outflow in the individual pools.

3.2. Pool and sub-pool budgets

3.2.1. Atmosphere

The N flow calculation for the pool *Atmosphere* combines different methods. The data on emission of NO_x, N_2O and NH_3 from the economic sectors is taken from the National Inventory Report (CLRTAP Reports; tables S2–S4, S2–S5 (available online at stacks.iop.org/ERC/3/095004/mmedia)), the deposition of NH_y and NO_x for various receptor surfaces is modelled by the LOTOS-EUROS model (Schaap *et al* 2018; tables S2–S6 in supplementary material) and the transboundary transport of NH_y and NO_x is given by the EMEP Source-Receptor-Tables (Norwegian Meteorological Institute 2017, tables S2–S8). There is a difference of 210 kt N a^{-1}

Table 6. Nitrogen inflow and outflow in *Energy and Fuels* pool (mean 2010–2014).

Nitrogen flow	kt N a^{-1}
Fuels—domestic extraction	694
Net import of fuels and mineral oil products	1,672
Wood (for combustion)	19
Formation of thermal NO_x	192
NH_3 used for denoxing of flue gases	85
Total inflow	2,662
NH_3, N_2O and NO_x emissions to atmosphere	−355
Conversion of N_r to N_2 with fuel combustion and denoxing	−1,418
N_r loss with crude oil refining	−818
Refined mineral-oil products for processing in the chemical industry	−32
Solid waste and wastewater	−10
Total outflow	−2,632
Net inflow	30

between total atmospheric N_r inflow and outflow (tables 5, S2–S10). Since no stock change occurs in the atmosphere, the difference might be due to disparate assumptions when modelling atmospheric NO_x and NH_y flow with the LOTOS-EUROS model on the one hand and the EMEP model on the other. Excluding N_2O, which is not deposited and for which none of the models include imports or exports, the difference between total inflow and total outflow is reduced to 126 kt N a^{-1}, or some 10% of the total atmospheric N_r turnover. N_2O can be taken up by soils, but the uptake rate is assumed to be marginal compared to the emission (Syakila *et al* 2010). According to our mass balance approach, about 60% of the national NH_3 emissions and 45% of the national NO_x emissions are redeposited in Germany. Overall, Germany is a net exporter of air pollutants, mainly due to its high spatial density of emissions (especially in the north-west and south-east region due to livestock farming). A large proportion of the emissions are transported in the atmosphere over long distances and carried beyond national borders.

3.2.2. Energy and fuels

The NB for the *Energy and Fuels* pool is based on the Energy Balance for Germany (AGEB 2017; tables S3–2) which is structured primarily to register the conversion and use of energy. The material flow data of the Energy Balance for Germany include double counting and uncertainties for various positions, as is pointed out in the comments (AGEB 2017). Double counts in the statistics cannot be corrected by an external user and as a consequence these effects carry over to the N flow calculation. With a difference of 30 kt N a^{-1}, the budget seems quite well balanced (tables 6, S3–S8). However, this is primarily due to the fact that we calculate two key N outflows as differences: (i) The N inflow with the combustion of fossil fuels (lignite, coal, mineral-oil products), wood for power and heat generation (*Energy Conversion* sub-pool) and the NH_3-consumption for flue gas denoxing, which totals 1,606 kt N a^{-1} (tables S3–S6). Of this, 188 kt N a^{-1} is converted to non-thermal NO_x. For the remaining 1,418 kt N a^{-1}, we assume that the combustion residues (ashes, filter dust, wastewater, etc.) are nitrogen-free and this share of N_r in fuels is completely converted to N_2. (ii) A statistical difference of 818 kt N a^{-1} occurs between the 932 kt N a^{-1} in crude oil refined for domestic consumption and the 144 kt N a^{-1} in the resulting mineral-oil products (tables S3–3). We assume that this N loss in refining of crude oil can also be regarded as an N_r-neutral process, because the oil is hydrotreated for removal of mainly sulfur, in which case probably most of the N flows out as NH_3 in the sour gas and is subsequently oxidized to N_2 in a Claus process burner. However, German petroleum companies are not able to provide any evidence for this assumption. Overall, the calculation of the N_r turnover with combustion and crude oil refining depends to a large extent on the assumptions about the nitrogen contents of the fuels. For coal and mineral oil, this depends strongly on the origins (deposits) of the fuels; the data on this vary widely.

3.2.3. Materials and products in industry

Materials and Products in Industry is by far the largest pool for N_r turnover due to the large amount of nitrogen used in chemical processes (tables 7, S4–S9). For the *Food and Feed Processing* sub-pool, the N inflows and outflows are nearly balanced (tables S4–S10). Obviously, the statistics on food and feed production and consumption quantities and the data on N content in products correspond quite well. For the manufacturing industry and the associated *Nitrogen Chemistry* sub-pool, the German Production Survey (Statistisches

Table 7. Totals of nitrogen inflow and outflow in *Industry* sub-pools and pool (mean 2010–2014).

Pool/Sub-Pool	Inflow kt N a^{-1}	Outflow kt N a^{-1}	Difference kt N a^{-1}
Food and Feed Processing	2, 568	−2,684	−116
Nitrogen chemistry	4, 989	−4,989	0[a]
Other Producing Industry	2, 078	−558	1,520
Material and Products in Industry, without internal N flows between sub-pools	8, 245	−6,841	1,404

[a] Sub-pool budget is based on the premise that the total outflow is equal to the total inflow

Table 8. Nitrogen inflow and outflow in *Humans and Settlement* pool (mean 2010–2014).

Nitrogen flow	kt N a^{-1}
Atmospheric N deposition (NO_x, NH_y) on urban land	60
Food and pet feed consumption	668
Consumption of commodities	166
Use of N mineral fertiliser, wood and sludge in settlements	76
Nitrate in groundwater abstracted for drinking water supply	15
Total inflow	985
Solid waste	−174
Wastewater (from households and from sealed areas)	−380
Nitrate leaching (from settlement areas)	−36
Total outflow	−590
Net inflow	395

Bundesamt 2020) provides data on production, import and export of commodities. The survey distinguishes between 'initial products' and 'products intended for sale'. However, an evaluation of the German Production Survey raises problems. It is not possible to rule out double counting in the statistics; all products that are not 'intended for sale' are initial products for further processing, but they may re-occur in a number of subsequent production steps. Also, items which are 'intended for sale' may nevertheless be used as initial products in another production process. Finally, for various types of goods, the data on production quantities are not published for data protection reasons. Despite these problems, inflow and outflow to the *Nitrogen Chemistry* sub-pool is calculated based on the German Production Survey. For this, the individual items are grouped in accordance with the Eurostat classification of commodities (at the 4-figure code level) to 28 classes of chemicals containing nitrogen (tables S4–S7). For each group, average N content is calculated from the chemical structure of a typical compound or is estimated according to the UNECE (2013), Annex 6. Key process in the nitrogen chemistry is ammonia synthesis with 2,695 kt N a^{-1}. Together with 2,262 kt N a^{-1} in imported chemical products (plus 32 kt N a^{-1} in mineral-oil products for processing in chemical industry), the inflow in the *Nitrogen Chemistry* sub-pool totals 4,989 kt N a^{-1}. Given the known outflows of −1,664 kt N a^{-1} with mineral N fertilizer, −1,797 kt N a^{-1} in chemical products and −217 kt N a^{-1} in N emissions and waste, we define the remaining 1,311 kt N a^{-1} as inflow in the *Other Producing Industry* sub-pool as precursors and chemicals for the production of consumer goods.

Production, import and export of commodities for use by consumers in the *Other Producing Industry* sub-pool is also derived from the German Production Survey (AGEB 2017). Note that the groups of manufactured commodities contain very heterogeneous materials (in terms of N contents; tables S4–S8). Furthermore, quantities of consumer goods may be expressed in various units, e.g. numbers of items, square meters, or cubic meters. These reasons may in part explain the discrepancy of 1,520 kt N a^{-1} between the calculated inflow and outflow in this sub-pool.

3.2.4. Humans and settlements

The calculation of N flows to *Humans and Settlement* pool links data from several sets of statistics and from other pools (tables 8, S5–2). The inflow of 668 kt N a^{-1} in food and pet feed consumption (including uptake and kitchen wastes) and of 166 kt N a^{-1} in consumption of commodities stems from the *Materials and Products in Industry* pool. Outflow of -590 kt N a^{-1} only takes place in form of solid waste and wastewater, and nitrate leaching. The difference of 395 kt N a^{-1} represents some 40% of the total N inflow, which is more than twice the inflow of 166 kt N a^{-1} with consumer goods (tables S5–S3). Since no other N outflow comes into question for

Table 9. Nitrogen inflow and outflow in *Agriculture* pool (mean 2010–2014).

Nitrogen flow	kt N a^{-1}
Mineral fertiliser	1,619
Feed (from industrial production)	1,102
Atmospheric N deposition (NO_x, NH_y) on agricultural land	276
Biological N fixation (legumes cropping)	195
Other inflows[a]	127
Total inflow	3,320
Marketed plant and livestock products	−1,548
NH_3, NO_x and N_2O emissions	−659
Denitrification in soils (root zone)	−234
Subtotal outflow	−2,441
N discharge into surface waters via run-off, erosion and tile drainage	−122
Nitrate leaching (below the root zone)	−757
Total outflow	−3,320
Net inflow	0[b]

[a] Seed and planting material, manure import, biogas co-substrates, compost, sludge, meat- and-bone-meal

[b] Budget is based on the premise that the total outflow equals to the total inflow

consumer goods apart from solid waste, these two quantities ought to be nearly equal. An increase in N stock in the *Humans and Settlements* pool is not plausible on this scale. Obviously, the uncertainties in calculating the N flow with nitrogenous products for sale to end consumers in the *Materials and Products in Industry* pool carry over to the *Humans and Settlements* pool. Furthermore, the N inflow with wastewater from households and run-off from sealed areas are only rough estimates. Finally, the N outflow of only −46 kt N a^{-1} in solid waste derived from waste generation statistics (section 3.2.7.) is somewhat lower than the N inflow of 166 kt N a^{-1} with consumption of commodities included in the consumer goods.

3.2.5. *Agriculture*

The *Agriculture* pool forms the second largest pool of N_r turnover in Germany. The calculation of NBs for German agriculture is well-established (Bach *et al* 2011, Häußermann *et al* 2019, 2020) and state-of-the-art data is annually published by the German Federal Ministry of Agriculture (BMEL 2020). The BMEL budget scheme is more differentiated than the OECD/EUROSTAT approach, furthermore the BMEL surplus figures are the reference values for the German nitrate report to the EU Commission, for the calculation of the nitrate river load within the Water Framework Directive reporting, and for the implementation of the Integrated National Nitrogen Target for Germany (Geupel *et al* 2021). Inflow of 1,619 kt N a^{-1} with mineral N fertilizer and 1,102 kt N a^{-1} with feed from industry production together account for 82% of the total N input (tables 9, S6–S5). The withdrawal of −1,548 kt N a^{-1} in marketed plant and livestock products represents the utilized part of the N outflow, while a substantial share of −659 kt N a^{-1} gets lost to the atmosphere as gaseous N_r species. Denitrification in the root zone of crops and grasses may vary widely from nearly zero up to a complete nitrate degradation. As an average denitrification rate, Well *et al* (2016) estimated 14 kg N ha^{-1} a^{-1}, corresponding to a denitrification rate of −234 kt N a^{-1}. Only one soil survey for the agricultural land in Germany has been carried out just once do date (Jacobs *et al* 2018). Thus, data on soil N stock changes are currently unavailable. According to a modelling approach based on long-term soil monitoring sites (Jacobs *et al* 2018), German cropland mineral soils probably show a moderate loss rate in soil organic substance over the past 20 years, and thus a slight decrease in soil N stock. In peat soils used for cropping in Germany, a preliminary estimate suggests that soil N stock depletion could be in the order of −500 kt N a^{-1} (Jacobs *et al* 2018). However, due to the large uncertainty, this value is not taken into consideration for the *Agriculture* N fluxes.

We calculate the agricultural N budget under the premises that (i) there are no further N outflows from the *Agriculture* pool than marketed products, gaseous N emissions (including N_2) and transport by water, and (ii) the soil N stock remains unchanged. Given this, the difference of 879 kt N a^{-1} between total inflow and subtotal of 'known' (directly calculated) outflows is interpreted as the N_r emissions into the hydrosphere from agricultural land (tables S6–S6). The emissions cover the nitrate leaching from the soil root zone (as system boundary of the *Agriculture* pool) towards groundwater, as well as the N discharge into surface waters via run-

Table 10. Nitrogen inflow and outflow in *Forest and Semi-natural vegetation* pool (mean 2010–2014).

Nitrogen flow	kt N a^{-1}
Atmospheric N deposition (NO_x, NH_y) on forest and semi-natural land	190
Biological N fixation (natural vegetation)	113
Change (reduction) in N stock of forest soils	293
Total inflow	598
Denitrification	−14
N_2O and NO_x emissions	<−1
Nitrate leaching	−63
Wood withdrawal (all uses)	−45
Increase in timber stocks	−17
Total outflow	−140
Net inflow	458

off, erosion and tile drainage. From the *Hydrosphere* pool −122 kt N a^{-1} is modelled as N in lateral discharge into surface waters from agricultural land. Subtracting this N amount from the difference of 879 kt N a^{-1} leaves as residual a N outflow of −757 kt N a^{-1} for nitrate leaching towards groundwater as the second pathway from agricultural land into the hydrosphere.

3.2.6. Agriculture forests and semi-natural vegetation

The *Forest and Semi-natural Vegetation* budget shows the largest relative inflow-outflow discrepancy, with a difference of 458 kt N a^{-1}, corresponding to 77% of the N inflow (tables 10, S7–7, S7–8). Currently, major processes such as biological N fixation, denitrification, and nitrate leaching cannot be quantified with the accuracy needed to close the NB for this pool. Only rough estimates are available for average biological N fixation (10 kg N ha^{-1} a^{-1}; Cleveland *et al* 1999), denitrification (1 kg N ha^{-1} a^{-1}; Andreae *et al* 2016) and nitrate leaching (5 kg N ha^{-1} a^{-1}; Beisecker and Evers 2012) in forest soils. The humus status of forest soils was surveyed representatively in Germany in 1987–1993 and again in 2006–2008 (Fleck *et al* 2019). An average annual loss in forest soil N stock (0–60 cm depth) of 26.5 kg N ha^{-1} a^{-1} was measured between the two monitoring periods, which results in a decrease in forest soil N in Germany of −293 kt N a^{-1} (tables S7–5). Related to the total N stock of ~6, 000 kg N ha^{-1} in forest soil humus (0–60 cm) the decrease is equivalent to a loss of around −7.5% over a period of ~17 years (estimate based on numbers from Fleck *et al* 2019). With respect to the NB, a soil N decrease represents a mobilization of N_r, and therefore an N inflow within the budget. Even if the soil stock decrease is not taken into account, a rather large difference of 165 kt N a^{-1} remains in the budget of the *Forest and Semi-natural Vegetation* pool.

3.2.7. Waste

The *Waste* pool is characterized by a high degree of data uncertainty, despite its overall rather small share of the NB. For solid waste, the German statistics on waste generation (Statistisches Bundesamt 2016) deviates considerably from the waste balance statistics (Statistisches Bundesamt 2020) which records the disposal and recycling of wastes. The waste generation statistics list 15 classes of waste materials (potentially) containing nitrogen to which we assigned N contents according of UNECE (2013), accounting for a total N inflow of 249 kt N a^{-1} (tables 11, S8–2). However, the flow of primary solid waste materials through the various sorting and treatment stages, the recycling of materials, and the quantities of final disposal and incineration cannot be traced transparently on the basis of the waste balance statistics. Furthermore, double counting occurs at all stages to an unknown extent. Assessed by the waste balance statistics (tables S8–3), the outflow with material recycling and landfill deposition of solid wastes from households and industry totals −349 kt N a^{-1}, while the solid waste incineration additionally converts −182 kt N a^{-1} in organic substances to N_2 (sludge not included). Thus, between the calculated inflow and outflow for the *Solid Waste* sub-pool there is a substantial gap of −282 kt N a^{-1}, which is more than the total inflow according to the waste generation statistics (tables S8–9). The difference illustrates the discrepancies in the two underlying statistics.

Nitrogen inflow in the wastewater system was calculated as part of the MoRE model (Fuchs *et al* 2017) resulting in 514 kt N a^{-1} (tables S8–5). The calculation is based on data on wastewater discharge from households, industry and sealed areas and their mean N contents. The outflow with treated wastewater discharge is calculated to −144 kt N a^{-1}, and the denitrification in wastewater plants to −211 kt N a^{-1}. For the *Wastewater* sub-pool, the difference between inflow and outflow amounts only to 37 kt N a^{-1} (tables S8–10). Since the *Wastewater* sub-pool does not contain any N stocks, the difference is due to the uncertainties in the calculation.

Table 11. Nitrogen inflow and outflow in *Waste* pool (mean 2010–2014).

Nitrogen flow	kt N a^{-1}
Solid waste (households and industry)	249
Wastewater (households, industry and sealed areas)	514
Total inflow	763
Material recycling of solid waste; compost and sludge used in agriculture	−423
Landfill of solid waste and sludge	−85
Conversion of N_r to N_2 with incineration of waste, meat-and-bone meal and sludge	−273
Discharge of wastewater treatment plants and sewer system into rivers	−114
Denitrification in wastewater treatment plants	−211
NH_3, NO_x and N_2O emissions	−4
Total outflow	−1110
Net outflow	−347

Table 12. Nitrogen inflow and outflow in *Hydrosphere* pool (mean 2010–2014).

Nitrogen flow	kt N a^{-1}
N inflow via run-off, erosion and tile drainage from agricultural land	122
Inflow via discharge of wastewater treatment plants and sewer system	114
Nitrate leaching (from all types of land use)	857
River load from upstream neighbouring countries	67
Atmospheric N deposition on inland surface waters	7
Total inflow (including	1,167
Nitrate removed with water abstraction	−15
Denitrification in the unsaturated zone and in groundwater	−572
Denitrification (retention) in surface waters	−76
River load to downstream neighbouring countries and into coastal seas	−500
Total outflow (including sea fishing)	−1,164
Net inflow	3

3.2.8. Hydrosphere

The nutrient flow in the hydrosphere on the national level is calculated regularly in the context of the EU Water Framework Directive implementation, in recent years using the MoRE model (Fuchs *et al* 2017). Assessed from the NBs for the *Agriculture*, *Humans and Settlement* and *Forest and Semi-natural Vegetation* pools, the leaching of 857 kt NO_3-N a^{-1} below the root zone forms the major N inflow to the *Hydrosphere* pool (tables 12, S9–2). MoRE estimates the denitrification (termed as N-retention by the MoRE model) along the water flow from the root zone through the vadose zone and the groundwater and finally into the river system up to the mouth of the North Sea and Baltic Sea to −648 by kt N a^{-1}. Deducting this estimata from the leached 857 kt N a^{-1} nitrate (below the root zone), −209 kt N a^{-1} (24%) effectively reaches the surface water system. This approach results in an almost balanced NB for the *Hydrosphere* pool (tables S9–7). It should be noted, however, that this approach is only valid under the assumption that there is no change in the N stock in the aquifers, i.e. that the groundwater nitrate concentration in Germany shows no change over time.

3.2.9. Transboundary nitrogen flows

Summed up over all pools, the German import-export budget is balanced (tables 13, S10–1; excluding the N_r import in fuels which is mainly converted to N_2 by fuel combustion). However, this result is due to two opposing factors. With the atmospheric transport of gaseous N species and the N river transport, −744 kt N a^{-1} of reactive nitrogen leaves the German territory into the biosphere of neighboring countries and the seas. On the other

Table 13. Nitrogen import and export from and to Germany (mean 2010–2014).

Pool / Sub-Pool	Import kt N a^{-1}	Export kt N a^{-1}	Budget kt N a^{-1}
Atmosphere	218	−529	−311
Food and Feed Processing	904	−635	269
Nitrogen chemistry	2,262	−1,797	465
Other producing industry	325	−314	11
Surface waters	67	−500	−433
Totals	3,776	−3,775	1

hand, there are budget-closing net imports of 745 kt N a^{-1} by food and feed products and for material for the chemical industry (fuels not included).

4. Discussion

We applied the UNECE (2013) NNB calculation scheme to quantify the N inflows and outflows and the N budget on the national level and for eight pools in Germany. Anthropogenic activities introduce a total of 6,275 kt N a^{-1} reactive N corresponding to annually 76 kg N per capita in Germany. As with all NNBs, ammonia synthesis is the largest N_r source, followed by the release of N_r from the organic N compounds in fuels. With the decision of the German government to phase out power generation from coal and lignite by 2038, the dimension of this N_r source will decline significantly in the next two decades. Due to the small proportion of legume cropping, biological N fixation is currently only of minor importance in German agriculture. The N_r net import consists mainly of nitrogen chemistry products, followed by food and feed. For manufactured goods, the N_r import-export budget is nearly balanced, but must be interpreted with caution in view of the uncertainties in the statistics. Considering the output side, from the total 4,471 kt N a^{-1} quantified final N_r sinks, some ~82% is converted back to molecular nitrogen by combustion and denoxing, refining of crude oil, and denitrification in soils, waters and wastewater treatment plant. Only 18% remains in the form of reactive N species, of which the largest proportion leaves Germany via the atmosphere and as river load. Only a very small amount remains in Germany with an increased disposal of waste and timber stock. However, there is a considerable difference of ~29% of the inflow between the N_r sources and the known or estimated sinks. With the current state of knowledge, we cannot judge to what extent this difference of 1,804 kt N a^{-1} is due to an overestimation of N_r releases from individual sources or to an underassessment of the N_r fluxes on the side of sinks. In the case of under-reporting of sinks, the question is whether these N_r quantities are also entirely converted to N_2 by combustion or denitrification, or whether there are additional releases of N_r that have not yet been recorded in the specific emission reports.

The NBB provides quantitative information on N_r emissions and N_r sinks. However, this does not yet evaluate the environmental impacts of N_r emissions and does not indicate the extent to which they must be reduced. For this, the NNB for Germany is linked to the 'Integrated National Nitrogen Target' implemented by the German Federal Environment Agency (Geupel *et al* 2021). The target value is based on six environmental impact indicators: nitrogen sensitive vegetation, terrestrial ecosystems, surface water quality, groundwater quality, climate change and human health. To protect these environmental goods the national N target quantifies the maximum amount of total acceptable N_r losses in Germany to nearly 1,000 kt N a^{-1}. Compared to the estimated 1,574 kt N_r a^{-1} losses into susceptible environmental sectors in 2015, the N_r losses in Germany have to be reduced by approximately one third.

A basic finding of our study is the rather large inflow-outflow differences discovered both for the NNB Germany amounting to 9% of the total inflow, as well as for the individual pools. Similar to results for other countries, the NNBs are not closed. Rather, we find even larger ranges of NNB imbalances in several cases. Positive differences, with the sum of inflows greater than the sum of outflows, are given by Houlton *et al* (2013) for the US with a surplus of 12% to 25%, for Austria 27% (Pierer *et al* 2015), for the Netherlands 8% (Olsthoorn and Fong 1998) and for China 28% (Gu *et al* 2013). In contrast, a compilation of NNBs for six European countries by Leip *et al* (2011) indicates larger outflows than inflows for all cases, ranging from −8% for Germany and the UK up to −25% for the Netherlands. However, one has to note that the values by Leip *et al* (2011) are based on a different methodological approach than that used by the other studies. The compilation of Leip *et al* (2011) further illustrates the large variability of results for identical sectors between the different approaches. For example, the authors report a balance surplus of 2,534 kt N a^{-1} for the agricultural sector in France, but only 62 kt N a^{-1} for the German agriculture, which is close to the assumption of a balanced *Agriculture* pool in our study. Obviously, the NNB calculation methods are handled very differently, which leads to considerable biases in the results.

To build an NNB is a challenging task. Several elements of the nitrogen budget are only quantifiable with some uncertainty, and the magnitude of this uncertainty is often not quantified as stated by Leip *et al* (2011). Consistent with this, only a few studies quantify the uncertainties of their NNB. Doering III *et al* (2011) estimated the uncertainties of +/−50% for emission and deposition and terms that derived by differences and Worrall *et al* (2016) assumed a percentage error of +/−80% of the median for data sources without providing an explicit uncertainty estimate. A spatialized European wide estimate of the N surplus by four models indicated an uncertainty close to 50% for individual countries (de Vries *et al* 2011). We estimate uncertainty ranges according to the EEA and EMEP (2013) scheme (ref. Supplement) for the individual N flows of our NNB. However, similar to Doering III *et al* (2011) and Worrall *et al* (2016), these are more or less speculative and should be interpreted with reservation.

Imbalances in the NNB and the different pools of the N budgets are caused mainly by three components, namely the uncertainties in our knowledge of the rates of biological nitrogen fixation, the conversion of N_r to N_2 by denitrification and combustion, and the changes over time in N_r stocks in all NNB pools. The information on these three components is insufficient and often contradictory. Some authors attribute the differences in their NNB mainly to the uncertainties in the calculation of the output and then explain a budget surplus with denitrification losses and an accumulation in the N stocks. For example, Janzen *et al* (2003) estimated that the 200 kt N a^{-1} surplus in their budget for Canada is stored in the agricultural soils. To balance the NNB for the Spanish agricultural and food system, Lassaletta *et al* (2014) ascribe 50% of the total inflow of 1,810 kt N a^{-1} to the potential retention within the hydrosystem, while 35% leave the county by products and N_r emissions, and 15% is input-output difference. However, they do not discuss whether associated N transformation processes (denitrification and others) within the hydrosystem could realistically cause an N loss rate of this magnitude. Olsthoorn and Fong (1998) attributed 12% of the Dutch NNB inflow to an N loss via soils, which covers nitrate leaching and denitrification, or to changes in the soil's N stock. The comparison of four NNBs for China illustrates the wide range of the differences between inflow and outflow estimate and their interpretation even for a single country. The NNB by Luo *et al* (2018) specifies 70.1 Mt N a^{-1} inflow but only 3.1 Mt N a^{-1} outflow for China in 2014 and explains the difference as the result of denitrification and accumulation in various N stocks. Ti *et al* (2012) took the difference of 30.1 Mt N a^{-1} between total N input and the accounted outputs in 2007, which corresponds to 58% of the input, and assigned them to denitrification and N storage changes without further distinction. In contrast, the study of Cui *et al* (2013) suggests that only around 20% of the annual N_r production was denitrified, while a total of 49% (31 Mt N a^{-1}) was stored in soil, biomass, products and inland water in 2010. For the same year, Gu *et al* (2013) estimated the total N accumulation to 22.5 Mt N a^{-1} in China, most of it in overfertilized cropland.

The studies cited rarely address the question of whether the N accumulations in the stocks of soils, forest, groundwater, landfill and/or human settlement (calculated as a difference term) are in realistic ranges. For the German NNB an increase in N stocks plays only a minor role, if any. There are no significant increases or decreases of stocks in the pools *Agriculture* (except soils), *Energy and Fuels* or *Material and Products in Industry*, as all related statistics indicate. For the *Forest* pool in Germany, a decrease of the N stock in forest soils was observed at a mean rate of −293 kt N a^{-1}, corresponding to −7.5% loss of the N in soil humus, which is attributed to climate change by Fleck *et al* (2019). The increase in the timber stock of 17 kt N a^{-1} in no way compensates for this N loss in soil. A decrease of the soil N stock can also be assumed for mineral soils and especially for peat soils used as arable land in Germany, although the magnitude cannot be quantified precisely.

In terms of industrial products in China, Gu *et al* (2013) assumed that 25% of these products tend to accumulate in human settlements due to their long service lives. This estimate could have some justification on a global scale for emerging economies, where urban areas are growing rapidly and construction activities (residential buildings, industrial plants and infrastructure) as well as the furnishing of households with durable consumer goods are considerably expanding. For Germany, however, there is no evidence that N is accumulated on a large scale in the long term with the use of materials containing nitrogen in the construction sector or the household consumption of consumer goods.

The largest uncertainty in N stock changes concerns the root zone-unsaturated zone-groundwater system as the main domain of nitrate turnover: neither the total amount of nitrate in these compartments in Germany is known, nor can its change due to seepage water exchange and/or denitrification be estimated plausibly. According to the four-yearly Member State Reports on the implementation of the EU Nitrate Directive since 2012, the nitrate concentration is nearly constant over time at the 697 groundwater monitoring sites in Germany (BMUB and BMEL 2020). As an approximation, we assume for our NNB that the difference of 648 kt N a^{-1} between the nitrate leaching from soil root zone (857 kt N a^{-1}) and the nitrate load into the surface waters from groundwater effluents (209 kt N a^{-1}) is entirely denitrified and thus contributes substantially to neutralizing the N_r emissions. However, the denitrification capacity of aquifers mainly depends on iron disulfide and organic carbon which, being finite resources, are susceptible to depletion (Knoll *et al* 2020). This will generally result in future risk of increasing nitrate concentrations in the groundwater and subsequent higher loads to surface water via the groundwater pathway. The status of denitrification capacity in aquifers and the consequences of its possible decline have been studied in Germany for some time (Wilde *et al* 2017) and are the subject of intense debate.

The *Waste* pool is the only sector of Germany's NNB for which a negative inflow-outflow difference in our estimate. The uncertainty is mainly attributed to the *Solid Waste* sub-pool due to the shortcomings and contradictory data in the two underlying statistics on waste generation and waste balance. The Federal Statistical Office has no sound information about the composition and the further treatment of the solid waste materials. The outputs from waste are classified as 'waste for recycling' and 'waste for disposal' by the plant operators without further verification. It is not possible to determine valid quantities for the individual types of waste or to quote their material recycling. Furthermore, the assumptions about the N contents of the types of waste are speculative and the N fluxes in waste treatment as well as the N accumulation by waste landfilling are therefore generally subject to large uncertainties, as also illustrated by the discrepancy for consumer products. According to the statistics, 166 kt N a^{-1} enters households in non-food consumer goods, but the waste statistics calculates only an outflow of −46 kt N a^{-1} with solid waste. Since there are no appreciable increases in stocks of commodities in the private households and no other outflow for consumer goods apart from waste is known, these two figures do not match in any way. For future calculation of N flows for the solid waste sector, the material flows must be broken down further to separate various material groups whose generation and final sinks are traced clearly by the statistics. Further gaps in the information relate to appropriate mean N contents that can be allocated to these material groups. A similar discrepancy was also found by Pierer *et al* (2015) for Austria. The mismatch between inflow of non-food industrial products and outflow of waste material there amounts to 83% of inflow. The authors assume streams of material waste, which are not accounted for by the statistical survey.

The divergent approaches and the partly contradictory results of the above-mentioned studies underline the urgency to standardize NNB calculations. With the development of the guideline EEA and EMEP (2013) such a standardized methodology is actually available. However, to the best of our knowledge, no NNB in accordance with the methodology of the EEA and EMEP (2013) has yet been established, except our study presented here. While we could make use of many of the equations and the wealth of underlying detail material provided, there were several instances that requires work-arounds because the NNB guidance still shows considerable room for further improvements. Specifically, we had to (i) adjust for some heterogeneous calculation schemes offering different levels of detail, (ii) add the N contents of material flows when unavailable, or use specific German data when the default value seemed implausible. Furthermore, (iii) the flow description and coding had to be adjusted occasionally as being incoherent between pools, and (iv) sink terms and stock changes had to be added into the concept in order to cover situations when flow balances did not match for a specific pool or the total NNB.

5. Conclusions

Our work provides a comprehensive reactive nitrogen data set for Germany. It summarizes the latest knowledge of emissions, production, flows and sinks of reactive nitrogen. It is the most complete dataset of reactive nitrogen data in Germany and therefore is a valuable database for policymaking and scientific activities. However, the quantification of the flows and the closure of the NNB for Germany, i.e. balancing the inflows and outflows, like the other cited NNBs, is characterized by a high degree of uncertainty. Especially the closure of the national budget or the budgets of the individual pools is not possible due to uncertainties in quantification of the numerous N fluxes, the sources of N_r emissions and their final sinks, and the changes in N stocks. In particular, further studies are needed on the magnitude of denitrification in soils and waters, which is the most important conversion process of N_r to N_2 in the biosphere. The possible accumulation of N in stocks (soil, water, products) is, in our opinion, overestimated in some studies and should be critically reviewed. Additionally, major deficits in the statistical recording of material flows can also be observed with regard to the German NNB, especially in production statistics and waste statistics. Without improvements in the statistical database, N flows in the *Material and Products in Industry* pool and the *Waste* pool cannot be captured reliably.

In terms of the EEA and EMEP (2013) initiative, it should be noted that the EEA and EMEP (2013) guidance on NNB calculation needs to be further harmonized and elaborated to facilitate future international comparability.

Data availability statement

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

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