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Estimating environmentally relevant fixed nitrogen demand in the 21st century

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Summary

Human activities affect the impact of the nitrogen cycle on both the environment and climate. The rate of anthropogenic nitrogen fixation from atmospheric N₂ may serve as an indicator to the magnitude of this impact, acknowledging that relationship to be effectdependent and non-linear. Building on the set of Representative Concentration Pathway (RCP) scenarios developed for climate change research, we estimate anthropogenic industrial nitrogen fixation throughout the 21st century. Assigning characteristic key drivers to the four underlying scenarios we arrive at nitrogen fixation rates for agricultural use of 80 to 172 Tg N/yr by 2100, which is slightly less to almost twice as much compared with the fixation rate for the year 2000. We use the following key drivers of change, varying between scenarios: population growth, consumption of animal protein, agricultural efficiency improvement and additional biofuel production. Further anthropogenic nitrogen fixation for production of materials such as explosives or plastics and from combustion are projected to remain considerably smaller than that related to agriculture. While variation among the four scenarios is considerable, our interpretation of scenarios constrains the option space: several of the factors enhancing the anthropogenic impact on the nitrogen cycle may occur concurrently, but never all of them. A scenario that is specifically targeted towards limiting greenhouse gas emissions ends up as the potentially largest contributor to nitrogen fixation, as a result of large amounts of biofuels required and the fertilizer used to produce it. Other published data on nitrogen fixation towards 2100 indicate that our high estimates based on the RCP approach are rather conservative. Even the most optimistic scenario estimates that nitrogen fixation rate will remain substantially in excess of an estimate of sustainable boundaries by 2100.

Key words:

Reactive nitrogen, scenarios, projection to 2100, environmental impact, climate change, nitrogen fixation

1. Introduction

Human alteration on the natural cycle of nitrogen has long been recognized causing major environmental impacts (Galloway et al. 2003, 2008; Elser, 2011; Sutton et al. 2011a). Anthropogenic activities are able to fix atmospheric di-nitrogen (N₂) either as a side effect (in high-temperature combustion processes) or with the purpose to produce nitrogen compounds, first of all to be used as agricultural fertilizers. Both the industrial activity (the Haber-Bosch process) and the "biological nitrogen fixation" (BNF) by agricultural cultivation of leguminous plants need to be regarded as human activities. Fixed or "reactive" nitrogen (N_r) comprises all forms of nitrogen except the unreactive gas N₂. Upon its release into the environment, e.g. after application of fertilizers in agriculture or emission of combustion byproducts, it causes a cascade of negative effects, ranging from local (smog) to regional (such as acid deposition, terrestrial and aquatic eutrophication) and to global scales (climate change, stratospheric ozone depletion). Rockström et al. (2009) listed this anthropogenic extension of the nitrogen cycle as one of the key global environmental challenges for maintaining human "operating space". Already the present level of anthropogenic nitrogen fixation substantially exceeds their estimate of a sustainable planetary boundary.

Nitrogen is closely linked to food production and the "green revolution" (Pimentel et al. 1973; Tilman 1998), being an essential component to improve agricultural productivity. The increased productivity has nourished a growing world population, despite only modest change in the global area of agricultural land. In contrast to many of the other challenges posed by Rockström and colleagues, policy efforts to curb nitrogen pollution have been limited to a regional scale. Current policy efforts recognize nitrogen pollution indirectly at the global level, as contributing to greenhouse gas formation (specifically in the form of nitrous oxide, but interacting also in many other ways: see Butterbach Bahl et al. 2011) and for endangering

biodiversity (e.g. Bleeker et al. 2011), both of which are topics for which global conventions have been forged.

Scientific evidence for the role of nitrogen compounds in climate change is available in the literature (e.g., Forster et al. 2007). Fig. 1 compares current radiative forcing, the increments in the atmosphere derived from observed concentrations, and emissions from anthropogenic sources (the latter two normalized by the "global warming potential" over 100 years, GWP) of N₂O and CO₂. Following data available from the EDGAR emission database (version 4.2, <u>http://edgar.jrc.ec.europa.eu</u>) fertilizer related emissions comprise about two thirds of all current N₂O emissions (assuming that also much of nitric acid produced is used in fertilizers). Climate related effects extend beyond N₂O (Butterbach Bahl et al., 2011), including NO_x-triggered formation of ozone as well as the formation of particles from ammonium- or nitrate-compounds. Thus, for the purpose of this paper, we focus on an indicator, nitrogen fixation, rather than on an individual compound.

Addressing the future challenges created by nitrogen release to the environment needs to consider the basis of expected developments, especially the main drivers of food production and fuel combustion. Without further intervention, e.g., more stringent laws limiting emissions or deposition of N compounds, it is foreseen that nitrogen will remain to cause important impacts, including economic costs associated with environmental damage as has been quantified by Sutton et al. (2011b) and the U.S. Environmental Protection Agency Science Advisory Board (USEPA-SAB, 2011) for Europe and the U.S., respectively.

This paper addresses the challenge to project the future of global anthropogenic nitrogen fixation as follows: In section 2, we will describe different approaches to develop environmental scenarios. Section 3 focusses on the methods applied to evaluate nitrogen-

related developments over the 21st century based on scenarios prepared for IPCC's 5th assessment report, the "representative concentration pathways" (RCPs). In section 4 we present the results and discuss their implication with respect to other available work before concluding in section 5.

2. Environmental scenarios

Scenarios have long been used successfully to provide scientifically based development options on environmental issues. The main reason for creating such scenarios is to support present decision making rather than to look into the future. Thus, scenarios typically are not limited to one instance of a future development, but instead allow for a variety of potential fates. Evaluation is performed along the differences between available different scenarios ("possible futures") and of course against a current situation.

One of the first exercises to develop long-range global environmental scenarios was the "Limits to Growth" (LtG) report prepared for the Club of Rome (Meadows et al., 1972). Key scenarios contained in this report are i) a "standard run", reflecting a continuation of present business-as-usual behavior from the time of scenario development; ii) one case of "comprehensive technology", providing technological solutions for any challenges to shift environmental problems into the future as much as possible, and iii) a case of a "stabilized world", which deliberately attempts to achieve equilibria for key parameters.

An approach to compare a "reference" situation (e.g. based on current legislation which may become effective in the future only) to one "with action" is also taken in shorter scale scenarios on air pollution (see Winiwarter et al., 2011). As scenario development at the same time is linked to mitigation of adverse effects (hence "effect based" scenarios are developed here) the need for the development of more stringent abatement scenarios may arise when reduction targets are not yet achieved ("with improved action").

The scenarios prepared for the Special Report on Emission Scenarios (SRES) by the Intergovernmental Panel on Climate Change (IPCC: Nakicenovic and Swart 2000) further develop the LtG approach, by differentiating two sets of parameters along two extremes, i.e., global vs. local trade patterns, and development vs. sustainability orientation. These authors used storylines to represent a consistent set of future events which cover also the potential socio-economic development. The approach provided the basis to establish four families of scenarios that were used in IPCC's Fourth Assessment Report, then with different integrated assessment models yielding a set of results for each of the scenario types "A1, "A2", "B1" and "B2", where the "A" scenarios refer to a development orientation, while the "1" scenarios assume global dissemination ("B" and "2" referring to the respective opposite). Similar approaches have i.a.been used for the Millennium Ecosystem Assessment (MEA: Carpenter et al 2005).

These scenario categories build on a "line" of events, so-called storylines. Storylines comprise the socio-economic backdrop that constitutes the economic development as well as the boundary conditions of technological changes. The linear build-up means that dramatic system breaks caused from outside the modeling system cannot be identified. Also short term fluctuations as from variations in economic growth would not emerge when the storylines use average growth rates as a basis. This means that variations on a short time scale between a scenario and an actual development may also occur, which do not invalidate the results.

For the next generation of climate scenarios in IPCC, a scheme was devised to first provide input data to global circulation models (GCMs). This input for the first time

considered emission mitigation scenarios incorporating the result of global climate policies assumed to be in place later this century. The "parallel process" (Moss et al. 2010) would allow two time consuming activities organized simultaneously, the computer runs of the GCMs projecting the global climate conditions into 2100, and developing the storylines for the future socio-economic conditions.

The first part of this approach has led to the development of so-called "Representative Concentration Pathways" (RCPs), which use a nomenclature indicating the radiative forcing exerted in the year 2100 (e.g., RCP8.5 resulting in additional anthropogenic forcing of 8.5 W/m²). Four such RCP scenarios have been developed, each based on a different integrated assessment model, and each with their own set of input assumptions that were not harmonized between models, but rather based on pre-existing information within the respective model (van Vuuren et al., 2011a). Meant as an input to GCMs, the level of radiative forcing seemed sufficient as a describer, so these sets do not contain coordinated storylines describing the socio-economic pathways, and knowledge on the philosophy underlying the scenarios is rather limited.

The parallel development of the socio-economic storylines is leading to a complementary set of scenarios termed the Shared Socioeconomic Pathways (SSPs: van Vuuren et al., 2012), a process not yet completed. Merging of the RCPs and SSPs will only be performed at a later stage, but it is expected that the RCP-based runs of global circulation models can be matched to the specific SSPs. Currently that is not yet possible. The disadvantage of delayed availability of coherent emission scenarios and socio-economic pathways is more than compensated by being able to feed the results of the GCMs back into the IPCC process in time to meet other operational needs of that process (Moss et al., 2010).

Each RCP derives atmospheric concentration patterns in line with the prescribed forcing values, and then estimates the emission patterns that are consistent with these atmospheric concentrations. In such a development the obvious focus is on providing an adequate representation on CO₂ emissions and sinks – basically energy and land use, because of its dominant contribution to climate forcing. The nitrogen cycle is covered in the analysis as much as it is considered influential on radiative properties of the atmosphere, but only to supplement the information provided to the carbon cycle (see van Vuuren et al., 2011b). While recognizing these limitations, the RCP scenarios are of specific interest not only as they represent the most recent set of scenarios, but also as they (together with SSPs) have been prepared as an input to IPCC's Fifth Assessment Report.

3. Extending global scenarios to nitrogen

In this paper, we analyze available information on the assumptions underlying the RCP scenarios in order to extrapolate the rates of nitrogen fixation throughout the 21st century. As mentioned above, we use nitrogen fixation as a more general indicator to represent different compounds and effects. While some non-linearities will arise (e.g., so that the indicator is not proportional to an effect in question), we believe the approach provides an informative basis to consider the future environmental impact of nitrogen.

Consistent with the concepts developed by Erisman et al. (2008), who estimated the development of N fixation in agriculture starting from the SRES scenarios, we assign five basic drivers to be used as archetypes of future change, and then analyze RCP scenarios whether a specific driver seems applicable. This will only in part reflect the assumptions contained within the respective RCP estimates, but will make them comparable in terms of N fixation.

Table 1 compares the respective scenario concepts of the RCP papers. With much of the storyline information of the SSPs not yet available, looking into the external population development scenarios that have been used in the RCPs may hint on the socio-economic conditions used to establish the scenario. While this interpretation possibly extends beyond the considerations of the RCP authors, at least it provides a consistent way of treating the respective scenarios. Table 1 also provides the respective suggestions which other driver influencing N consumption may be applied on what scenario. These drivers are presented individually below, while the Supplementary Material explains in detail the algorithms applied.

We start at a mineral fertilizer nitrogen demand of 94.2 Tg N/yr for 2005, for a world population of 6.5 billion (UN, 2007), and scale the population-dependent agricultural nitrogen fixation according to the population projections linked to the respective scenarios. Using data of industrial nitrogen fixation only as indicator necessarily neglects the more uncertain estimate of BNF, which we assume to be covered implicitly and to proportionally follow the trends of our indicator.

While external population projections (consistent with RCPs, see above) are used as a first proxy to nitrogen, we use four more major factors of influence. Depending on the respective development scenarios, these factors may or may not need to be considered and this interpretation adds the "storylines" to the scenarios, which we start with the year 2000. These factors (described in detail in the Supplementary Material) specifically are:

Efficiency increase: extrapolation according to population neglects agronomic changes that may occur over time. Here we use the Nitrogen Use Efficiency (NUE) of agricultural soils as defined by OECD (2008), being the ratio of N removed in crops divided by the N applied to soil in all forms, to indicate such economic changes. We implement an increase in efficiency as a relative reduction of nitrogen demand by 0.5% each year, until a level of NUE at 66.6% has been reached at which point improvement is assumed to halt. Improved efficiency is assumed to occur for all food production in each of the RCP scenarios, but not for biofuels (see below).

<u>Food equity</u>: This option assumes diet improvements in large parts of the world which now lack of sufficient animal protein. We set the level of European consumption of animal protein as the standard to be achieved globally by 2100. Animal production requires feed production, which in turn needs to be driven by mineral fertilizer. At the assumptions given, an increase of mineral fertilizer consumption of 69% would occur progressively to materialize fully at the end of the scenario (year 2100). Food equity is assumed to be consistent only with the globalized and environmentally considerate scenario underlying RCP4.5.

Diet optimization: Efficiency of N conversion is different in different animal systems. If human diets are made up from animal products that more efficiently make use of N, this will decrease the amount of nitrogen needed to produce the animal protein. In consequence the need for animal feed decreases as well as the nitrogen demand. We estimate diet optimization may allow a 12% decrease in mineral fertilizer by 2100. Also we understand "diet optimization" to be consistent with all SRES type "B" scenarios (sustainability oriented) and apply it in all RCP scenarios except for RCP8.5.

<u>Biofuels</u>: Increased production of biofuels will require additional nitrogen fertilizer to maximize the outputs on limited area. The amount needed will depend on the climate, soil conditions and the agricultural practice implemented. Little experience is available regarding optimized fertilizer levels because fertilizer inputs are not taken into account in biofuel

policies. Furthermore, it has not been assessed what the optimal fertilizer uses should be for the energy crops grown to produce second generation biofuels. Tilman et al. (2006) report biofuels production in principle is possible without fertilization – but that may be unrealistic when attempting to produce biomass quickly. We account for substantial additional biofuel production in RCP2.6 only, and also derive the underlying N_r demand from the RCP literature (see Supplementary Material for details).

4. Results and discussion

While population projections as drivers are based on the intrinsically provided numbers for each RCP scenario, for all other drivers we only distinguish whether they are applicable or not applicable. We do not test the intermediate option space (e.g., half of the efficiency increase as stated). It may be argued that such additional assumptions would more closely reflect a probable future condition, but for the purpose of this paper we believe it is more interesting to build on these characteristic features as archetypes to explain the direction of developments.

The resulting trajectories of anthropogenic N fixation in agriculture are our interpretation based on the RCP scenarios (Fig. 2). On the left panel, the temporal development over a 200 year period is shown for the respective RCP scenarios, while the right panel disassembles the totals for 2100 into the respective drivers for each of the scenarios. Moreover, alternate estimates from the literature of fixed N generation in agriculture (displayed as dots or asterisks for specific years) are compared with the curves of temporal developments.

The largest population – and the corresponding need for nitrogen – is associated with RCP8.5. At more than 12 billion inhabitants, in 2100 the world accommodates about twice as many people as at the beginning of this century. The expected improvements in nitrogen use

efficiency in the order of 60 Gg N or half of the final estimate limits the extension of the nitrogen cycle. The population influence is much smaller for RCP6.0, as this scenario (as with the two remaining scenarios) projects an increase to 9.5 billion inhabitants only. Moreover, RCP6.0 benefits (again like the other two scenarios) from an improved lifestyle which permits consumption of less animal protein and thus decreases nitrogen demand. RCP6.0 ends up at an anthropogenic impact on the N cycle slightly smaller than today, and is the lowest estimate for the year 2100. Since it depends on substantial improvement in NUE, diet optimization and limited increase in human population, this may be considered as the most optimistic of the scenarios in regards of N impacts.

The two final scenarios, which are those scenarios that extend furthest in climate mitigation, are both associated with elements of additional nitrogen application. For RCP4.5 we expect "food equity", i.e. better protein supply for most of the world, would require a considerable extension in fertilizer nitrogen for availability of animal feed. The increase is somewhat lessened due to efficiency improvements and better diets which also affect the additional nitrogen applied. In consequence the impact on the nitrogen cycle is very similar to RCP8.5, the scenario with the largest population. For RCP2.6 biofuel production leads to the additional N needs. In line with the descriptions by van Vuuren et al. (2010) we do not assume any efficiency improvements. The evolution of nitrogen fixation, with a peak around 2025, reflects the assumed change from first generation biofuels (which need much more nitrogen) to second generation biofuels, while biofuel demand increases continuously. While van Vuuren and coworkers argue that the additional greenhouse gases (N₂O) released due to cultivation of second generation biofuel crops are small compared to the savings in fossil CO_2 , the impact of biofuel production on the nitrogen cycle would be significant, as has also been pointed out by Davidson (2012). The biofuel demand drives this scenario to become the largest in terms of N fixation. As an interesting note, also third-generation biofuel production,

biofuels from algae, has been associated with considerable additional nitrogen demand (Wijfels and Barbosa, 2010).

Our interpretation of N fixation for the RCP's seems to differ to some extent to the original RCPs' published N₂O emission data (e.g. as displayed by Riahi et al., 2011), with RCP8.5 providing highest and RCP2.6 lowest global N₂O emissions in 2100. We conclude that the original RCP8.5, in their baseline, may not even have considered efficiency increase. Thus our interpretation of future N fixation may be considered rather a low estimate. For RCP2.6 and biofuels, where we actually apply closely the authors' understanding of N demand, the difference indicates nitrogen being moved into a different environmental pool.

In order to understand scenario limitations in general, we look into an available retrospective analysis of scenarios. The "Limits to Growth" (LtG) is the only set of long-term environmental scenarios established early enough to allow for current investigation of scenario performance. Analysis of the first 40 years until 2010 (Turner, 2012) suggests that:

- For many key parameters (population, food availability, industrial output, nonrenewable resources, pollution) the real development seems to follow the LtG "standard run" reasonably closely.
- The expectation of a general environmental improvement, as a consequence of perceived damage and political action, seems not evident, at least at the global scale. This is in contrast to the well-known "DPSIR" concept fostered by the EEA (EEA, 1998), bearing the assumption that environmental policies as a response to observed impacts promote environmental improvement. This assumption of improvement typically represents the central rationale in effect-oriented scenarios (see Winiwarter et al., 2011).

- The effective growth limits in this "standard run" scenario are set to appear around the year 2020 in LtG, which is beyond the time range so far considered. Therefore, even while a considerable stretch of the overall scenario period can be compared with real data, the striking path changes of the growth limits cannot. Thus no validation of the most important scenario conclusion is yet possible.
- Any difference in the timing of a systems transformation event between scenario and observation could not disprove the general assumption of the LtG approach. The general concept of a growth limitation may still remain valid even if the actual effects occur somewhat later than anticipated 40 years ago.

Considering the nitrogen scenarios of Fig. 2, we note the difficulty in exact interpretation of the scenario timelines. We therefore focus on comparing the ranges between the scenarios developed and the differences to alternate estimates by other authors. The overall spread of scenario results is almost a factor of 2, which is clearly larger than the range of population projections, indicating that the future N demand later this century will more strongly depend on agricultural practice and the use made of agricultural products than population alone. While the underlying scenarios differ, the range of results obtained by Erisman et al. (2008) for the year 2100 is quite close to the one presented here. This indicates an obvious relatedness of the assumptions taken, even if the earlier publication refers to SRES scenarios.

An alternate interpretation of SRES scenarios has been presented by Bodirsky et al. (2012), whose lower estimates range close to the central RCP-based estimates of this paper both in 2100 and in 2050. Much higher impacts to the global nitrogen cycle are seen in their upper end. This may indicate that, in contrast to our work, their model provides little feedback of parameter changes within the system, while in our work we argue that high N use in one of our elements causes a high probability that N use becomes low for the other elements, thus

moderating any excessive (but also any extremely low) N use. This is a consequence of our interpretation of storylines, which implicitly or explicitly take account of other limitations such as area competition.

Further available developments of fertilizer application, while only extending towards 2050, derive from a refined extrapolation of past trends. Especially of interest is the latest projection developed by FAO (Alexandratos and Bruinsma, 2012) which takes into account recent developments of fertilizer consumption. In contrast to the assumptions developed here, Alexandratos and Bruinsma argue that developing countries will still strongly increase their fertilizer needs, based on these authors' experience over the recent years after 2000. Thus the FAO projection (as well as a much earlier one by Tilman et al., 2001) indicate there still may the possibility for even larger impact on N cycles, such that our approach represents a rather moderate and conservative result, consistent with our assumption of an anticipated improvement in NUE. By contrast, estimates performed within the Millennium Ecosystems Assessment (Bouwman et al., 2009) consider a smaller impact and lower nitrogen fertilizer demand by 2050, since they assume human excreta would in future also be reclaimed as nutrients for agricultural purposes. The difference between these scenarios illustrate the substantial potential that future policies may have in achieving improvements in NUE and in recycling of all available Nr resources.

In order to extend the indicator of anthropogenic nitrogen fixation beyond fertilizer N, Fig. 3 presents the fixation contributions of fertilizer N, combustion NO_x -N and other reactive nitrogen fixed in 2000 vs. the respective figures in 2100 for the different RCPs. In this case "other N" for 2000 estimates the difference to the anthropogenic subtotal (Galloway et al. 2008, provide data for 1995 and 2005 which we interpolate), which covers N used in industrial practices including plastics and explosives as well as cultivation-induced BNF. The

mineral fertilizer N in Fig. 3 derives from previously described assumptions, while estimated NO_x emissions are directly taken from the RCP database (version 2.0.5,

<u>http://www.iiasa.ac.at/web-apps/tnt/RcpDb/dsd?Action=htmlpage&page=compare</u>). Note that no scenarios are available for "other N", which includes industrial use that at least in 2000 contributed a smaller amount only (Winiwarter et al., 2011).

It can be seen that all of the RCP scenarios assume successful NO_x abatement in the future at a global scale. By comparison, the contribution of agricultural N tends to rise, continuing as the largest anthropogenic N impact. While NO_x emissions and to a large extent also fertilizer additions to soil are clearly released to the environment, this may not be the case for the industrial products orBNF contained in "other N" in Fig 3. Thus the environmental impact of that part of fixed N may be much smaller than assumed from using the indicator. In consequence the difficulty in projecting "other N" might not strongly affect our understanding of environmental impacts.

In this paper we operate on global averages only, acknowledging that considerable regional differences exist. The major part of mineral fertilizer use and thus also of the Nr impact occurs in regions of easy access to fertilizers. Here improvement of NUE will be possible. In contrast, there are areas in the world, which lack of fertilizer availibility and in which agricultural improvement to nourish the growing population will most probably lead to decreasing NUE (Bodirsky et al 2012 provide regional figures also on scenarios). Still for the global situation, areas of high N_r use will weigh significantly stronger, for which NUE increase is realistic. Considering the experience of LtG, however, there is a possibility that such improvements will just not materialize. In case NUE efficiency does not improve, nitrogen fixation rates as shown in Fig. 2 for agriculture would increase to 120-210 Tg N in 2100, and total N_r fixed by anthropogenic activities as shown in Fig. 3 may get as high as 200

to 300 Tg N per year, assuming also the foreseen NO_x emission reductions do not work out. This may be a matter of policy implemented, and again policies may differ strikingly on the regional scale. So any emission reductions or efficiency improvements may work generally, not at all, or in larger or smaller parts of the world. In the approach used here we believe efficiency improvements can happen generally. These reflections indicate that the nitrogen fixation rates used as indicators in Figs. 2 and 3, while exhibiting a tendency to increase, provide a rather careful and conservative view of the future situation.

5. Conclusions

Assessing the future rates of nitrogen fixation provides fundamental information on potential environmental effects of fixed nitrogen. Water quality and eutrophication, soil quality, air quality, biodiversity and climate change are all issues that have been clearly brought into connection with excess reactive nitrogen (Sutton et al., 2011a). Taking advantage of the scenarios used as RCPs and providing our own interpretation of some of the nitrogenrelated consequences of these scenarios, we obtain a considerable range of plausible future anthropogenic contributions to the global nitrogen cycle. None of these markedly reduces the human impact from the current condition. Based on our interpretation of the RCP2.6 and 4.5 scenarios, a doubling of nitrogen fixation for agricultural purposes seems a realistic possibility, especially if the improvements in nitrogen use efficiency assumed in the scenarios are not achieved.

The range presented for agricultural nitrogen using the RCP scenarios is similar to that developed for the earlier SRES scenarios (see Erisman et al., 2008). While this range is larger than the range of population projections used in the underlying scenario, it is smaller than one might expect from looking at all of the respective elements leading to change. Here we understand high-nitrogen cases will not all occur simultaneously, but rather exclude each

other as a consequence of perceived or modeled (in the referenced work) resource limitations, thus moderating any differences between scenarios. Other interpretations that do not have this restriction, or projections that are more strongly based on extrapolation of current trends, extend their ranges of nitrogen impacts to considerably higher values. So the result presented here seems to represent rather conservative and optimistic assumptions.

One specific aspect influencing the nitrogen cycle is the influence of agricultural production increases. Despite of possible optimization, a production increase will more likely be coupled also with increased N demand. Even if, e.g., biofuel production of second generation biofuel can be performed very efficiently improving the greenhouse gas balance, its effects on the nitrogen cycle may remain considerable.

Agricultural nitrogen trends, as presented here, do not rely on distinct measures describing a specific way of abatement. Rather, measures are incorporated in the overall assumption of improved NUE. By contrast, for N fixation due to combustion, the available technical fixes are more specific and have been used in the projections. In consequence, combustion related N is assumed to decrease in all scenarios. However, the LtG ex-post analysis indicates that improvements required and expected as a consequence of observed pollution may not always occur, which may apply both for implementation of low emission NO_x technologies and improvements in agricultural NUE. If in contrast to the scenarios shown in Figure 3, the expected improvements for NO_x and NUE were not achieved, then the total reactive N fixation in the four scenarios for 2100 could be as high as 200-300 Tg N/yr.

Expectations regarding the future of the nitrogen cycle in the 21st century therefore range from a slight overall decrease of the anthropogenic impact to a strong increase. Despite the nitrogen related problems already experienced, we need to expect the situation to deteriorate

rather than to improve. This is the result of a rather cautious and optimistic approach to estimate future directions of anthropogenic nitrogen fixation.

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Wijffels RH, Barbosa MJ (2010) An Outlook on Microalgal Biofuels. Science 329: 796-799

Winiwarter W, Hettelingh JP, Bouwman AF et al. (2011) Future scenarios of nitrogen in Europe. In: Sutton MA, Howard CM, Erisman JW, Billen G, Bleeker A, Grennfelt P, van Grinsven H, Grizzetti B (eds) The European Nitrogen Assessment, Cambridge University Press, Cambridge, UK, pp. 551-569 Table 1: Comparison of the Representative Concentration Pathway (RCP) scenarios with the IPCC Special Report on Emissions Scenarios (SRES), including comments on the relationships with nitrogen fixation.

RCP name	RCP reference	Population projection	Interpretation of storyline and relationships to nitrogen scenarios
RCP8.5	Riahi et al. (2011)	Strong population growth scenario as in SRES A2, but revised projections according to Grübler et al., 2007	Development oriented, regionalized scenario (A2r) will not attempt to improve the diet in the western world; a nitrogen efficiency increase is needed to feed the strongly increasing population, but there will be no incentive to "food equity".
RCP6.0	Masui et al. (2011)	Population following UN (2007) before 2050, and then trends from UN (2004)	Updated SRES B2 scenario includes climate policy intervention. Environmentally considerate "B"-type scenarios (B1, B2) all include diet optimization for the overfed rich countries.
RCP4.5	Thomson et al. (2011)	Population as listed by Clarke et al. (2007) from UN (2005) before 2050, thereafter following O'Neill (2005)	Stabilization scenario following a "Techno-Garden" millennium ecosystem assessment scenario (globalized, environmentally considerate storyline). We assume poor countries better supplied with food nitrogen, strongly increasing N release to the environment.
RCP2.6	van Vuuren et al. (2011c)	Population taken directly from UN (2004)	Based on IMAGE 2.4 B2 scenario (van Vuuren, 2010). Minimizes fossil carbon use and thus is strong on biofuels – we extrapolate fertilizer N application from the biofuel-induced N_2O emissions reported.



Fig. 1: Comparison of the contributions of N_2O vs. CO_2 to radiative forcing (Forster et al., 2007), their increment in the atmosphere derived from concentration increase (Forster et al., 2007) and the anthropogenic emissions to the atmosphere (EDGAR vs. 4.2).



Fig. 2: Global agricultural demand for industrial N fixation (Tg N/yr), projected till 2100. Lines in the left panel reflect trends attributed in this paper to the respective RCP scenarios, while dots and asterisks show other assessments. The asterisks express the ranges (maximum and minimum) out of several scenarios based on storylines, with Erisman et al. (2008) using a methodology very similar to the one applied here for RCPs. Global population numbers used for 2100 are 12.4 billion, 9.34 billion, 8.6 billion and 9.06 billion (RCP8.5, RCP6.0, RCP4.5 and RCP2.6, respectively).



Fig. 3: Amounts of N fixed due to anthropogenic activities comparing estimates for 2000 with estimates based on different RCPs in 2100 (Tg N/yr). Combustion NO_x is taken from RCP directly, amounts of "other N" are shown as fade-out bars for the scenarios in 2100 as they have not been quantified. While all N may have some potential for release into the environment, combustion NO_x and fertilizer N are clearly linked to such a fate.

Supplementary Material:

Here we present the detailed algorithm to estimate agricultural demand for industrial N fixation, based on a few key parameters. As described in section 3, the initial scaling parameter is <u>population</u>, such that we model population induced N fixation (N_p) in proportion to the population estimate of the respective scenario. The sources for the respective population projections are also shown in Table 1 in the main text. These original sources needed to be consulted in order to obtain a full time series; data were also maintained in case of inconsistencies to the values presented in the RCP papers. Matsui et al. (2011) report for 2100 a global population of "9.8 billion persons", while following their described procedure we end up in 9.34 billion. Similarly, Thompson et al. (2011) report for 2100 "8.7 billion", while their source lists 8.6 billion (Clarke et al. 2007). We use the respective underlying figures from the original sources, such that for 2100 the population projections of 12.40 billion, 9.34 billion, 8.60 billion and 9.06 billion are applied for RCP8.5, RCP6.0, RCP4.5 and RCP2.6, respectively.

$$N_{P}(yr) = N(2005) * population (yr) / population (2005)$$
(1)

where population (yr) stands for the estimated global population number in the year yr. Population projections are taken, at 10 year intervals, as provided in the papers describing the respective RCP. N(2005) is the mineral fertilizer N produced in 2005, 94.23 Tg N. The parameter is also explicitly calculated for the year 2000, the starting year for all other driving factors.

<u>Efficiency increase</u>: Prior to the widespread availability of mineral fertilizers, there was a strong need to keep agronomic nutrient cycles closed. With the availability of fossil fuels, bulk industrial production of mineral fertilizers increased substantially (Smil 2001), allowing

N becoming a plentiful resource in many countries. Koning et al. (2008) provide a relationship based on historic data where each additional kg N harvested in crops comes at a cost of 2 kg mineral fertilizer N added to soils. The inefficiency of this system is one of the key reasons for nitrogen pollution (Galloway et al., 2004; 2008). It also reflects the challenge to produce even more food for a global population, in the context of the parallel challenge to mitigate nitrogen pollution (e.g. Mueller et al. 2012).

Based on OECD's concept of Nitrogen Use Efficiency (NUE) and following Cassman et al. (2002) as well as Balasubramanian et al. (2004), we find global NUE to be below 40% in practice, with a considerable potential for improvement being discussed (see also the NUE improvements demonstrated in OECD, 2008). Accounting for reported efficiency levels of 60-80 % in research trials, and considering the need of large production amounts while keeping the environmental impact low, we assume 60% as a reasonable estimate of a future optimized NUE. Changing from 40% NUE to 60% means for each kg of N in product that instead of 2.5 kg only 1.67 kg N input will be required, which is one third less for the same amount of production. As for a global average, NUE will be determined by areas of high production, but not by parts of the world where there is nitrogen shortage, and where also no input reduction is expected (e.g., Africa). While not all previous authors share the assumption on globally improved NUE's in the future (Tubiello and Fischer, 2007, rather assume constant ratios of cereal production and fertilizer input between 2000 and 2080), improvements of 1% per year are also being discussed based on past experience in a number of countries (Dobermann and Cassman, 2005).

For this parameter, we thus apply a correction factor based on an assumed general improvement in NUE of 0.5% per year. This factor has a lower limit of 0.666:

 $F_{\text{NUE}}(\text{yr}) = \text{minimum} [0.995^{(\text{yr-}2000)}, 0.666]$ (2)

<u>Food equity</u>: Raising the protein availability of the global population to the same level as now available in Europe would require increases in animal production, resulting in more animal feed to be produced. We assume additional animal protein adds to rather than replaces plant protein, and additionally required fertilizer N would strictly be replenished from mineral fertilizer. No additional atmospheric deposition (due to changed NO_x emissions) or BNF is considered. Using the parameters for a global nitrogen cycle taken from Smil (1999), we estimate a feedback loop of animal manure influencing mineral fertilizer demand. Considering all the losses involved in the process, an increase in animal protein (and thus animal production) of 78% would thus need about 69% more mineral fertilizer (of the total for both food and feed production). The change would occur progressively to materialize fully at the end of the scenario (year 2100).

Again a correction factor is being used for implementation. The correction factor describes a geometric interpolation of an expected change, which in 2100 will become 1.69 or a 69% increase.

$$F_{\text{equity}}(\text{yr}) = 1.69^{(\text{yr}-2000)/100}$$
(3)

Diet optimization: As one of several options to improve the efficiency in protein production, we envisage a change of the current European ratio of meat to milk from 2:1 to 1:2. Following Smil (2001) and the efficiencies provided for milk and meat, this would increase overall efficiency from 23 to 30% in animal production, which we extrapolate globally. Using the same feedback loop as discussed for food equity results in a 12% reduced need in mineral fertilizer. Like in the "food equity" case above, we assume a progressive change over the whole scenario period. The final factor in 2100 would then approach 0.88 (12% less than in 2000).

$$F_{diet_optim} (yr) = 0.88^{(yr-2000)/100}$$
(4)

Biofuel related nitrogen demand is considered for the RCP2.6 only, following the temporal development as obtained from the original literature (van Vuuren et al., 2011). Biofuel production in the RCP scenario follows a previous publication of the same authors (van Vuuren et al. 2010). The authors of that study provide details on the additional primary energy produced in 2050 and 2100 from biofuels (we interpolate linearly), loss rate between feedstock and primary energy, and energy-related emission factors. Furthermore, they also inform on the share of second generation biofuels for specific years which we extend to cover each ten-year period. This is important as for second generation biofuels, covering practically all production from 2050 onwards, the whole plants are used for energy and thus less N_r is wasted to grow unused plant material. Using the authors' original methods (which are described by Harmelink and Hoogwijk 2008, who refer back to the IPCC greenhouse gas inventory guidelines, Houghton et al. 1997) we are able to trace back the nitrogen fertilizer demand as originally established, which does not include any efficiency improvement over time. Globally, this results in 95 Tg additional N for biofuels by 2100. Note that a previous estimate (Erisman et al., 2008), assuming 100 kg N addition per ha applied to 0.74 Gha additional agricultural land reserved for biofuels (about half to the current agricultural area) ended up in a similar order of magnitude, at 74 Tg additional N.

Thus industrial reactive nitrogen to be used for biofuel production is just an additive term, N_B .

 $N(yr) = N_P(yr) * F_{NUE}(yr) * F_{equity}(yr) * F_{diet_optim}(yr) + N_B(yr)$ (5)

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