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Future riverine inorganic nitrogen load to the Baltic Sea from Sweden: An ensemble approach to assessing climate change effects

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Key Points:

- A systematic ensemble approach based on empirical inorganic nitrogen models provides robust estimates of future inorganic nitrogen dynamics.
- The amount and seasonal variability of future inorganic nitrogen loads are controlled by shifts in growing season and streamflow patterns.
- Annual inorganic nitrogen loads will rise as a result of rising winter streamflow, which will outweigh the decline in spring flood volume.
Abstract

The dramatic increase of bioreactive nitrogen entering Earth’s ecosystems continues to attract growing attention. Increasingly large quantities of inorganic nitrogen are flushed from land to water, accelerating freshwater and marine eutrophication. Multiple, interacting, and potentially countervailing drivers control the future hydrologic export of inorganic nitrogen. In this paper, we attempt to resolve these land-water interactions across boreal/hemiboreal Sweden in the face of a changing climate with help of a versatile modeling framework to maximize the information value of existing measurement time series. We combined 6962 spatially distributed water chemistry observations spread over 31 years with daily streamflow and air temperature records. An ensemble of climate model projections, hydrological simulations and several parameter parsimonious regression models was employed to project future riverine inorganic nitrogen dynamics across Sweden. The median predicted increase in total inorganic nitrogen export from Sweden (2061-2090) due to climate change was 14% (interquartile range 0-29%), based on the ensemble of 7500 different predictions for each study site. The overall export as well as the seasonal pattern of inorganic nitrogen loads in a future climate are mostly influenced by longer growing seasons and more winter flow, which offset the expected decline in spring flood. The predicted increase in inorganic nitrogen loading due to climate change means that the political efforts for reducing anthropogenic nitrogen inputs need to be increased if ambitions for reducing the eutrophication of the Baltic Sea are to be achieved.

1 Introduction

Multiple ongoing global changes have reshaped the pools and fluxes of biogeochemical elements in terrestrial and aquatic ecosystems. Of these, dramatic increases in the loading of bioreactive nitrogen (N) to terrestrial ecosystems during the 20th century have drawn particular attention (Galloway et al., 2008) and are linked to multiple environmental problems, ranging from declines in species diversity to stratospheric ozone loss (Gruber & Galloway, 2008). Large quantities of N are also flushed from land to water (Seitzinger et al., 2005) which contribute to freshwater and marine eutrophication (Conley et al., 2009). These mounting water quality concerns are linked to hydrological patterns that are themselves sensitive to climate drivers (IPCC, 2014). Concurrent to these global increases in N inputs, warming temperatures, longer growing seasons, and rising atmospheric CO₂ concentrations may lead to increased plant growth (Richardson et al., 2010), greater N uptake and accumulation in terrestrial ecosystems (Luo et al., 2004) and, in some cases, reduced N losses to surface waters (Lucas et al., 2016). Thus, future change in hydrologic export of bioreactive N from catchments will reflect multiple, interacting, and sometimes countervailing drivers. Resolving these land-water interactions and predicting future dynamics in N export is an important goal that requires a new set of modelling tools and approaches.

The boreal forest, which represents one of the largest biomes on Earth (Scheffer et al., 2012), plays a significant role in global elemental cycles. Boreal forest landscapes typically support organic rich soils (Harriss, 1989) and N limited vegetation (Högberg et al., 2017) and thus efficiently retain and recycle inorganic forms of N (iN) on land while exporting less useable forms of dissolved organic N (DON) to aquatic systems (Sponseller et al., 2016). However, key processes regulating the N balance of forests, including plant uptake, immobilization/mineralization by soil microbes, and hydrologic flushing are all strongly seasonal and sensitive to a range of environmental changes (Bai et al., 2013). In this context, the internal
cycling and retention of iN in the boreal zone could be strengthened by increases in biotic demand as plant-soil systems respond to warmer temperatures (Barichivich et al., 2013), elevated CO$_2$ (Graven et al., 2013), or changing precipitation (Lim et al., 2017). Alternatively, loss of consistent winter snowpack may constrain microbial N immobilization in soils and foster greater hydrologic exports during spring flood (Brooks et al., 1998). More generally, changes in the timing and magnitude of flushing events can potentially override biotic controls and promote hydrologic losses, particularly when these occur during periods of weaker biotic N demand (Dittman et al., 2007). Given that the boreal zone is warming more rapidly than other biomes (IPCC, 2007), and in some regions is predicted to have altered precipitation patterns, this is an important biome to explore future changes in hydrologic iN export.

In addition to climate change, parts of the boreal zone are also subject to a variety of additional anthropogenic forces (e.g., related to land management) that may have additional consequences for landscape nutrient balance (Gauthier et al., 2015). For example, more than two thirds of Sweden is currently covered by forests, the vast majority of which are subject to forestry (SLU, 2015). A continued intensification of the forest industry (Helmisaari et al., 2014), in particular extensions of managed forest land and increasing use of fertilization (Rytter et al., 2013), could increase the risk of iN leaching from catchments (Sponseller et al., 2016). However, at broader scales, the long-term transformation toward actively managed forests has increased terrestrial N storage in parts of northern Sweden, and is potentially responsible for recent reductions in hydrologic export of inorganic forms (Lucas et al., 2016). In addition, agricultural activity in this region, focused largely in the south, represents an obvious source of anthropogenic N (Hägg et al., 2011), one that potentially leaves a long legacy in soil pools that are connected to surface waters (Basu et al., 2010). Regardless of source, iN losses from managed lands, and the resulting downstream consequences, have increasingly been the focus of attention (Williams & Tonnessen, 2000), as these forms of nitrogen could potentially contribute to eutrophication of the Baltic Sea (Brandt et al., 2009; Conley et al., 2009).

Future changes in riverine iN export in the boreal zone may depend upon how seasonal hydrological changes interact with the characteristic timing of iN use in terrestrial ecosystems. Across much of the northern hemisphere, concentrations of iN in streams and rivers are greater during the dormant compared to growing seasons, when both vegetation demand and terrestrial nutrient retention are high (e.g., Sponseller et al., 2014). In Sweden, this seasonality is superimposed upon a hydrologic regime characterized by rather low winter streamflow with a dominating snowmelt-driven spring flood peak (mainly in central and northern Sweden), followed by low summer flows and/or a somewhat lower precipitation-induced flood peaks in the fall (mainly in southern Sweden). In a future climate, however, streamflow is projected to change to a regime where large winter streamflow dominates and there is no spring flood peak at all (Arheimer & Lindström, 2015; Donnelly et al., 2013; Teutschbein et al., 2011, 2015). Thus, the combination of higher winter streamflow with rather high winter iN concentrations could lead to substantially more iN loads in a future climate.

We evaluate the hypothesis that iN loading to the Baltic Sea will accelerate in the future despite efforts to reduce the input to receiving water bodies in Sweden because of hydrologic change. To test this, we present a versatile framework, which employs an ensemble of climate model projections, hydrological simulations and regression models to simulate historical and future iN loads across Sweden. Using an empirical regression approach to establish statistical relationships between iN and hydro-climatic variables (i.e., streamflow and air temperature) is a
vital conceptual element in this study, which fully utilizes existing measurements and provides
systematic estimates of future iN loads for stationary landscape ecosystems.

2 Materials and Methods

2.1 Study Area

According to the Köppen-Geiger classification (Kottek et al., 2006), Sweden stretches
over four different climate classes (Figure 1a): Southern Sweden is located within the oceanic
climate zone (Cfb) with warm summers and cool winters, Central Sweden is part of the subpolar
oceanic climate zone (Cfc) with cool summers and cool winters, Northern Sweden experiences a
subarctic boreal climate (Dfc) with cool summers, very cold winter, persistent seasonal snow
cover and soil frost during winters, and the Scandinavian Mountains in Northwestern Sweden are
located in the polar tundra climate zone (ET) with monthly mean temperatures below 10°C.
During the period 1961-2010, the annual mean temperature was 2.5°C while the annual
precipitation averaged 733 mm. Temperature and precipitation are projected to increase more in
high-latitude regions such as Sweden than in the rest of Europe (IPCC, 2014; Jacob et al., 2014).
Due to the fact that high-latitude ecosystems have adapted to low natural energy flows, they are
relatively more sensitive to a given shift in climate, physical and biogeochemical conditions
(Roots, 1989).

Sweden has a land area of approximately 408,000 km² (SLU, 2015) with elevations
ranging from -2 to 2100 m.a.s.l.. About 69% of the land area are covered by forest (SLU, 2015),
9% by wetlands, 8% by shrubs and grass land, 8% by agriculture (mostly in Southern Sweden),
3% by human settlements and the remaining 3% are open land.

2.2 Observed Data

2.2.1 Inorganic Nitrogen

Inorganic nitrogen (iN) data was available for 440 stream/river sites distributed over the
Swedish main land (Figure 1a) and were collected as part of the national environmental
monitoring program (Fölster et al., 2014) financed by the Swedish Agency for Marine and Water
Management. The data was downloaded from a water chemistry database
(http://webstar.vatten.slu.se/db.html) that is currently hosted by the Swedish University of
Agricultural Sciences (SLU). Water samples in this particular database were collected from a
well-mixed portion of each stream (SLU, 2009) at a depth of 0.5 m (Fölster, 2010). Total
inorganic nitrogen (TIN) was calculated as the sum of ammonium nitrogen (NH₄-N) and the sum
of oxidized nitrogen (NO₂-N+NO₃-N).

2.2.2 Streamflow

Daily streamflow measurements for 320 Swedish catchments (Figure 1b) were
downloaded from a publicly accessible streamflow database (http://vattenwebb.smhi.se/)
provided by the Swedish Meteorological and Hydrological Institute (SMHI). About two thirds of
the streamflow stations within this database are owned and operated by SMHI and additionally
about 100 stations are operated by energy companies (Bergström & Wennerberg, 2008).
Geospatial data for the streamflow stations was obtained from SMHI’s SVAR database (Eklund,
2011), which offers information on more than 100,000 lakes, 28,000 streams and roughly 52,000
catchments (Henestål et al., 2015).

2.2.3. **Landuse data**

Catchment-specific landuse characteristics were included in SMHI’s streamflow database
and were based on national maps over agricultural land areas provided by the Swedish Board of
Agriculture, lake information from SMHI’s geospatial SVAR database (Eklund, 2011), as well
as landuse information provided by the CORINE land cover project (CLC2000). Landuse
characteristics were assumed to not change over time.

2.2.4. **Meteorological Data**

For each catchment, gridded daily mean values of temperature and precipitation were
obtained from SMHI’s PTHBV database (SMHI, 2005), which provides a spatially interpolated 4
km x 4 km national grid for the period 1961-2010 (Johansson, 2002). Catchment-specific
temperature and precipitation values were calculated by an area-weighted average of all grid
cells partly or fully lying within the catchment boundaries. Long-term monthly mean potential
evaporation (pET) values for each catchment were obtained through Kriging interpolation
(Isaaks & Srivastava, 1989) of 152 meteorological stations across Sweden which had available

2.3. **Selection of Suitable Study Sites**

In this study, we tried to match the location of the 440 TIN sampling sites (Figure 1a)
with the position of the 320 streamflow stations (Figure 1b) under the conditions that (1) TIN
sites must have at least 3 years of available data with a minimum of 36 measurements, (2) both
TIN and streamflow stations should be located within a distance of 3 km on the same stream (not
on tributaries), and (3) streamflow stations should drain catchments with a degree of regulation
(i.e., the percentage of annual streamflow that could be stored in upstream reservoirs) of less than
25% to ensure a reasonable performance of the hydrological model used in this study. The 19
study sites (Figure 1c) that fulfilled these selection criteria drain together roughly 13% of
Sweden’s land area. With an average forest cover of 71.6% (Table 1), these catchments were
considered to be representative for entire Sweden, which is covered by 69% forest. They further
span a latitudinal gradient from 56.4° N to 68.4° N and, therefore, represent all four Swedish
climate zones (Figure 1a). The average period of TIN measurements of the 19 chosen sites was
31 years with approximately 370 measurement points per catchment in time. The longest TIN
record dating back to 1962 (station ‘Sävjaån Kuggebro’) consisted of more than 600 monthly
TIN samples, while the shortest record (station ‘Skeingesjön’) covered 6 years with 37
bimonthly samples. The chosen sites had daily streamflow measurements available for periods
between 28 and 93 years with an average length of 47 years. The catchment with the longest
record (station no. 1160 ‘Kukkusjärvi’) dated back to 1923.

The selected 19 study sites exhibit distinctly different present-day hydro-climatic
conditions, landscape characteristics and TIN signatures (Table 1). Depending on the location of
the study sites, annual mean air temperature ranges from -3.4 to +6.7 °C with a total annual
precipitation of 613-1082 mm, of which 210-846 mm (32%-84%) are transformed to streamflow
(Table 1). Rainfall-runoff ratios range from 0.32 to 0.84. All study sites show similar seasonal
patterns (but different orders of magnitude) of air temperature, precipitation, streamflow and TIN
concentrations (Figure 2). Air temperature reaches its maximum in the summer months June-August, while it is lowest in winter (December-February). The highest precipitation typically occurs in late summer or early autumn (July-September). Due to cold temperatures and snow accumulation in winter, all selected study sites are characterized by a snow-melt-driven spring flood peak in spring or early summer (April-July). TIN concentrations typically reach their highest levels in late winter or early spring (dormant season), while they are rather low during growing season (May-October), highlighting the potential for changes in biological and/or hydrological processes to influence seasonal concentrations (Sponseller et al., 2014).

2.4. Climate Data

The public web portal of the ENSEMBLES EU project (Van der Linden & Mitchell, 2009) provided 31 transient regional climate model (RCM) simulations forced by various global climate models (GCMs). Daily temperature and precipitation simulations for a reference control period (1961-2010) and a future scenario period (2061-2090) were downloaded from 15 selected simulations that (1) were run at a resolution of 25 km x 25 km, (2) had projections until year 2100 and (3) were based on the Special Report on Emissions Scenarios (SRES) A1B scenario, which assumes very rapid economic growth and a balance between fossil and non-fossil energy sources (Nakicenovic et al., 2000). For each of the selected 19 study sites, RCM precipitation and temperature values were averaged over all grid cells containing parts of the catchment. To correct for systematic errors (so-called biases) in RCM simulations (Ehret et al., 2012; Muerth et al., 2013; Teutschbein & Seibert, 2010, 2012, 2013), a post-processing procedure (i.e., bias correction) was applied. Bias correction typically involves the identification of differences between observations and RCM simulations for a control period, which are used to calibrate a correction algorithm. This correction algorithm is then applied to both the control and the future period of the RCM simulations. We used the so-called distribution scaling algorithm (Boe et al., 2007; Déqué et al., 2007; Ines & Hansen, 2006) to adjust the systematic biases in RCM-simulated temperature and precipitation on a monthly basis, which has been proven to be most suitable for Swedish catchments under current and future climate conditions (Teutschbein & Seibert, 2012, 2013). This method essentially aims at matching the theoretical cumulative distribution function (CDF) of RCM-simulated data with the observed CDF in three steps: (1) For precipitation, an RCM-specific threshold was introduced to avoid substantial distortion of the distribution by too many RCM-simulated days with low precipitation, (2) the parameters of the theoretical CDFs (e.g., Gamma for precipitation or Gaussian for temperature data) of both the observations and the RCM-simulated control run were calculated, and (3) these distribution parameters were then used to adjust the CDFs of both the RCM-simulated daily control (1961-2010) and future (2061-2090) variables. For a more detailed mathematical description of the procedure the reader is referred to Teutschbein and Seibert (2012).

2.5. Hydrological Modeling

2.5.1. The HBV model

We applied the conceptual hydrological model HBV (Bergström, 1976) to simulate daily streamflow during the control (1961-2010) and the future period (2061-2090) separately for each of the 19 catchments. HBV is a rainfall-runoff model that has been used in more than 90 countries and in various versions throughout the years (Bergström & Lindström, 2015). In the present study, we used the HBV-light software package (Seibert & Vis, 2012), which was driven
by daily temperature, daily precipitation and long-term monthly mean values of potential evaporation (pET). The model and its several subroutines for calculating snow, soil moisture, evapotranspiration, groundwater and channel routing were controlled by twelve lumped parameters that needed to be calibrated. For further detailed information about HBV we refer the reader to papers on its model structure and parameter uncertainty (Bergström, 1976, 1992; Bergström & Lindström, 2015; Harlin & Kung, 1992; Lindström et al., 1997; Seibert, 1999).

2.5.2. HBV Model Calibration and Validation Procedure

To assess how well the HBV model can describe streamflow in the 19 catchments, the split-sample technique proposed by Klemeš (1986) was used, which involved splitting the available observed streamflow series into a calibration and validation set. The HBV model was first calibrated separately for each catchment using observed meteorological data during the period 1991-2000. Parameter ranges were defined based on earlier model applications (Seibert, 1999; Seibert et al., 2000; Seibert & McDonnell, 2010). A built-in Genetic Algorithm and Powell’s Method Optimization (GAP) function (Seibert, 2000) were used to calibrate the model 100 times, which, due to its stochastic components, resulted in an ensemble of 100 different optimal parameter sets allowing the consideration for parameter uncertainty. To evaluate the model’s performance, a fuzzy measure \(X\) was calculated by combining three differently weighted objective functions (Table 2) as proposed by Seibert (1997): the Nash-Sutcliffe efficiency \(R_{\text{eff}}\) (Nash & Sutcliffe, 1970), Nash-Sutcliffe efficiency with logarithmic values \(L_{\text{eff}}\) and the volume error \(VE\). Additionally, we also considered seasonal correlation (SC) and anomaly correlation (AC) for further model evaluation as suggested by Orth et al. (2015):

\[
SC = \text{corr}(Q_{\text{obs}}, Q_{\text{mod}})
\]

where \(Q_{\text{obs}}\) is the observed and \(Q_{\text{mod}}\) the modeled streamflow series.

\[
AC = \text{corr}(Q'_{\text{obs}}, Q'_{\text{mod}})
\]

with \(Q'_{\text{obs}} = Q_{\text{obs},d,y} - \overline{Q_{\text{obs},d}}\) and \(Q'_{\text{mod}} = Q_{\text{mod},d,y} - \overline{Q_{\text{mod},d}}\)

where \(Q'_{\text{obs}}\) and \(Q'_{\text{mod}}\) are the anomaly time series (i.e., seasonal patterns in both observed and modeled streamflow series are eliminated). Subscript \(d\) indicates the day of the year and subscript \(y\) the actual year.

The calibrated model parameters were then validated using observed hydro-meteorological data during the period 2001-2010 to assure the HBV model is also valid for conditions different from those that it was calibrated to.

2.5.3. Simulations of Historical and Future Streamflow

The calibrated HBV model was then applied to simulate daily streamflow in each of the 19 catchments for the entire reference control period (1961-2010) and the future scenario period (2061-2090) using the bias-corrected precipitation and temperature series from the 15 RCMs as input. By combining an ensemble of 100 optimized HBV parameter sets with an ensemble of 15 RCMs (resulting in 1500 streamflow simulations for each time period), we ensured coverage of a wide range of possible outcomes. But we did not account for the influence of different bias correction methods or different hydrological models, both of which are likely to add further
uncertainties to the projections (Ehret et al., 2012; Teutschbein & Seibert, 2012; Wilby & Harris, 2006). Moreover, we made no attempt to weight climate models or regression models according to their skill level.

Teutschbein & Seibert (2010) showed that the median of such an ensemble is likely to fit observations better than each individual simulation. The first year of each period was considered as warm-up time of the HBV model and therefore disregarded. This means we ended up with 1500 streamflow simulations of 49 years each for the reference period (in total 73,500 annual streamflow series) and with 1500 streamflow simulations of 29 years each for the future period (in total 43,500) for each catchment (Table 3).

2.6. Nitrogen Modeling

2.6.1. The Fluxmaster Tool

Fluxmaster (Schwarz et al., 2006) is a regression tool, which typically combines available streamflow measurements with water-quality observations at a particular site to establish a regression model (RM), which can provide more accurate load estimates than single water quality point-measurements (Robertson & Saad, 2011). In the past, Fluxmaster has been used to simulate a variety of water-quality parameters, such as riverine carbon (Hood & Scott, 2008), nitrogen (Benoy et al., 2016; Kaushal et al., 2008, 2011; Richards et al., 2013; Robertson & Saad, 2011), phosphorus (Johnson et al., 2013; Richards et al., 2013; Robertson & Saad, 2011; Scott et al., 2007) or silica (Swaney et al., 2010) for sites in the USA. In Sweden, Fluxmaster has so far only been used in connection with simulating total organic carbon (TOC), as well as absorbance and iron in streams (Hytteborn et al., 2015; Temnerud et al., 2014). Here we used Fluxmaster to estimate detrended long-term fluxes for TIN and to predict long-term mean annual TIN loads for each monitored site in a future climate in Sweden.

2.6.2. Regression Models within Fluxmaster

One of the most basic built-in RMs in Fluxmaster relates the logarithm of TIN concentration \( \ln(TIN_t) \) to logarithmic daily streamflow \( \ln(Q_t) \) (Robertson & Saad, 2011). However, the strong seasonality of TIN across Sweden shows that concentrations are not only sensitive to flow, but also to seasonal changes in air-temperature. Thus, we extended the basic RM to include the logarithm of daily air temperature \( \ln(T_t) \) into our initial regression equation:

\[
\ln(TIN_t) = a + b \cdot \ln(Q_t) + c \cdot \ln(T_t)
\]  

(3)

where the lowercase letters \( a \), \( b \) and \( c \) are coefficients to be calibrated for each study site; \( t \) indicates the time step (here one day).

Fluxmaster allows for modifying the built-in equations and extending them with other variables in order to consider different log-linear RMs. We identified the following five additional variables (cf. Table 4) to potentially drive TIN concentrations in Sweden: (1) decimal time \( t \) to account for temporal trends (Robertson & Saad, 2011), (2) squared logarithmic daily streamflow \( \ln(Q_t)^2 \), (3) logarithm of streamflow with a one-day lag \( \ln(Q_{t-1}) \), (4) squared logarithmic daily air temperature \( \ln(T_t)^2 \), and (5) logarithm of air temperature with a seven-day (one-week) lag \( \ln(T_{t-7}) \). For the decision on which of these variables to add to equation (3), a model selection procedure was needed. Since the number of contemplated variables (i.e., five) was relatively small, we chose an all-possible-regressions procedure (Gatu et al., 2007; Hocking & Leslie, 1967; LaMotte
& Hocking, 1970; Schatzoff et al., 1968) as an alternative to stepwise regression, which selects a
model (or a subset of models) that optimizes some statistical criterion (Gatu et al., 2007). All-
possible-regression literally bases the selection of a model on the calculation of all regressions of
a dependent variable upon all possible $2^k-1$ subsets of $k$ independent variables (Schatzoff et al.,
1968). Given the $k=5$ potential independent variables suggested above (i.e., $\ln(Q_t)^2$, $\ln(Q_{t-1})$,
$ln(T_t)^2$, $ln(T_{t-7})$ and $t_t^2$), this resulted in $2^5-1=31$ distinct equations (Table 4) with a varying degree
of complexity to be tested in addition to equation (3).

2.6.3. Regression Model Calibration and Validation Procedure

Fluxmaster was used to calibrate the coefficients of the 32 pre-defined RMs (Table 4) for each of
the 19 chosen study sites using the entire records of observed daily streamflow, observed air
temperature and measured TIN samples. The models were then evaluated and ranked for each
individual study site based on the optimization of four evaluation criteria: the coefficient of
determination ($R^2$), the probability of outperforming another RM ($p_i$), the Akaike criterion (AIC)
and the Bayesian information criterion (BIC).

$R^2$ is defined as the proportion of variance explained by the RM (Nagelkerke, 1991) and
measures the goodness of fit in the sense of comparing a model including the independent
variables with a model in which none of the independent variables appear (Barrett, 1974). A
large $R^2$ typically reflects an increase in predictive precision for a RM (Barrett, 1974). For each
study site, we therefore compared all possible 496 combinations of 2 individual models out of all
32 RMs (i.e., $\binom{32}{2} = 496$) and decided with help of an F-test (significance level of 5%) which of
the two compared RMs gave a significantly better fit. This procedure made a ranking of the RMs
based on their probability $p_i$ of being chosen over another model possible.

Both AIC (Akaike, 1974) and BIC (Schwarz, 1978) are parsimony estimators (Aho et al.,
2014) and useful tools to evaluate the performance of RMs whose parameters are estimated with
the maximum likelihood method (Konishi & Kitagawa, 2008):

$$AIC = -2 \cdot \ln(L) + 2 \cdot k$$  \hspace{1cm} (4)

where $L$ is the maximum value of the model’s likelihood function and $k$ is
the number of independent RM variables.

$$BIC = -2 \cdot \ln(L) + k \cdot \ln(n)$$  \hspace{1cm} (5)

where $n$ is the sample size (i.e., the number of observations).

Both criteria do not only take the goodness of fit (measured by the maximized likelihood)
into account, but also consider simplicity in terms of the number of model variables (Aho et al.,
2014). According to Aho et al. (2014) the decision of whether to use AIC or BIC for a given
application depends on the research question: In exploratory analyses with multiple incompletely
specified or infinite RMs (“Which model will best predict the data?”), AIC seems to be more
appropriate, while BIC should be the first choice in confirmatory analyses with a small number
of completely defined models (“Which model generated the data?”). Since the goal of this study
is to produce future predictions as accurate as possible and not finding the one-and-only correct
model, AIC was favored.
Based on these four criteria $R^2$, $p_i$, AIC and BIC averaged over all study sites (Table 4), the best performing RM structures were selected. Additionally, their performance under conditions different from those that they were calibrated to (so-called ‘operational adequacy’) was evaluated with help of a split-sample test (Klemeš, 1986): for the five study sites with the longest complete records of observed data (cf. Table 3, Figure 1c), the best RMs were calibrated based on the period 1981-1995, while they were validated on the period 1995-2010. The validation period was on average 0.6°C warmer and slightly wetter (+2% precipitation) with 7% less streamflow than the calibration period.

### 2.6.4. Simulations of Historical and Future Nitrogen

The best performing RMs were then used to create an ensemble of daily nitrogen load simulations at each of the 19 study sites for the entire reference control period (1961-2010) and the future scenario period (2061-2090) using the bias-corrected temperature simulations from the RCMs and the 1500 HBV-modeled streamflow series as input. Such a large multi-model-multi-catchment ensemble provides more robust results as it considers a more realistic range of uncertainties (Teutschbein et al., 2011). Simulations of the reference control period (1961-2010) were compared to observations to analyze the ability of the climate-model-data-driven Fluxmaster setup to represent present-day conditions.

We would like to emphasize that the results presented are mostly based on median values of the ensemble simulations, which are considered statistically to be the most likely future outcomes as they attach reduced importance to outliers.

### 2.6.5. Upscaling of Simulated TIN Loads to the Baltic Sea

In terms of their landuse, the chosen study sites can be considered to be representative for most areas in Sweden, except for intensively farmed areas in southern Sweden. To provide a more realistic nationwide estimate of future annual TIN loads from Swedish terrestrial areas (30,080 km$^2$ agricultural land, which equals 7.37% agriculture) to the Baltic Sea, a regression approach was used. First, a linear RM was fitted (for each simulation ensemble member) using the fraction of agriculture in the individual catchments as explanatory variable and the simulated TIN loads during the reference period as response variable. Then, another linear RM was fitted (again for each ensemble member) using the fraction of agriculture in the individual catchments as explanatory variable and the projected change in TIN loads as response variable. Finally, the resulting linear regression equations and their coefficients were used to estimate an ensemble of present-day and future TIN loads exported from the entire Swedish land mass with 7.37% agriculture.

### 2.6.6. Effects of Land Use Change

The linear RMs that were previously established to upscale TIN loads to the entire Swedish land mass (section 2.6.5.) can also be used to estimate present-day and future TIN loads exported from Sweden assuming alternative futures with different fractions of agricultural land. This was utilized here to find the required reduction of agricultural land area that would have to occur to either keep the current TIN export rates to the Baltic Sea at a constant level or to cut them by 6% respective 13%, which are the latest HELCOM waterborne-nitrogen reduction goals for Sweden respective the entire Baltic Sea Basin (HELCOM, 2015).
2.6.7. Separating the Effects of Future Streamflow and Concentration Changes

Changing climate conditions affect both the actual streamflow and the iN concentrations, which in turn influence the nitrogen loads exported to the Baltic Sea. In an attempt to distinguish between these two effects, a simple experiment was set up as follows.

First, long-term monthly mean concentrations ($\overline{TIN}_m$) were estimated both for the entire reference control period (1961-2010) and the future period (2061-2090) using the daily nitrogen concentration simulations by Fluxmaster at each of the 19 study sites. Similarly, long-term monthly mean streamflow values ($\overline{Q}_m$) were calculated using the daily HBV-simulated values for both periods. Given that TIN loads are calculated as the product of TIN concentrations and streamflow, an estimate of average historical TIN loads (annual) can be calculated as follows:

$$L_{TIN,1961-2010} = \sum_{m=1}^{12} (\overline{TIN}_{m,1961-2010} \cdot \overline{Q}_{m,1961-2010})$$ (6)

where $L_{TIN}$ is the annual TIN load/export, $TIN_m$ the long-term monthly mean TIN concentrations, and $Q_m$ the long-term monthly mean streamflow.

Thereafter, three scenarios were considered for future TIN loads: Scenario 1 accounts for changing TIN concentrations with no shift in streamflow (equation 7), scenario 2 assumes shifting streamflow with no change in concentrations (equation 8), and scenario 3 considers both concentration and streamflow changes (equation 9).

$$L_{Scenarios,1} = \sum_{m=1}^{12} (\overline{TIN}_{m,2061-2090} \cdot \overline{Q}_{m,1961-2010})$$ (7)

$$L_{Scenarios,2} = \sum_{m=1}^{12} (\overline{TIN}_{m,1961-2010} \cdot \overline{Q}_{m,2061-2090})$$ (8)

$$L_{Scenarios,3} = \sum_{m=1}^{12} (\overline{TIN}_{m,2061-2090} \cdot \overline{Q}_{m,2061-2090})$$ (9)

The final step involved the comparison of the three different calculated future loads with the historical TIN loads to estimate the projected change for each of these scenarios.

2.6.8. Quantification of Uncertainty Sources

To quantify the uncertainty sources in our projections of future riverine inorganic nitrogen yield, we followed the procedure suggested by Bosshard et al. (2013) that decomposes the sum of squares as described within the analysis of variance (ANOVA) theory. Thus, an ANOVA-based approach was used to separate the total ensemble uncertainty into the contributions from (1) the different driving GCMs, (2) the RCM simulations, (3) the 100 HBV parameterizations and (4) the different RMs used for TIN modeling. This approach also enables
analysis of interactions between these three uncertainty sources and is feasible for any relevant variable (Bosshard et al., 2013).

Using ANOVA theory we split the total sum of squares (SST) into sums of squares (SS) caused by each individual source (i.e., GCM, RCM, HBV and RM) and into the remaining sum of squares due to their interactions (SSI):

\[
SST = SS_{GCM} + SS_{RCM} + SS_{HBV} + SS_{RM} + SSI
\]  

As proposed by Bosshard et al. (2013), a variance fraction \( \eta^2 \) was then derived for each individual source and their interactions as follows:

\[
\eta^2_{GCM} = \frac{SS_{GCM}}{SST}, \eta^2_{RCM} = \frac{SS_{RCM}}{SST}, \eta^2_{HBV} = \frac{SS_{HBV}}{SST}, \eta^2_{RM} = \frac{SS_{RM}}{SST} \text{ and } \eta^2_{interactions} = \frac{SSI}{SST}
\]  

Therefore, the contribution of an individual source to the total modeling uncertainty of the ensemble is reflected by the values of the variance fraction ranging from 0 to 1 (corresponding to 0%-100%).

First, we performed the ANOVA approach for the future TIN loads estimated for entire Sweden, followed by an ANOVA for each study site individually.

3 Results

3.1. Projected Climate Change

Regional climate model simulations indicated a considerable warming combined with a precipitation increase throughout the entire year in a future Sweden. More specifically, annual mean air temperature in the selected study sites was projected to increase on average by 2.6 to 4.3°C, with a more pronounced increase in the North (Figure 3a). Similarly, total annual precipitation was estimated to increase by 10 to 23 %, with a stronger change signal in the North (Figure 3b). Not only did the magnitude of future changes in temperature and precipitation depend on the catchment location (i.e., latitude), it also featured distinct seasonal patterns (Figure 3a,b) with a larger increase during the colder season (October-March). The rising temperatures in the selected study sites will inevitably push the spring thaw, the start of spring and the beginning of the growing season earlier. According to SMHI’s national definition of spring (i.e., the first day on which the average temperature is between 0°C and 10°C for at least seven consecutive days), spring currently starts on average between February in southern Sweden and May in northern Sweden, but was projected to begin 14-40 days (2-6 weeks) earlier in a future climate. Rising temperatures also imply that the winter period with temperatures below the freezing point will shorten considerably, while the growing season (defined as part of the year with daily mean temperatures above +5°C) will lengthen by 35-50 days (5-7 weeks) in the 19 study catchments (with a stronger extension in the northernmost sites).

3.2. Hydrological Modeling

3.2.1. HBV Performance

The calibration-validation procedure against observed streamflow using measured precipitation and streamflow as input to the HBV model highlighted only small differences between calibration and validation period: The calibrated HBV model simulated present-day
observed streamflow during the calibration period (1991-2000) with fuzzy measure \( (X) \) values ranging from 0.74 to 0.96 (where 1.0 specifies a perfect fit) combined with high \( R_{eff} \), \( L_{eff} \) and \( VE \) values (Table 2) for the different catchments. Observed and simulated streamflow (both with and without consideration of seasonal fluctuations) were strongly positively correlated, which was reflected in a high \( SC \) (0.81-0.98) and \( AC \) (0.59-0.91). During the validation period (2001-2010), the model performance decreased somewhat with \( X \) values lying between 0.59 and 0.91 (Table 2). Both \( SC \) and \( AC \) decreased only slightly during the validation period (\( SC: 0.78-0.96, AC: 0.56-0.96 \)).

The good performance of the HBV model persisted even in more extreme situations that are more similar to a future climate (Figure 4a,b). For example, in 2008, a rather wet and warm year with precipitation 12% above average and temperature 1.2 °C above average in our study sites, there was no significant loss of model performance (\( X \) ranging from 0.59 to 0.92) compared to the entire validation period 2001-2010 for any of the study sites.

3.2.2. Simulations of Historical and Future Streamflow

When driven with bias-corrected precipitation and temperature series from the 15 RCMs, the calibrated HBV model simulated historical long-term monthly mean streamflow values reasonably well (Figure 5), indicating that the climate data used in this study is suitable for hydrological modeling of the selected study sites. Simulations of future streamflow (2060-2090) in the study sites indicated a total annual increase in the range of 19-119 mm/d with a median of 50 mm per year, which equals about 5-29% (on average 13 %) more streamflow. Future annual hydrographs were characterized by noticeably higher winter streamflow and considerably lower spring floods (Figure 3c,d), leading to a lower amplitude and a phase shift. On average, streamflow during the winter months December-February was projected to increase by 0.6 mm/d (70%), but the range of change varied in the selected study sites between 0.3 and 0.9 mm/d (30-160%). During the month of the spring flood, projected streamflow decreased on average by 0.5 mm/d (-22%), with the different study sites ranging from -2.1 to +0.2 mm/d (-64% to +15%). Projected streamflow changes indicated a clear pattern along the latitudinal gradient with changes in northern Sweden (snow-melt driven) being more pronounced than in southern Sweden (rainfall-driven). Winter streamflow in the northern study sites is projected to increase more than in the southern study sites (+127% versus +56%), while spring streamflow decreases more (-41% versus -17%).

3.3. Nitrogen Modeling

3.3.1. Fluxmaster Performance

All 32 RMs provided good simulations of TIN concentrations after calibration. Averaged over all 19 study sites, \( R^2 \) values ranged from 0.42 to 0.55, \( p \) varied from 1.1% to 5.1%, AIC values were within the interval 539 and 605, and BIC values lay within 567 and 615. Based on these four evaluation criteria (Table 4), the following top five RM structures (no. 21, 26, 30, 31 and 32 in Table 4) were chosen to predict future TIN with Fluxmaster:

\[
\ln(TIN_t) = a + b \cdot \ln(Q_t) + c \cdot \ln(T_t) + d \cdot t_t + f \cdot \ln(Q_{t-1}) + h \cdot \ln(T_{t-7})
\]

(12)

\[
\ln(TIN_t) = a + b \cdot \ln(Q_t) + c \cdot \ln(T_t) + f \cdot \ln(Q_{t-1}) + g \cdot \ln(T_t)^2 + h \cdot \ln(T_{t-7})
\]

(13)
\[ \ln(TIN_t) = a + b \cdot \ln(Q_t) + c \cdot \ln(T_t) + d \cdot t_t + f \cdot \ln(Q_{t-1}) + g \cdot \ln(T_t)^2 + h \cdot \ln(T_{t-7}) \] (14)

\[ \ln(TIN_t) = a + b \cdot \ln(Q_t) + c \cdot \ln(T_t) + e \cdot \ln(Q_t)^2 + f \cdot \ln(Q_{t-1}) + g \cdot \ln(T_t)^2 + h \cdot \ln(T_{t-7}) \] (15)

\[ \ln(TIN_t) = a + b \cdot \ln(Q_t) + c \cdot \ln(T_t) + d \cdot t_t + e \cdot \ln(Q_t)^2 + f \cdot \ln(Q_{t-1}) + g \cdot \ln(T_t)^2 + h \cdot \ln(T_{t-7}) \] (16)

According to the split-sample test (Klemeš, 1986), there were no distinct differences in the performance of these five models for a given study site (Table 5). However, the general suitability of RMs strongly depended on the study site, with Kåfalla and Sävja featuring rather high \( R_{eff} \) values (i.e., 0.7-0.9) and Edsforsen Krv rather low \( R_{eff} \) values (i.e., 0.4-0.5) during the calibration period (Table 5). During the independent validation period, \( R_{eff} \) values were only somewhat lower for four of the study sites indicating an acceptable operational adequacy, while Edsforsen Krv featured a considerable drop in \( R_{eff} \) values (Table 5). The five RMs also showed a good performance in more extreme situations that better resemble future climate conditions. For example, in the unusually warm and wet year 2008, the simulated TIN loads captured the observed dynamics well (Figure 4c,d).

### 3.3.2. Simulations of Historical and Future Nitrogen

For each catchment we simulated an ensemble of 7500 TIN loads (15 RCMs x 100 HBV parameter sets x 5 RMs) for the reference periods and an ensemble of 7500 TIN loads for the future period. When driven with historical climate model data, Fluxmaster was able to reproduce the seasonal pattern of observed loads and of Fluxmaster simulations driven with observational data for most catchments (Figure 6). TIN load patterns showed generally high loads in winter with a subsequent peak in spring and considerably lower loads during summer. Climate model driven Fluxmaster simulations tended to provide somewhat lower load estimates than Fluxmaster driven with observed streamflow and temperature except for Kåfalla (Figure 6).

Both the simulated historical TIN concentrations (Figure 7a) and historical TIN loads (Figure 7b) were linearly related to the percentage agriculture in a catchment. Fluxmaster driven with future climate model data simulated a future decrease in TIN concentrations (Figure 7c) of -22 µg/l (-20%) on average, but an increase in annual TIN loads (Figure 7d) of +0.07 kg/ha/yr (+6%) on average for all study sites. The magnitude of projected future change in TIN loads was linearly dependent on the percentage of agricultural area in a catchment (Figure 7e).

Not only annual TIN values were projected to change in a future climate, also seasonal patterns will likely be altered under changing climate conditions (Figure 8). In terms of concentration changes, only a weak pattern was seen: while the majority of study sites (17 out of 19) featured some decrease during warmer months (Figure 8a,b), in winter the direction of changes varied with some (mostly southern) study sites pointing towards higher concentrations and some towards lower concentrations. For TIN loads, however, we got a much clearer signal: winter TIN load will increase considerably while summer loads were projected to decrease (Figure 8c,d).
3.3.3. Upscaling of Simulated TIN Loads to the Baltic Sea

The selected study sites covered a combined total area of 52,554 km$^2$, which exported on average 2.50 kt/yr TIN (ensemble median) to the Baltic Sea during the reference period (Figure 9a, left). In the future, total TIN export was projected to reach 2.64 kt/yr (Figure 9a, middle), which equals a 6% increase (+140 t/yr) by the end of the century (Figure 9a, right).

After fitting a linear RM to the relationship between the percentage of agriculture and the simulated present-day TIN loads (equation 17, Figure 7b), the total TIN load exported from Sweden (7.37% agricultural land area) to the Baltic Sea during the reference period was estimated to range from 30 to 36 kt/yr, with a median value of 32.8 kt/yr (Figure 9b, left).

$$TIN\ load\left(\frac{kg}{ha\cdot yr}\right) = c_1 \cdot \text{percentage}\ agriculture + c_2$$  \hspace{1cm} (17)

where $c_1$ and $c_2$ were the regression coefficients (i.e., slope and intercept).

Based on another linear RM fitted to the relationship between agriculture and the simulated future change in TIN loads (equation 17, Figure 7e), future changes were projected to vary between -24% and +53% (+14% on average, interquartile range 0-29%), which consequently led to future TIN export estimates of 25 kt/yr to 52 kt/yr (with a median value of 37.3 kt/yr) towards the end of this century (Figure 9b, middle). Even though the majority of simulations (75%) projected an increase in TIN (Figure 9), it should be noted that 25% of the simulations indicated a possible reduction in TIN. Not all projected changes were, however, significant: A two-sample t-test indicated that only one third of all simulations projected significant changes (25% significant increase and 8% significant decrease in TIN load), while the remaining 67% were not changing significantly.

$$\Delta TIN\ load\left(\frac{kg}{ha\cdot yr}\right) = c_3 \cdot \text{percentage}\ agriculture + c_4$$ \hspace{1cm} (18)

where $c_3$ and $c_4$ were the regression coefficients (i.e., slope and intercept).

3.3.4. Effects of Land Use Change

Based on equations (16) and (17), we estimated hypothetical future TIN loads from Swedish terrestrial areas to the Baltic Sea assuming different fractions of agricultural area (Table 6). We found that the agricultural area would have to be reduced by approximately 15% (from 30,080 km$^2$ to 25,568 km$^2$) to keep the TIN export rate at the same level as today’s load of 32.8 kt/yr (Table 6, bold/underlined). In order to reach the HELCOM goal of cutting TIN exports by 6% down to 30.8 kt/yr (Swedish reduction target) respective 13% down to 28.5 kt/yr (Baltic Sea Basin reduction target), the agricultural area would have to be cut by 21% respective 29% (Table 6, italic/underlined). In other words, anthropogenic TIN sources from agricultural areas need to be reduced by at least 21% to reach acceptable future loads to the Baltic Sea, if reduction in area of agriculture is the only measure taken to reduce loads.
3.3.5. Separating the Effects of Future Streamflow and Concentrations Changes

In scenario 1, which assumed only changes in concentrations but not in streamflow, the TIN exported on average by the selected study sites (covering an area of 52,554 km$^2$) would decrease by 297 t/yr (-12%). According to scenario 2, which is based on increased streamflow in the future but stationary concentrations, the selected study sites would export 494 tons more per year, which equals an increase by 20%. In scenario 3, which represented the actual climate change impacts with changes in both streamflow regimes and TIN concentrations, the selected study sites would export 117 t more per year (+5%) compared to the 140 t/yr (+6%) that were projected based on our more complex modeling procedure (cf. section 3.3.3). Consequently, the decreasing TIN concentrations considerably mitigate the effects of increasing streamflow: Instead of a 20% increase, TIN loads increase by only 5% in this simple experiment.

3.3.6. Quantification of Uncertainty Sources

In the present study, the choice of climate models (both GCM and RCM) caused by far the largest uncertainties. When looking at the projected annual TIN changes for entire Sweden, GCMs contributed 53%, RCMs 26% and the combined interactions another 21% of the uncertainties, while the importance of the different HBV parameterizations (0.4%) and the chosen RMs (0.3%) became relatively small. However, when studying the future projections of a single study site, HBV parameterizations (contribution of 0.5-21%) and the different RMs (contribution of 0.1-19%) gained importance. It should be noted that we did not account for prediction and confidence intervals of the RMs, which might have diminished the real contribution of the RMs to uncertainty to some degree.

4 Discussion

4.1. Projected Climate Change

In this study, we incorporated simulations of 15 RCM simulations under GHG emissions scenario A1B provided by the ENSEMBLES project (Van der Linden & Mitchell, 2009), which covers a wide range of possible future climate conditions. The modeled increases in temperature and precipitation are of similar magnitude and follow similar patterns as projections in other recent climate change studies over Scandinavia (Arheimer & Lindström, 2015; Beldring et al., 2008; Koca et al., 2006; Moore et al., 2008; Teutschbein et al., 2011), as well as the results of the ClimateCost project (Christensen et al., 2011) and the Fifth Assessment Report (AR5) of the United Nations Intergovernmental Panel on Climate Change (IPCC, 2014).

4.2. Hydrological Modeling

The simulation of current and future hydrological regimes of 19 different catchments in Sweden was based on 73,500 simulated streamflow time series for the reference control period 1961-2010 and on 43,500 series for the future period 2061-2090 that were simulated with the HBV software package. Such a large streamflow ensemble for each of the selected study sites provided a wide variety of possible outcomes, but also more robust results, because we considered uncertainties in climate model data by employing 15 different RCM projections and uncertainties in hydrological model parameters by using 100 optimal parameter sets.

The applied HBV model effectively simulated the key features, such as water balance, seasonal flow patterns, as well as high and low flow events for each study site when driven by
observed meteorological data. When forced with climate data provided by RCMs during the
reference period (1961-2010), the relatively good performance was successfully validated, which
enabled us to draw robust conclusions about potential future streamflow changes from the
subsequent analyses.

Our hydrological simulations confirmed previous climate change impact studies
(Andréasson et al., 2004; Arheimer & Lindström, 2015; Bergström et al., 2001; Graham et al.,
2007; Teutschbein et al., 2011; Teutschbein & Seibert, 2012) by showing that changing climate
conditions will perturb current annual and seasonal streamflow in Sweden. Accordingly, in
northern Sweden, streamflow regimes are projected to change from snow-melt driven regimes
with very low winter flow and a dominant spring flood peak to regimes with much higher winter
flows, as well as spring floods with earlier initiation and lower peaks. Consequently, future flow
regimes in these catchments are likely to resemble today’s streamflow regimes of regions further
south (e.g., central or southern Sweden). Such strong hydrological shifts will potentially
influence stream water levels and velocities, which in turn are important for aquatic ecology,
water quality, riparian zone processes, stream bed erosion and sediment transport.

4.3. Nitrogen Modeling

Nitrogen simulations were produced with different selected RMs in Fluxmaster using
available measurements of streamflow and air temperature. Our model selection approach based
on four optimization criteria ($R^2$, $p_i$, AIC and BIC) resulted in five equally likely RM structures.
The parsimony, fast run times, and low calibration efforts of simple linear RMs are great
advantages over complex process-oriented models. However, simple RMs are likely not able to
capture complex processes in a catchment such as shifting vegetation behavior or in-stream
processes as a response to changing climate conditions. Furthermore, the projections were made
under the assumption of constant nitrogen input from landuse activities over time. This
assumption is supported by Basu et al. (2010), who demonstrated that anthropogenic impacts
have contributed to an effective biogeochemical stationarity in managed catchments in the Baltic
Sea Drainage Basin. In this sense, stationarity implies temporal invariance in the annual flow-
weighted concentration and suggests that load variations are mainly controlled by streamflow.
Indeed, all RMs relied on the assumption of stationarity, in that the statistical relationship (i.e.,
the coefficients) established between predictor (i.e., streamflow and air temperature) and
predictand (i.e., TIN load) does not change over time. Consequently, the more heuristic the
predictor-predictand relationship becomes, the less confident we can be that the established
statistical relationship holds under changing climate conditions. But climate change impact
studies require the chosen approach to function in a perturbed climate (Maraun et al., 2010).
Therefore, we tested the RMs operational adequacy (Table 5), and showed that all five RMs
perform reasonably well (when driven with observed data) under conditions different from those
that they were calibrated to, as well as during a period in the observational record that was
markedly wetter and warmer (Figure 4). Even when driven with climate model data, observed
values were sufficiently reproduced during the reference period (1961-2010). Thus, we
concluded that the RM approach is valid as an initial effort to maximize the information value of
existing observed data and to draw reliable conclusions about potential future TIN load changes.
Still, it is likely that the assumption of stationarity is more appropriate for our catchments that
have a history of agricultural activity and thus a larger accumulation of soil N that can be acted
upon by future alterations to the flow regime.
4.4. Present and Future Nitrogen Loads

Our simulations of TIN loads during the reference period were in line with approximations provided in the literature. For instance, data provided by Savchuk et al. (2012) through their decision support system Nest (http://nest.su.se/nest/) suggest a total TIN export from Sweden to the Baltic Sea for the period 1971-2000 of 38 kt/yr including point sources discharging directly to the sea. Compared to our estimate of 33 kt/yr for 1961-2010, which does not include point sources, this adds more confidence to the reliability of our modeling approach. In terms of future TIN loadings, the overall simulated signal was positive and implied an average increase of roughly 14%. This supports our hypothesis that increasing winter streamflow (caused by future alterations to precipitation and temperature patterns) will cause TIN export rates to increase. It should however be noted, that not all of the 7500 ensemble members projected a future increase: one quarter of the simulations indicated a possible reduction in TIN load.

We have shown that TIN export is tightly linked to the amount of agriculture in a catchment, but the controls over these losses may be complex. In agricultural systems, fertilizer use and animal wastes from farming are well known sources of iN in surface waters in Sweden (Hägg et al., 2011), yet nutrient delivery to streams is further modified by various processes (e.g., denitrification and plant uptake) that remove nitrate in riparian soils (Ranalli & Macalady, 2010). It is difficult to predict how these processes may change under future hydrological regimes, but in the case of denitrification, there is consensus that N removal increases with longer residence time in carbon-rich, anoxic environments (Pinay et al., 2015). Thus, with predicted shifts toward higher and more intense flow regimes, hydrologic transport rates through riparian soils may more frequently outpace removal rates, reducing the efficiency of these buffer zones (Ocampo et al., 2006). Under such circumstances, our projections may therefore underestimate future N export in agricultural systems.

As indicated above, our assumption of stationarity is likely weaker for forested catchments that do not have a strong legacy of soil N enrichment (Basu et al., 2010), and as such we may be overestimating future N losses from these systems. Indeed, there are reasons to expect that terrestrial N demand and limitation may increase in response to climate change (Wieder et al., 2015), thereby reducing the size of the iN pool subject to hydrologic losses. For example, Sweden has experienced continuously increasing levels of forest growth and biomass - under fairly low and relatively stable inputs of nitrogen (SLU, 2016). Such a trend could lead to progressive nitrogen limitation (Luo et al., 2004), which may ultimately influence hydrologic losses in the Baltic proper (Lucas et al., 2016). Indeed, theory predicts that under severe N shortage, seasonal patterns in stream DIN should become greatly diminished (Stoddard, 1994).
In addition, while N export displays seasonal variation that is potentially linked to biological demand on land (Birgand et al., 2007; Sponseller et al., 2014), different plant species differ in their ability to respond to changes in growing conditions including temperature and season length. Norway spruce, for instance, is able to photosynthesize whenever there is sufficient temperature, water and light (Bergh & Linder, 1999), while deciduous trees such as birch and aspen are confined to periods of leaf out. In catchments dominated by coniferous forest, warmer winters may thus extend the period under which plant nutrient uptake occurs, with potential implications for hydrologic losses.

Ultimately, it remains an important challenge to understand how forest ecosystem responses to future climate change, including a potential ‘tightening’ of the N cycle (cf. McLauchlan et al., 2017), will interact with hydrologic changes to shape patterns of nutrient loss from boreal catchments. Perhaps the greatest unknown in high latitude catchments is the effect of changing winter conditions on soil processes, which can be important for the regulation of ecosystem nitrogen retention (Groffman et al., 2001). As winter temperatures warm, large areas of Sweden are likely to move from continuous to discontinuous snow pack, thus increasing soil freezing depth, and freeze-thaw cycles, with profound effects on soil microbial processes (Öquist et al., 2009) and material fluxes (Haei et al., 2010). Ecosystem responses to these disturbances are complex, affected by multiple factors, and remain a challenge to predict (Groffman et al., 2011). However, Brooks et al. (1998) suggested that microbial biomass in soils below a continuous snow cover provides a buffer, which limits TIN export to streams during spring snow-melt. Consequently, a future loss in snow pack potentially increases the nitrogen losses (Brooks & Williams, 1999), which in turn indicates an underestimation of our projections. Adding to this complexity, a recent global analysis suggested that the soil carbon (C) pool in boreal ecosystems is particularly sensitive to temperature change (Crowther et al., 2016). Given clear coupling between soil C and N cycles (Högberg et al., 2017), the implications of changes to this large organic matter pool for ecosystem N losses merit further attention.

Despite these uncertainties, our projections that are essentially a result of streamflow shifts are valuable and the projected range of change (-24% to +53%) with a median of 14% and an interquartile range of 0-29% seems reasonable. While we could not find any study on future TIN export to directly compare with these values, recent studies by Meier et al. (2012) and Hägg et al. (2014) provided estimates for future total nitrogen (i.e., the sum of organic and inorganic nitrogen) in this region. For example, Meier et al. (2012) suggested a change in total nitrogen export (though for the entire Baltic Sea drainage basin) within the range of -10% to +64%. Similarly, Hägg et al. (2014) projected changes in total nitrogen export of -10% to +26% for the entire Baltic Sea drainage basin. In both of these studies, a reduction of nitrogen loads could only be obtained in the most optimistic scenarios, which assumed targeted management strategies to be applied to achieve immense reductions of riverine nitrogen concentrations.

Our present study indicates that a considerable increase in TIN loads from the Swedish land area to the Baltic Sea is likely based on changing climate conditions. The projected rise in winter streamflow and winter TIN loads is greater than the reductions observed during spring flood, which consequently leads to an overall increase of TIN loading. But our simulations revealed not only a shift in the total amount of TIN, but also suggested a shift in the seasonal pattern. At present, the annual cycle of TIN loads is characterized by high values during winter, an even higher peak at the end of the winter (spring flood), and rather low values during summer, which start to rise in autumn as the dormant season starts. In a future climate, our projections
indicated even lower summer TIN loads, a longer period of low summer loads, a faster increase
during autumn, and much higher loads during the somewhat shorter winter period. Given that
inorganic nitrogen is already an important limiting nutrient in streams and lakes during
summer/autumn in the boreal region (Bergström et al., 2013; Burrows et al., 2015), decreasing
TIN loads and concentrations during the time of year when resources are in greatest demand
could have important consequences for aquatic ecosystems and food webs in the future.
Concurrently, higher winter loads entering the Baltic Sea could, in combination with higher sea
surface temperatures, have harmful effects. In the past, Baltic Sea surface concentrations of
nitrate in winter increased threefold from 1970 to 1985 as a direct response to doubling riverine
nitrogen loads (Neumann et al., 2002). Meier et al. (2011) demonstrated in a modeling study that
increased future nitrogen loads to the Baltic Sea would cause higher phytoplankton
concentrations, leading to earlier and more intense spring blooms in regions where light is not
limiting. Additionally, warmer future temperatures may significantly amplify the effect of
increased nutrient loads on phytoplankton production (Meier et al., 2011). It should, however, be
noted that nutrient limitation of phytoplankton varies within the Baltic Sea basin: Rolff and
Elfwing (2015) found that spring bloom in the Bothnian Sea is more nitrogen-limited than in
other basins. Consequently, this basin is likely to be more sensitive to increasing riverine
nitrogen loads.

Given that future changes in phytoplankton biomass in the Baltic Sea are expected to
arise from changes in nutrient availability (Meier et al., 2011), reducing TIN loading has become
a priority. One suggested goal is to reduce waterborne N export from Sweden to the Baltic Sea
by 6% (HELCOM, 2015). In modeling experiments, such reductions have been shown to reduce
biomass of diatoms, flagellates and zooplankton with large spatial variations in the Baltic proper
(Neumann et al., 2002). Typically, these results do not account for climate change effects and
related flow-driven losses, which could cause – as suggested in this paper - higher TIN loadings,
which in turn could diminish the effects of such politically agreed nutrient reduction input goals.
Consequently, there is potentially a need to reduce anthropogenic nitrogen from sources such as
agriculture, forestry, peat mining, sewage and other industry beyond the reduction targets to
reach acceptable future loads to the Baltic Sea (Enell & Fejes, 1995).

4.5. Uncertainties

Assessments of future climate change impacts on ecosystems, people’s lives and
livelihoods all over the world are typically impeded by three different sources of uncertainty:
internal climate variability, model uncertainty and scenario uncertainty (Hawkins & Sutton,
2009). Climate projections with longer time horizons (like the ones in the present study) are
dominated by model and scenario uncertainty (Hawkins & Sutton, 2009), whereas internal
variability (i.e., natural fluctuations of the climate system) is only of importance for shorter
decadal predictions (Foley, 2010). For the latter reason and due to a lack of transient RCM
simulations based on SRES scenarios other than A1B, we focused solely on model uncertainty
and addressed neither scenario uncertainty nor internal climate variability.

Model uncertainties can arise from various sources (Kay et al., 2009; Wood et al., 1997),
because the modeling of individual impacts relies on long serial and rather complex modeling
chains, which can cause propagation and interference of uncertainties (Teutschbein et al., 2011).
This was also true for our particular study, because we had to make choices regarding
greenhouse gas emission scenarios, driving GCMs, RCM simulations, downscaling/bias
correction methods, hydrological models and their parameterizations as well as suitable TIN models and their parameterization. In addition, other factors such as vegetation responses to climate change (Ström et al., 2012), changes in water demand (Middelkoop et al., 2001; Vörösmarty et al., 2000) or landuse changes (Vörösmarty et al., 2004) can also contribute to the total uncertainty, but were not directly considered in this study.

Though we mostly presented median values in the results section, our simulations resulted in 7500 possible outcomes for each of the catchments, which enabled us to further analyze and quantify the spread of uncertainties with help of an ANOVA analysis. We showed that the driving climate models affected the resulting impact simulations the most. This finding agrees with previous studies that clearly identified climate models (and especially GCMs) as the main cause of model uncertainty (Barron et al., 2010; Bosshard et al., 2013; Dankers et al., 2014; Kay et al., 2009; Prudhomme & Davies, 2009; Raje et al., 2013). But when analyzing an individual study site, the hydrological model parameterization and the choice of an appropriate nitrogen model was of much greater importance than when upscaling to the entire Swedish land area.

Whether these results are also valid for other geographic regions or other experimental approaches remains to be seen. More detailed process-oriented nitrogen models are required to evaluate the stationarity assumption of landscape ecosystems (i.e., whether the statistical relationship between streamflow/air temperature and TIN load stays the same over time), though these models face considerable challenges in reliably simulating nitrogen limitation and require a better conceptual understanding of the main triggers (Thomas et al., 2015). Meanwhile, this study presents a benchmark against which observed divergence from the predictions in future observations can be used to explore eventual shortcomings in the assumption of stationarity. We also argue that further work is needed to increase our understanding of the propagation and interactions of uncertainties in such complex modeling setups, which might help to conduct climate change impact studies with a more complete uncertainty assessment in the future.

5 Conclusions

Spatial and temporal variation in riverine TIN loads are controlled by hydro-climatic and biogeochemical factors. By making maximal use of observed TIN measurements, this study presents an attempt to systematically quantify how future changes in these factors will affect the rate at which TIN is exported from the Swedish boreal landscape. We hypothesized that higher winter streamflow and warmer temperatures in a future climate will cause higher annual TIN loads entering the Baltic Sea, despite lower concentrations. A systematic ensemble approach was presented to assess climate change impacts on riverine TIN and to quantify associated modeling uncertainties. To our best knowledge, this study is the first one to evaluate empirical TIN models in such a comprehensive framework for multiple sites along a regional gradient. By including seasonal temperature and streamflow effects, our approach offers additional insights to seasonal variability of TIN and provides more robust estimates on future TIN loads exported from Sweden’s land areas in comparison to those obtained from simple linear trend analyses. We demonstrated that the most important consequences of a changing climate manifest themselves in longer growing season and more freshwater (~13%) flowing into the Baltic Sea (especially during winter), both of which directly influence the total amount and the seasonal pattern of TIN loads. Overall, a median increase of 14% (interquartile range 0-29%) in annual TIN loads was projected for 2061-2090. Consequently, we argue that politically agreed nutrient reductions input
goals will be less effective under future climate conditions and that anthropogenic nitrogen sources have to be reduced even further to reach acceptable future loads to the Baltic Sea. Although the results of this study might not be directly transferable to areas with different climate, landuse or catchment conditions, the established framework is versatile and can thus be adjusted to study other water chemistry variables in other regions.

6 Acknowledgements

The Uppsala University Department of Earth Sciences, Program for Air, Water and Landscape Sciences has funded the lead author. Collection and maintenance of the water chemistry database (http://webstar.vatten.slu.se/db.html) has been supported by the Swedish Environmental Protection Agency. The Swedish Meteorological and Hydrological Institute (SMHI) is also acknowledged for maintenance of the PTHBV database with meteorological data and for making both streamflow and geospatial data available on their web page, which has been funded by the Swedish water authorities. The ENSEMBLES data used in this work (http://ensemblesrt3.dmi.dk/) were funded by the EU FP6 Integrated Project ENSEMBLES (contract 505539) whose support is gratefully acknowledged.

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Table 1. Present-day hydro-climatic and landscape characteristics of the 19 selected study sites.

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>Mean</th>
<th>Median</th>
<th>Min</th>
<th>Max</th>
</tr>
</thead>
<tbody>
<tr>
<td>Drainage area [km$^2$]</td>
<td>2,766</td>
<td>413</td>
<td>4</td>
<td>26,885</td>
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<tr>
<td>Annual mean temperature [°C]</td>
<td>2.9</td>
<td>2.7</td>
<td>-3.4</td>
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<tr>
<td>Total annual precipitation [mm/yr]</td>
<td>759</td>
<td>765</td>
<td>613</td>
<td>1,082</td>
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<tr>
<td>Total annual streamflow [mm/yr]</td>
<td>409</td>
<td>384</td>
<td>210</td>
<td>846</td>
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<tr>
<td>Mean annual streamflow [m$^3$/s]</td>
<td>37.9</td>
<td>5.5</td>
<td>0.1</td>
<td>328.8</td>
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<tr>
<td>Rainfall-runoff ratio [-]</td>
<td>0.52</td>
<td>0.50</td>
<td>0.32</td>
<td>0.84</td>
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<tr>
<td>TIN concentration* (µg/l)</td>
<td>226</td>
<td>114</td>
<td>44</td>
<td>1188</td>
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<tr>
<td>Lakes [%]</td>
<td>4.5</td>
<td>4.2</td>
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<tr>
<td>Forest [%]</td>
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<td>77.9</td>
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<tr>
<td>Agriculture [%]</td>
<td>5.3</td>
<td>1.4</td>
<td>0.0</td>
<td>27.5</td>
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</table>

*long-term average values derived from monthly sampling (n = 36–642 sampling dates)

Table 2. Summary of objective functions used for the HBV model calibration and validation procedure. Their applied weights, mathematical descriptions and corresponding ranges for both calibration and validation period are shown.

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<tbody>
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<td>Nash-Sutcliffe efficiency</td>
<td>50</td>
<td>$R_{eff} = 1 - \frac{\sum(Q_{obs} - Q_{sim})^2}{\sum(Q_{obs} - \text{mean}(Q_{obs}))^2}$</td>
<td>0.962$^{\text{max}}$</td>
<td>0.924$^{\text{max}}$</td>
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<td>0.858$^{\text{med}}$</td>
<td>0.781$^{\text{med}}$</td>
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<td></td>
<td>0.719$^{\text{min}}$</td>
<td>0.539$^{\text{min}}$</td>
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<tr>
<td>Nash-Sutcliffe efficiency with log. values</td>
<td>40</td>
<td>$L_{eff} = 1 - \frac{\sum(\ln Q_{obs} - \ln Q_{sim})^2}{\sum(\ln Q_{obs} - \text{mean}(\ln Q_{obs}))^2}$</td>
<td>0.957$^{\text{max}}$</td>
<td>0.895$^{\text{max}}$</td>
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<td>0.685$^{\text{min}}$</td>
<td>0.456$^{\text{min}}$</td>
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<tr>
<td>Volume error</td>
<td>10</td>
<td>$VE = \max \left( 0, 1 - \frac{\sum(Q_{obs} - Q_{sim})^2}{\sum Q_{obs}} \right)$</td>
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<td>0.988$^{\text{max}}$</td>
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<td>0.999$^{\text{med}}$</td>
<td>0.956$^{\text{med}}$</td>
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<td>0.937$^{\text{min}}$</td>
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<td>Fuzzy measure</td>
<td>100</td>
<td>$X = R_{eff} \cdot 0.5 + L_{eff} \cdot 0.4 + VE \cdot 0.1$</td>
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<td>0.744$^{\text{min}}$</td>
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Table 3. Setup of the modeling procedure with consideration of various sources of variability.

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<th>Sources of Variability</th>
<th>Used in this study</th>
<th>Ensemble Size</th>
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<tr>
<td>(1) Greenhouse Gas Emission Scenario</td>
<td>1. A1B</td>
<td>(1)</td>
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<td>(2) Climate Model Combinations</td>
<td>Global Climate Model (GCM)</td>
<td>Regional Climate Model (RCM)</td>
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<td>1. ARPEGE</td>
<td>CNRM-RM5.1</td>
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<td>2. ARPEGE</td>
<td>DMI-HIRHAM5</td>
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<td>3. BCM</td>
<td>DMI-HIRHAM5</td>
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<td>4. BCM</td>
<td>SMHI-RCA3.0</td>
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<td>5. ECHAM5 r3</td>
<td>DMI-HIRHAM5</td>
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<td>6. ECHAM5 r3</td>
<td>ICTP-RegCM3</td>
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<td>7. ECHAM5 r3</td>
<td>KNMI-RACMO2.1</td>
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<td>8. ECHAM5 r3</td>
<td>MPI-REMO5.7</td>
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<td>SMHI-RCA3.0</td>
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<td>HC-HadRM3Q0</td>
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<td>11. HadCM3Q0</td>
<td>ETHZ-CLM2.4.6</td>
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<td>12. HadCM3Q3</td>
<td>HC-HadRM3Q3</td>
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<td>14. HadCM3Q16</td>
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<td>(3) Bias Correction</td>
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<td>(4) Hydrological Model</td>
<td>1. HBV (with 100 optimal parameter sets)</td>
<td>x 100 = 1500</td>
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<tr>
<td>(5) Nitrogen Model</td>
<td>1. Regression Model 21 (6 variables)</td>
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<td>2. Regression Model 26 (6 variables)</td>
<td>x 5 = 7500</td>
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<td>3. Regression Model 30 (7 variables)</td>
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<td>4. Regression Model 31 (7 variables)</td>
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<td>5. Regression Model 32 (8 variables)</td>
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<td>(6) Catchment (SMHI station name/number)</td>
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<td>3. Övre Lainsjärv (No. 1740)</td>
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<td>5. Otvik (No. 2234)</td>
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<td>(7) Time Period</td>
<td>1. Reference Period (1961-2010)</td>
<td>x 2 = 285,000</td>
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<td>2. Future Period (2061-2090)</td>
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* Five study sites with longest observational records that were used for additional split-sample testing
Table 4. Overview of the 32 tested regression models (RMs), their 8 variables and coefficients (a-h) as well as their evaluation criteria: the coefficient of determination ($R^2$), the probability ($p_i$) in % of a model being selected over another model, the Akaike information criterion (AIC) and the Bayesian information criterion (BIC).

<table>
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<tr>
<th>Number of RM</th>
<th>Number of Variables</th>
<th>Intercept</th>
<th>Logarithmic streamflow</th>
<th>Logarithmic air temperature</th>
<th>Decimal time</th>
<th>Squared log. streamflow</th>
<th>Squared log. streamflow with time lag of 1 day</th>
<th>Squared log. air temperature</th>
<th>Squared log. air temperature with time lag of 7 days</th>
<th>$R^2$</th>
<th>$p_i$</th>
<th>AIC</th>
<th>BIC</th>
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* Best 20% values for specific evaluation criterion
† Best RMs that were chosen for all further modeling and analysis
Table 5. $R_{adj}$ values for both calibration (‘cal’) and validation (‘val’) period of the split-sample test for the five best performing regression models (columns) and the five study sites with the longest observed records (rows) when regression models are driven with observed streamflow and temperature.

<table>
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<th>Study sites</th>
<th>Regression model structure</th>
<th>No. 21 (Equ. 12)</th>
<th>No. 26 (Equ. 13)</th>
<th>No. 30 (Equ. 14)</th>
<th>No. 31 (Equ. 15)</th>
<th>No. 32 (Equ. 16)</th>
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Table 6. Simulated present-day and future TIN loads from Swedish terrestrial areas to the Baltic Sea assuming different fractions of agricultural area.

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<tr>
<th>Reduction in agricultural area [%]</th>
<th>Total agricultural area [km²]</th>
<th>Agricultural area as a fraction of total Swedish land mass [%]</th>
<th>Control period (1961-2010) [kt/yr]</th>
<th>Future period (2061-2090) [kt/yr]</th>
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Figure Captions

**Figure 1.** Overview of (a) 440 inorganic nitrogen measurement sites (orange circles), (b) 320 streamflow stations (orange circles) and (c) 19 chosen sites (orange circles) that fulfill the selection criteria in Sweden, with the five sites with longest observation records being highlighted in pink.

**Figure 2.** Observed seasonal patterns of (a) temperature, (b) streamflow, (c) precipitation and (d) TIN concentrations (standardized z-scores) in the 19 selected catchments. Light shades show the 10th-90th percentile range (i.e., middle 80% of the study sites), dark shades show the 25th-75th percentile range (i.e., middle 50% of the study sites) and the dark curve is the median of all study sites.

**Figure 3.** Projected changes in long-term monthly mean values of (a) temperature, (b) precipitation, (c) streamflow expressed as absolute change, and (d) streamflow expressed as relative change as a function of study site latitude.

**Figure 4.** Comparison of simulated and observed streamflow (a and b) and TIN loads (c and d) for stations no. 1911 - Näskv (a and c) and no. 2243-Sävja (b and d) for 2008, which was an exceptionally warm and wet year.

**Figure 5.** Streamflow as simulated by HBV driven with bias-corrected precipitation and temperature series from the 15 RCMs (light/dark blue curves) in comparison with observed streamflow (black dots) for the five study sites with the longest complete records of observed data (cf. Table 3, Figure 1c).

**Figure 6.** Comparison of observed monthly mean TIN loads (black dots) with estimates provided by Fluxmaster driven with observed data (gray/black curve) and provided by Fluxmaster driven with RCM simulations (light/dark blue curves) during the reference period for the five study sites with the longest complete records of observed data.

**Figure 7.** Relationships of simulated TIN concentrations (left panels) and loads (right panels) with agricultural area in each catchment. The median values of Fluxmaster-simulated historical TIN concentrations (a) and loads (b) are shown as a function of agricultural area in each catchment for the reference period 1961-2010. A comparison of present-day (black circles) and future (white circles) median Fluxmaster-simulated TIN concentrations (c) and loads (d) is displayed for each catchment (sorted by agricultural area). The median of projected absolute change in TIN loads (e) is shown as a function of agricultural area in each catchment.

**Figure 8.** Projected median changes in long-term monthly mean TIN as a function of study site latitude: (a) TIN concentration in absolute values, (b) TIN concentration in percentage values, (c) TIN load expressed as absolute change, and (d) TIN load in percent.

**Figure 9.** Overview of simulated TIN loads for the combined area of the selected 19 study sites and upscaled for the entire Swedish land area.
Figure 1.
Figure 2.
Figure 5.
Figure 6.
Figure 8.
Figure 9.