Quantification of the German nitrogen cycle

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Abstract
Nitrogen and its compounds behave very differently in the environment. While atmospheric nitrogen is practically inert, the oxidised and reduced compounds are reactive. Human activity has led to massive changes to the natural nitrogen cycle over the past century and a drastic increase has been seen in the amounts of reactive nitrogen in the environment. Also in Germany the excessive release of reactive nitrogen compounds into the environment leads to a series of problems which must be urgently addressed. These include the loss of aquatic and terrestrial biodiversity, the impairment of air quality, the increased release of greenhouse gases, and constraints on the use of groundwater as drinking water. To improve knowledge on reasons for this situation we reviewed literature and databases to quantify the German nitrogen cycle. In Germany, some 4.2 million tonnes of reactive nitrogen enter into the nitrogen cycle annually, corresponding to some 50 kg per person. While considerable reductions have been achieved in oxidised nitrogen emissions from fossil fuel burning and also from wastewater management, reductions in the agricultural sector have been much less successful. The failure to meet targets is due in part to the fact that a comprehensive solution to the problems posed by nitrogen is hardly possible by implementing separate technical measures in individual areas. Therefore, it is necessary to adopt an integrated approach to the various problems in all relevant policy areas.

Key Words
reactive nitrogen, nitrogen budget, ammonia, nitrate, nitrogen oxide, nitrous oxide

Introduction
Nitrogen (N) can take various forms. Mostly it occurs in its molecular form as a relatively inactive gas in the atmosphere. However, it also occurs as reactive nitrogen in various compounds. Paradoxically, these reactive nitrogen compounds can be both essential nutrients and harmful pollutants. Over the past century, human activities have more than doubled the amounts of inactive atmospheric nitrogen being converted every year into reactive nitrogen (Fowler et al., 2013), and in Europe the amount has quadrupled (Sutton et al., 2011). This was either intentional, e.g. in order to produce artificial fertilisers and thus ensure the food production for the growing world population, or was an unintended by-product from the combustion of fuels.
In addition to the desirable effect of increasing agricultural production, the intensified nitrogen cycle today leads to many negative environmental impacts (Galloway et al., 2003). According to Rockström et al. (2009) and Steffen et al. (2015), the global boundaries of ecosystems have been exceeded by anthropogenous impacts on these cycles. International scientists therefore recommend a marked reduction in the conversion of atmospheric nitrogen into reactive nitrogen (Fowler et al., 2013; Rockström et al., 2009; Steffen et al., 2015). Discussions are ongoing to establish a target value which would ensure nutrition and at the same time remain within environmental boundaries. Recent contributions state that the global conversion of atmospheric nitrogen to reactive nitrogen should be limited to about half the current level (De Vries et al., 2013). At the German national level the German Advisory Council for the Environment (SRU) suggest reductions of German reactive nitrogen emissions at the same level of magnitude (Salomon et al., 2016; SRU, 2015).
In Germany, the intensified nitrogen cycle leads to unwanted effects at different scales and in relation to all environmental sectors. Several environmental quality and action goals such as concentration limits in hydrosphere and atmosphere or target loads and emission ceilings remain unattained (SRU, 2015). The continuously high deposition of reactive nitrogen to natural and semi-natural ecosystems leads to risks for the biodiversity especially for the vegetation adapted to nutrient poor conditions (Bundesamt für Naturschutz (BfN), 2004, 2012; Schaap et al., 2015). Air Quality is affected by reactive nitrogen through increased NO_2-concentrations at traffic related monitoring stations (Umweltbundesamt, 2016) and by increased secondary particulate matter as a result from agricultural ammonia emissions (Stern, 2013). Water quality is impacted negatively through increased nitrate levels in groundwater, which is an important source for drinking water in Germany (Bundesministerium für Umwelt & Bundesministerium für Ernährung, 2012). As a result of this,

the increased outflow of reactive nitrogen to coastal waters (LAWA, 2014) in combination with nitrogen deposition from short- and long-range nitrogen deposition (Bartnicki et al., 2013) leads to a unfavourable eutrophication status of coastal ecosystems. Effects on the climate have to be dealt with at the global scale. However nitrous oxide concentrations measured at German background stations show a continuous increase since 2001(Umweltbundesamt, 2015b).

Based on this situation we tried to quantify the German nitrogen cycle to identify main polluters, most important nitrogen flows and most promising intervention points. The quantification of a national nitrogen cycle or the inventory of a national nitrogen budget is a recommended tool for policy support under Geneva Convention on Long-Range Transboundary Air Pollution (Convention on Long-Range Transboundary Air Pollution (CLRTAP), 2013). This present study was part of a policy support of the German Environment Agency to the German Federal Ministry for the Environment forming the basis for a policy integration and an integrated solution with respect to the German nitrogen problem.

Methods

The quantification of the German national nitrogen budget was performed mainly as a review process based on data from statistical and research literature completed by data from inventories as a part of reporting obligations. Few numbers were computed following simplified approaches. The applied method is a national approach. It does not follow the standardized international procedure for setting up national nitrogen budgets as drafted by the Geneva Convention on Long-Range Transboundary Air Pollution as these guidelines had not been available by the time setting up the budget. However the general approach is very similar but less elaborated.

Nitrogen flows leaving or meeting pools of nitrogen such as industry and energy, transport, agriculture, natural and semi-natural ecosystems, waste and wastewater, atmosphere, hydrosphere and urban systems were put together. Nitrogen pools are elements in a nitrogen budget, which serve to store quantities of nitrogen. Exchange of nitrogen occurs between different pools via nitrogen flows.

Only flows amounting to more than 1 Gg/yr have been considered and rounded to entire numbers. Reviewed data is valid for a year between 2005 to 2010. If possible an average value for the years 2008 to 2010 has been calculated. Pooled nitrogen has not been quantified.

Results

The most important nitrogen flows in Germany are shown in Figure 1. About 4200 Gg of reactive nitrogen find their way into the national nitrogen cycle in Germany every year, either because molecular nitrogen is converted into a reactive form or because reactive nitrogen is imported. The most important inputs into the national cycle are in firsthand the industrial production of ammonia from atmospheric nitrogen for chemical production, accounting for approximately 2700 Gg N/yr (Verband der Chemischen Industrie e. V. (VCI), 2012). Secondly there are emissions of reactive nitrogen from the power industry, manufacturing, households, and the transport sector amount to approximately 440 Gg N/yr (Umweltbundesamt, 2012). The import of animal feed amounts to approximately 370 Gg N/yr (Bundesministerium für Ernährung und Landwirtschaft (BMEL), 2013). Biological fixation of nitrogen in agriculture (Bundesministerium für Ernährung und Landwirtschaft (BMEL), 2013) and in terrestrial ecosystems increase nitrogen availability by about 275 Gg N/yr. Finally the transboundary import of nitrogen within rivers is as big as 320 Gg N/yr (Fuchs et al., 2010; Umweltbundesamt, 2014). Within the atmosphere it amounts about 250 Gg N/yr (Fagerli, 2012). Important flows within the budget are the 890 Gg N applied to agricultural areas every year as manure and the national sale of chemical fertiliser of 1640 Gg N/yr (Bundesministerium für Ernährung und Landwirtschaft (BMEL), 2013), which corresponds to most of the industrially fixed nitrogen. 665 Gg N is available in agricultural products for food consumption or for the production of food products.

Quantifications of the nitrogen cycle are also available for the global and the European levels (Fowler et al., 2013; Sutton et al., 2011). A comparison of the estimates for the entry of reactive nitrogen into the cycle in Germany, with the European and global levels (Table 1) shows that natural processes make a much greater contribution to the nitrogen cycle at a global level, due to the large areas covered by oceans, than they do in Europe and Germany, where anthropogenous effects are more intensive.

In addition to the intensity of the nitrogen cycle, the emissions of reactive nitrogen are an important parameter in view of the direct relationship to the effects. Table 2 presents the average annual emissions in Germany for the most important nitrogen compounds and the key emitting groups. Table 3 shows that agriculture has meanwhile become the most important sector for the release of reactive nitrogen into the environment. Due to the greater reductions achieved in other sectors, the relative share of remaining emissions caused by agriculture increased over the past 20 years, despite the fact that measures adopted to reduce agricultural emissions have shown some effects. In the 1990s, less than half of the total emissions of reactive nitrogen compounds were from agriculture (Eichler & Schulz, 1998).

Figure 1: Key flows of reactive nitrogen in Germany [Gg N/yr] aggregated to total nitrogen and given in rounded values of 5

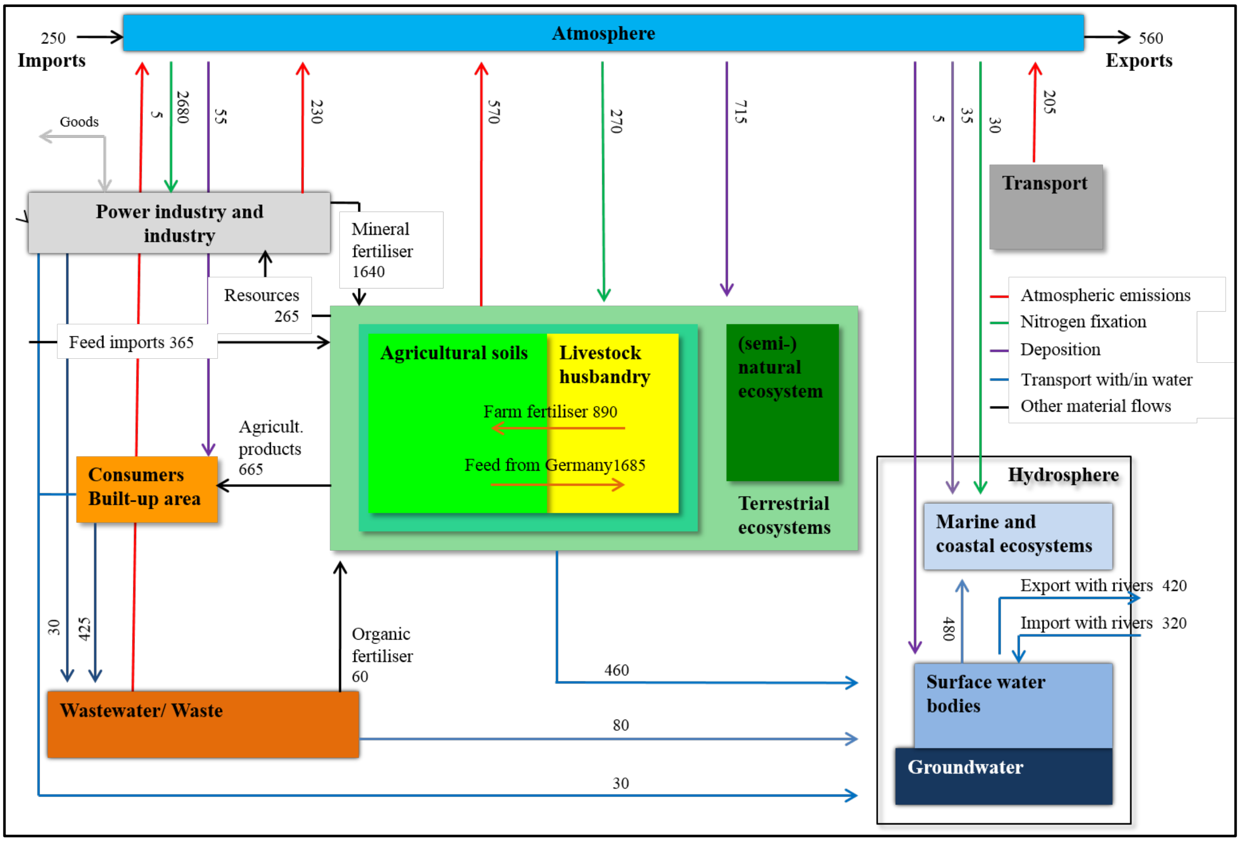

Table 1: Total annual input of reactive nitrogen into the nitrogen cycle at different scales [Tg N /yr] by different natural and human activities and their share to the total fixation (in brackets)

		Global	Europa (EU-27)	Germany
Biological fixation	Terrestrial ecosystems	58.0 (14.1 %)	0.3 (1.3 %)	0.1 (2.6 %)
	Lightning	5.0 (1.2 %)	n. q.	n. q.
	Marine ecosystems	140.0 (33.9 %)	0.5 (2.2 %)	n. q.
Anthropogenous fixation	Haber Bosch	120.0 (29.1 %)	16.6 (73.5 %)	2.7 (71.1 %)
	Biological fixation in agriculture	60.0 (14.5 %)	1.0 (4.4 %)	0.2 (5.2 %)
	Fodder imports	n.q.	0.5 (2.2 %)	0.4 (10.5%)
	Emissions related to fossil fuel combustion	30.0 (7.3 %)	3.7 (16.4 %)	0.4 (10.5 %)
	Total fixation	413.0 (100.0 %)	22.6 (100 %)	3.8 (100 %)
	area [ha]	51,007,200,000	430.000.000	35.700.000
	Area related [kg ha^{-1}]	8	53	103

Table 2: Average annual emissions of the main N-compounds affecting atmosphere and hydrosphere in the current budget period by different sectors [Gg/yr] and their shares to the total emissions [%]

	NO_x	NH_3	N_2O	NO_3^-/NH_4^+	Total	Share [%]
Agriculture	33	435	88	424	980	63
Traffic	192	13	2		207	13
Industry and energy	166	15	27	10	218	14
Wastewater / Surface runoff	21	1	6	135	163	10
Total	412	464	123	569	1568	100
Share [%]	26	30	8	36	100	

Table 2: Development of sector related annual nitrogen emissions [Gg N/yr] and their share to the total emissions [%] over three budgeting periods in the last 20 years

	1995		2000 – 2004		2005 – 2010	
	(Eichler & Schulz, 1998)		(Umweltbundesamt, 2009)		(Umweltbundesamt, 2015a)	
	Emission	Share	Emission	Share	Emission	Share
Agriculture	1330	48 %	1055	57 %	980	63 %
Transport	595	22 %	270	15 %	207	13 %
Industry and energy	354	13 %	244	13 %	241	15 %
Wastewater / surface runoff	474	17 %	270	15 %	140	9 %
Total emissions	2753		1839		1568	

Discussion

A thorough review analyses of statistical literature, literature on reporting obligations and research literature from modelling has been performed. Uncertainty of statistical and reported data is small to acceptable. Larger uncertainties go along with modelled data. However, there remain data gaps that hasn't been able to close. Especially the removal of reactive nitrogen from the cycle has not been sufficiently recorded. While it is estimated that some 900 Gg N of dissolved nitrogen compounds are introduced annually into coastal ecosystems via rivers (Fuchs et al., 2010), some 560 Gg N are transported transboundary via the atmosphere (Fagerli, 2012) and about 340 Gg N/yr are released into the atmosphere as molecular N_2 after denitrification in wastewater treatment (Deutsche Vereinigung für Wasserwirtschaft, 2011), denitrification and retention in surface waters, in agro-ecosystems, in natural and semi-natural ecosystems and in groundwater we could only approximately estimate based on current data. Older evaluations show that, depending on the distance flowed and other factors, retention (and in particular denitrification) can account for up to 50 per cent (Fuchs et al., 2010). This goes along with the difference of approximately 750 Gg N/yr based on the comparison between the nitrogen surplus in the agricultural sector (Bundesministerium für Ernährung und Landwirtschaft (BMEL), 2013) and the reviewed inputs from agriculture into atmosphere and surface waters, of which a large part is probably denitrified or immobilized. However, this estimate represents only a first approximation because data from various sources are being combined, and the possible accumulation of reactive nitrogen and the time-delay for the passage into the groundwater has not been taken into account. Furthermore, a number of industrial products also contain reactive nitrogen. According to first estimates about 10 kg reactive nitrogen per person per year are used for the production of these products (Gu et al., 2013; Sutton et al., 2011). In Germany, every year nearly 2700 Gg N is industrially fixed and only 1640 Gg N is marketed as chemical fertiliser. The remaining 1000 Gg N/yr (about 12.5 kg N per person per annum) are used for further industrial processing. However, the material flows of the reactive nitrogen contained in these products (international trade, product storage, waste disposal, recycling) have so far hardly been documented. Finally the transboundary trade of manure and of biomass as fermentation substrate is not contained in the current budget. Over the observation period this probably represents an omission of at least 20 Gg N/yr. Flows of plant fermentation residues within the agricultural sector are also not taken into account, yet. Closing the above mentioned data gaps would certainly lead to larger robustness of the performed quantification of the German nitrogen cycle. Especially improved quantification of the fate of reactive nitrogen being introduced into the nitrogen cycle would strengthen validity of the budget approach. However with the distilled information on inputs to the nitrogen cycle and emissions to the environment, a very valuable set of information is at hand to further develop nitrogen related policies in Germany.

Conclusion

It could be shown, that observed nitrogen induced environmental effects in Germany go along with an intensified nitrogen cycle. Nitrogen is emitted to the environment in similar shares as NH_3, NO_x and NO_3. A minor part is emitted as nitrous oxide. One third is released to the hydrosphere, two thirds are released to the atmosphere. Though agriculture with nearly two thirds of the emissions is the dominant polluter while all the other sectors contribute equally to the problem, the results of the quantified nitrogen cycle highlight that the problem is complex. It underpins significantly that an integrated approach, equally addressing the reduction of different gaseous and dissolved nitrogen compounds in the different interlinked sectors at the same time is necessary and has the potential to avoid pollution swapping. However from our point of view in the first place it is tremendously important to pick up the low hanging fruits and to use the existing reduction potential in agriculture.

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