Malve, Olli; Tattari, Sirkka; Riihimarki, Juha; Jaakkola, Elina; Voss, Anja; **Williams, Richard**; Baerlund, Ilona. 2012 Estimation of diffuse pollution loads in Europe for continental scale modelling of loads and in-stream river water quality. *Hydrological Processes*, 26 (16). 2385-2394. 10.1002/hyp.9344

Estimation of agricultural non-point load at the European scale

Olli Malve[1], Sirkka Tattari[1], Juha Riihimäki[1], Elina Jaakkola[1], Anja Voβ[2], Richard Williams[3], Ilona Bärlund[4]

[1]Finnish Environment Institute, Finland
[2]Centre for Environmental Systems Research, University of Kassel, Germany
[3]Centre for Ecology and Hydrology, Natural Environment Research Council, UK
[4] Helmholtz Centre for Environmental Research-UFZ, Germany

Abstract

Regression equations for BOD_5, total nitrogen and total phosphorus were developed to estimate loads at the European scale. Annual loads from between 79 and 106 large European river basins for three different time periods (1988 – 1992, 1993 -1997, 1998-2002) were regressed against a range of catchmentment characteristics. BOD_5 was best related to runoff, number of livestock units and point load, while for total diffuse nitrogen load, also the lake and cropland area were influential. For diffuse phosphorus load, the factors were almost similar to nitrogen, but instead of cropland area, the combined factor of average slope steepness multiplied by runoff was influential. In the both nutrient equations, the lake area had a negative impact describing the retention character of the lakes. All the regression equations gave good statisical fits to the estimated catchment loads. When model results were compared with the estimates of diffuse polution from independent sources the equations performed resonably well.

1. Introduction

Diffuse nutrient loading typically originates from agriculture and deposition but locally also managed forestry and urban areas contribute to the diffuse load. In some studies, the natural background leaching which refers to natural erosion and leaching of nutrients from unmanaged areas, is separately calculated while in others not. Studies on so called source apportionment where different pollutant sources, e.g. deposition, agriculture and forestry have been calculated separately have mostly been performed in Northern and Central Europe, while little information is available for Southern European countries. The methods for analyzing nutrient concentrations and loads vary between countries and have changed over time. A simpler source apportionment method to estimate terrestrial gross diffuse nutrient losses is based on export coefficients. The export coefficient gives the rate at which nitrogen or phosphorus is exported from each land use type within the catchment. The losses from all terrestrial sources are calculated by summing up losses from different land use types and estimated losses from scattered settlements. The nitrogen and phosphorus load from households that are not connected to the sewerage system can be estimated by considering specific emission factors for the non-urban population (Grizzetti & Bouraoui, 2006).The export coefficient for each land use type can be based either on measurements, modeling or expert judgment. The system utilizes estimated values of nutrient loads for a set of field properties, e.g. soil type, slope and vegetation. For agricultural land, statistical information about the area of different crops is needed as input.

Catchment scale modeling is a widely used technique to estimate water and nutrient (nitrogen and phosphorus) transport and transformations as well as effects of policies and mitigation measures (e.g. Hesse et al. 2008, Tattari et al. 2009, Wade et al. 2008, Zhang et al. 2008). However, there are only few examples of larger scale modeling tools on nutrient loading (e.g. Behrendt & Schreiber 2004, Grizzetti & Bouraoui 2006, Pettersson et al. 2000). The GREEN model developed by the European Commission's Joint Research Institute has been calibrated in seven large European river basins and diffuse load estimates of nitrate nitrogen has been extrapolated at the European scale. Calibration was implemented using a database compiled in the FATE (Fate of Agrochemicals in Terrestrial Ecosystems) research program (Mulligan et al. 2006, Bouraoui & Aloe 2007 and Pistocchi et al. 2006). The driver behind the nutrient losses in GREEN model is the annual precipitation and the retention in water is linked to the river length. According to this approach, diffuse sources (e.g. fertilizer applications, scattered dwellings, and atmospheric deposition) are first reduced in the soil matrix and then once in the stream they undergo further reduction due to in-stream retention processes. According to the study, agriculture was the dominant contributor in diffuse losses of both nitrate nitrogen and phosphorus. Diffuse phosphorus loads are the highest in the regions of intensive animal farming, whereas the highest diffuse nitrogen losses occur in intensive cultivated areas and also where rainfall is rather high. Relatively, diffuse sources contributed less to the total phosphorus load than for nitrogen.

Diffuse BOD load data on European scale is sparse. Loading estimates are available as country specific total loads (HELCOM 2004) and export coefficients specific to certain geographic areas. Typical non-point sources of BOD include livestock, scattered settlements not connected to sewage systems and urban development. There has been a decrease in BOD concentrations in the EU, reflecting implementation of the urban wastewater treatment directive (EEA 2005). There are models available for calculating non-point BOD loads originating from US and Japan. For example, PLOAD-model is a simplified GIS based model to calculate pollutant loads to watersheds on an annual average basis. Annual pollutant loads may be calculated using e.g. export coefficient method (EPA 2001). STEPL- model (Spreadsheet Tool for Estimation of Pollutant load) computes surface runoff, nutrient and BOD_5 losses, and sediment delivery based on different land uses and management practices. The land use types considered are urban land, cropland, pastureland, feedlot and forest (EPA 2006). Shrestha et al. (2008) used a seven parameter log-linear model to estimate loadings from the sub-catchments of the Fuji river. In the equation the load is related to discharge and parameters that describe seasonal variations and trends in concentrations.

This paper describes how the diffuse load equations for BOD_5, total nitrogen and total phosphorus are estimated using available Pan-European databases. Catchments are selected from around the Europe where both measured annual average concentrations for BOD, nitrogen or phosphorus and the discharge data are adequate. The calculated load is then regressed against a range of different catchment characteristics that can be considered as explanatory variables. In the statistical analyses used here the load from scattered settlements is combined to point load (Williams et al. in this volume). The developed annual equations are further modified to be able to be used in the WaterGAP -model (Alcamo et al. 2003, Döll et al. 2003, Flörke & Alcamo 2004), which originally calculates water use and availability on global scale. With the new equations developed here and by Williams (in this volume), a new water quality module WorldQual was developed to calculate point and diffuse source pollutant loading on a continental scale (Voβ et al., in this volume).

2. *Material and methods*

Terrestrial non-point load were estimated using a linear export coefficient model and data from a set of observed river basins. An export coefficient *(e_i)* gives the rate at which BOD, N or P is exported from each catchment given the characteristics of land use type. Total load transported out of observed catchments was calculated by summing up the loads from all land uses together with estimated losses from scattered settlement and point sources, by multiplying it with retention coefficient and by subtracting the resulting amount with retention in lakes according to following equation:

$$L_j = r_1 * \{\sum_{i=1}^{n}(e_i * C_{i,j}) + (S_j + P_j)\} - r_2 * lake_j \qquad (1)$$

where:
L = Total load from terrestrial sources (kg km^{-2}a^{-1})
$C_{i,j}$ = Characteristic of land use type i of catchment j (char)
e_i = Export coefficient for land use i (kg a^{-1}char^{-1})
S_j = Load from scattered settlement in a catchment j (kg km^{-2} a^{-1})
P_j = Load from point sources in a catchment j (kg km^{-2} a^{-1})
r_1 = Retention coefficient – other than lake (-)
r_2 = Retention per lake percentage (kg km^{-2}a^{-1}%$^{-1}$)
$lake_j$ = Lake percentage of catchment j (%)

Export and retention coefficients were estimated using data from the set of observed catchments described below. Loads from scattered settlements and point sources were fixed using the load estimates obtained from Williams et al. (in this volume).

Equation 1 was reformulated to make it feasible for the linear regression analysis:

$$L_j = \sum_{i=1}^{n}(\beta_i * C_{i,j}) + r_1 * (S_j + P_j) - r_2 * lake_j \qquad (2)$$

where
$\beta_i = r_1 * e_j \iff e_j = \beta_i / r_1$

The data of BOD, nitrogen and phosphorus concentration were collated and used to fit the linear export coefficient model (equation 2). The European Environment Agency (EEA) assembles its water quality data on a representative sub-sample of national monitoring results, which EEA member countries report voluntarily each year to the EEA. EEA has mainly collected annual values (e.g. average, median, minimum and maximum). In this study, group of water quantity (discharge) stations from EEA WISE database (EEA 2007) were selected with nearby WISE stations where also measured annual average water quality data (BOD, phosphorus and nitrogen) was available. The criterion for selecting a station was at the minimum 6 measurements per year. With more than 12 measurements, the amount of selected stations would have been too low. To examine changes in recent history, three time-steps were selected: 1988–1992, 1993–1997, and 1998–2002.

Watersheds above the selected station were determined with ESRI's GIS-software ArcMap Version 9.2 using the drainage direction data of WaterGAP3 (CESR 2007-2010, Flörke & Alcamo 2004). For each watershed, a dataset of different variables was collated (Figure 1 and in supplemental material). This data included the following variables: Areas of the basin, lakes and cropland (km^2), river channel length (m), livestock units (LSU), discharge (m^3 s^{-1}), year, type of BOD (BOD_5 or BOD_7), BOD concentration (mg l O^2), calculated flux (t a^{-1}), total phosphorus and total nitrogen concentration (mg l^{-1}), temperature (°C), slope (m/m), calculated loads from scattered and urban settlements as well as from point sources (manufacturing and domestic) for total phosphorus, total nitrogen and BOD (t a^{-1}) and the fertilizer use of mineral and manure phosphorus and nitrogen and the deposition of nitrogen (t a^{-1}).

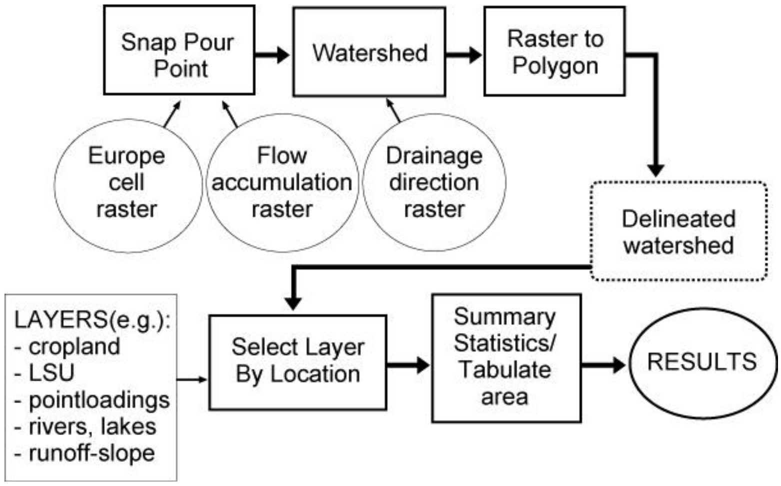

Figure 1. Scheme of the ArcGis macro that was used to delineate watersheds and to calculate data from different layers. Highlighted boxes are ArcMap tools.

The data sources are presented in Table 1. Lake area and length of river channels in the watersheds were calculated from the River and Catchment database provided by Joint Research Centre (JRC 2008). Slope, livestock units, temperature and loads from the scattered settlements as well as loads from the point sources were calculated from WaterGAP3 data layers (CESR 2007-2010, Williams et al., in this volume). The fertilizer use of mineral and manure P and N (t a^{-1} ; original resolution 10 km) and the deposition of N (t a^{-1}; original resolution 50 km) were scanned, georeferenced and reshaped with ArcMap's Multivariate-toolbox from Mulligan et al. (2006), ending up to a technical resolution of 100 m for fertilizers and 400 m for deposition. In addition, drained area (ha) (Goethe Universität 2008) and prevailing Köppen – Geiger climate class (Institute for Veterinary Public Health 2008) was added to the model. Furthermore, the area of cropland was reconstructed with Corine land cover 2000 (CLC2000 2009) and Global land cover characterization (GLCC 2008). CLC2000 was of 100 m resolution and categorized to 44 classes, whereas GLCC was of 1000 m resolution and categorized to 24 classes. The two land use maps were joined to one raster dataset and reclassified into 17 land use classes, one of them being class 'cropland'. The cropland area corresponded rather well to the statistics of the Food and Agriculture Organization of the United Nations (FAO, FAOSTAT 2010), which provides official data relating to food and agriculture for some 200 countries (Figure 2).

Table 1. Reference table to the collected input data.

INPUT DATA	SOURCE
Water quantity and quality	EEA WISE database (EEA, 2007)
Lake percentages, river lengths	JRC 2008
Livestock units, temperature	WaterGAP3 data (CESR 2007-2010)
Load from point sources and scattered settlements	WaterGAP3 data (CESR 2007-2010)
Drainage direction, slope	WaterGAP3 data (CESR 20012010)
Fertilization and deposition maps	(Mulligan et al. 2006)
Köppen-Geiger climate classification	Institute for Veterinary Public Health 2008
Land use	CLC2000 2009 & GLCC 2008

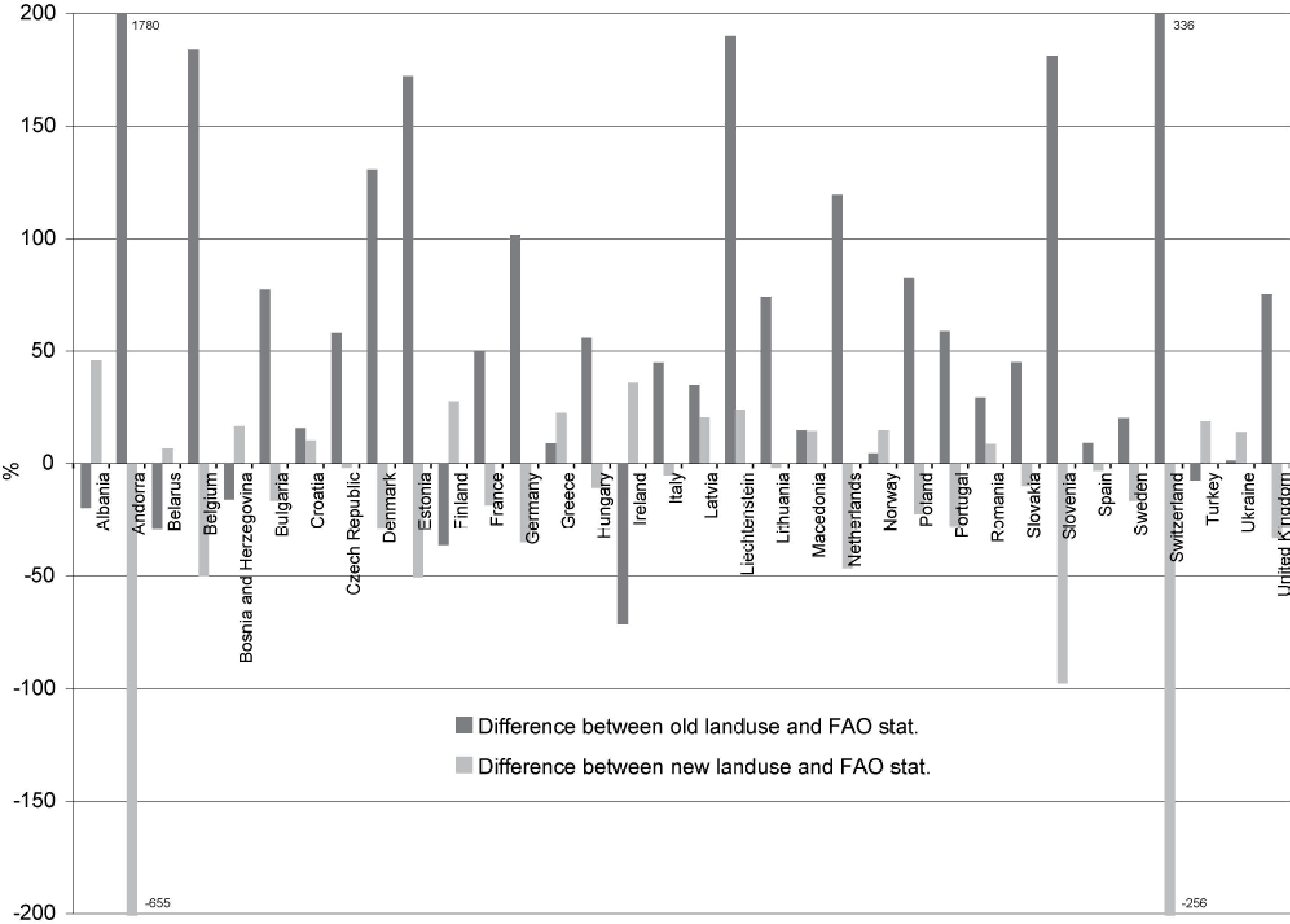

Figure 2. Differences between old and new cropland data layers compared to FAO statistics (FAOSTAT 2010).

The collected data consisted of altogether 79 different river basins from around the Europe. Not all of the basins had information for BOD, nitrogen and phosphorus concentrations and therefore the number of basins behind each regression model varies (See appendices 1 – 3). The characteristics of the river basins are presented in Figure 3. The figure shows the median value of each factor including the relative range in the y-axis.

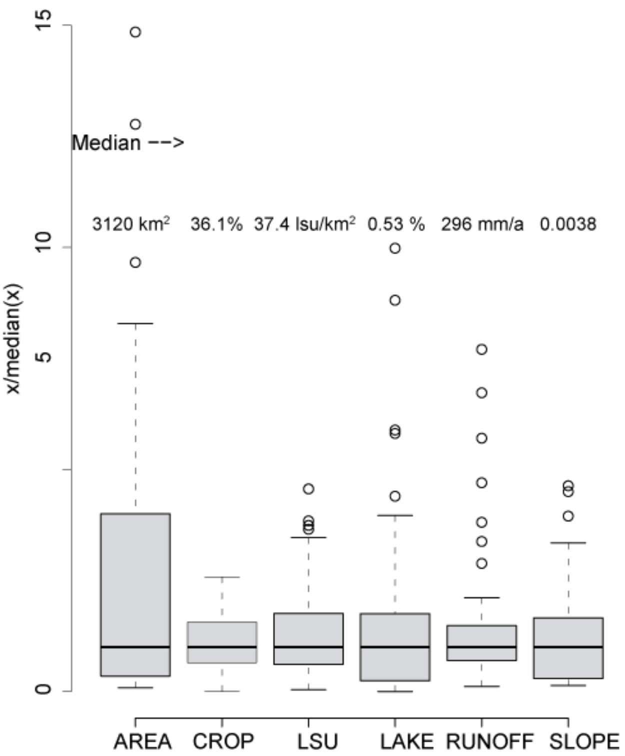

Figure 3. Characteristics of observed river basins; the median value of each factor and the relative range in the y-axis.

3. *Model fit and validation*

BOD_5

A linear export coefficient model using data (in supplemental material 1) from three time periods 1988-1992, 1993-1997, and 1998-2002 was formulated using R software (R Development Core Team 2009). Only statistically significant factors ($P<0.05$) were included in a model. Because there were no significant differences between the equations developed for different time periods the data was compiled together and the final equation included all the years from 1988 to 2002. The delineated watersheds for the BOD_5, nitrogen and phosphorus calculations are presented in Figure 4.

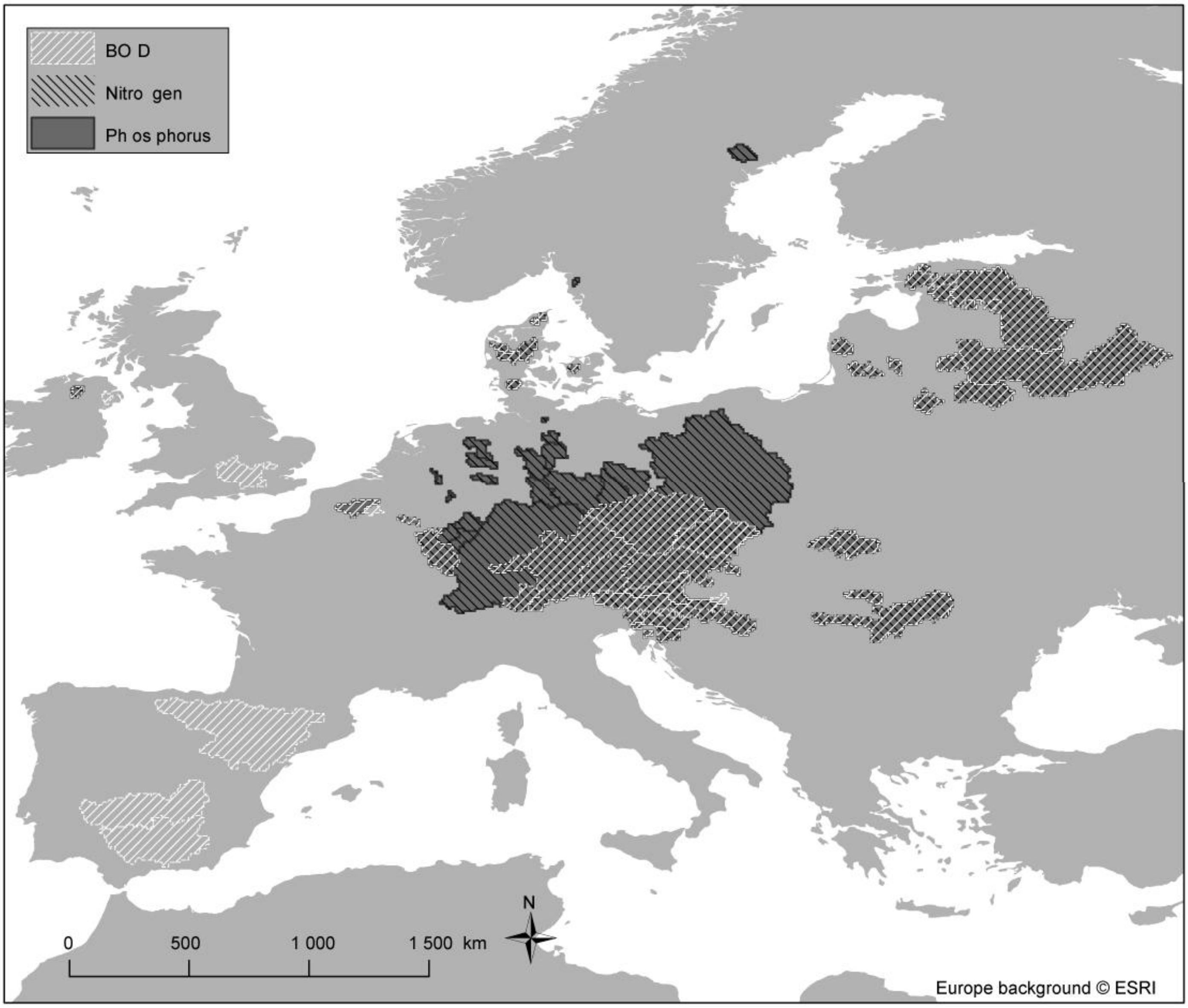

Figure 4. The delineated watersheds for the BOD_5, nitrogen and phosphorus regression models.

A linear export coefficient model for ***BOD_5* flux *f*** out from an experimental catchment area [*tn km*$^{-2}$*a*$^{-1}$] was:

$$f \sim 3.245e^{-3} \cdot lsu + 5.393e^{-1} \cdot point + 1.517e^{-3} \cdot r \quad (3)$$

where *lsu* is number of livestock units [lsu km^{-2}], *point* is load from point sources and scattered settlements [*tn km*$^{-2}$*a*$^{-1}$] and *r* is runoff [*mm a*$^{-1}$]. From the equation 3, we can conclude that export e_j due to a *lsu* is (β_i/ r_1=3.24/0.539=) 6,02 *kg a*$^{-1}$ *lsu*$^{-1}$, the retention of *point load* is 0.539 and export e_j due to *r* is (β_i/ r_1=1.517/0.539=) 2.81 *kg km*$^{-2}$ *mm*$^{-1}$. Simulated effects of all factors are clearly positive (Table 1). Figure 5 illustrates the model fits for BOD_5.

Table 1.Statistical summary of the BOD_5 model.

Coefficients:				
	Estimate	**Std. Error**	**t value**	**Pr(>|t|)**
lsu	3.245e-03	1.433e-03	2.265	0.0261 *
point	5.393e-01	1.079e-01	4.999	3.16e-06 ***
r	1.517e-03	8.871e-05	17.097	< 2e-16 ***

Residual standard error: 0.4171on 83degrees of freedom
Multiple R-squared: 0.9329, Adjusted R-squared: 0.930
F-statistic: 384.8 on 3 and 83 DF, p-value: < 2.2e-16
Significance codes: *0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1*.

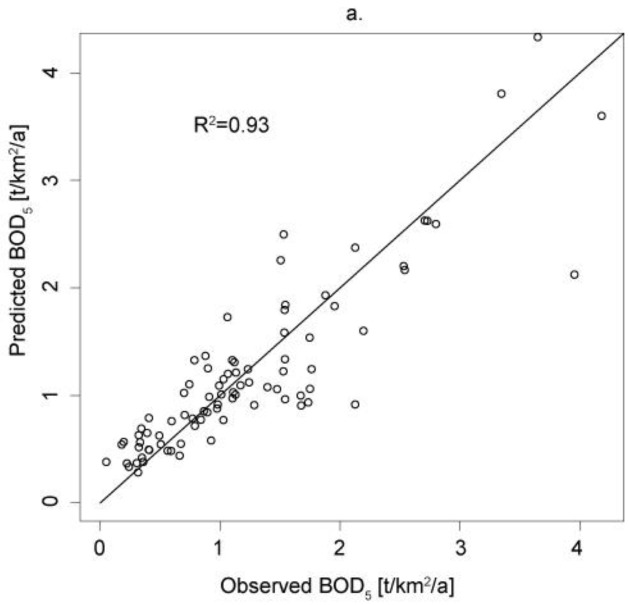

Figure 5. Model fits for BOD_5.

The median diffuse load calculated with BOD_5 equation for the selected river basins was 16 kg ha^{-1} a^{-1}. The corresponding calculated point load estimate for these river basins was 2.1 kg ha^{-1} a^{-1}. The diffuse load here included the load from scattered settlements. The calculated median BOD load at the outlet of the basin was 10.5 kg ha^{-1} a^{-1} (Figure 6).

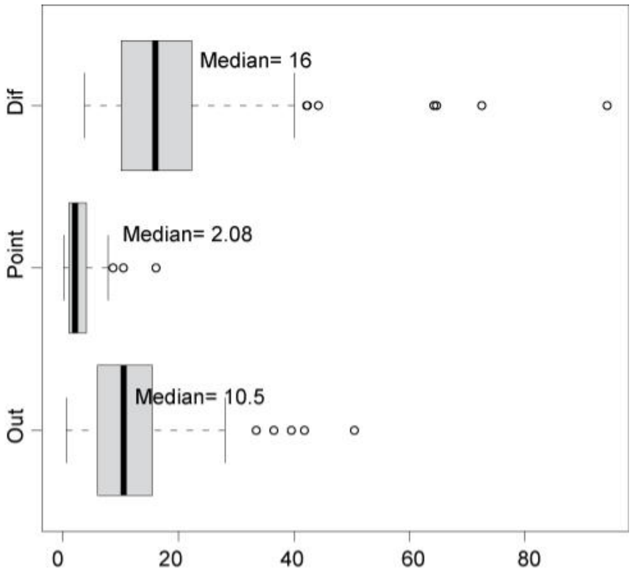

Figure 6. BOD balance of observed river basins. Box and whiskers: Min, Q1 (25%), Median, Q3 (75%) and Q3 + 1.5*(Q3-Q1). Dots are outliers.

Sensitivity of equation within the observational range was studied one factor at a time (Figure 7). The other factors were fixed to median. The study indicated high sensitivity to runoff – less to LSU and point load.

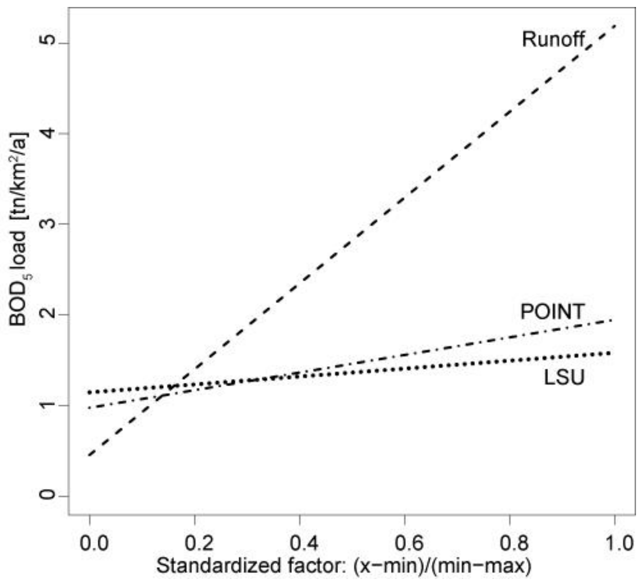

Figure 7. Sensitivity of BOD-equation. Predicted BOD as a function of one factor at a time (the other factors were fixed to median) within the observational range (Runoff: 33-3154 mm/a, LSU: 5-140 lsu/km2 and Point load: 0-1.8 t/km2/a). Note that the factors are standardized between 0 and 1.

The diffuse loads derived from the equations for BOD_5, total nitrogen and total phosphorus were compared with the data available from international pollution load reports. Only HELCOM reports diffuse BOD_5 loads every 5^{th} year for countries around the Baltic Sea. The sampling frequencies in monitored rivers are at least 12 times per year (HELCOM 2004). The regression equation was used in the WaterQual model (Voβ et al. in this volume) together with scattered settlements (Williams et al., in this volume), and resulting loads were compared with loads reported by HELCOM. Results show that the diffuse BOD_5 load was highly underestimated in all countries except Poland (Fig 8). The average difference between reported and modeled diffuse BOD_5 load was 3.2 kg ha^{-1} a^{-1}.

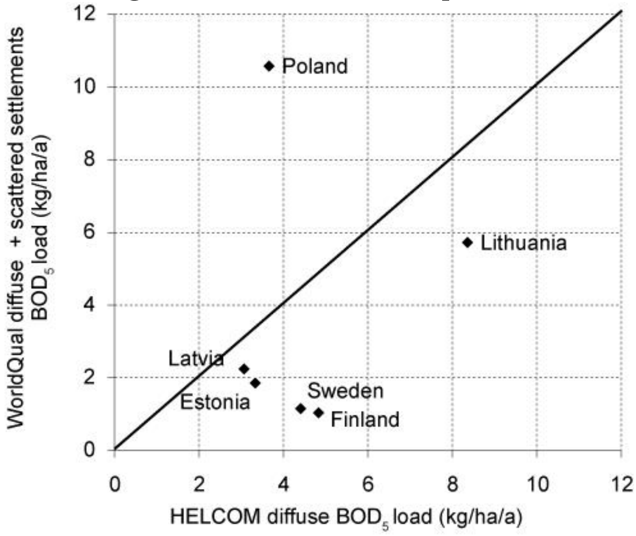

Figure 8. Predicted (WorldQual) and reported (HELCOM 2004) annual BOD loads.

Total phosphorus

As for the BOD_5 model, also for the nutrients (phosphorus and nitrogen) there were no significant differences between the equations developed for different time periods and the final equation included all the years from 1988 to 2002. The delineated watersheds for the total phosphorus (TP) calculations are presented in Figure 4.

A linear export coefficient model for ***TP* flux *f*** out from an experimental catchment area [*tn* $km^{-2}a^{-1}$] was:

$$f \sim 2.14e^{-4} \cdot lsu + 3.92e^{-1} \cdot point + 8.03e^{-5} \cdot r - 4.94e^{-3} \cdot lake - 4.69e^{-3} \cdot r \cdot slope \quad (4)$$

where *lsu* is number of livestock units [lsu km^{-2}], *point* is load from point sources and scattered settlements [*tn* $km^{-2}a^{-1}$], *r* is runoff [*mm* a^{-1}], *lake* is lake percentage [%] and *slope* is slope of terrain [%]. From the equation 4, we can conclude that the export e_j due to a *lsu* is (β_i/ r_1=0.2142/0.392=) 0.546 *kg* a^{-1} lsu^{-1}, the retention of *point* load is 0.3916, the export e_j due to runoff is (β_i/ r_1=0.08027/0.392=) 0.205 *kg* km^{-2} mm^{-1} and retention due to *lake* percentage is -4.941 kg $km^{-2}a^{-1}$ $\%^{-1}$. The export e_j due to the interaction of *slope* and runoff is (β_i/ r_1=-4.686/0.392=) -11.9 *kg* km^{-2} mm^{-1} (Table 2). Figure 9 illustrates the model fit for total phosphorus.

Table 2. Statistical summary of the total phosphorus model.

Coefficient	Estimate	Std. Error	t value	Pr(>|t|)
lsu	2.14e-04	8.27e-05	2.59	0.0111 *
point	3.92e-01	5.85e-02	6.70	1.22e-09 ***
r	8.03e-05	1.28e-05	6.26	9.54e-09 ***
lake	-4.94e-03	1.95e-03	-2.53	0.0130 *
r · slope	-4.69e-03	8.34e-04	-5.61	1.73e-07 ***

Residual standard error: 0.03041 on 101 degrees of freedom
Multiple R-squared: 0.785, Adjusted R-squared: 0.7743
F-statistic: 73.74 on 5 and 101 DF, p-value: < 2.2e-16
`Significance codes: 0 ‘***’ 0.001 ‘**’ 0.01 ‘*’ 0.05 ‘.’ 0.1.`

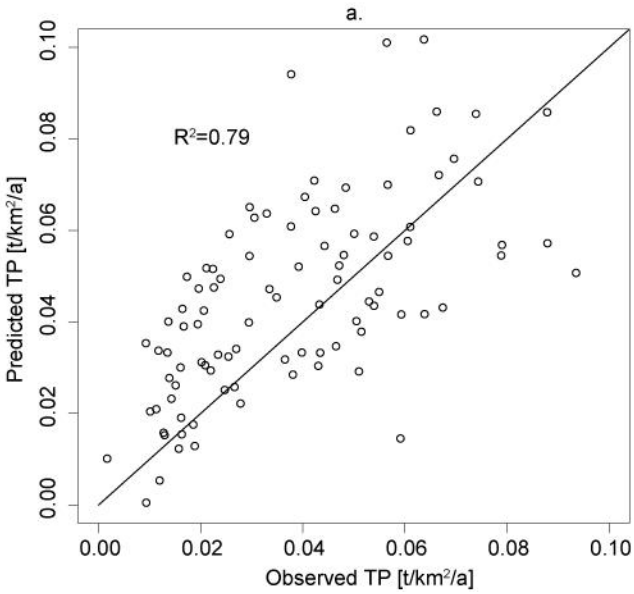

Figure 9. Model fit for total phosphorus.

The median diffuse load calculated with P equation for the selected river basins was 0.681 kg ha^{-1} a^{-1}. The corresponding calculated point load estimate for these river basins was 0.267 kg ha^{-1} a^{-1}. The diffuse load here included the load from scattered settlements. The calculated median P load at the outlet of the basin was 0.395 kg ha^{-1} a^{-1} (Figure 10).

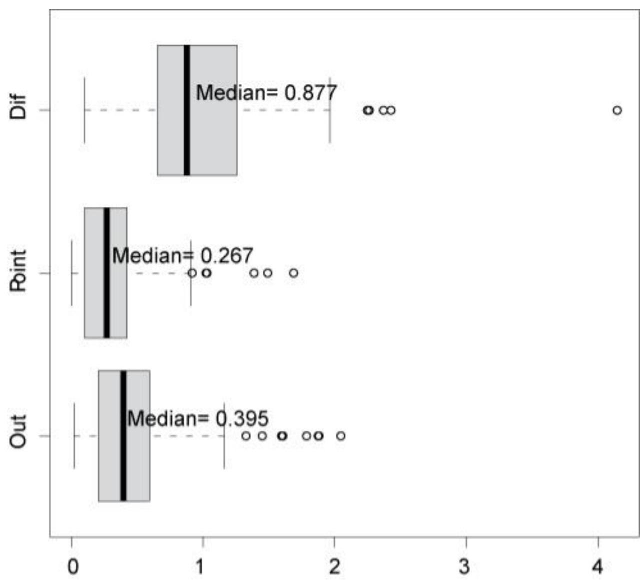

Figure 10. P balance of observed river basins. Box and whiskers: Min, Q1 (25%), Median, Q3 (75%) and Q3 + 1.5*(Q3-Q1). Dots are outliers.

Sensitivity analysis (Figure 11) revealed a strong positive effect of runoff and point load and a clear negative impact of lake percentage.

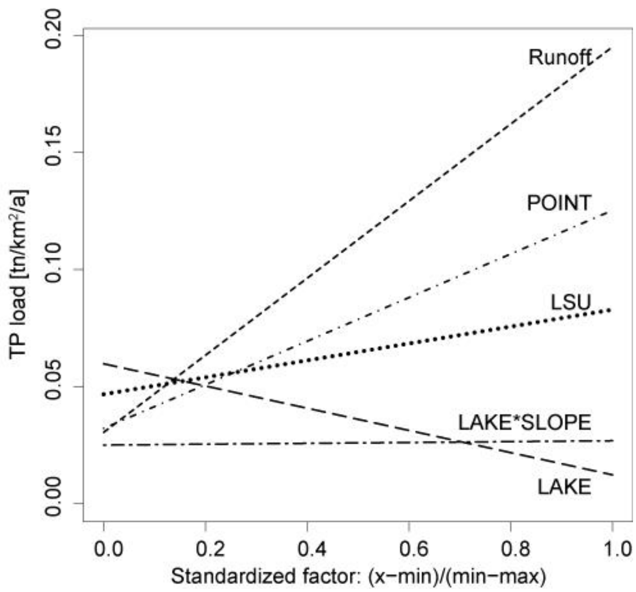

Figure 11. Sensitivity of TP-equation. Predicted TP as a function of one factor at a time (the other factors were fixed to median) within the observational range (Runoff: 33-3154 mm/a, LSU: 1-170 lsu/km2, Point load: 0-0.2 t/km2/a, Lake: 0-10 %). Note that the factors are standardized between 0 and 1.

The regression equation was used in the WaterQual model (Voβ et al. in this volume) together with scattered settlements (Williams et al., in this volume), and resulting loads were compared with country loads reported by HELCOM (2004), Schreiber et al. (2003) and Behrendt et al. (2003). For other countries, the loads were calculated based on river basin specific loads reported by EUROHARP (2003-2005). Comparing the predicted and reported diffuse phosphorus loads, an underestimation of modeled values was seen. The reported average diffuse phosphorus load including background load was 0.4 kg ha^{-1} a^{-1}. The corresponding average WorldQual estimate was only 0.25 kg ha^{-1} a^{-1} (Figure 12).

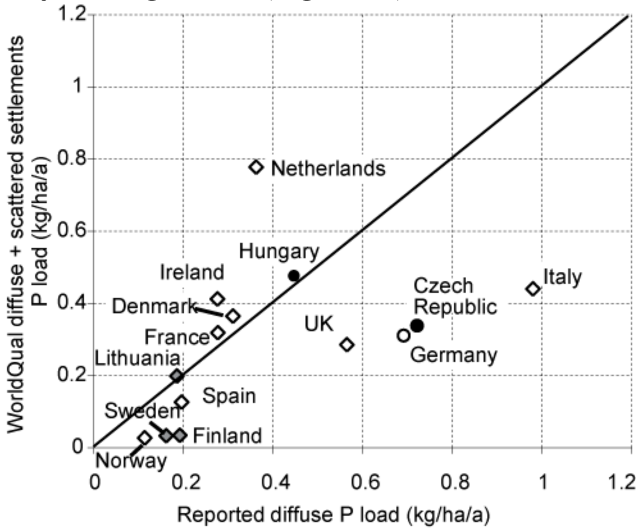

Figure 12. Predicted (WorldQual) and reported (●=Schreiber et al. (2003), o = Behrendt et al. (2003), ◆= HELCOM (2004), ◇= EUROHARP (2003-2005) annual diffuse P loads.

Total nitrogen

For the total nitrogen (TN) equation there was not as much concentration information available as there was for total phosphorus. The delineated watersheds for the total nitrogen (TN) calculations are presented in Figure 4.

A linear export coefficient model for ***TN* flux *f*** out from a experimental catchment area [*tn* $km^{-2}a^{-1}$] was:.

$$f \sim 5.58e^{-3} \cdot lsu + 5.91e^{-1} \cdot point + 1.07e^{-3} \cdot r - 6.13e^{-2} \cdot lake + 7.10e^{-3} \cdot crop \qquad (5)$$

where *lsu* is number of livestock units [lsu km^{-2}], *point* is load from point sources and scattered settlements [*tn* $km^{-2}a^{-1}$], *r* is runoff [*mm* a^{-1}], *lake* is lake percentage [%] and *crop* is percentage of cropland [%]. From the equation 5, we can conclude that the export e_j due to *lsu* is (β_i/ r_1= 5.58/0.591=) 9.44 *kg* a^{-1} lsu^{-1}, the retention of *point* load is 0.59, the export e_j due to runoff is (β_i/ r_1= 1.07/0.591=) 1.81 *kg* km^{-2} mm^{-1}, retention due to *lake* percentage is -61.32 kg $km^{-2}a^{-1}$ $\%^{-1}$ and the export e_j due to the percentage of crop area is (β_i/ r_1= 7.103/0.591=) 12.0 kg $km^{-2}a^{-1}$ $\%^{-1}$ (Table 3). Figure 13 illustrates the model fit for total nitrogen.

Table 3. Statistical summary of the total nitrogen model.

Coefficient	Estimate	Std. Error	t value	Pr(>|t|)
lsu	5.58e-3	1.84e-3	3.031	0.00336 **
point	5.91e-1	2.06e-1	2.869	0.00536 **
r	1.07e-3	1.23e-4	8.635	8.15e-13 ***
lake	-6.13e-2	2.95e-2	-2.080	0.04101 *
crop	7.10e-3	2.24e-3	3.167	0.00224 **

Residual standard error: 0.4385 on 74 degrees of freedom
Multiple R-squared: 0.8944, Adjusted R-squared: 0.8873
F-statistic: 125.4 on 5 and 74 DF, p-value: < 2.2e-16
Significance codes: 0 ‘***’ 0.001 ‘**’ 0.01 ‘*’ 0.05 ‘.’ 0.1

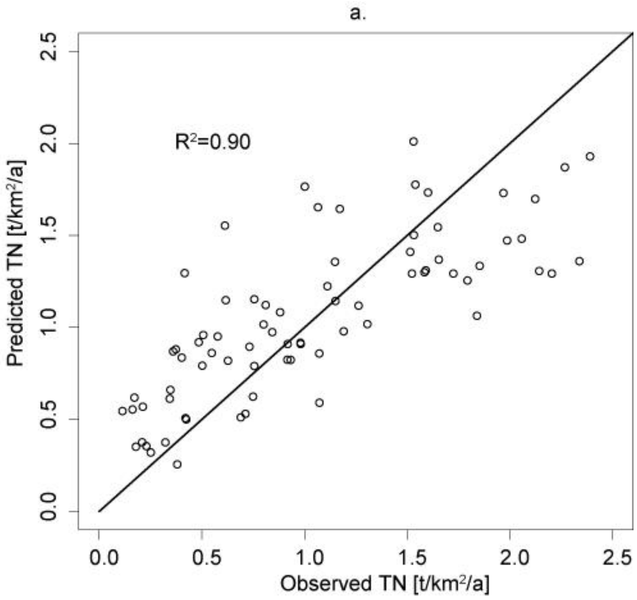

Figure 13. Model fit for total nitrogen.

The median diffuse load calculated with N equation for the selected river basins was 10.7 kg ha^{-1} a^{-1}. The corresponding calculated point load estimate for these river basins was 1.92 kg ha^{-1} a^{-1}. The diffuse load here included the load from scattered settlements. The calculated median N load at the outlet of the basin was 9.32 kg ha^{-1} a^{-1} (Figure 14).

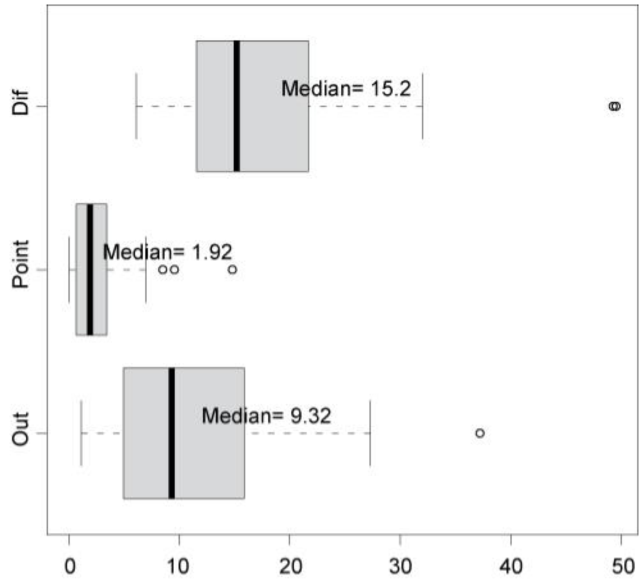

Figure 14. N balance of observed river basins. Box and whiskers: Min, Q1 (25%), Median, Q3 (75%) and Q3 + 1.5*(Q3-Q1). Dots are outliers.

Sensitivity analysis (Figure 15) revealed a strong positive effect of runoff, moderate effects of point load, LSU and percentage of crop land and a negative impact of lake percentage.

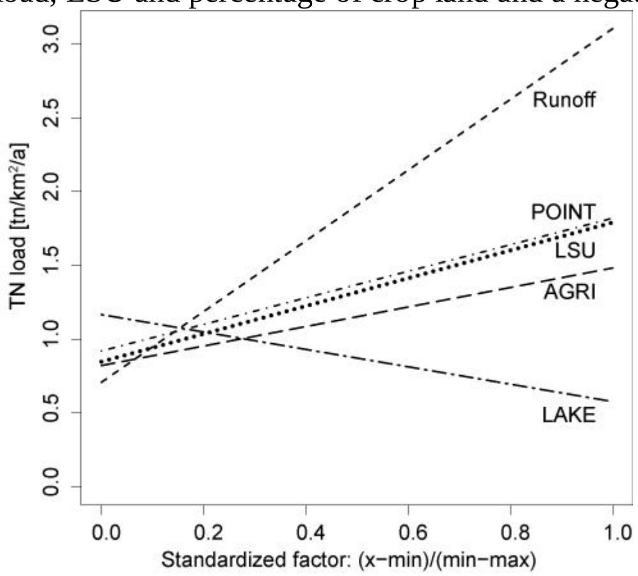

Figure 15. Sensitivity of TN-equation. Predicted TN as a function of one factor at a time (the other factors were fixed to median) within the observational range (Runoff: 33-2281 mm/a, LSU: 1-170 lus/km2, Point load: 0-1.5 t/km2/a, Crop: 0-93 % and Lake: 0-10 %.). Note that the factors are standardized between 0 and 1.

The WorldQual model predicted quite satisfactorily the country nitrogen loads of Italy, Czech Republic and the Netherlands (Figure 16). Here and also for phosphorus, the reported national diffuse loads were taken from HELCOM (2004), Schreiber et al. (2003), Behrendt et al. (2003) and EUROHARP (2003-2005). The diffuse load of N was underestimated in all Nordic countries. The biggest difference, 17.4 kg ha^{-1} a^{-1} between modeled and reported loads occurred in United Kingdom. The average difference between reported and modeled loads was 7.2 kg ha^{-1} a^{-1}.

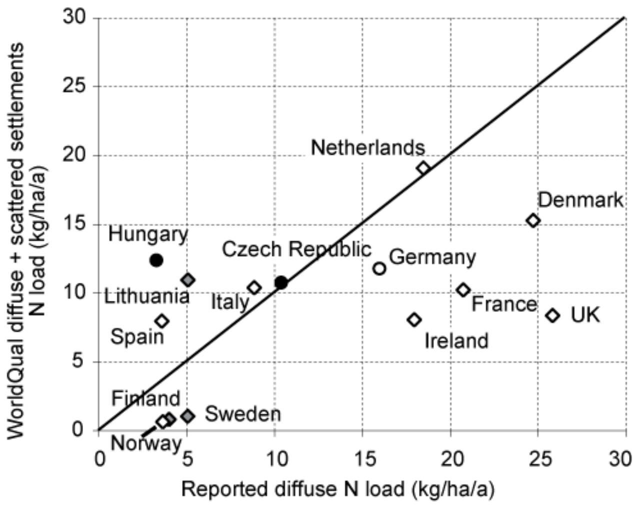

Figure 16. Predicted (WorldQual) and reported (●= Schreiber et al. (2003) , ○ = Behrendt et al. (2003), ◆= HELCOM (2004), ◇= EUROHARP (2003-2005)) annual diffuse N loads.

4. *Discussion and conclusions*

This paper presents the European diffuse export coefficient equations for BOD_5, total nitrogen and total phosphorus. The analysis was based on EU EEA databases of 79-106 selected river basins around the Europe depending on the variable in question. Estimated export coefficients were on a reasonable level. BOD_5 was best related to runoff, number of livestock and point load, while for total diffuse nitrogen load, also the lake and cropland area were influential. For diffuse phosphorus load, the factors were almost similar to nitrogen, but instead of cropland area, the combined factor of the average slope steepness multiplied by runoff had a negative impact on load. In both nutrient equations, the lake area had a negtive impact describing the retention character of lakes.

It was also showed that European wide or even global datasets can be used in the modeling of annual average diffusive nutrient flux at the larger basin scale. However, the imprecision of these equations should be seriously taken into account when applied for a smaller river basin. In addition, it should be noticed that these equations were validated using country scale loading data which are also based on indirect measurements and calculation methods. Furthermore, these methods differ from country to country. Thus, more data is also necessary for decisive validation.

Based on this study, the following conclusions were drawn: (i) The set of export coefficients for BOD, TP and TN were estimated based on European scale data to be used in the WorldQual model for the estimation of country specific diffuse loads. (ii) Comparison of the resulting loads with reported estimates from different countries revealed considerable error variance. For example, BOD load was underestimated and phosphorus and nitrogen estimates were scattered on both sides of 1:1line, with a slight underestimation of loads from the Nordic countries. (iii) If the model is applied in a river basin scale, more specific local data would improve the precision of the diffuse load estimates. In addition, larger set of data with higher spatial and temporal resolution, and partitioning of the data based on e.g. climate or spatial patterns would further improve the precision of the estimates.

References

Alcamo, J., Döll, P., Henrichs, T., Kaspar, F., Lehner, B. Rösch, T., Siebert, S. 2003. Development and testing of the WaterGAP2 global model of water use and availability. *Hydrol. Sci. J.* 48, 317-337.

Behrendt, H., Bach, M., Kunkel, R., Opitz, D., Pagenkopf, W.-G., Scholz, G., Wendland, F., 2003. Nutrient emissions into surface waters of Germany on the base of a harmonised procedure Summary. Environmental research Plan of the Federal Environmental Ministry, Water Management, Project No_29922285.

Behrendt, H. & Schreiber, H. 2004. Point and diffuse nutrient emissions and loads in the transboundary Danube river basin – a modelling approach. *Limnological Reports*, Vol. 35, 139-147.

Bouraoui, F. & Aloe, A., 2007. *European Agrochemicals Geospatial Loss Estimator: Model Development and Applications*. Institute of Environment and Sustainability, European Comission Joint Research Center. EUR 22690 EN.

CESR, 2007-2010. Center for Environmental Research. <*http://www.usf.uni-kassel.de/cesr*>. University of Kassel, Germany.

CLC2000, 2009. Corine Land Cover 2000 100 m, Version 12/2009. *http://www.eea.europa.eu/data-and-maps/data/corine-land-cover-2000-clc2000-100-m-version-12-2009*. European Environment Agency.

Döll, P., Kaspar, F., Lehner, B. 2003. A global hydrological model for deriving water availability indicators: model tuning and validation. *Journal of Hydrology* 270:105-134.

EEA, 2005. European Environment Agency. The European environment – State and outlook 2005. EEA, Copenhagen. ISBN 92-9167-776-0

EEA, 2007. Waterbase – Rivers Dataset, Version 20 Apr 2007 - Waterbase – Rivers. <*http://www.eea.europa.eu/data-and-maps/data/waterbase-rivers-6*>. The European Environment Agency, Denmark.

EPA, 2001. PLOAD version 3.0. An Arc View GIS Tool to Calculate Nonpoint Sources of Pollution in Watershed and Stormwater Projects, User's Manual.

EPA, 2006. User Guide, Spreadsheet Tool for the Estimation of Pollution Load (STEPL), Version 4.0.

EUROHARP, 2003-2005. Towards European Harmonised Procedures for Quantification of Nutrient Losses from Diffuse Sources. <*http://www.iis.niva.no/php/euroharp/diss/rep.htm*>

FAOSTAT, 2010. <*http://faostat.fao.org/default.aspx*> Food and Agriculture Organization of the United Nations (FAO).

Flörke, M. & Alcamo, J. 2004. European Outlook on Water Use. Center for Environmental Systems Research, University of Kassel, Final Report, EEA/RNC/03/007, 83 pp.

GLCC, 2008. Global Land Cover Characterization, Version 2.0. *http://edc2.usgs.gov/glcc/tablambert_euras_eur.php*. U.S. Geological Survey.

Goethe-Universität, 2008. Global map of artificially drained agricultural areas. *http://www.geo.uni-frankfurt.de/ipg/ag/dl/forschung/Global_Drainage_Map/index.html*. Goethe-Universität Frankfurt am Main, Germany.

Grizzetti, B. & Bouraoui, F. 2006. Assessment of nitrogen and phosphorus environmental pressure at European scale. *EUR Report* 22526 EN, 66p.

HELCOM, 2004. *The Fourth Baltic Sea Pollution Load Compilation (PLC-4).* Baltic Sea Environment Proceedings No. 93, Baltic Marine Environment Protection Commission - Helsinki Commission, ISSN 0357-2994

Hesse, C., Krysanova, V., Päzolt, J. & Hattermann, F.F. 2008. Eco-hydrological modelling in a highly regulated lowland catchment to find measures for improving water quality. *Ecological modeling* 218:135-148.

Institute for Veterinary Public Health, 2008. World Map of the Köppen-Geiger climate classification. *http://koeppen-geiger.vu-wien.ac.at/*. University of Veterinary Medicine Vienna, Austria.

JRC, 2008. River and Catchment database, Version 2.0, CCM2 Download. *http://ccm.jrc.ec.europa.eu/php/index.php?action=view&id=24*. Institute for Environment and Sustainability, Joint Research Centre, European Commission.

Mulligan, D., Bouraoui, F., Grizzetti, B., Aloe, A., Dusart, J., 2006. *An Atlas of Pan- European Data for Investigating the Fate of Agrochemicals in Terrestrial Ecosystems*. European Comission, Joint Research Centre, Institute of Environment and Sustainability. EUR 22334 EN.

Pettersson, A., Brandt, M. & Lindström, G., 2000. Application of the HBV-N model to the Baltic Sea drainage basin. *Vatten,* 56:7-13.

Pistocchi, A., Vizcaino, P. & Pennington, D, 2006. *Analysis of Landscape and Climate pArameters for Continental Scale Assessmentof the Fate of Pollutants*. Institute of Environment and Sustainability, European Comission Joint Research Center. EUR 22624 EN.

R Development Core Team, 2009. R: A language and environment for statistical computing. Vienna, Austria: R Foundation for Statistical Computing.

Rantakari, M. Kortelainen, P. Vuorenmaa, J. Mannio, J. Forsius, M., 2004. Finnish lake survey: the role of catchment attributes in determining nitrogen, phosphorus, and organic carbon concentrations. Water, Air and Soil Pollution: Focus 4: 683-699.

Schreiber, H, Constaniescu, L. T., Cvitanic, I., Drumea, D., Jabucar, D., Juran, S., Pataki, B., Snishko, S., Zessner, M. & Behrendt, H. 2003. Harmonised Inventory of Point and Diffuse

Emissions of Nitrogen and Phospsorus for a Transboundary River Basin, Final version of the delivery 5.5 of the EU-project DANUBS.

Shrestha, S., Kazama, F. and Nweham, L.T.H., 2008. A framework for estimating pollutant export coefficients from long-term in-stream water quality monitoring data. – Environmental Modeling & Software 23 (2008) 182 – 194.

Tattari, S., Koskiaho, J., Bärlund, I. & Jaakkola, E., 2009. Testing a river basin model with sensitivity analysis and autocalibration for an agricultural catchment in SW Finland. *Agricultural and Food Science* 18, 3-4: 428-439.

Voß, A., J. Alcamo, I. Bärlund, F. Voß, E. Kynast, R. Williams & O. Malve., in this volume. Continental scale modelling of in-stream river water quality: A report on methodology, test runs, and scenario application.

Wade, A.J., Jackson, B.M., Butterfield, D., 2008. Over-parametrised, uncertain 'mathematical marionettes' – How we can best use catchment water quality models? An example of an 80-year catchment-scale nutrient balance. *Science of Total Environment* 400:52-74.

Williams, R., Keller, V., Voß, A. Bärlund, I., Malve, O., Riihimäki, J., Tattari, S. & Alcamo, J., in this volume. Assessment of current water pollution loads in Europe: Estimation of gridded loads for use in global water quality models.

Zhang, X., R. Srinivasan, & M. Van Liew., 2008. Multi-Site Calibration of the SWAT Model for Hydrologic Modeling, *Transactions of the ASABE*. 51(6): 2039-2049.

Supplemental material

Appendix 1. Catchment data available from 1988-2002 for the BOD regression model (n=104).

Year	Basin	River	Country	Area (km^2)	Lake-%	Cropland (km^2)	Livestock (LSU)	Temperature (°C)	Runoff (m^3/s)	BOD type	co (r
2000	Barta	Barta	LV	2273	0.3	1010	21740	8.1	15.2	BOD_7	
1990	Bodrog_90	Bodrog	HU	8649	0.6	2887	237037	9.0	80.1	BOD_5	
1992	Danube1	Bodrog	SK	8535	0.6	2837	232730	8.7	105.0	BOD_5	
1995	Danube1	Bodrog	SK	8535	0.6	2837	146078	8.4	112.8	BOD_5	
2000	Danube1	Bodrog	SK	8535	0.6	2837	126546	9.2	136.8	BOD_5	
1994	Danube2	Inn	AT/GE	26444	0.7	3587	912246	6.5	708.0	BOD_5	
1999	Danube2	Inn	AT/GE	26444	0.7	3587	1126427	5.5	875.0	BOD_5	
1990	Danube3	Sebes-Körös	HU	3431	0.5	1066	141666	9.5	10.1	BOD_5	
1995	Danube3	Sebes-Körös	HU	3431	0.5	1066	90580	9.1	27.0	BOD_5	
2000	Danube3	Sebes-Körös	HU	3431	0.5	1066	77068	10.0	28.2	BOD_5	
1995	Danube4	Danube4	HU	135111	0.7	47859	6733927	7.7	2180.0	BOD_5	
2001	Danube4	Danube4	HU	135111	0.7	47859	6713490	7.6	2140.0	BOD_5	
1995	Danube5	Danube5	SK	133971	0.7	47088	6687310	7.7	2329.0	BOD_5	
2000	Danube5	Danube5	SK	133971	0.7	47088	6695003	8.5	2338.0	BOD_5	
1995	Danube6	Danube6	SK	25847	0.5	13346	853597	8.4	107.4	BOD_5	
2002	Danube6	Danube6	SK	25847	0.5	13346	845774	9.1	122.5	BOD_5	
2000	Daugava	Daugava	LV	70953	3.1	7305	791640	6.8	552.0	BOD_7	
1991	Drava_90	Drava	SI	13363	0.8	1049	289470	4.6	282.0	BOD_5	
1991	Drava3	Drava3	SI	16667	0.7	1992	429942	5.6	323.0	BOD_5	
1995	Drava3	Drava3	SI	16667	0.7	1992	385853	6.0	262.0	BOD_5	
2001	Drava3	Drava3	SI	16667	0.7	1992	501220	6.8	296.0	BOD_5	
1995	Drava4	Drava4	HU	39850	0.5	9340	932249	7.8	503.4	BOD_5	
2000	Drava4	Drava4	HU	39850	0.5	9340	1003260	8.4	540.7	BOD_5	
2000	Ebro	Ebro	ES	84265	0.4	34328	2764315	12.2	243.3	BOD_5	
2001	Escaut	Escaut	BE	4213	0.4	2971	403338	10.6	48.0	BOD_5	
1991	Foyle4	Mourne river	GB	1036	0.1	0	154769	8.4	55.9	BOD_5	
2000	Foyle4	Mourne River	GB	1036	0.1	0	144668	8.4	65.4	BOD_5	
2000	Guadalquivir	Guadalquivir	ES	47799	0.5	16387	676720	16.2	33.3	BOD_5	
2000	Guadiana	Guadiana	ES	46742	0.9	24130	998908	15.8	36.8	BOD_5	
2001	GulfOfFinland	Keila	EE	1012	0.3	274	18671	6.1	6.9	BOD_7	
2001	GulfOfRiga	Pärnu	EE	5486	0.1	1182	28191	6.1	51.5	BOD_7	
2001	GulfOfRiga4	Pärnu	EE	2204	0.0	544	13784	6.1	23.5	BOD_7	
2001	GulfRiga2	Kasari	EE	2913	0.1	680	19779	6.3	27.2	BOD_7	
1990	Gyongyos	Balaton	HU	13049	0.1	2269	431319	7.7	156.0	BOD_5	
1995	Gyongyos	Balaton	HU	13049	0.1	2269	389753	7.4	171.0	BOD_5	
2001	Gyongyos	Balaton	HU	13049	0.1	2269	498611	7.6	126.0	BOD_5	
1990	Hornad3	Hornad	SK	4152	0.3	1156	241865	8.5	20.6	BOD_5	
2002	Hornad3	Hornad	SK	4152	0.3	1156	127356	8.2	18.0	BOD_5	
1991	Kolpa	Kolpa	SI	3120	0.1	231	52103	8.6	69.3	BOD_5	

1995	Kolpa	Kolpa	SI	3120	0.1	231	40122	9.0	82.3	BOD_5
1999	Kolpa	Kolpa	SI	3120	0.1	231	56437	9.5	84.4	BOD_5
1994	Labe	Labe	CZ	51714	0.9	22095	1654439	9.0	278.0	BOD_5
2001	Labe	Labe	CZ	51714	0.9	22095	1666796	8.0	315.0	BOD_5
1991	Lagan	Lagan	UK-IRL	899	0.6	71	181699	8.9	7.4	BOD_5
1995	Lagan	Lagan	UK-IRL	899	0.6	71	178001	9.4	8.4	BOD_5
2000	Lagan	Lagan	UK-IRL	899	0.6	71	169721	9.1	10.9	BOD_5
1995	Merkys	Merkys	LT	3300	1.6	832	51755	6.7	34.9	BOD_5
2000	Merkys	Merkys	LT	3300	1.6	832	42564	7.5	30.1	BOD_7
2000	Meuse1	Sambre	FR/BE	1485	0.1	443	176769	10.7	16.7	BOD_5
1995	Meuse2	Semois	FR/BE	1320	0.1	238	29023	9.5	37.8	BOD_5
1990	Moselle_90	Moselle	FR	11928	0.6	4141	679700	10.2	123.0	BOD_5
1990	Mura_1990	Mura	SI	10099	0.1	835	338573	6.6	130.9	BOD_5
1990	Mures	Maros	HU	30144	0.1	7502	1164554	9.0	95.0	BOD_5
1995	Mures	Mures	HU	30144	0.1	7502	748766	8.5	182.0	BOD_5
2002	Mures	Mures	HU	30144	0.1	7502	637956	9.0	181.9	BOD_5
2001	narva1	Narva	EE	46329	9.6	6068	279017	5.8	303.0	BOD_7
2001	narva4	Väike-Emajõgi	EE	906	1.2	185	6689	6.0	8.4	BOD_7
2001	narva5	Võhandu	EE	1221	1.2	260	9173	5.7	8.0	BOD_7
1994	Nemunas10	Jura	LT	1832	0.6	814	23694	7.0	26.3	BOD_5
1999	Nemunas10	Jura	LT	1832	0.6	814	22091	7.5	21.1	BOD_7
1995	Nemunas11	Sventoji	LT	3600	4.7	1360	39414	6.5	32.2	BOD_5
2002	Nemunas11	Sventoji	LT	3600	4.7	1360	36101	7.2	29.2	BOD_7
1994	Nemunas6	Sesuvis	LT	1171	1.0	636	15712	7.2	20.3	BOD_5
2001	Nemunas6	Sesuvis	LT	1171	1.0	636	14546	7.3	18.2	BOD_7
2001	Nemunas8	Susve	LT	1299	2.0	778	20241	7.3	1.4	BOD_7
1995	Nemunas9	Streva	LT	296	5.3	114	5377	6.6	1.9	BOD_5
1999	Nemunas9	Streva	LT	296	5.3	114	7432	7.4	1.7	BOD_7
1995	Neris	Neris	LT	10840	2.1	1919	279257	6.7	59.6	BOD_5
2000	Neris	Neris	LT	10840	2.1	1919	245340	7.5	51.3	BOD_7
1994	Nordjylland2	Uggerby Å	DK	460	0.0	316	36223	8.4	5.0	BOD_5
1999	Nordjylland2	Uggerby Å	DK	460	0.0	316	42075	8.1	5.8	BOD_5
1995	Nordjylland3	Ry Å	DK	368	0.0	342	28724	8.0	3.2	BOD_5
1999	Nordjylland3	Ry Å	DK	368	0.0	342	36745	8.0	4.5	BOD_5
1992	Raba_1990	Raba	HU	1403	0.4	648	40037	11.4	3.7	BOD_5
1991	Reka_1990	Reka	SI	540	0.2	51	6402	8.4	8.4	BOD_5
1992	Rhein_1990	Lustenauer	AT	6315	0.3	726	152665	4.9	217.4	BOD_5
1995	Rhine2	Mosel	FR/GE	11928	0.6	4141	660108	10.1	202.0	BOD_5
1991	Ringkob_90	Skern Å	DK	1872	0.3	1170	77856	8.2	21.7	BOD_5
1999	Ringkobing	Storå	DK	1137	0.2	813	84322	8.7	21.8	BOD_5
1990	Sava	Sava	SI	8642	0.1	1712	364059	9.7	258.0	BOD_5
1995	Sava	Sava	SI	8642	0.1	1712	302322	9.4	299.0	BOD_5
2001	Sava	Sava	SI	8642	0.1	1712	369069	9.5	274.0	BOD_5
1991	Sava2	Sava2	SI	531	0.4	28	10829	5.8	53.1	BOD_5
1995	Sava2	Sava2	SI	531	0.4	28	8978	6.3	35.2	BOD_5
1999	Sava2	Sava2	SI	531	0.4	28	36511	7.0	38.4	BOD_5

1999	Savinja2	Savinja2	SI	1239	0.3	161	89641	8.5	44.3	BOD_5
1991	Soca	Soca	SI	2124	0.2	60	33693	7.7	89.5	BOD_5
1995	Soca	Soca	SI	2124	0.2	60	27951	8.1	84.5	BOD_5
2001	Soca	Soca	SI	2124	0.2	60	49046	7.1	93.7	BOD_5
1995	Soenderjylland	Groenå	DK	1078	0.4	810	57523	8.6	7.8	BOD_5
1998	Soenderjylland	Groenå	DK	1078	0.4	810	56671	8.5	8.4	BOD_5
2002	Thames1	Thames	GB	10197	0.7	6000	482694	10.8	100.3	BOD_5
2002	Thames2	Lee (R.Ash)	GB	689	0.2	607	11149	11.1	7.5	BOD_5
1995	Vestsjaelland	Tude Å	DK	673	3.1	487	56960	8.6	2.1	BOD_5
1998	Vestsjaelland	Tude Å	DK	673	3.1	487	56849	8.7	2.3	BOD_5
1991	Vipava	Vipava	SI	717	0.1	105	21010	10.6	18.4	BOD_5
1995	Vipava	Vipava	SI	717	0.1	105	17442	10.9	22.2	BOD_5
1999	Vipava	Vipava	SI	717	0.1	105	29692	10.7	14.2	BOD_5
1990	Zala	Zala	HU	986	0.0	356	25557	10.1	1.2	BOD_5
1995	Zala	Zala	HU	986	0.0	356	22788	9.4	2.4	BOD_5
1998	Zala	Zala	HU	986	0.0	356	32522	10.1	2.4	BOD_5
1991	Århus Amt	Guden Å	DK	2519	2.3	1553	204465	8.1	28.2	BOD_5
1994	Århus Amt	Guden Å	DK	2519	2.3	1553	215007	8.5	41.1	BOD_5
1999	Århus Amt	Guden Å	DK	2519	2.3	1553	214558	8.3	37.4	BOD_5

Appendix 2. Catchment data available from 1988-2002 for the phosphorus regression model (n=106).

Year	Basin	River	Country	Area (km^2)	Lake-%	slope (%)	Cropland (km^2)	Livestock (LSU)	Temperature (°C)	Runo (m^3/s
2000	Aller	Aller	DE	3545	0.2	0.005	1854	508814	9.7	35.3
2000	Aller2	Leine2	DE	6105	0.9	0.004	3404	1039812	9.9	51.1
2000	Barta	Barta	LV	2273	0.3	0.002	1010	21740	8.1	15.2
1990	Bodrog_90	Bodrog	HU	8649	0.6	0.006	2887	237037	9.0	80.1
1992	Danube1	Bodrog	SK	8535	0.6	0.006	2837	232730	8.7	105.
1995	Danube1	Bodrog	SK	8535	0.6	0.006	2837	146078	8.4	112.
1994	Danube2	Inn	AT/GE	26444	0.7	0.019	3587	912246	6.5	708.
1995	Danube3	Sebes-Körös	HU	3431	0.5	0.006	1066	90580	9.1	27.0
2000	Danube3	Sebes-Körös	HU	3431	0.5	0.006	1066	77068	10.0	28.2
1995	Danube4	Danube4	HU	135111	0.7	0.008	47859	6733927	7.7	2180
2001	Danube4	Danube4	HU	135111	0.7	0.008	47859	6713490	7.6	2140
1995	Danube5	Danube5	SK	133971	0.7	0.008	47088	6687310	7.7	2329
2000	Danube5	Danube5	SK	133971	0.7	0.008	47088	6695003	8.5	2338
1995	Danube6	Danube6	SK	25847	0.5	0.005	13346	853597	8.4	107.
2002	Danube6	Danube6	SK	25847	0.5	0.005	13346	845774	9.1	122.
2000	Daugava	Daugava	LV	70953	3.1	0.001	7305	791640	6.8	552.
1991	Drava_90	Drava	SI	13363	0.8	0.020	1049	289470	4.6	282.
1991	Drava3	Drava3	SI	16667	0.7	0.018	1992	429942	5.6	323.
1995	Drava3	Drava3	SI	16667	0.7	0.018	1992	385853	6.0	262.
2001	Drava3	Drava3	SI	16667	0.7	0.018	1992	501220	6.8	296.
1995	Drava4	Drava4	HU	39850	0.5	0.013	9340	932249	7.8	503.
2000	Drava4	Drava4	HU	39850	0.5	0.013	9340	1003260	8.4	540.
2000	Elbe	Elbe	DE	1441	0.0	0.001	816	74377	10.3	7.7
2000	Elbe2	Elbe2	DE	61207	1.0	0.005	26803	1975228	9.1	348.

2000	Elbe3	Saale	DE	17599	0.8	0.005	10321	974896	9.4	89.2
2000	Elbe4	Bile	DE	255	0.9	0.001	115	34775	10.2	2.4
2000	Ems	Hase	DE	3380	0.1	0.001	2498	355221	10.7	27.9
2000	Ems2	Ems2	DE	2952	0.3	0.001	2263	214715	10.8	35.5
2000	Enningdalsälven	Enningdalsälven	SE	353	8.7	0.005	33	1470	7.6	18.9
2000	Erft	Saale	DE	432	0.7	0.005	250	21662	11.0	0.7
2001	Escaut	Escaut	BE	4213	0.4	0.001	2971	403338	10.6	48.0
2000	Foyle4	Mourne River	GB	1036	0.1	0.003	0	144668	8.4	65.4
2001	GulfOfFinland	Keila	EE	1012	0.3	0.001	274	18671	6.1	6.9
2001	GulfOfRiga	Pärnu	EE	5486	0.1	0.001	1182	28191	6.1	51.5
2001	GulfOfRiga4	Pärnu	EE	2204	0.0	0.001	544	13784	6.1	23.5
2001	GulfRiga2	Kasari	EE	2913	0.1	0.001	680	19779	6.3	27.2
1990	Gyongyos	Balaton	HU	13049	0.1	0.013	2269	431319	7.7	156.
1995	Gyongyos	Balaton	HU	13049	0.1	0.013	2269	389753	7.4	171.
2001	Gyongyos	Balaton	HU	13049	0.1	0.013	2269	498611	7.6	126.
2002	Hornad3	Hornad	SK	4152	0.3	0.007	1156	127356	8.2	18.0
1991	Kolpa	Kolpa	SI	3120	0.1	0.008	231	52103	8.6	69.3
1995	Kolpa	Kolpa	SI	3120	0.1	0.008	231	40122	9.0	82.3
1999	Kolpa	Kolpa	SI	3120	0.1	0.008	231	56437	9.5	84.4
1994	Labe	Labe	CZ	51714	0.9	0.005	22095	1654439	9.0	278.
2001	Labe	Labe	CZ	51714	0.9	0.005	22095	1666796	8.0	315.
2000	Maas	Niers	DE	798	0.8	0.001	521	62144	11.4	8.1
2000	Merkys	Merkys	LT	3300	1.6	0.001	832	42564	7.5	30.1
1995	Meuse2	Semois	FR/BE	1320	0.1	0.004	238	29023	9.5	37.8
2000	Meuse2	Semois	FR/BE	1320	0.0	0.004	238	29001	10.0	36.5
2000	Mosel1	Sar	DE	6886	0.4	0.003	2409	360111	10.7	90.5
1990	Moselle_90	Moselle	FR	11928	0.6	0.003	4141	679700	10.2	123.
2000	Moälven	Moälven	SE	2318	2.0	0.005	34	3267	2.9	28.9
2000	Mulde	Vereinig. Mulde	DE	7375	0.4	0.006	4000	418407	9.2	66.8
1990	Mura_1990	Mura	SI	10099	0.1	0.015	835	338573	6.6	130.
1995	Mures	Mures	HU	30144	0.1	0.008	7502	748766	8.5	182.
2002	Mures	Mures	HU	30144	0.1	0.008	7502	637956	9.0	181.
2001	narva1	Narva	EE	46329	9.6	0.001	6068	279017	5.8	303.
2001	narva4	Väike-Emajõgi	EE	906	1.2	0.002	185	6689	6.0	8.4
2001	narva5	Võhandu	EE	1221	1.2	0.001	260	9173	5.7	8.0
1994	Nemunas10	Jura	LT	1832	0.6	0.002	814	23694	7.0	26.3
1999	Nemunas10	Jura	LT	1832	0.6	0.002	814	22091	7.5	21.1
2002	Nemunas11	Sventoji	LT	3600	4.7	0.001	1360	36101	7.2	29.2
1994	Nemunas6	Sesuvis	LT	1171	1.0	0.002	636	15712	7.2	20.3
2001	Nemunas6	Sesuvis	LT	1171	1.0	0.002	636	14546	7.3	18.2
2001	Nemunas8	Susve	LT	1299	2.0	0.001	778	20241	7.3	1.4
1999	Nemunas9	Streva	LT	296	5.3	0.002	114	7432	7.4	1.7
2000	Neris	Neris	LT	10840	2.1	0.001	1919	245340	7.5	51.3
1994	Nordjylland2	Uggerby Å	DK	460	0.0	0.001	316	36223	8.4	5.0
1999	Nordjylland2	Uggerby Å	DK	460	0.0	0.001	316	42075	8.1	5.8
1995	Nordjylland3	Ry Å	DK	368	0.0	0.001	342	28724	8.0	3.2
1999	Nordjylland3	Ry Å	DK	368	0.0	0.001	342	36745	8.0	4.5
2000	Oder	Oder	DE	110628	1.3	0.002	56573	3780156	9.9	494.
1991	Reka_1990	Reka	SI	540	0.2	0.011	51	6402	8.4	8.4
2000	Rhine	Rhine	DE	94780	1.6	0.009	38285	5546431	9.3	1770
2000	Rhine1	Saar	GE	3756	0.6	0.003	1242	234409	10.6	53.6
1995	Rhine2	Mosel	FR/GE	11928	0.6	0.004	4141	660108	10.1	202.
2000	Rhine2	Mosel	FR/GE	11928	0.6	0.004	4141	708576	10.8	92.6
2000	Rhine3	Ruhr	DE	1806	0.8	0.004	517	67465	9.7	27.5

2000	Rhine4	Nahe	DE	3637	0.1	0.004	1401	103370	10.3	43.3
1991	Ringkob_90	Skern Å	DK	1872	0.3	0.001	1170	77856	8.2	21.7
1999	Ringkobing	Storå	DK	1137	0.2	0.001	813	84322	8.7	21.8
1990	Sava	Sava	SI	8642	0.1	0.009	1712	364059	9.7	258.
1995	Sava	Sava	SI	8642	0.1	0.009	1712	302322	9.4	299.
2001	Sava	Sava	SI	8642	0.1	0.009	1712	369069	9.5	274.
1991	Sava2	Sava2	SI	531	0.4	0.017	28	10829	5.8	53.1
1995	Sava2	Sava2	SI	531	0.4	0.017	28	8978	6.3	35.2
1999	Sava2	Sava2	SI	531	0.4	0.017	28	36511	7.0	38.4
1999	Savinja2	Savinja2	SI	1239	0.3	0.011	161	89641	8.5	44.3
1991	Soca	Soca	SI	2124	0.2	0.015	60	33693	7.7	89.5
1995	Soca	Soca	SI	2124	0.2	0.015	60	27951	8.1	84.5
2001	Soca	Soca	SI	2124	0.2	0.015	60	49046	7.1	93.7
1995	Soenderjylland	Groenå	DK	1078	0.4	0.001	810	57523	8.6	7.8
1998	Soenderjylland	Groenå	DK	1078	0.4	0.001	810	56671	8.5	8.4
2000	Weser	Werra	DE	6040	0.1	0.005	2203	306407	9.2	48.4
2000	Weser2	Aller	DE	4644	0.4	0.003	2600	601011	10.0	22.0
1995	Vestsjaelland	Tude Å	DK	673	3.1	0.001	487	56960	8.6	2.1
1998	Vestsjaelland	Tude Å	DK	673	3.1	0.001	487	56849	8.7	2.3
1991	Vipava	Vipava	SI	717	0.1	0.009	105	21010	10.6	18.4
1995	Vipava	Vipava	SI	717	0.1	0.009	105	17442	10.9	22.2
1999	Vipava	Vipava	SI	717	0.1	0.009	105	29692	10.7	14.2
1990	Zala	Zala	HU	986	0.0	0.005	356	25557	10.1	1.2
1995	Zala	Zala	HU	986	0.0	0.005	356	22788	9.4	2.4
1998	Zala	Zala	HU	986	0.0	0.005	356	32522	10.1	2.4
1991	Århus Amt	Guden Å	DK	2519	2.3	0.001	1553	204465	8.1	28.2
1994	Århus Amt	Guden Å	DK	2519	2.3	0.001	1553	215007	8.5	41.1
1999	Århus Amt	Guden Å	DK	2519	2.3	0.001	1553	214558	8.3	37.4

Appendix 3. Catchment data available from 1988-2002 for the nitrogen regression model (n=79).

Year	Basin	River	Country	Area (km^2)	Lake-%	Cropland (km^2)	Livestock (LSU)	Temperature (°C)	Runoff (m^3/s)	N conce (mg/
2000	Aller	Aller	DE	3545	0.2	1854	508814	9.7	35.3	5.1
2000	Aller2	Leine2	DE	6105	0.9	3404	1039812	9.9	51.1	5.8
2000	Barta	Barta	LV	2273	0.3	1010	21740	8.1	15.2	1.6
1990	Bodrog_90	Bodrog	HU	8649	0.6	2887	237037	9.0	80.1	3.4
1995	Danube3	Sebes-Körös	HU	3431	0.5	1066	90580	9.1	27.0	2.2
2000	Danube3	Sebes-Körös	HU	3431	0.5	1066	77068	10.0	28.2	2.4
1995	Danube4	Danube4	HU	135111	0.7	47859	6733927	7.7	2180.0	3.4
2001	Danube4	Danube4	HU	135111	0.7	47859	6713490	7.6	2140.0	3.6
1995	Danube5	Danube5	SK	133971	0.7	47088	6687310	7.7	2329.0	3.4
2000	Danube5	Danube5	SK	133971	0.7	47088	6695003	8.5	2338.0	2.9
1995	Danube6	Danube6	SK	25847	0.5	13346	853597	8.4	107.4	3.7
2002	Danube6	Danube6	SK	25847	0.5	13346	845774	9.1	122.5	2.4
2000	Daugava	Daugava	LV	70953	3.1	7305	791640	6.8	552.0	1.6
2001	Drava3	Drava3	SI	16667	0.7	1992	501220	6.8	296.0	1.0
1995	Drava4	Drava4	HU	39850	0.5	9340	932249	7.8	503.4	2.7
2000	Drava4	Drava4	HU	39850	0.5	9340	1003260	8.4	540.7	1.7
2000	Elbe	Elbe	DE	1441	0.0	816	74377	10.3	7.7	3.0
2000	Elbe2	Elbe2	DE	61207	1.0	26803	1975228	9.1	348.0	5.2
2000	Elbe3	Saale	DE	17599	0.8	10321	974896	9.4	89.2	7.2

2000	Elbe4	Bile	DE	255	0.9	115	34775	10.2	2.4	3.6
2000	Ems	Hase	DE	3380	0.1	2498	355221	10.7	27.9	4.5
2000	Ems2	Ems2	DE	2952	0.3	2263	214715	10.8	35.5	6.0
2000	Enningdalsälven	Enningdalsälven	SE	353	8.7	33	1470	7.6	18.9	0.7
2000	Erft	Saale	DE	432	0.7	250	21662	11.0	0.7	7.7
2000	Foyle4	Mourne River	GB	1036	0.1	0	144668	8.4	65.4	1.9
2001	GulfOfFinland	Keila	EE	1012	0.3	274	18671	6.1	6.9	3.5
2001	GulfOfRiga	Pärnu	EE	5486	0.1	1182	28191	6.1	51.5	2.3
2001	GulfOfRiga4	Pärnu	EE	2204	0.0	544	13784	6.1	23.5	3.2
2001	GulfRiga2	Kasari	EE	2913	0.1	680	19779	6.3	27.2	2.4
1990	Gyongyos	Balaton	HU	13049	0.1	2269	431319	7.7	156.0	2.6
1995	Gyongyos	Balaton	HU	13049	0.1	2269	389753	7.4	171.0	2.2
2001	Gyongyos	Balaton	HU	13049	0.1	2269	498611	7.6	126.0	1.3
2002	Hornad3	Hornad	SK	4152	0.3	1156	127356	8.2	18.0	3.7
1999	Kolpa	Kolpa	SI	3120	0.1	231	56437	9.5	84.4	1.0
2001	Labe	Labe	CZ	51714	0.9	22095	1666796	8.0	315.0	4.8
2000	Maas	Niers	DE	798	0.8	521	62144	11.4	8.1	8.6
2000	Merkys	Merkys	LT	3300	1.6	832	42564	7.5	30.1	1.5
2000	Mosel1	Sar	DE	6886	0.4	2409	360111	10.7	90.5	4.0
2000	Moälven	Moälven	SE	2318	2.0	34	3267	2.9	28.9	0.6
2000	Mulde	Vereinig. Mulde	DE	7375	0.4	4000	418407	9.2	66.8	7.5
2002	Mures	Mures	HU	30144	0.1	7502	637956	9.0	181.9	1.8
2001	narva1	Narva	EE	46329	9.6	6068	279017	5.8	303.0	0.6
2001	narva4	Väike-Emajõgi	EE	906	1.2	185	6689	6.0	8.4	1.5
2001	narva5	Võhandu	EE	1221	1.2	260	9173	5.7	8.0	1.0
1999	Nemunas10	Jura	LT	1832	0.6	814	22091	7.5	21.1	2.1
2002	Nemunas11	Sventoji	LT	3600	4.7	1360	36101	7.2	29.2	1.3
2001	Nemunas6	Sesuvis	LT	1171	1.0	636	14546	7.3	18.2	1.7
2001	Nemunas8	Susve	LT	1299	2.0	778	20241	7.3	1.4	4.9
1999	Nemunas9	Streva	LT	296	5.3	114	7432	7.4	1.7	1.0
2000	Neris	Neris	LT	10840	2.1	1919	245340	7.5	51.3	1.5
1994	Nordjylland2	Uggerby Å	DK	460	0.0	316	36223	8.4	5.0	6.8
1999	Nordjylland2	Uggerby Å	DK	460	0.0	316	42075	8.1	5.8	5.2
1995	Nordjylland3	Ry Å	DK	368	0.0	342	28724	8.0	3.2	5.6
1999	Nordjylland3	Ry Å	DK	368	0.0	342	36745	8.0	4.5	5.2
2000	Oder	Oder	DE	110628	1.3	56573	3780156	9.9	494.0	2.7
2000	Rhine	Rhine	DE	94780	1.6	38285	5546431	9.3	1770.0	2.8
2000	Rhine1	Saar	GE	3756	0.6	1242	234409	10.6	53.6	3.5
2000	Rhine2	Mosel	FR/GE	11928	0.6	4141	708576	10.8	92.6	3.6
2000	Rhine3	Ruhr	DE	1806	0.8	517	67465	9.7	27.5	4.6
2000	Rhine4	Nahe	DE	3637	0.1	1401	103370	10.3	43.3	4.9
1991	Ringkob_90	Skern Å	DK	1872	0.3	1170	77856	8.2	21.7	3.5
1999	Ringkobing	Storå	DK	1137	0.2	813	84322	8.7	21.8	3.5
2001	Sava	Sava	SI	8642	0.1	1712	369069	9.5	274.0	1.5
1999	Sava2	Sava2	SI	531	0.4	28	36511	7.0	38.4	0.9
1999	Savinja2	Savinja2	SI	1239	0.3	161	89641	8.5	44.3	2.1
2001	Soca	Soca	SI	2124	0.2	60	49046	7.1	93.7	0.7
1995	Soenderjylland	Groenå	DK	1078	0.4	810	57523	8.6	7.8	2.7
1998	Soenderjylland	Groenå	DK	1078	0.4	810	56671	8.5	8.4	3.1
2000	Weser	Werra	DE	6040	0.1	2203	306407	9.2	48.4	4.7
2000	Weser2	Aller	DE	4644	0.4	2600	601011	10.0	22.0	4.1
1995	Vestsjaelland	Tude Å	DK	673	3.1	487	56960	8.6	2.1	8.1
1998	Vestsjaelland	Tude Å	DK	673	3.1	487	56849	8.7	2.3	12.[illegible]
1999	Vipava	Vipava	SI	717	0.1	105	29692	10.7	14.2	1.8

1990	Zala	Zala	HU	986	0.0	356	25557	10.1	1.2	3.0
1995	Zala	Zala	HU	986	0.0	356	22788	9.4	2.4	2.7
1998	Zala	Zala	HU	986	0.0	356	32522	10.1	2.4	2.2
1991	Århus Amt	Guden Å	DK	2519	2.3	1553	204465	8.1	28.2	4.3
1994	Århus Amt	Guden Å	DK	2519	2.3	1553	215007	8.5	41.1	3.9
1999	Århus Amt	Guden Å	DK	2519	2.3	1553	214558	8.3	37.4	3.2