# Nutrient cycling and $N_2O$ emissions in a changing climate: the subsurface water system role



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# Nutrient cycling and $N_2O$ emissions in a changing climate: the subsurface water system role

**Georgia Destouni and Amélie Darracq**

Department of Physical Geography and Quaternary Geology, Stockholm University, SE-10691 Stockholm, Sweden

E-mail: georgia.destouni@natgeo.su.se

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**Abstract**

This study has quantified the subsurface (groundwater, soil, sediment) water system role for hydrological nitrogen (N) and phosphorus (P) loading to the coast and agricultural $N_2O$ emissions to the atmosphere in a changing climate. Results for different climate and hydrological model scenarios in the Swedish Norrström drainage basin show that the subsurface water system may largely control a long-term increase in the coastal nutrient loading, in particular for P, irrespectively of the realized future climate change scenario and our uncertainty about it and its water flow effects. The results also indicate an important subsurface water system role for current atmospheric $N_2O$ emissions from agriculture, and an even greater role for future ones. The current $N_2O$–N emissions from agriculture are quantified to be about 0.05 g $m^{-2}$ $yr^{-1}$ over the basin surface area, or 3% of the direct N mass application on the agricultural land. These results are consistent with recent global emission estimates, and show how the latter can be reconciled with previous, considerably smaller subsystem emission estimates made by the IPCC (Intergovernmental Panel on Climate Change).

**Keywords:** groundwater, climate change, hydrology, nutrient cycling, $N_2O$ emissions, earth system feedbacks

## 1. Introduction

Eutrophication of inland, coastal and marine waters is a problem in many parts of the world (Turner and Rabalais 1994, Smith *et al* 1999, Beman *et al* 2005, Conley *et al* 2009). This problem depends significantly on excess nutrient loads that are carried by inland water flows toward and into the sea from various human activities, such as agriculture and wastewater discharges, as well as the atmospheric deposition on land from fossil fuel combustion.

Recent studies have shown that nutrient pools and moving plumes in the subsurface water system (groundwater, soils and sediments and their pore water) of hydrological catchments may significantly affect, delay and modify the loads of nutrients to inland and coastal waters (Baresel and Destouni 2005, 2006, Lindgren *et al* 2007, Darracq *et al* 2008). However, these studies did not account for how changes in climate will interact with this subsurface water system role to determine the development of future nutrient loads. In this paper, we have investigated such climate change effects by use of reported regional climate change projections (Mattson and Rummukainen 1998) in a hydrological simulation model (Lindgren *et al* 2007, Darracq *et al* 2008) of water and nutrient (nitrogen and phosphorus) mass flows in the Swedish Norrström drainage basin (NDB, figure 1).

The NDB is a well-investigated hydrological basin, used as a large-scale field laboratory for quantification of water and nutrient cycling in several previous studies (Baresel and Destouni 2005, 2006, Darracq and Destouni 2005, 2007, Darracq *et al* 2005, 2008 Lindgren *et al* 2007). The present study has built on the process understanding and dynamic

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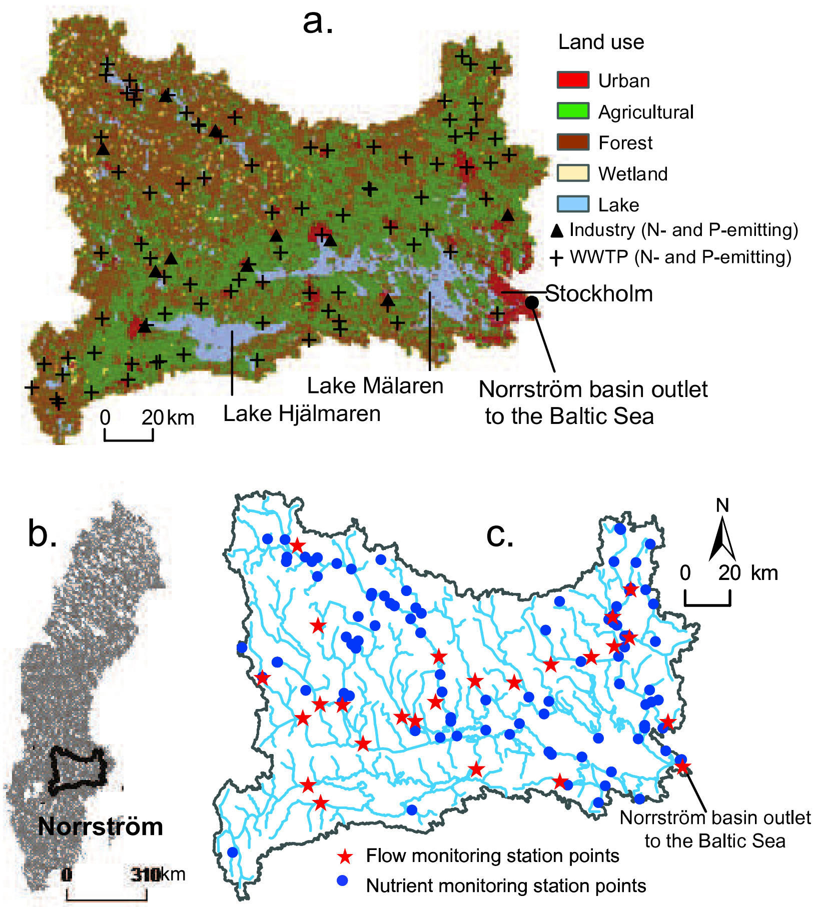


**Figure 1.** (a) Point and land-use sources of nutrients and other land cover in the Norrström drainage basin (NDB); (b) the NDB location within Sweden; and (c) the water flow and nutrient monitoring stations and stream network in the NDB.

model developments of the previous NDB studies by Lindgren *et al* (2007) and Darracq *et al* (2008) to investigate how climate change and our uncertainty about it and its water flow effects may interact with the hydrological transport of nutrient mass in the basin, and in particular its subsurface water system, to determine the development and our projections of the future nutrient loading to the coast.

Furthermore, we have in this study also used a complementary modelling approach, quantifying the total nitrogen cycling between different socio-economic sectors and engineered and natural water systems in the NDB (Baresel and Destouni 2005, 2006) to address the subsurface water system role for another important question related to climate change, posed by a recent study by Crutzen *et al* (2008). That study showed that an increase in the use of biofuels instead of fossil fuels as energy sources might increase rather than decrease global warming, due to increased $N_2O$ emissions to the atmosphere associated with nitrogen (N) fertilization. $N_2O$ is a by-product of N fertilization in agriculture and a greenhouse gas with a 100 year average global warming potential that is 296 times larger than an equal mass of $CO_2$ (Prather *et al* 2001). The result of Crutzen *et al* (2008) depended on an overall conversion factor of 3–5% from newly fixed agricultural N to $N_2O$–N, which was arrived at in their study by accounting for global-scale changes in synthetic N fertilizer production. However, the factor found by Crutzen *et al* (2008) was much larger than the corresponding 1% conversion factor used by IPCC (2006) for $N_2O$–N emissions from agriculture, and an explicit process explanation for this discrepancy remained as an important open question for further investigation.

Crutzen *et al* (2008) argued that this discrepancy depends on indirect $N_2O$ emissions related to agriculture, which were not accounted for in the direct, bottom-up conversion methodology of IPCC (2006), but were implicitly included in the global-scale, top-down method in their study. In the present study, we have used available data-driven model results for the NDB to investigate the possibility of reconciling the different agricultural N to $N_2O$–N conversion factors of IPCC (2006) and Crutzen *et al* (2008), by explicit account of $N_2O$ emissions from agricultural N transported through the subsurface and surface water of drainage basins. Specifically, we have here addressed this question by use of the previously quantified and reported scenario results of the total (i.e., both direct and indirect) nitrogen mass flow from agriculture through the different socio-economic sectors and engineered and natural water systems of the NDB (Baresel and Destouni 2005, 2006).

## 2. Material and methods

In order to quantify the future development of N and phosphorus (P) loading from the NDB to its recipient coastal waters, we have used regional projections of climate change (Mattson and Rummukainen 1998) together with the spatially distributed, dynamic modelling approach to hydrological nutrient transport that also underlies a series of previous NDB studies (Darracq and Destouni 2005, 2007, Darracq *et al* 2005, 2008, Lindgren *et al* 2007). We have then assumed that the various external point sources and diffuse land-use inputs of nutrients in the NDB (figure 1(a)) remain constant after 2005. Previous simulation results of long-term nutrient load development in the NDB, which made the same source assumption but neglected climate change (Lindgren *et al* 2007, Darracq *et al* 2008), showed that N and P loads continue to increase after external source stabilization because the nutrient transport is not at steady-state. We have therefore quantified the future water and nutrient mass flows on both a decadal short term (from 2000 until 2030) and with regard to the long term when steady-state mass flow conditions are reached.

Furthermore, in order to account for possible uncertainty in the regional climate modelling, we have investigated and compared three different climate change scenarios: (a) no change from the 2000 climate conditions, (b) change according to reported regional climate model projections for the period 2000–2100 (Mattson and Rummukainen 1998), and (c) twice as large climate change between 2000 and 2100 as in (b). The climate change scenarios have been implemented in the long-term dynamic hydrological modelling in terms of changes in average annual temperature, winter temperature and precipitation, linearly interpolated between the implications of the climate scenarios for 2000 and 2100. According to the regional climate model results of Mattson and Rummukainen (1998), the average annual temperature, winter temperature and average annual precipitation in the basin are in the projected climate change scenario (b): 6.5 °C, −1.9 °C and 630 mm $yr^{-1}$ in 2000, and 9.9 °C, 2.4 °C and 700 mm $yr^{-1}$ in 2100, respectively. In the double projected climate change scenario (c), these climate measures are the same as in (b) in 2000, and 13.3 °C, 6.8 °C and 770 mm $yr^{-1}$ in 2100. After 2100, until a long-term steady-state is reached in nutrient mass fluxes, the climate measures have been assumed to remain constant at their 2100 levels.

The hydrological modelling that propagates the climate change effects through the NDB has been described in detail in previous studies (Darracq and Destouni 2005, 2007, Darracq *et al* 2005, 2008, Lindgren *et al* 2007) and a summary is provided here in the appendix. In order to also account for possible uncertainty in the hydrological modelling of the climate change effects on water flows, for each climate scenario simulation, we have used two different evapotranspiration (ET) parameterization schemes, which can both equally well explain the available observation data of inland water flows (see observation points in figure 1). The two ET schemes are: (i) the temperature-based quantification approach of Meinardi *et al* (1994), and (ii) the quantification approach of Wendland (1992) based on prevailing soil texture and land cover. In summary, the result differences between the two ET schemes provide some measure of hydrological model uncertainty, in addition to the climate model uncertainty implied by the three different climate change scenarios (a)–(c). Such ET-related hydrological model uncertainty has also been considered and used in hydrological studies of other catchment areas (Shibuo *et al* 2007, Jarsjö *et al* 2008).

To quantify the direct and indirect atmospheric emissions of $N_2O$ related to agriculture, we have further used the reported input–output mass flow modelling scenarios and results of Baresel and Destouni (2005, 2006) for N in the NDB. These scenarios and results all fit to and can explain the relevant data available for the basin and quantify the total (direct and indirect) N mass flows between the different socio-economic sectors and land uses (agriculture, urban and forest land, industry, households and municipalities), engineered water systems (wastewater treatment plants, urban storm water handling, municipal water supply, private sewage systems) and natural water systems (surface and subsurface) of the NDB. The input–output modelling approach requires, includes and interprets, lumped over the whole basin and for present-time conditions, more types of data and data sources (outlined in detail by Baresel and Destouni (2005, 2006)) than the above-discussed, spatially distributed, dynamic hydrological modelling of the direct source-to-recipient nutrient transport in the NDB (Darracq and Destouni 2005, 2007, Darracq *et al* 2005, 2008, Lindgren *et al* 2007); see also the associated summary in the appendix. With regard to the subsurface water system, however, both modelling approaches have yielded consistent results, implying that the nutrient accumulation–remobilization dynamics in subsurface nutrient pools and the slow-moving subsurface nutrient plumes may significantly affect and modify present and future loads of nutrients to inland and coastal waters (Baresel and Destouni 2005, 2006, Lindgren *et al* 2007, Darracq *et al* 2008).

In the present study, we have extracted and summarize below the reported Baresel and Destouni (2005, 2006) scenario range of N mass flows from agriculture to and among the subsurface and surface water systems and nutrient pools in the NDB. We have further combined these N mass flow quantifications with the $N_2O$–N conversion factors of IPCC (2006) for different subsystems, in order to investigate whether an explicit account of the agricultural N mass flows through different subsystems can reconcile the IPCC (2006) with the lumped global-scale conversion factor of Crutzen *et al* (2008).

## 3. Results

Figure 2 shows the modelled short- and long-term effects of climate change on the total water flow and nutrient loads to the sea from the NDB. For the modelled water flow, the response and the response uncertainty related to climate change generally lies within the hydrological uncertainty range implied by the two different ET quantification schemes. That is, due to this hydrological uncertainty, one cannot conclusively expect the total water flow to increase due to climate change in this basin. The temperature-based ET scheme (i) of Meinardi *et al* (1994) yields a decreasing water

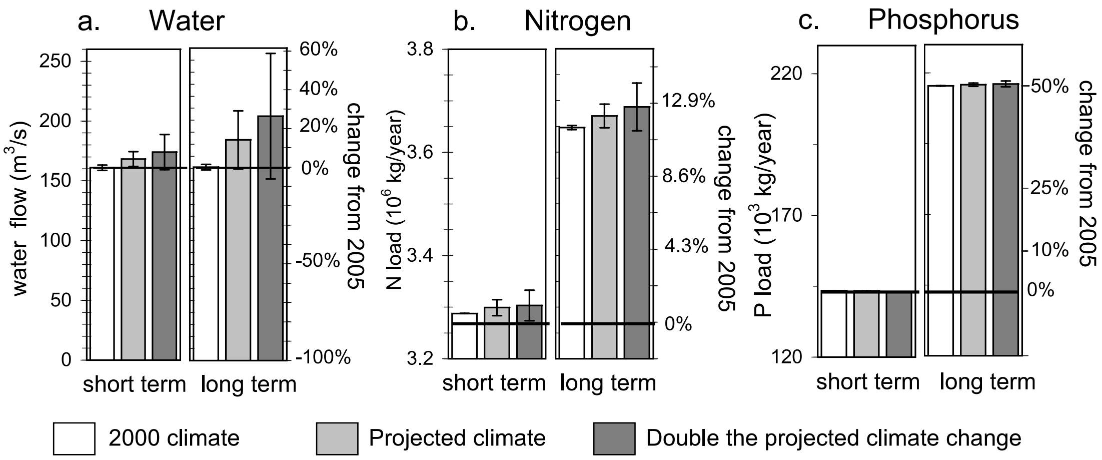


**Figure 2.** Short- and long-term changes of (a) total water flow, (b) coastal nitrogen N mass flow, and (c) and coastal phosphorus P mass load, at the Norrström basin outlet. Error bars show the result range for the two different model schemes for evapotranspiration, described in the main text. For direct comparison, the black line shows 2005 values of total water flow and coastal nutrient loads.

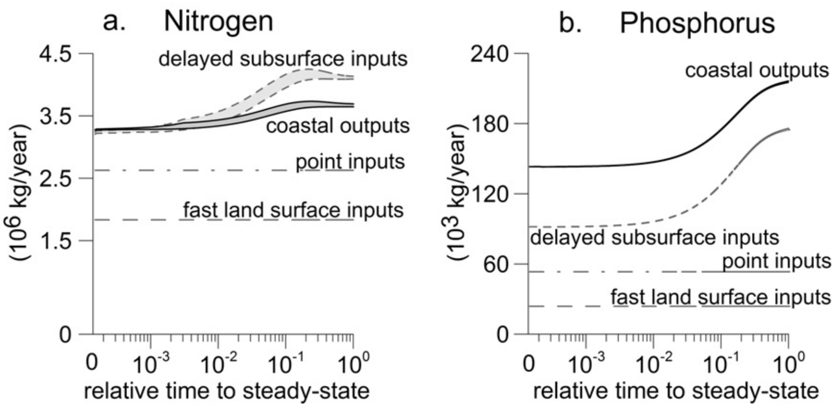


**Figure 3.** Temporal development from 2005 until steady-state is reached for (a) nitrogen and (b) phosphorus mass inputs from different source contributions (described in the main text) to surface waters, and total outputs to the coast for the Norrström drainage basin. Filled areas between curves show the result range of the two different model schemes for evapotranspiration, described in the main text.

flow because ET is predicted to increase more than the climate model projected increase in precipitation in the warming regional climate. In contrast, the soil and land cover based ET scheme (ii) of Wendland (1992) predicts unchanged ET and therefore increasing water flow as precipitation increases in the changing climate.

The mid-range water flow, however, does increase, and the increase ranges from 0% to 8% in the short term and 26% in the long term, depending on climate change scenario. Also the coastal nutrient loads increase, by 0.5–1% for N and 0–0.5% for P in the short term, and by 11–13% for N and around 50% for P in the long term. However, the nutrient load increases are almost independent of climate change scenario and the uncertainty about its realization and water flow effects. The nutrient load changes are thus relatively insensitive to climate change and its uncertainties, and must be controlled by other factors.

Figure 3 identifies and quantifies the main controlling factor for the large projected long-term increases of nutrient loads, by showing the modelled future development of the coastal outputs of nutrients, and the contributions to this development from different components of nutrient mass inputs to surface waters. These components are: (1) the direct point source inputs to surface waters from present wastewater treatment plants and industrial discharges (point inputs, constant by assumption); (2) the diffuse source inputs to surface waters by the direct atmospheric deposition on and the relatively fast hydrological transport (with a less-than-5 year delay) to surface water from urban surface runoff and leaching from agriculture, private sewers and deforestation (fast land surface inputs, constant by assumption); and (3) the delayed diffuse inputs from the subsurface to the surface water system, from earlier inputs of the same types of sources as in (2), where the delay is due to the nutrient immobilization–remobilization and slow transport processes in the subsurface (delayed subsurface inputs). The component (3) increases and controls the future increase of coastal nutrient loads, due to the long memory of earlier nutrient releases, which remain for long time in and are only slowly propagated through the subsurface water system, irrespectively of climate change scenario.

The effect of the delayed, component (3), inputs to surface waters is greater for P than for N, because some of the N

**Table 1.** Nitrogen (N) mass flows and $N_2O$ emissions to the atmosphere from agriculture in the Norrström drainage basin.

| | Total agricultural N mass flow through the system (t $yr^{-1}$) | Conversion factor for $N_2O$–N emissions to atmosphere (%) (default IPCC (2006) value) | Absolute $N_2O$–N emissions to atmosphere (t $yr^{-1}$) | $N_2O$–N emissions relative to the direct N mass flow through agriculture (%) (mid-range value) |
|---|---|---|---|---|
| Primary, directly from agricultural land | 45 760[a] | 0.3–3[b] [1] | 137–1373 | 0.3–3 [1.7] |
| Secondary, from volatilized and re-deposited N on agricultural land (10–20% of direct N flow[b]) | 4576–9152 | 0.2–5[b] [1] | 9–458 | 0.02–1 [0.5] |
| Secondary, from agricultural N mass flow in surface water | 4259–6689[a] | 0.5–2.5[b] [0.75] | 21–167 | 0.05–0.4 [0.2] |
| Secondary, from agricultural N mass flow in subsurface water[c] | 757–7860[a] | 0.5–2.5[d] | 4–197 | 0.01–0.4 [0.2] |
| Total secondary from agricultural N transport in the subsurface and surface waters | 5016–14 549 | 0.5–2.5[d] | 25–364 | 0.05–0.8 [0.4] |
| Total primary and secondary agricultural N in the basin | 55 352–84 010 | — | 171–2195 | 0.4–5 [3] |

[a] Total N mass flows from agriculture (directly and indirectly through other socio-economic sectors and engineered and natural water systems) to the surface and subsurface waters, as derived from the basin-scale input–output flow analysis and data-driven scenarios of Baresel and Destouni (2005, 2006).
[b] Range given by Crutzen *et al* (2008).
[c] Including a delayed N mass flow contribution of 399–1203 t $yr^{-1}$ from remobilization of N in subsurface accumulation pools, in addition to the delayed contribution of the also relatively slow advective subsurface transport, according to the different input–output flow scenarios of Baresel and Destouni (2005, 2006).
[d] Neither IPCC (2006) nor Crutzen *et al* (2008) accounted for $N_2O$–N emissions from the subsurface water system. The conversion factors for the $N_2O$–N emissions from subsurface water to the atmosphere have here been assumed to be in the same range as that reported for surface water by Crutzen *et al* (2008).

input mass undergoes truly irreversible denitrification, while the remaining N mass is less affected than P by further delaying sorption–desorption and reaction processes in addition to the slow physical transport along the subsurface pathways. As a consequence, the delayed subsurface inputs at the long-term steady-state are for P about 2.5 times greater than, while for N about the same as the sum of the point inputs and the fast land surface inputs (figure 3).

Table 1 finally synthesizes and summarizes all the different reported (Baresel and Destouni 2005, 2006) scenarios and their associated results with regard to the total N mass flows from agriculture in the NDB. Table 1 also lists the conversion factors for $N_2O$–N emissions from IPCC (2006) and their uncertainty ranges as reported by Crutzen *et al* (2008), the present extended use of these factors, and the resulting implications for the absolute and relative agricultural $N_2O$–N emissions from the NDB to the atmosphere.

The direct N mass flow through agricultural land in the NDB is around 46 kt $yr^{-1}$. The total (direct and indirect) N mass flow of agricultural origin in the whole basin, however, is in the range of 55–85 kt $yr^{-1}$, i.e., 20–83% greater than the direct mass flow. This yields a total conversion factor for agricultural $N_2O$–N emissions relative to the direct N mass flow through agriculture in the range of 0.4–5%, with a mid-range value of 3%. This result is consistent with the large global conversion factor of 3–5% total agricultural $N_2O$–N emissions by Crutzen *et al* (2008), as well as locally with the various, smaller subsystem conversion factors of IPCC (2006).

Baresel and Destouni (2005, 2006) quantified the fraction of N fertilizer application to be 86% of the total direct N mass flow through agricultural land in the NBD. Combined with the results in table 1, this implies total $N_2O$–N emissions from N fertilizer application in agriculture of about 1017 t $yr^{-1}$ (range: 147–1888 t $yr^{-1}$), or 0.046 g $m^{-2}$ $yr^{-1}$ averaged over the NDB surface area of about 22 000 $km^2$. Also this land area averaged estimate of agricultural $N_2O$–N emissions is consistent with the global land area averaged result of 0.04 g $m^{-2}$ $yr^{-1}$ from the entirely different, top-down quantification approach of Crutzen *et al* (2008).

## 4. Conclusions

This study has built on and combined the process understanding and model developments of different previous NDB studies and extended the use of this basin as a large-scale field laboratory for quantification of climate and other environmental change effects. The results show that the regional nutrient loading to inland and coastal waters is likely to increase in the future, in particular for phosphorus. The regional nutrient load increases are quite insensitive to the future realization of climate change and our uncertainty about it and its effects on total water flow. The long-term nutrient load development is instead largely controlled by the delayed load contributions from earlier nutrient mass inputs, which are slowly propagated, retained and remobilized in the subsurface water system, almost irrespectively of prevailing climate change scenario.

Corresponding results may turn out differently under different regional conditions. A changing climate may also affect the nutrient attenuation and transformation rates, an effect that has not been accounted for here. Nevertheless, the present study emphasizes that the subsurface transport effects on the long-term development of nutrient and pollutant loading cannot just be *a priori* neglected in any region. Such neglect must be appropriately and quantitatively justified, by investigations that explicitly account for and combine the long-term effects of subsurface transport with those of climate change, similarly to this study.

The present results further indicate an important subsurface water system role also for the agricultural $N_2O$ emissions to the atmosphere. The current total (primary and secondary) $N_2O$–N emissions from agricultural N fertilization are quantified to be on average 0.05 g $m^{-2}$ $yr^{-1}$ over the total basin surface area, or 3% of the direct N mass flow through the agricultural land in the basin. These results are consistent with corresponding global emission estimates of Crutzen *et al* (2008), and show how they can be reconciled with the smaller IPCC (2006) estimates of different subsystem emissions. The subsurface water system is shown to contribute as much to the total $N_2O$–N emissions as the surface water system, and both these water systems together contribute as much, and with similar quantification uncertainty, as the secondary $N_2O$–N emissions from volatilized and re-deposited N on agricultural land.

Among the different secondary contributions to the agricultural $N_2O$ emissions, those of the subsurface water system have been neglected both in the subsystem estimates of IPCC (2006) and in the discussion of implicit secondary contributions to the larger global estimate of Crutzen *et al* (2008). The present simulations for different climate change scenarios indicate that the already considerable, yet so far neglected, subsurface water system role for agricultural $N_2O$ emissions is likely to increase in the future, regardless of actual climate change realization and its total water flow effects.

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## Appendix

For the present quantification of climate change effects, we have used the same, spatially distributed, dynamic hydrological approach as Darracq and Destouni (2005, 2007), Darracq *et al* (2005, 2008) and Lindgren *et al* (2007) for the direct nutrient transport from the various sources to the coastal recipient of the NDB. This approach is based on the POLFLOW modelling framework for water, total nitrogen (N) and total phosphorus (P) mass flows (de Wit 2001), using the same input data sources, calibrated parameter values and validation results for the present simulations as for the N and the P study of Darracq and Destouni (2007). The POLFLOW model is embedded in the geographic information system (GIS) tool PCRaster (Van Deursen 1995). The different POLFLOW water and nutrient modules estimate the water and the N and P mass flows, respectively, including for the latter the transport and attenuation processes that occur from the nutrient sources to the basin outlet. The spatial resolution used in the NDB modelling is 1 $km^2$ and the time step for the present simulation of the long-term dynamics of inland and coastal nutrient loads is 5 years.

As described in more detail for both the N and the P modelling by Darracq and Destouni (2007), the input data for the POLFLOW-based water flow and nutrient transport–attenuation modelling of the NDB include: meteorological data (Swedish Meteorological and Hydrological Institute), digital elevation model (HYDRO1K, US Geological Survey), basin landscape characteristics (soil and hydrogeological maps, Geological Survey of Sweden), land use (BALANS maps and statistical database, UNEP/GRIP-Arendal), river network (Digital Chart of the World Server; map of Water systems in Sweden, Swedish Meteorological and Hydrological Institute), point source emissions and diffuse source emissions (TRK project, Brandt and Ejhed 2003). Furthermore, measurements of water discharges (Swedish Meteorological and Hydrological Institute) and nutrient concentrations (Swedish University of Agricultural Sciences, Environmental monitoring and assessment database) are used for model calibration.

Water flow and nutrient mass flow calculations honour the water and nutrient mass balances, respectively, over each grid cell as well as over subcatchments and the whole drainage basin, with the various subsurface and surface water transport, mass transfer and attenuation processes being semi-empirically quantified following de Wit (2001). The water flow module yields annual average (over 10 years) runoff as the difference between annual average precipitation and actual ET; the latter is empirically estimated according to two different ET schemes described in the main text.

The POLFLOW model conceptualization assumes that groundwater and its advected solutes flow through two different physical subsystems of the subsurface: a relatively highly conductive (possibly shallow) groundwater system and a less conductive (possibly deeper, or just a relatively immobile part of the shallow) groundwater system. Calculated groundwater recharge indices are related to topographic slope, soil type, texture, aquifer type, groundwater level, land cover and January temperature, and groundwater residence times are related to aquifer conductivity, porosity, thickness, local slope and groundwater recharge.

Nutrient transport pathways are assumed to follow the water flow paths (vertical or horizontal), over and/or through the soil surface, into underlying groundwater before reaching the stream network. In each cell of the rasterized drainage basin, the contributing net nutrient mass inputs from various sources are given from independently reported source release values (TRK project, Brandt and Ejhed 2003). Nitrogen loss to the atmosphere by denitrification in the soil and groundwater systems is represented following Wendland (1992), based on groundwater residence times and other transport characteristics of the basin's soils and aquifers.

Topography-induced local drainage direction networks direct the water and nutrient mass flows towards the basin outlet, and a nutrient immobilization–remobilization parameterization at constant rates is applied to all surface water cells, accounting for their hydraulic characteristics (surface slope and water flow). This determines the nutrient mass that is transported further downstream along the stream drainage network.

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