Nitrogen surface water retention in the Baltic Sea drainage basin

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Abstract

In this paper, we estimate the surface water retention of nitrogen (N) in all the 117 drainage basins to the Baltic Sea with the use of a statistical model (MESAW) for source apportionment of riverine loads of pollutants. Our results show that the MESAW model was able to estimate the N load at the river mouth of 88 Baltic Sea rivers, for which we had observed data, with a sufficient degree of precision and accuracy. The estimated retention parameters were also statistically significant. Our results show that around 380 000 tons of N are annually retained in surface waters draining to the Baltic Sea. The total annual riverine load from the 117 basins to the Baltic Sea was estimated to 570 000 tons of N, giving a total surface water N retention of around 40%. In terms of absolute retention values, three major river basins account for 50% of the total retention in the 117 basins; i.e. around 104 000 tons of N is retained in Neva, 55 000 tons in Vistula and 32 000 tons in Oder. The largest retention was found in river basins with a high percentage of lakes as indicated by a strong relationship between N retention (%) and share of lake area in the river drainage areas. For example in Göta älv, we estimated a total N retention of 72%, whereof 67% of the retention occurred in the lakes of that drainage area (Lake Vänern primarily). The obtained results will hopefully enable the Helsinki Commission (HELCOM) to refine the nutrient load targets in the Baltic Sea Action Plan (BSAP), as well as to better identify cost-efficient measures to reduce nutrient loadings to the Baltic Sea.

1 Introduction

Expanding human activities have had a great impact on nutrient dynamics and nutrient export from watersheds (Hill and Bolgrien, 2011; Mayorga et al., 2010). Increased population densities, food production, sewage emissions and fossil fuel combustion are among the driving forces causing increased nutrient mobilisation and alterations to hydrological systems (Mayorga et al., 2010). Increased nutrient export from coastal watersheds has had severe impacts on the ecological functions and community composition of estuaries, with algal blooms, increased water turbidity, oxygen depletion, and severe fish deaths as the most prominent consequences (Kellogg et al., 2010; Mayorga et al., 2010; Hoffmann et al., 2009).

Several geomorphic, hydraulic and biological factors may interact to reduce nutrient export from watersheds (Wollheim et al., 2006). Hejzlar et al. (2009) define retention as the fraction of external nutrient inputs that is retained within watersheds, either in absolute values or relative to the input. For nitrogen (N), the term retention is widely used to describe the processes leading to a temporary immobilisation of reactive (non-N_2) N by incorporation into biomass or sedimentation, or the permanent loss of reactive N by conversion into the non-reactive atmospheric form (N_2) by denitrification (Billen et al., 2009). Nitrogen is primarily removed (or retained) from surface water by denitrification (i.e., the microbial production of N_2 from fixed N), followed by processes such as sorption to sediment or organic matter, and biological uptake (Hejzlar et al., 2009).Results from mass-balance studies across a wide range of geographic scales indicate that watersheds could retain as much as 60-90% of total N inputs (Kellogg et al., 2010). Reduced N export can be achieved by increasing N retention in soils, sediments and biomass, reducing atmospheric and terrestrial N sources, and increasing in-stream N removal and retention processes (Hill and Bolgrien, 2011).

Water residence time is a major factor determining the retention of nutrients in watersheds (Hejzlar et al., 2009), while Hesse and co-workers emphasised the need for better understandding of terrestrial retention (i.e., in soils; Hesse et al., 2013). Watershed characteristics, such as hydrology and geomorphology, strongly control water residence time, and increased water residence time can enhance denitrification processes and thereby reduce N loads to coastal waters (Kellogg et al., 2010; Behrendt and Opitz, 2000). Total N inputs influence denitrification rates, whereas hydrology and geomorphology (or water residence time) influence the proportion of N inputs that are denitrified (Seitzinger et al., 2006). Certain areas within watersheds can be identified as sink areas with regard to N export, often being areas with a relatively long water residence time where biogeochemical processes can transform reactive N into organic N in biomass, or N gases via denitrification (Kellogg et al., 2009), or burial of N in sediments (Harrison et al., 2009). The mitigating effect of these sink areas could in some cases be negligible, especially in cases where such areas are bypassed by N-carrying water flows due to specific land management practices (e.g. tile drains or storm water overflows) (Kellogg et al., 2009). Denitrification processes are favoured in sediments and hypoxic or anoxic bottom waters, particularly in systems with abundant organic carbon (C) and nitrate (Harrison et al., 2009; Mulholland et al., 2008).

The question on how to quantify the retention of nutrients from source to river mouth remains one of the largest uncertainties in river basin management. Several authors (e.g. Mayorga et al., 2010; Seitzinger, 2008) emphasise the need for advances in methods and models for determining the impacts of human activities on nutrient inputs to coastal waters, and a better understanding of the processes leading to retention of N in watersheds. Seitzinger et al. (2002) argue that studies generally have focused on N removal in shorter sub-sections of rivers and emphasise the need for a river network approach if we are to quantify the retention of nutrients relative to total inputs. In later years, a number of models of different complexity have been developed for estimating surface water N retention (e.g. Billen et al., 2009;

Grimvall and Stålnacke, 1996; Hejzlar et al., 2009; Hill and Bolgrien, 2011; Jung and Deng, 2011; Mayorga et al., 2010; Seitzinger et al., 2002).

In this paper, we estimate the surface water retention of N in the Baltic Sea drainage basin with the use of a statistical model for source apportionment of riverine loads of pollutants, the MESAW model (Grimvall and Stålnacke, 1996). Scientifically, estimation of retention is one of the largest challenges in river basin nutrient accounting (i.e. source apportionment and budget calculations), and so is also the case in the Baltic Sea drainage basin.

2 Materials and methods

The Baltic Sea, together with the lakes and watercourses in its drainage basin, represents one of the most intensively monitored aquatic systems in the world, and eutrophication has been identified as a major threat to this system. The total area of the Baltic Sea drainage basin is 1 745 000 km^2, which is around four times the area of the sea itself. The long-term average inflow of freshwater with the rivers is 475 km^{3}yr^{-1} or 15 130 m^{3}s^{-1} (Bergström and Carlsson, 1994; Mörth et al., 2007). Details on population and land use characteristics in the Baltic Sea drainage area can be found in Mörth et al. (2007).

2.1 MESAW input data

Model input data included:

1. Land cover, including cultivated land, wetland, lake area, other land (mainly forest), and total drainage area (Corine Land Cover 2006 raster data)
2. Atmospheric N wet deposition (EMEP; http://emep.int/publ/helcom/2012/index.html)
3. Point source emissions, including emissions from waste water treatment plants (WWTPs) and industry (data from HYDE, EUROSTAT and OECD)

4. Observed annual riverine N load (kg N yr^{-1}) as estimated from riverine N concentration and water discharge data for the time period 1994-2006 (PLC database by HELCOM (www.helcom.fi) and data from Denmark from NERI)

The input data for all basins is found in Table A1.

For the estimation of WWTP emissions, we created a spatially distributed data set of people 'connected' or 'not connected' to WWTPs (primary, secondary and tertiary) within the Baltic Sea river basins. For this, we used spatially distributed population data and national level statistics on WWTP connection. Population numbers for the year 2005 divided into urban and rural population were obtained from the HYDE database (http://themasites.pbl.nl/en/themasites/hyde/). These data were redistributed into a 10x10 km grid. Percentages of population 'connected' and type of waste water treatment were compiled from EUROSTAT (European Commission) and the Organisation for Economic Co-operation and Development (OECD). For Russia, Belarus, Ukraine and Slovakia, only percentage of people 'connected' to any type of waste water treatment was available, so the distribution between primary, secondary and tertiary treatment was based on assumption and expert judgement. Based on these national statistics, the total number of 'connected' people in each country was calculated. The number of 'connected' people was then spatially distributed to the grid cells. The distribution was made based on the assumption that urban population and grid cells with higher population numbers would be more likely to have a municipal WWTP connection than rural and smaller populations. Applying this principle, the grid cells for each country were classified as 'connected' starting with urban populations in a descending order, and continuing with rural population in the same way until the number of 'connected' people reached the number specified by the national statistics. This procedure was carried out for all three treatment types; first tertiary, then secondary and last primary. The number of people 'not connected' to any type of treatment plant was also calculated for each grid cell. Total N emission from WWTPs was then calculated for each grid cell based on the approach of Mörth et al. (2007).

2.2 The MESAW model and model parameterisation

MESAW is a statistical model for source apportionment of riverine loads of pollutants developed by Grimvall & Stålnacke (1996). This model-approach uses non-linear regression for simultaneous estimation of export coefficients to surface waters for the different specified land cover or soil categories and retention coefficients for pollutants in river basins. Examples of application of the MESAW model are given in Lidèn et al. (1999), Vassiljev and Stålnacke (2005), Vassiljev et al. (2008) and Povilaitis et al. (2012). To its character, MESAW, have many common features with the more well-known SPARROW model developed in the U.S. (Smith et al., 1997; Alexander et al., 2000).

The basic principles and major steps in the procedure included: (i) estimation of mean annual riverine N loads for a fixed time period (i.e., the years 1994-2006) at each of the 88 monitoring sites, (ii) derivation of statistics on land cover, lake area, point source emissions and atmospheric deposition (see Section 2.1) for each river basin, and (iii) use of a general non-linear regression expression with N loads at each river basin as the dependent/response variable and basin characteristics as covariates/explanatory variables. This gave the following generalised form of the model (Eq. (1)):

$$L_i = \sum_{i=1}^{n} (1-R)S_i + (1-R)P_i + (1-R)D_i + \varepsilon_i \qquad i = 1,2,...,n \qquad (1)$$

where L_i is the load at outlet of basin i;

S_i is total losses from soil to water in basin *i*;

P_i is the point source discharges (WWTP and industry) to waters in basin *i*;

D_i is the atmospheric deposition on surface waters in sub-basin i;

R denote the retention for the source emissions S, P and D, respectively;

n is the number of basins, and

ε_I is the statistical error term.

The parameterisation of the model is flexible and study area specific depending on the data and expert knowledge. The model is fitted by minimising the sum of squares for the differences between observed and estimated loads. The model can be run based on absolute or relative values. If based on absolute values, the optimisation procedure finds the minimum sum of squares of the absolute differences between observed and estimated transport. This procedure implies that the influence of the different rivers/basins will be a function of size. If relative values are used, the optimisation procedure finds the minimum sum of squares of relative differences between observed and estimated transport. This procedure assumes that all rivers have the same weight in the optimisation routine. In this study, we used relative values in order to give equal weight to small and large river basins.

The total diffuse loss of N from soil to water, S_i, in the i^{th} sub-basin was assumed to be a function of the land cover (Eq. (2)):

$$S_i = (\theta_1 a_{1i} + \theta_2 a_{2i} + \theta_3 a_{3i}) \qquad (2)$$

where a_{1i}, a_{2i} and a_{3i} in our study refer to the areas of three land cover classes, i.e. cultivated land, wetlands and other land (mainly forests), respectively. θ_1, θ_2 and θ_3 are unknown emission coefficients for the three land use categories that are statistically estimated in MESAW jointly with the retention (see Eq. (3) below). The point source emissions, P_i, and atmospheric deposition on surface waters, D_i, were assumed to be known (see Section 2.1).

Throughout the exploratory analysis we found that certain basins deviated from the relationship and in most cases also where geographically located near to each other. Thus we introduced a ‘grouping variable’ according to the following:

$$S_i = (\theta_1 a_{1i} + \theta_2 a_{2i} + \theta_3 a_{3i}) * \omega_j \qquad (2b)$$

where each group j consisted of 2 or more basins depending on the model run (see Table 1) and where ω is the unknown coefficient(s). The model was run with different combinations of basin sub-groups in order to obtain reasonable model coefficients and load estimates (i.e. little deviation between predicted and observed loads). The grouping of basins was based on prior knowledge of similarities between basins as well as geographic location. For example, the 10 smaller Danish sub-basins formed one group, as a residual analysis showed that these sub-basins deviated from the general relationships. In its practical meaning, we simply adjusted the ‘global’ diffuse emission coefficients to the local conditions (despite we don’t know the underlying causes). This can be justified since applying the same coefficient to such a large drainage basin (1 745 000 km^2) seem less logic.

Retention was in our study used as a summarising expression for all hydrological and biogeochemical processes that may decrease or retard the transport of N, e.g. denitrification, sedimentation and biological uptake. Irrespective of the exact retention mechanism, the parameterisation of the retention in the different basins was after several exploratory runs with alternative models done with the following empirical function (Eq. (3)):

$$R_i = 1 - \frac{1}{1+\lambda_1\sqrt{drainagearea_i}} * \frac{1}{1+\lambda_2 \frac{lakearea_i}{drainagearea_i}} \quad i = 1,2,...,n \qquad (3)$$

where λ_1 and λ_2 denotes a non-negative parameter and R_i denote the retention in the i^{th} basin. The empirical function were in our case derived from the conception that the removal of N takes place primarily in the surface waters (both instream and in lakes). The first part of the function reflects the instream retention whereas the second part reflects the retention in lakes and reservoirs. Our assumption was that the removal rate is proportional to drainage basin size and the ratio of the lake to the total drainage area. Subsequently this can be seen as an indirect expression of the water residence time in the river basin.

Moreover, for the sake of simplicity, we assumed that the retention is the same for source categories D, S and P. Finally, by combining the parametric expressions of losses from soil to waters and retention with the empirical data, the θ_1, θ_2, θ_3, $\lambda_1\lambda_2$ and ω parameters were estimated simultaneously.

3 Results and discussion

3.1 Parametrisation results

For estimation of total N retention, the model was first run including only 88 river basins for which we had observed annual N load (Table 1). Among these 88 were 10 smaller Danish sub-basins, all with available monitoring data, but which only constitute parts of the major Danish river basins draining to the Baltic Sea. As given in Section 2.2, the model was run exploratory in order to obtain reasonable parameter coefficients and load estimates (i.e. little deviation between predicted and observed loads). In the final model run (#4 in Table 1 with 9 estimated parameters), including all the 88 basins with observed N load, both retention parameters (λ_1 and λ_2) and land use category 'cultivated' (i.e., θ_1) were statistically significant ($p<0.05$). The land use category 'other' (θ_2 which basically is the forest land) was very close to being statistically significant ($p<0.06$). 'Wetland' (θ_3) was not statistically significant, but this land use category accounts for less than 4% of the total drainage area in the Baltic Sea drainage basin. It should be noted that the classification of wetlands is rather rough from the data source and given as joint expression of all wetlands ranging from marches to peatland bogs. All the four grouping parameters ω_1- ω_4 were statistically significant.

Worthwhile to notice is that these diffuse losses parameters (θ_1 - θ_3) all are given in kg km^{-2} and thus can be interpreted as export or unit-area loss coefficients. Interestingly, our estimates corroborate well with the results of monitored losses from small catchments with relative uniform landuse. For example, the point estimate and standard error for cultivated land gave an estimate of 1073 kg km^{-2} and 109 kg km^{-2}, respectively (Model run #4; Table 1). Stålnacke and co-workers compiled data from 35 small agricultural catchments in the Nordic and Baltic region (Stålnacke et al. 2014). They found that a majority of these catchments had a unit-area loss between 600-2500 kg km^{-2}. In addition, our results showed that the nitrogen losses from agricultural land were almost four times higher than the corresponding losses from forested land (Table 4) which is found to be realistic and in line with other results (Lidèn et al., 1999; Vassiljev and Stålnacke, 2005, Vassiljev et al., 2008)

3.2 Major retention estimate results

The final model parameterisation using the 88 river basin data (i.e. Model run #4 in Table 1) was used to determine the surface water retention of N in all the 117 major river basins in the Baltic Sea drainage area. This included 78 river basins with observed N load (excluding the 10 smaller Danish sub-basins), and also an additional 39 unmonitored river basins.

The total annual riverine load from the 117 basins to the Baltic Sea was estimated to 570 000 tons of N compared to the model-estimated gross load of 950 000 tons of N (Figure 2). Thus, our results show that around 380 000 tons of N are annually retained in surface waters draining to the Baltic Sea (streams, rivers, reservoirs and lakes; Fig. 2), giving a total surface water N retention of around 40%. This is substantially higher than given by Mörth et al (2007) who reported a mean N retention of 15% in the Baltic Sea rivers. The spatial distribution of the relative surface water retention is shown in Fig. 3. Averaged over all basins, mean lake retention is 25% whereas the estimated in-stream retention is 5% (Table A2). In terms of absolute retention values, three major river basins account for 50% of the total retention in the 117 basins; i.e. around 104 000 tons of N is retained in Neva, 55 000 tons in Vistula and 32 000 tons in Oder (Table A2).

Most of the retention occurs in lakes, as indicated by a strong relationship between N retention (%) and share of lake area in the river drainage areas (up to 20% lake area; Fig. 4). In Göta älv, we estimated a total N retention of 72%, whereof 67% occurred in the lakes of that drainage area (Lake Vänern primarily). Other river basins with high retention were Kymijoki (70%), Motalaström (73%) and Neva (74%). All these basins are characterised by a high percentage of lakes. Low retention was estimated for lake-poor basins, e.g. Aurajoki (2%), Kasari (4%) and Kelia (3%). This is in accordance with earlier studies, where the highest N retention has been found in river basins with a large proportion of lakes. In a comparison of N retention in four selected watersheds in Europe, representing a wide range in climate, hydrology and nutrient loads, Hejzlar et al. (2009) found the highest retention values in the two watersheds with lakes as compared to the two other mostly or entirely lake-less watersheds. A global-scale analysis by Harrison et al. (2009) indicated that lakes and reservoirs are important sinks for N in watersheds, with small lakes (<50 km^2) retaining about half of the global total. Despite the fact that reservoirs occupy only 6% of global lentic surface area, the reservoirs were estimated to retain about 33% of the total N retained by lentic systems.

3.3. Uncertainty aspects and outlook

It should be noted that there are considerable uncertainties related to estimates of nutrient loads and especially retention at the watershed scale. In a study comparing nutrient retention estimates by catchment-scale models of different complexity, Hejzlar et al. (2009) showed a large variation in nutrient retention values as estimated by the different models in four selected catchments in Europe. They further showed that retention values were directly proportional to nutrient sources within catchments, indicating a close relationship between uncertainties in quantification of diffuse nutrient sources and nutrient retention determination. They concluded that realistic modelling of nutrient export from large catchments is only possible with a certain level of measured data. However, modelling efforts that combine comprehensive datasets on population, land cover, water discharge and quality, etc., may serve as important tools for improved watershed management and for better identification of cost-efficient measures to reduce nutrient loading. In our study, the MESAW model was apparently able to estimate the N load at the river mouth of 88 Baltic Sea rivers for which we had observed data with a sufficient degree of accuracy (Fig. 1, upper panel; Table 1). However, when we show the obtained relationships using unit-area (specific) load, the model underestimates the load (Fig. 1, lower panel). Worth to notice is also that the 10 Danish sub-basins included (despite the effort with the grouping) deviate from the general relationship. These 10 smaller sub-basins have a high observed specific N load, which is not well predicted by the model.

Fig. 5a-d show the relationships between observed specific N load (kg N km^{-2}) and share of various land cover categories and lake area in the 88 (78 for wetland) Baltic Sea basins with observed N loads. A high specific N load was generally found in river basins with a large share of cultivated land, as indicated by a strong positive relationship between specific N load (kg N km^{-2}) and share of cultivated land (%; Fig. 5a). Opposite to cultivated land, specific N load was found to be negatively correlated with the share of ‘Other land’ (i.e., primarily forest; Fig 5d).

In their modeling of riverine N transport to the Baltic Sea, Mörth et al. (2007) found diffuse sources to contribute the most to the overall simulated riverine N loads. A review by Stålnacke et al. (2009) also emphasized the importance of diffuse sources (or share of cultivated land) in contributing to N loads in watersheds. HELCOM (2011) reports that 45-61% of the total waterborne inputs of N to the Baltic Sea are from diffuse sources. The importance of wetlands in determining N loads seems highly variable, with no apparent relationship between specific N load and share of wetland area in the river basins (Fig. 5b). This was less surprising since this land cover class included all kind of wetlands (from marshes to peatlands). The low specific load for drainages basin with a wetland coverage exceeding 15% are all located in middle/north Finland and also in the northern part of Sweden (Table A1). These basins are all characterised by low population density and low share of cultivated land. In a meta-analysis of the importance of wetlands for the removal of inorganic N and reduction of N export from watersheds, Jordan et al. (2011) found a large variation (0.25 to 100%) in N removal efficiency between individual wetlands. When grouped into different wetland classes, mean efficiency was highest for palustrine forested wetlands (63%) and lowest for estuarine emergent wetlands (33%).

Regarding statistical uncertainty in our study, the 3 land cover classes (and the surface water area) adds up to 100 % and apparently these explanatory variables are inter-correlated. This will have less influence on the method applied although there is always a risk of multicollinearity in these kinds of regression-type of models. It should be noted that the model inputs are areas of the land cover and not the percentages which will decrease the risk of multicollineariety. Experiences with the MESAW models as also given in the earlier quoted papers in different geographical areas (Lidèn et al., 1999; Vassiljev and Stålnacke, 2005;

Vassiljev et al., 2008; Povilaitis et al., 2012) have not indicated any problem with possible interrelated explanatory variables. In addition, the parameter estimates showed reasonable stability; little change occurred in the values of the most statistically significant model coefficients when additional variables were added in exploratory regressions (Table 1).

The predicted climate change is an additional factor that may significantly affect nutrient loads and retention in watersheds (Jeppesen et al., 2011). Changes in temperature and precipitation will most likely induce changes in agricultural land use, e.g. type of crops grown, rates and timing of fertiliser use etc., and thereby influence N cycling and export to coastal waters. However, given the uncertainties in predicting future climate and land use on a regional level, the predicted effects on nutrient budgets in watersheds remain highly uncertain (Jeppesen et al., 2011). Further studies on these issues is needed.

Despite these discerned uncertainties, it seems that the MESAW model seems to be a reliable tool for simultaneous estimation of sources and retention in a river basin. It was also evident that MESAW is flexible and can accommodate many functional relationships and explanatory variables. In addition, MESAW can be used to identify measurements and basins that are outside the general patterns and relationships. The main advantages with the model are: (i) the simple structure of the model (ii) the simple input data (iii) all unknown parameters are derived from empirical data, (iv) that information from all water quality monitoring sites are used in an optimal way, and (v) that the model give results on the base of all available measured data which is more optimal than applying emission coefficients received from literature; normally even extrapolated from other regions or up-scaled from small watersheds.

4 Conclusions

We claim that one of the largest scientific and management uncertainties are devoted to the question on how to quantify the retention from source to river mouth. In this study, we used the MESAW statistical model to estimate the surface water N retention in the 117 river basins draining to the Baltic Sea. The MESAW model was able to estimate the N load at the river mouth of 88 Baltic Sea rivers, for which we had observed data, with a sufficient degree of accuracy. The estimated retention parameters were also statistically significant. Our results show that around 380 000 tons of N are annually retained in surface waters draining to the Baltic Sea. The total annual riverine load from the 117 basins to the Baltic Sea was estimated to 570 000 tons of N, giving a total surface water N retention of around 40%. The largest retention was found in river basins with a high percentage of lakes.

The obtained results will hopefully enable the Helsinki Commission (HELCOM) to refine the nutrient load targets in the Baltic Sea Action Plan (BSAP), as well as to better identify cost-efficient measures to reduce nutrient loadings to the Baltic Sea.

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Figure captions

Figure 1 Relationship between observed and predicted annual N load (kt N yr^{-1}; upper panel) and specific observed and predicted N load (kg N yr^{-1} km^{-2}; lower panel) in the 88 Baltic Sea basins with observed N load (lower panel).

Figure 2 Total estimated nitrogen (N) load (kt N yr^{-1}) in the 117 basins of the Baltic Sea drainage area. Total retention is given as the difference between the estimated total load if no retention and the estimated total riverine net N load.

Figure 3 Relative total nitrogen (N) retention in the Baltic Sea drainage basins.

Figure 4 Relationship between estimated retention (%) and total drainage area (km^2; upper panel) and share of lake area (% of total drainage area; lower panel) for 117 Baltic Sea basins.

Figure 5 Relationship between specific N load (kg N km^{-2}) and share of (a) cultivated, (b) wetland, (c) lake and (d) other area (in % of total drainage area) in the 88 (78 for wetland) Baltic Sea basins with observed N load.

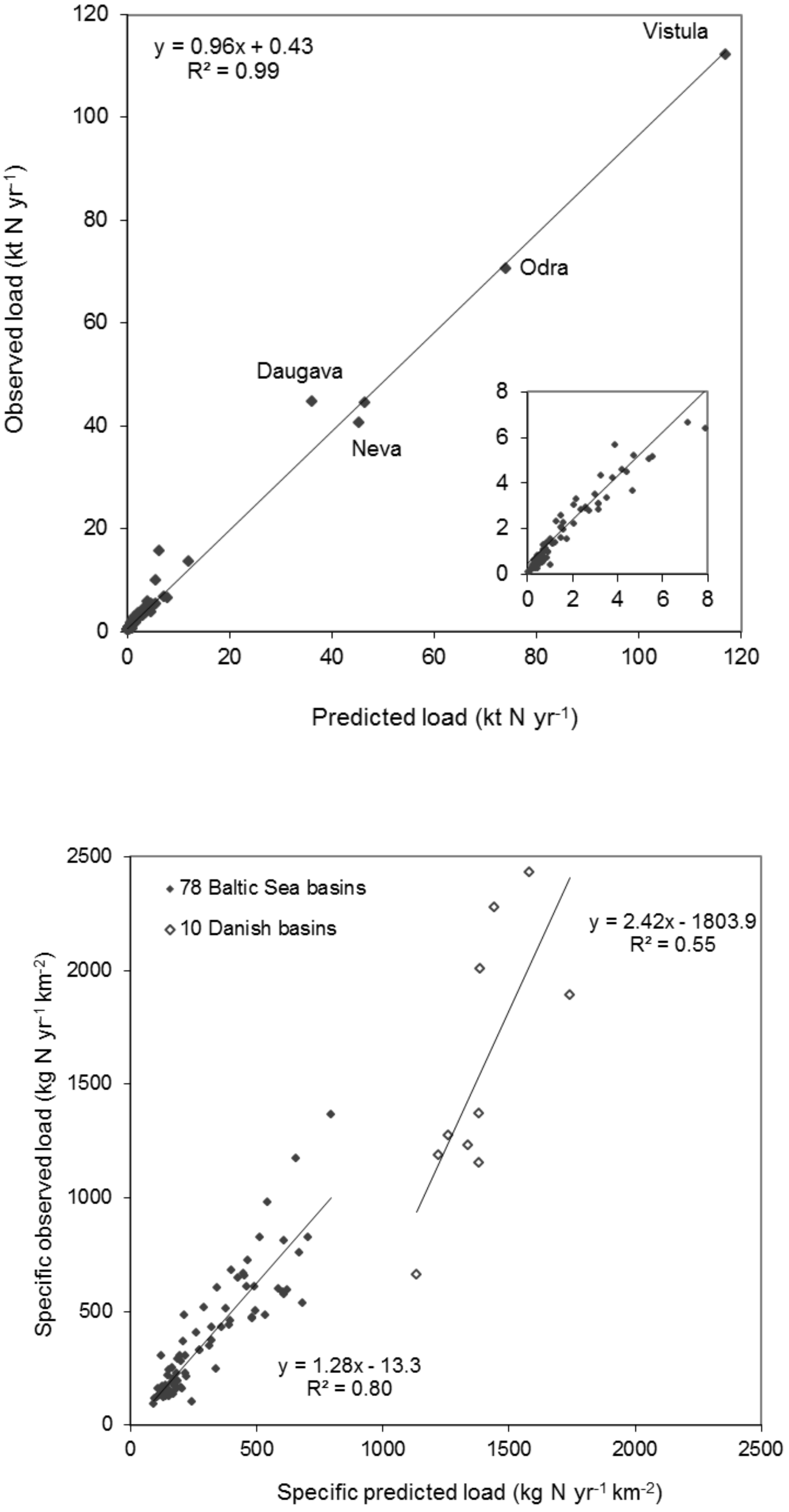

y = 0.96x + 0.43
R² = 0.99
Vistula
Odra
Daugava
Neva
Observed load (kt N yr-1)
Predicted load (kt N yr-1)
0
20
40
60
80
100
120
0
2
4
6
8
78 Baltic Sea basins
10 Danish basins
y = 2.42x - 1803.9
R² = 0.55
y = 1.28x - 13.3
R² = 0.80
Specific observed load (kg N yr-1 km-2)
Specific predicted load (kg N yr-1 km-2)
0
500
1000
1500
2000
2500

Figure 1

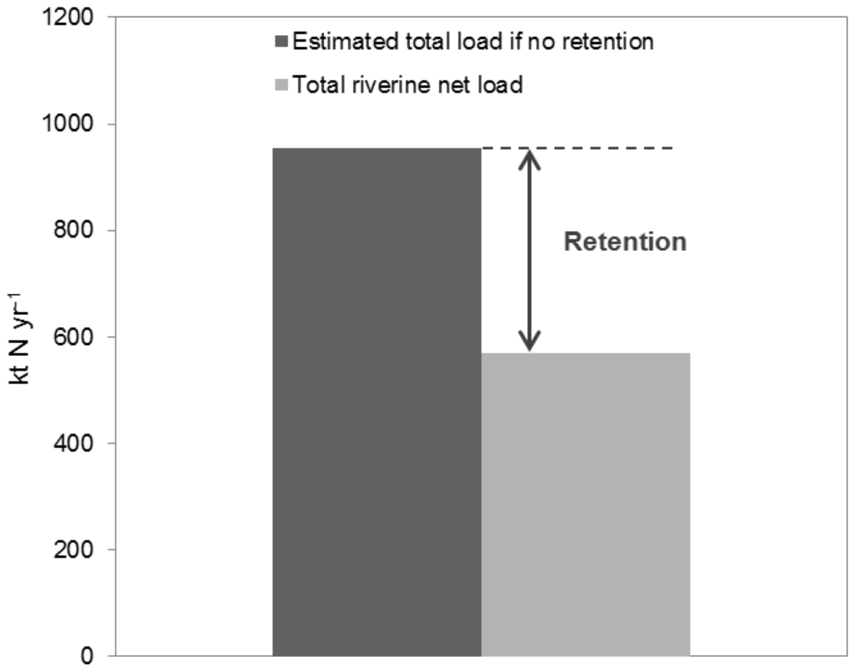
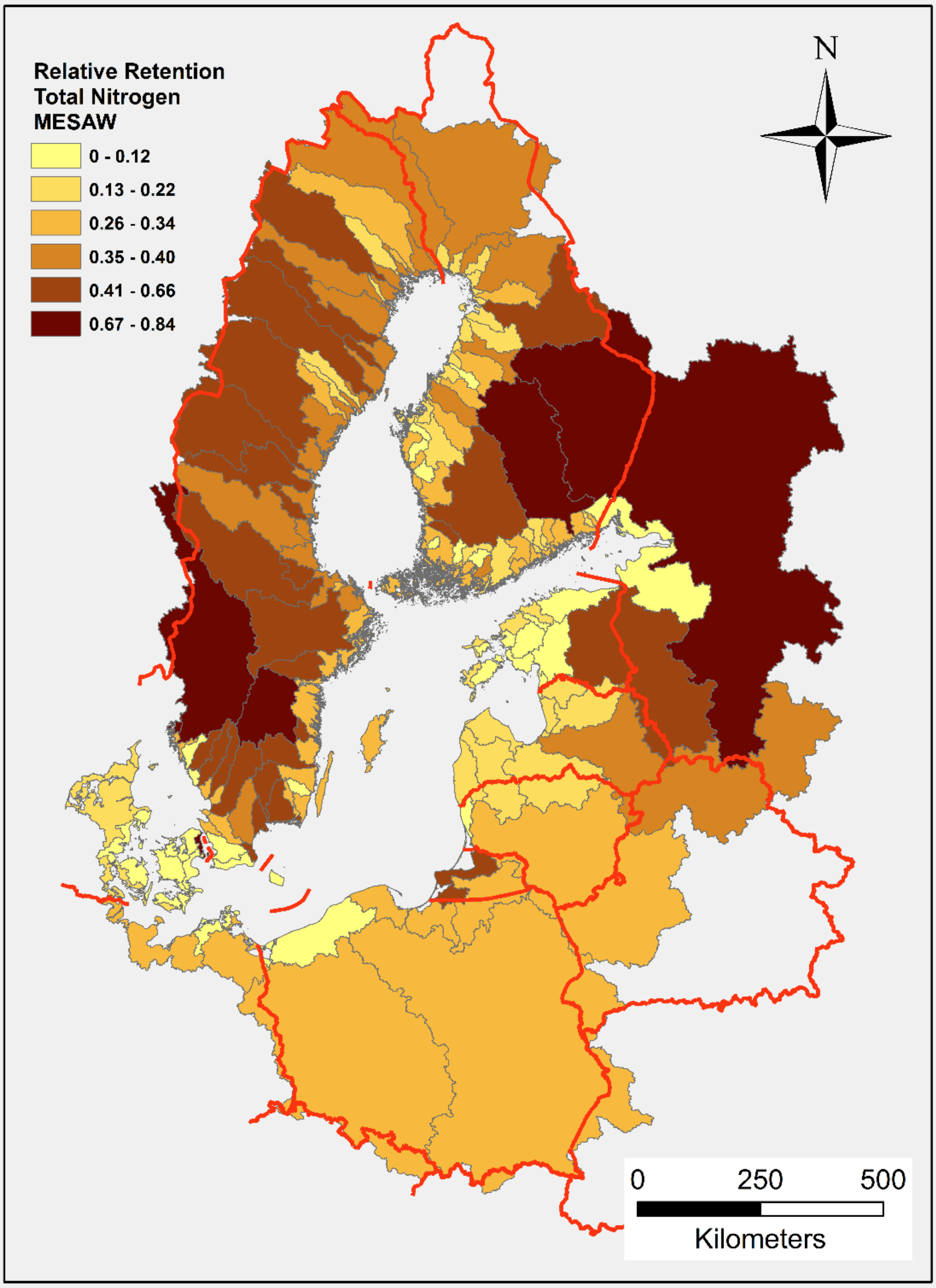

1200
1000
800
600
400
200
0
kt N yr-1
Estimated total load if no retention
Total riverine net load
Retention

Figure 2



475

476 Figure 3

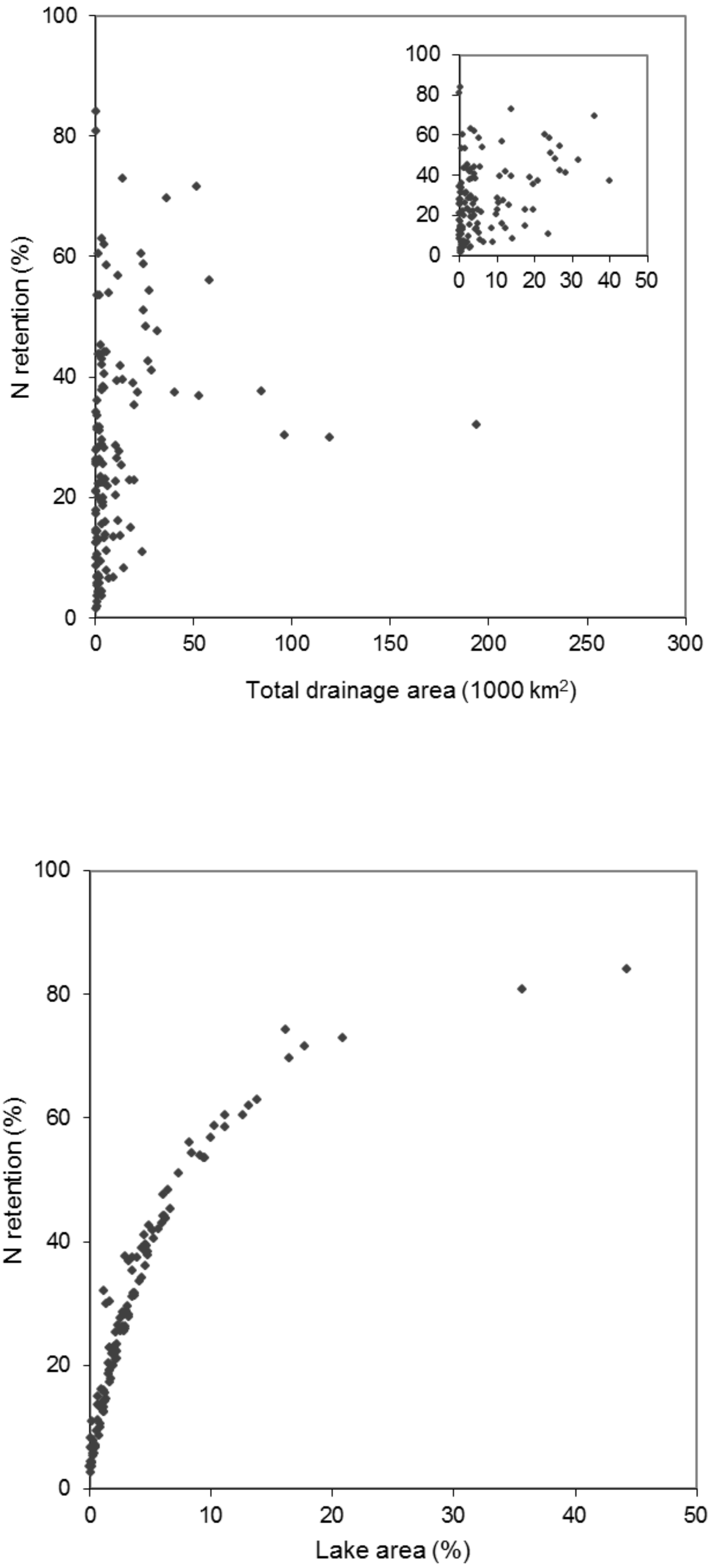

N retention (%)
Total drainage area (1000 km²)
0
50
100
150
200
250
300
20
40
60
80
100
10
30
40
50
Lake area (%)

Figure 4

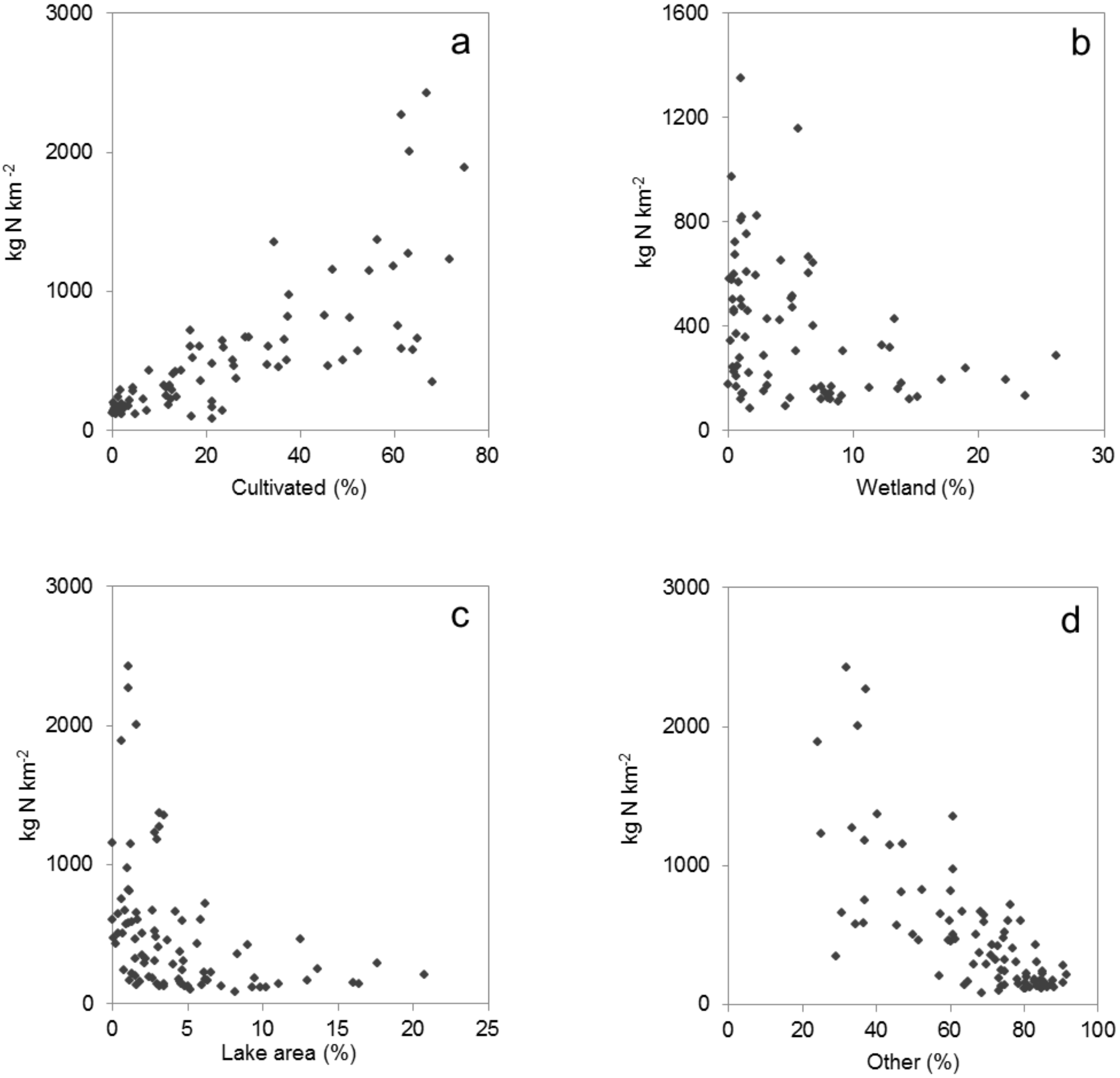

a
3000
2000
1000
0
kg N km -2
0
20
40
60
80
Cultivated (%)
b
1600
1200
800
400
0
kg N km-2
0
10
20
30
Wetland (%)
c
3000
2000
1000
0
kg N km-2
0
5
10
15
20
25
Lake area (%)
d
3000
2000
1000
0
kg N km-2
0
20
40
60
80
100
Other (%)

Figure 5

Table legends

Table 1 Results from the different MESAW model runs for estimation of total nitrogen (N) retention with different combinations of basin sub-groups. Results include estimated export coefficients (kg/km^2) from different land use classes (i.e., cultivated, wetland and other), estimated retention coefficients (dimension-less) for lake area and total drainage area, and the coefficient of determination (R^2) between observed and predicted annual loads. Standard error and t-ratio of the estimated coefficients are given for each model run.

Table 1

Model run		**Diffuse emissions**			**Retention**		**R^2 (observed vs. predicted)**
		Cultivated	*Wetland*	*Other*	*Lake*	*Instream*	
		θ_1	θ_2	θ_3	λ_2	λ_1	
		(kg/km^2)	*(kg/km^2)*	*(kg/km^2)*	*(dimension-less)*	*(dimensionless)*	
1 88 monitored basins (1 group)	Est. coeff.	1435	405	233	9	4E-03	0.94
	St. err.	929	2527	443	16	4E-03	
	t-ratio	1.54	0.16	0.53	0.57	1.03	
2 88 monitored basins (3 groups)	Est. coeff.	1440	386	185	8	2E-03	0.98
	St. err.	172	753	136	5	5E-04	
	t-ratio	8.38	0.51	1.36	1.78	3.60	
3 88 monitored basins (4 groups)	Est. coeff.	1137	208	220	11	8E-04	0.99
	St. err.	115	668	126	5	3E-04	
	t-ratio	9.88	0.31	1.75	2.16	2.62	
4 88 monitored basins (5 groups)	Est. coeff.	1073	158	225	12	7E-04	0.99
	St. err.	109	675	123	5	3E-04	
	t-ratio	9.85	0.23	1.83	2.23	2.27	

Estimated group ratios ± standard error for diffuse emissions coefficients:

Model run	**Basin sub-group**	**Ratio ± st. err.** ω_j
2	2 Pregolia, Narva	0.3± 0.2
	3 Daugava, Neva	2.2 ± 0.3
3	2 Pregolia, Narva	0.4 ± 0.2
	3 Daugava, Neva	2.0 ± 0.3
	4 10 Danish+6 Swedish (south-west coast) basins	2.0 ± 0.8

4	2	Pregolia, Narva	0.4 ± 0.2
	3	Daugava, Neva	2.0 ± 0.2
	4	10 Danish+6 Swedish (south-west coast) basins	2.1 ± 0.8
	5	27 Finnish basins	1.1 ± 0.2

Appendix

Table A1 Input data to the MESAW model for estimation of total nitrogen (N) retention. Input data include land cover (cultivated, wetland, lake area, other and total drainage area; km^2) and point source emissions (WWTP and industry; kg N yr^{-1}). Observed annual loads are given with the retention results in Table A2.

Table A2 Observed and predicted annual N loads (kg N yr^{-1}) and total N retention as estimated by the MESAW model.

Table A1.

River basin	ID	Land cover (km^2)					Point sources (kg N yr^{-1})	
		Cultivated	*Wetland*	*Lake area*	*Other*	*Total area*	*WWTP*	*Industry*
Alterälven	5	18	15	6	418	457	863	0
Aurajoki	231	388	20	0	448	856	49322	526
Botorpströmmen	98	120	1	94	772	986	0	0
Dalälven	25	1071	2158	1281	24128	28638	250825	393700
Daugava	62	18242	973	2480	62912	84608	2895199	83017
Delångersån	28	99	21	184	1665	1969	7258	0
Emån	97	559	23	272	3572	4427	73043	2200
Eurajoki	221	351	22	169	800	1342	0	24611
Forsmarksån	24	14	28	9	252	302	5	0
Gauja	61	3353	99	66	5432	8951	256499	0
Gavleån	26	169	44	165	2115	2494	85652	10600
Gideälven	341	50	277	103	3007	3437	37	0
Göta älv	151	6669	1484	9105	34206	51465	517002	882100
Helge å	91	1116	108	220	3237	4681	98586	0
Iijoki	14	193	1933	647	11369	14142	4400	0
Indalsälven	31	641	2162	1654	21382	25839	119903	35800
Kalajoki	173	771	234	127	3326	4457	17282	62
Kalix älv	8	92	3030	288	14286	17696	34519	55000
Karvianjoki	250	460	239	109	2673	3481	7550	0
Kasari	103	1085	211	2	1938	3236	34256	0
Kelia	47	335	40	1	336	712	29518	9300
Kemijoki	12	306	7633	1682	42892	52513	50135	125000

Kiiminkijoki	15	81	866	96	2850	3894	7666	0
Kiskonjoki	234	255	7	44	649	955	0	0
Kokemäenjoki	21	5180	402	2265	19281	27128	353484	201645
Koskenkylänjoki	404	338	5	35	571	950	0	114
Kuivajoki	132	25	355	30	942	1352	86	0
Kymijoki	41	2741	425	5971	27138	36275	189501	509272
Kyrönjoki	178	1442	320	46	3128	4936	45276	578
Lagan	143	903	275	598	4802	6579	104364	31000
Lapuanjoki	177	1052	206	83	2720	4060	12102	4650
Lestijoki	174	156	141	3	753	1053	0	0
Lielupe	63	10872	271	121	6549	17814	701774	66470
Ljungan	29	229	638	645	11092	12605	20964	0
Ljungbyån	96	140	4	8	854	1006	31242	0
Ljusnan	27	449	1645	705	17224	20024	57246	0
Luleälv	6	46	2016	1790	20702	24554	9045	0
Lyckebyån	95	36	8	33	721	797	6849	0
Lögdeälven	342	9	104	28	1361	1503	1600	0
Motala ström	99	3042	96	2937	8046	14121	341463	111400
Mustijoki	402	285	9	9	455	758	0	0
Mörrumsån	93	394	27	464	2490	3376	87147	2900
Narva	46	12437	1048	4789	39852	58126	1068418	132850
Neman	83	44359	554	1544	49469	95925	6206082	139420
Neva	42	7004	8126	45020	219436	279586	3522246	2586124
Nissan	145	254	100	179	2619	3152	62723	28000
Norrström	101	5457	302	2564	14754	23076	860051	379300
Nyköpingsån	100	952	32	579	2877	4440	61879	0

Närpiönjoki	202	241	70	5	706	1022	0	0
Odra	87	73524	225	1630	43559	118939	13758343	1133829
Oulojki	16	513	1828	2490	19411	24242	36077	50297
Paimionjoki	232	567	13	14	524	1118	0	0
Perhonjoki	175	313	327	57	1822	2519	0	15700
PiteÄlv	4	42	863	515	9732	11152	13057	0
Porvoonjoki	403	504	5	14	814	1337	38752	1135
Pregolia	84	9187	34	280	3919	13419	1276555	0
Pyhäjoki	172	440	204	179	2903	3727	2568	1412
Pärnu	601	2198	341	8	4053	6600	84972	0
Rickleån	1	49	52	104	1453	1658	824	0
Rönneå	142	661	21	67	1154	1903	46885	17100
Råneälven	71	24	994	70	3087	4175	3798	0
Salaca	602	1294	150	57	2007	3508	63498	0
Siikajoki	171	464	506	65	3074	4109	0	3629
Simojoki	131	47	597	148	2349	3141	501	0
Skellefteälv	2	98	1026	1152	9337	11613	23639	30500
Torne älv	10	264	6089	1405	32354	40112	52914	131000
Töreälven	72	3	70	14	417	505	719	0
Ume älv	35	252	2169	1318	23199	26939	38219	59100
Uskelanjoki	233	472	4	4	478	959	5763	26640
Vantaanjoki	401	541	12	52	1290	1895	39672	12032
Venta	80	6146	107	111	5328	11692	580890	0
Vironjoki	43	67	2	6	283	357	0	0
Viskan	149	365	14	136	1664	2178	44596	0
Vistula	85	124478	751	2268	66398	193894	20541873	547784

Ähtävänjoki	176	737	201	225	3155	4318	2349	14376
Ätran	147	558	50	196	2515	3320	6049	0
Öreälven	343	63	347	37	2600	3046	1801	0
Ångermanälven	33	398	2923	1921	26572	31815	36673	0
Ry å		214		2	69	285	23275	0
Lindenborg å		197		4	119	319	19624	0
Skals å		401		16	139	556	22700	0
Karup å		344		8	275	627	92696	0
Gudenå		1563		79	961	2603	404099	0
Århus å		183		10	130	324	134450	0
Kolding å		180		3	85	268	32126	0
Odense å		339		9	187	535	31007	0
Ndr. Halleby å		272		18	128	418	31204	0
Suså		478		24	254	756	107901	0
Coast DE &Arkona Basin	1011	1740	28	8	620	2395	74132	0
Coast DE & Bornholm Basin	1012	7858	81	191	2455	10585	336061	1535
Coast DE & Fehmarn Belt	1013	8041	52	280	2148	10522	377142	23830
Coast DK &Arkona Basin	2011	1109	14	3	500	1626	44132	55702
Coast DK & Bornholm Basin	2012	446	2	0	134	581	19962	9873
Coast DK & Central Kattegat	2018	9915	194	82	824	12459	71261	21316
Coast DK & Fehmarn Belt	2015	2471	14	62	1729	2961	228639	145739
Coast DK & Northern Kattegat	2017	376	16	13	463	629	78572	10873
Coast DK &Samso Belt	2014	7346	67	6	296	9204	0	2317
Coast DK & Southern Kattegat	2013	2141	27	0	237	3074	12299	183156
Coast DK & The Sound	2016	117	2	150	2199	422	370565	76197
Coast EE & Baltic Proper	3011	1102	201	40	3120	4463	38295	0

Coast EE & Gulf of Finland	3012	1953	181	16	3694	5843	90250	636400
Coast EE & Gulf of Riga	3013	1610	373	34	3375	5392	71976	0
Coast FI & Baltic Proper	4013	802	15	58	8112	3716	48827	1275161
Coast FI &Bothnian Bay	4011	1283	608	152	9272	10061	37028	393799
Coast FI &Bothnian Sea	4012	2255	165	292	2607	11844	5282	123771
Coast FI & Gulf of Finland	4014	1120	58	109	3999	5286	997	155412
Coast LT & Baltic Proper	5011	846	34	7	713	1599	23821	0
Coast LV & Baltic Proper	6011	2605	101	60	2491	5257	54410	0
Coast LV & Gulf of Riga	6012	1697	210	112	4052	6071	169910	0
Coast North of Northern Kattegat	9018	129	0	205	5857	464	178001	0
Coast PL & Baltic Proper	7012	7144	71	251	3313	10778	191663	14737
Coast PL & Bornholm Basin	7011	8207	64	10	2125	14333	67794	0
Coast RU & Baltic Proper	8011	3552	28	345	22109	5716	50496	334590
Coast RU & Gulf of Finland	8012	690	689	34	569	23832	50617	170000
Coast SE &Arkona Basin	9012	1182	0	2	154	1338	34653	0
Coast SE & Baltic Proper	9014	5022	66	315	4230	19925	54353	418800
Coast SE & Bornholm Basin	9013	1049	24	623	14215	5618	165765	223000
Coast SE &Bothnian Bay	9015	769	1195	821	16344	19129	105747	259100
Coast SE &Bothnian Sea	9016	1641	315	833	18550	21339	159982	1544000
Coast SE & Central Kattegat	9020	595	7	5	330	1878	9798	0
Coast SE & Northern Kattegat	9019	141	0	28	576	745	10765	7600
Coast SE & Southern Kattegat	9021	1054	26	80	1195	2234	21757	131000
Coast SE & The Sound	9011	2019	2	15	1138	2623	11479	11000
Laihianjoki	201	239	21	1	461	723	6010	0
Isojoki	205	176	80	3	895	1155	0	3000
Sirppujoki	222	143	7	3	270	424	0	0

Iilolanjoki	405	102	1	7	196	306	0	0

Table A2.

River basin	ID	Annual N load (kg N yr^{-1})		Retention (kg N yr^{-1})	Relative retention		
		Observed	*Predicted*		*Total surface water*	*Lake*	*In-stream*
Alterälven	5	96331	100515	17 290	0.15	0.134	0.014
Aurajoki	231	704385	600476	11 817	0.02	0.000	0.019
Botorpströmmen	98	173792	171655	197 934	0.54	0.526	0.021
Dalälven	25	5226692	4744728	3 296 123	0.41	0.343	0.102
Daugava	62	40351648	45292798	27 364 274	0.38	0.255	0.164
Delångersån	28	231231	255778	294 739	0.54	0.522	0.029
Emån	97	993846	953813	756 687	0.44	0.417	0.043
Eurajoki	221	639538	284749	434 449	0.60	0.594	0.024
Forsmarksån	24	90969	58633	20 485	0.26	0.250	0.012
Gauja	61	4467000	4443302	690 252	0.13	0.079	0.060
Gavleån	26	559846	456153	378 260	0.45	0.435	0.032
Gideälven	341	474231	568452	229 492	0.29	0.260	0.038
Göta älv	151	15496154	6222224	15 737 523	0.72	0.673	0.132
Helge å	91	2786308	2742915	1 697 732	0.38	0.354	0.044
Iijoki	14	2205385	2092250	1 372 973	0.40	0.348	0.074
Indalsälven	31	4321692	3260548	3 047 764	0.48	0.427	0.098
Kalajoki	173	2294615	1294900	507 684	0.28	0.249	0.043
Kalix älv	8	3505231	3021890	895 054	0.23	0.159	0.082
Karvianjoki	250	1398667	902551	378 474	0.30	0.267	0.038
Kasari	103	1949457	1593791	74 510	0.04	0.008	0.037
Kelia	47	831729	467939	12 609	0.03	0.009	0.018
Kemijoki	12	6372308	7897005	4 619 699	0.37	0.272	0.134

Kiiminkijoki	15	746000	719171	246 453	0.26	0.224	0.040
Kiskonjoki	234	351985	306363	173 663	0.36	0.349	0.020
Kokemäenjoki	21	9839231	5662122	6 747 574	0.54	0.493	0.100
Koskenkylänjoki	404	429846	376743	174 023	0.32	0.302	0.020
Kuivajoki	132	388538	252573	72 255	0.22	0.203	0.024
Kymijoki	41	5673077	3912199	8 968 328	0.70	0.657	0.114
Kyrönjoki	178	3274615	2201007	353 912	0.14	0.098	0.045
Lagan	143	2812308	2375686	2 785 262	0.54	0.515	0.052
Lapuanjoki	177	2052308	1525444	443 272	0.23	0.192	0.041
Lestijoki	174	448077	339396	20 572	0.06	0.037	0.021
Lielupe	63	13435786	11941659	2 100 218	0.15	0.073	0.082
Ljungan	29	1536538	1751972	1 256 416	0.42	0.374	0.070
Ljungbyån	96	241923	340462	40 219	0.11	0.087	0.021
Ljusnan	27	2822077	3149047	1 715 354	0.35	0.291	0.087
Luleälv	6	2920615	2572731	2 688 010	0.51	0.459	0.095
Lyckebyån	95	221462	158152	79 930	0.34	0.323	0.019
Lögdeälven	342	234077	272877	68 367	0.20	0.179	0.025
Motala ström	99	3017538	2067763	5 577 661	0.73	0.708	0.074
Mustijoki	402	620923	388273	59 428	0.13	0.117	0.018
Mörrumsån	93	834923	557809	951 339	0.63	0.616	0.038
Narva	46	5034077	5400364	6 902 138	0.56	0.490	0.140
Neman	83	44323731	46377160	20 173 375	0.30	0.158	0.172
Neva	42	44616846	36056404	104 559 254	0.74	0.652	0.262
Nissan	145	1368846	1231196	893 156	0.42	0.399	0.036
Norrström	101	3637692	4695747	7 182 353	0.60	0.564	0.093
Nyköpingsån	100	730077	792009	1 293 905	0.62	0.603	0.043

Närpiönjoki	202	657615	432931	32 255	0.07	0.049	0.021
Odra	87	70289195	73974593	31 717 905	0.30	0.138	0.188
Oulojki	16	2894615	2598002	3 708 470	0.59	0.545	0.095
Paimionjoki	232	900846	680805	114 622	0.14	0.125	0.022
Perhonjoki	175	815769	688291	209 547	0.23	0.208	0.033
PiteÄlv	4	1594231	1492285	966 100	0.39	0.350	0.066
Porvoonjoki	403	1303615	723513	107 528	0.13	0.108	0.024
Pregolia	84	4580143	4207286	1 429 755	0.25	0.195	0.072
Pyhäjoki	172	1127385	807208	504 218	0.38	0.359	0.039
Pärnu	601	3091070	3193671	219 537	0.06	0.013	0.052
Rickleån	1	283462	233266	181 333	0.44	0.422	0.027
Rönneå	142	2587846	1511841	682 779	0.31	0.291	0.029
Råneälven	71	540308	714315	177 607	0.20	0.164	0.042
Salaca	602	2287635	1585606	377 174	0.19	0.160	0.038
Siikajoki	171	1332615	1127084	266 866	0.19	0.157	0.041
Simojoki	131	748231	471133	286 563	0.38	0.355	0.036
Skellefteälv	2	1319385	1124731	1 475 798	0.57	0.536	0.068
Torne älv	10	5154615	5552103	3 319 728	0.37	0.290	0.119
Töreälven	72	89992	83532	28 567	0.25	0.244	0.015
Ume älv	35	3359846	3522076	2 619 449	0.43	0.363	0.099
Uskelanjoki	233	508182	652521	47 115	0.07	0.048	0.020
Vantaanjoki	401	1283000	753105	269 584	0.26	0.242	0.028
Venta	80	6649974	7118779	1 365 064	0.16	0.100	0.068
Vironjoki	43	213534	122758	26 503	0.18	0.167	0.013
Viskan	149	1568692	1016516	792 104	0.44	0.420	0.030
Vistula	85	112041104	116917897	55 292 179	0.32	0.120	0.229

Ähtävänjoki	176	419608	1049732	713 541	0.40	0.378	0.042
Ätran	147	2007769	1529797	1 155 539	0.43	0.408	0.037
Öreälven	343	491769	605790	110 818	0.15	0.123	0.036
Ångermanälven	33	4223154	3798849	3 450 419	0.48	0.413	0.107
Ry å		537807	495987				
Lindenborg å		724156	458904				
Skals å		683604	743902				
Karup å		720306	866486				
Gudenå		3075608	3177587				
Århus å		442719	446084				
Kolding å		651265	423277				
Odense å		1071416	740493				
Ndr. Halleby å		275250	473721				
Suså		961097	952650				
Coast DE & Arkona Basin	1011		1953735	139 436	0.07	0.036	0.032
Coast DE & Bornholm Basin	1012		7394183	2 176 860	0.23	0.174	0.065
Coast DE & Fehmarn Belt	1013		7094187	2 843 034	0.29	0.237	0.065
Coast DK & Arkona Basin	2011		1345482	61 852	0.04	0.018	0.026
Coast DK & Bornholm Basin	2012		529791	8 593	0.02	0.000	0.016
Coast DK & Central Kattegat	2018		9543967	1 505 255	0.14	0.071	0.070
Coast DK & Fehmarn Belt	2015		2715470	783 402	0.22	0.195	0.035
Coast DK & Northern Kattegat	2017		483611	127 542	0.21	0.195	0.017
Coast DK & Samso Belt	2014		7432683	537 534	0.07	0.007	0.061
Coast DK & Southern Kattegat	2013		2458534	91 878	0.04	0.000	0.036
Coast DK & The Sound	2016		232761	984 193	0.81	0.806	0.014
Coast EE & Baltic Proper	3011		1711747	261 982	0.13	0.094	0.043

Coast EE & Gulf of Finland	3012	3399885	289 177	0.08	0.031	0.049
Coast EE & Gulf of Riga	3013	2339545	295 661	0.11	0.068	0.047
Coast FI & Baltic Proper	4013	3285098	754 614	0.19	0.153	0.039
Coast FI &Bothnian Bay	4011	3211420	818 564	0.20	0.149	0.063
Coast FI &Bothnian Sea	4012	2382056	908 814	0.28	0.223	0.068
Coast FI & Gulf of Finland	4014	1783711	535 861	0.23	0.193	0.047
Coast LT & Baltic Proper	5011	1024349	78 227	0.07	0.046	0.026
Coast LV & Baltic Proper	6011	2919632	550 310	0.16	0.118	0.047
Coast LV & Gulf of Riga	6012	2347634	656 593	0.22	0.178	0.050
Coast North of Northern Kattegat	9018	291927	1 530 714	0.84	0.838	0.014
Coast PL & Baltic Proper	7012	6523722	2 349 513	0.26	0.213	0.065
Coast PL & Bornholm Basin	7011	8603413	770 891	0.08	0.008	0.075
Coast RU & Baltic Proper	8011	5280324	4 169 888	0.44	0.413	0.048
Coast RU & Gulf of Finland	8012	1082667	132 001	0.11	0.016	0.094
Coast SE &Arkona Basin	9012	1284175	55 321	0.04	0.018	0.024
Coast SE & Baltic Proper	9014	5408956	1 605 059	0.23	0.156	0.087
Coast SE & Bornholm Basin	9013	2193909	3 088 678	0.58	0.564	0.048
Coast SE &Bothnian Bay	9015	3191350	2 042 113	0.39	0.333	0.085
Coast SE &Bothnian Sea	9016	4986008	2 981 173	0.37	0.313	0.089
Coast SE & Central Kattegat	9020	687471	41 291	0.06	0.029	0.028
Coast SE & Northern Kattegat	9019	222276	101 656	0.31	0.301	0.018
Coast SE & Southern Kattegat	9021	1120775	520 464	0.32	0.295	0.031
Coast SE & The Sound	9011	2227338	232 256	0.09	0.063	0.033
Laihianjoki	201	356461	13 468	0.04	0.019	0.018
Isojoki	205	385234	21 891	0.05	0.032	0.022
Sirppujoki	222	195592	21 695	0.10	0.087	0.014

Iilolanjoki	405	124007	33 175	0.21	0.202	0.012