

Student thesis series INES nr 452

# Organic nitrogen and carbon in Swedish rivers: Increasing trends from 1987-2017

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2018  
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Maya Ahlgren (2018)

***Organic nitrogen and carbon in Swedish rivers: Increasing trends from 1987-2017***

***Organiskt kväve och kol i Sveriges floder: Ökad trend från 1987–2017***

Bachelor degree thesis, 15 credits in *Physical Geography and Ecosystem Science*

Department of Physical Geography and Ecosystem Science, Lund University

Level: Bachelor of Science (BSc)

Course duration: *March* 2018 until *June* 2018

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Bachelor thesis, 15 credits, in *Physical Geography and Ecosystem Science*

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## **Acknowledgement**

I would like to thank my supervisor, Martin Berggren, for the support and guidance through this thesis. I would also like to thank my classmates for encouraging and helping one another. We have been a small, but tight group and without you this thesis would not have been half the report it became to be. Finally, I would like to thank my family for believing in me on days when I did not believe in myself.

## Abstract

In the past decades, increased organic carbon concentrations have been reported from freshwaters across the Northern Hemisphere. However, there is little knowledge regarding whether or not these changes are linked to increasing concentrations of organic nitrogen. This poor understanding of nitrogen biogeochemistry is a problem in face of the potential eutrophication effects of nitrogen in receiving coastal ecosystems. The biogeochemical cause as well as the subsequent ecosystem impacts are well known when it comes to increased dissolved organic carbon, but trends in organic nitrogen concentration in rivers remain poorly explored. Recent studies show that organic nitrogen should be included in when discussing eutrophication impacted by runoff from land, which leads to the question of how much the Baltic Sea has been affected by organic nitrogen. Have the changes been proportional to the increase of organic carbon or are they not significant? Eutrophication is a known problem in the Baltic Sea and the impact of dead zones is a cause of the ecosystem changing. The aim of this report was to investigate the trends of organic nitrogen, organic carbon and water color in 30 streams across Sweden from 1987-2017. The correlation between total organic nitrogen and total organic carbon showed positive significant results in nine out of thirty rivers and ten rivers had positive correlations between total organic nitrogen and water color. The correlations were, however, only significant in the area of the Baltic Proper and on the west coast, which shows regional differences that require further research to explain the underlying reason. It is a relevant issue for the present and the future, and for the interest of the environment along Sweden's coastline. More research is needed to investigate the source of the increase and its consequences thoroughly.

**Keywords:** Total organic nitrogen • Total organic carbon • Eutrophication • Sweden • Dissolved organic carbon • Brownification • Pearson's correlation • Baltic Sea

## Populärvetenskaplig sammanfattning

Östersjön har under decennier påverkats av övergödning. Det är den stora tillförseln av näringsämnen och den långsamma vattenomsättning som gör att Östersjön är speciellt utsatt för övergödning. De näringsämnen som orsakar övergödning är vanligtvis fosfor och kväve. Dock finns en tveksamhet i forskningsvärlden om huruvida respektive ämne bidrar till övergödning. En fråga som forskarna är särskilt osäker på, är hur fosfor och kväve motverkar tillväxten av organismer. Både fosfor och kväve har beskrivits som en begränsande faktor för tillväxt av organismer. Forskare har fört fram argument kring denna påverkan, med tankar som baseras på säsong eller huruvida det är en kustmarin miljö. Oavsett vilket ämne och hur det påverkar bidrar till begränsningar, så är vetenskapen kring koncentrationen av ämnena i vattendrag en viktig insikt. Under senare tid har det rapporterats att en ökad koncentration av organiskt kol finns i sötvatten över det norra halvklotet. Organiskt kol är ett av de ämnen som man anser påverkar vattnets färg. Denna uppsats vill bidra med kunskap och data kring hur organiskt kväve och organiskt kol har utvecklats i floder i Sverige, som majoriteten av dem har utlopp till Östersjön.

På grund av den ökade övergödningen, orsakad av kväve eller fosfor, samt kunskapsbristen kring huruvida organiskt kol och kväve interagerar med varandra genomfördes en statistisk undersökning i denna studie. Trettio floder i hela Sverige analyserades i en stor datasamling som sträckte sig 30 år bakåt i tiden med start 1987. Årtalen för den data som används var 1987 till 2017 för att kunna få så mycket jämförelsebar data som möjligt över längsta möjliga tid. Som underlag har medelvärde per år använts, detta för att även kunna genomföra en jämförelse på en regional nivå. Underlaget som har använts har tittat på koncentration av organiskt kväve och organiskt kol samt vattnets färg.

Resultaten visar att det finns en relation mellan en ökad koncentration av organiskt kol och organiskt kväve samt en relation mellan organiskt kväve och vattnets färg. Av de 30 floder som används visar de sydligaste, samt västligaste, floderna högst mätvärden och den största korrelationen.

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# 1 Introduction

There is generally an increasing amount of dissolved organic carbon (DOC) in the watercourses of the Northern Hemisphere (Worrall et al. 2003; Monteith et al. 2007). There are different causes for this increase such as a warmer climate (Tranvik et al. 2009) driven by increased carbon in the atmosphere (Fransner 2018), changes in the soil pH and intensified land use (Worrall et al. 2003; Fransner 2018). The content of total organic nitrogen (TON) has not been studied to the same extent but is believed to show correlation to the increased DOC (Evans et al. 2005).

Increased human activity has strongly altered the global nitrogen (N) cycle in multiple ways (Vitousek et al. 1997). The accumulation of reactive nitrogen (Nr) in the ecosystems is believed to cause changes in estuarine and nearshore ecosystems regarding their composition and function, which leads to long-term decline in the fisheries (Vitousek et al. 1997). The alterations of the chemistry of the watercourses may not only influence the fisheries but also lead to eutrophication.

Eutrophication, as proposed by Nixon (1995) is defined as: “an increase in the rate of supply of organic matter to an ecosystem”. The increase in organic matter supply is usually caused by phosphorus or nitrogen that boost growth of algae but can also result from excessive decomposable organic carbon from other sources (Rabalais 2002). This is concise, but such a broad- open definition can lead to difficulties in a monitoring and management context (Andersen et al. 2006).

Another definition of eutrophication is: “a process in aquatic ecosystems initiated by a pulse of nutrients that results in an increase of algal growth that may lead to decomposition and depleted oxygen conditions (hypoxia)” (Sadava et al. 2016). In view of this definition, organic carbon is not a cause of eutrophication *per se*, but it exacerbates the symptoms of eutrophication because the organisms that break down the organic carbon consume large amounts of oxygen (Larsson et al. 1985). Eutrophication leads to tremendous consequences for the ecosystems, such as biodiversity loss, and dense algal blooms that can lead to anoxia in the deeper parts of the ocean (oxygen depletion) (Schindler et al. 2008).

In Sweden’s national water chemistry monitoring program, the measurements for total organic carbon in rivers became standard in 1987 and have been carried out since then. The method for measuring nitrogen has varied and so has the starting point for the time series, depending on location, hence to ensure a long time series and assuring data from each location this study has focused on the trend between 1987-2017.

Though there have been various studies on which nutrient that is the limiting nutrient for primary production, there is an ongoing debate whether phosphorus or nitrogen is the primary candidate for eutrophication (Conley et al. 2009).

Recent studies show that organic nitrogen should be included in the equation of eutrophication impacted by wastewater (Sun et al. 2017), which leads to the question of how much the eutrophication in the Baltic Sea has been impacted by organic nitrogen.

If organic material, containing both nitrogen and carbon, has increased this would not automatically mean that the components have increased proportionally. The composition



between nitrogen and carbon could change over time due to, for example, an increase in N poor humic material which contains 35-95 times more C than N (Mcknight DM and GR 1988). This would reflect in a correlation of lower nitrogen compared to carbon.

This thesis aims to investigate how the changes of organic components (C and N) have occurred spatiotemporally in Sweden. The question to be answered is:

*How have the concentrations of TON and TOC changed through the last 30 years in Sweden?*

The null hypothesis,  $H_0$ , assumes that there has not been an increase of total organic nitrogen, total organic carbon nor water color during the period. The null hypothesis will be tested against a couple alternative hypothesis;

$H_1$ : There has been an increase of total organic nitrogen, however it is not correlated to the total organic carbon nor water color.

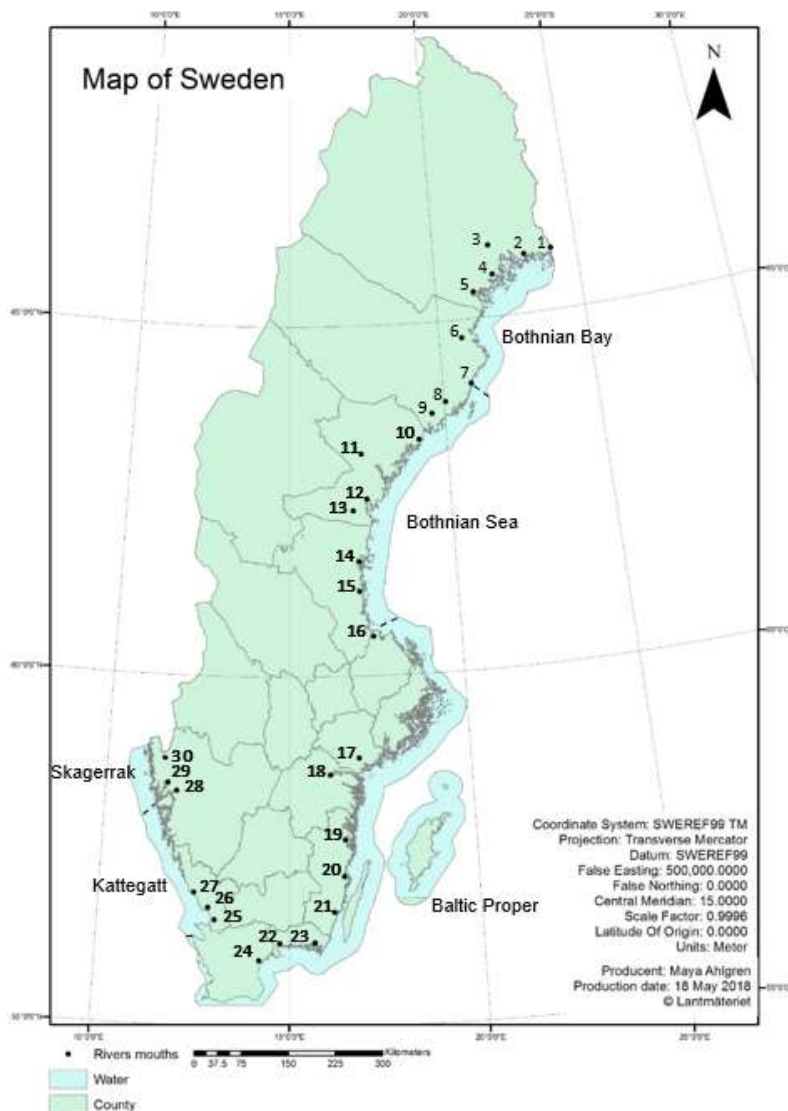
$H_2$ : The changes of organic nitrogen have increased proportionally to the organic carbon and there is a spatiotemporal difference within the country.

## 2 Background

### 2.1 Study area

The study area covers the country of Sweden, at latitude N 59-69° and longitude E 10-24°. The rivers (Table 1) are spread out, over the majority of the country and the river mouths are the locations denoted as numbers in Figure 1. The rivers provide drainage to marine recipients along the coastline, mainly on the east coast but also several on the west coast.

Sweden has four seasons per year and a temperate or boreal climate. The range from north to south changes from cold, snowy winters and cool summers to cool winters and warm summer (Kritzberg and Ekström 2012). According to the Köppen-Geiger climate classification Sweden



**Table 1.** Rivers in Sweden (“å”, “ström” or “älv” are words for river in Swedish).

Number	River
1	Torne älv
2	Kalix älv
3	Råne älv
4	Lule älv
5	Pite älv
6	Kvistforsen
7	Rickleån
8	Ume älv
9	Öre älv
10	Gideälv
11	Ångermanälven
12	Indalsälven
13	Ljungan
14	Delångersån
15	Ljusne Strömmar
16	Dalälven
17	Nyköpingsån
18	Motala ström
19	Botorpsström
20	Emån
21	Ljungbyån
22	Lyckebyån
23	Mörumsån
24	Helgeån
25	Lagan
26	Nissan
27	Ätran
28	Bäveån
29	Örekilsälven
30	Enningsdalsälven

**Figure 1.** Map of Sweden. River mouths are shown through numbers 1-30 and the sub-basins of the Baltic Sea are shown. The lines within the country are the divisions of counties.

is divided into the two main classifications; Dfc and Dfb, which are characterized to have no dry seasons and cold/warm summer (Peel et al. 2007). The season and temperature within the country causes variation in flood seasons and it is one of the factors that affect the river discharge.

The rivers can be divided into five major sub-basins that they drain into; the Bothnian Bay, Bothnian Sea, Bothnian Proper, Kattegatt and Skagerrak (Figure 1). These basins have different salinity, depths and water turnover but covers six to nine of the rivers each. This division is considered for the regional differences within the country. The drainage sizes of rivers varies, but in this study the drainage size and the river lengths are not considered in the analyses.

## 2.2 Nitrogen

The gas form of nitrogen makes up almost 80% of the world's atmosphere. Nitrogen (N) is also a crucial element for enzymes and other proteins as well as in nucleic acids (Galloway and Cowling 2002). The gas form of nitrogen must be fixated for organisms and vegetation to be able use the compound.

Reactive nitrogen ( $N_r$ ) is a collective term that describes nitrogen which is biological or photochemical active. To determine the organic nitrogen in the data set it is important to understand that the total nitrogen is a measure by the chemical compounds of nitrogen summed together. The main chemical constituents of N are nitrate ( $NO_3^-$ ), nitrite ( $NO_2^-$ ), ammonium ( $NH_4^+$ ) and the organic nitrogen (ON). The inorganic nitrogen components,  $NO_3^-$ ,  $NO_2^-$  and  $NH_4^+$  can be taken up directly by organisms, fully bioreactive, while the organic nitrogen is broken down by microorganisms to small molecules and then can be converted to other forms of inorganic nitrogen (Sadava et al. 2016). The ON is therefore partly bioreactive.

Nitrogen in the form of nitrate has been measured in water for decades due to the health concern in drinking water. The concerns basis is how microorganisms can convert nitrate to nitrite in the stomach. Nitrite in the blood system can convert hemoglobin to methemoglobin, which decreases the effectivity of transporting oxygen (Vitousek et al. 1997).

It has been debated whether phosphorus or nitrogen is the limiting nutrient for primary production in marine ecosystems (Howarth 1988). There are arguments that phosphorus is the primary limiting nutrient (Schindler et al. 2008) while some argue that the phosphorus and the nitrogen can be limiting dependent on season (Eppley et al. 1973). At the same time, it has been argued that the nitrogen is a limiting factor in coastal marine ecosystems (Graneli 1987; Bushaw-Newton and Moran 1999; Howarth and Marino 2006).

## 2.3 Carbon

Dissolved organic carbon (DOC) and its related constituents can be referred to by a variety of names such as dissolved organic matter (DOM), natural organic matter (NOM) and humus (Williamson et al. 1999), however, DOM concerns all organic material and their basic elements together. The NOM can also be referred to as the total organic carbon (TOC) if unfiltered or DOC if filtered, the difference being the particulate organic carbon (Sarkkola et al. 2009). In this report the TOC, the unfiltered total organic carbon, is considered and used for the analysis.

The color, (presented further in 2.4 below) can vary from yellow to brown or black and are organic compounds that are found in waterways (Williamson et al. 1999).

There have been studies that suggest that coniferous forests would provide greater amount of DOC than hard deciduous forests (Cronan and Aiken 1985). The temperate, boreal and the subarctic surface water showed a significant increase of organic carbon between 1990–2013 on the Nordic peninsula, indicating that the changes are global/regional rather than on local level (de Wit et al. 2016). The effected carbon cycle would hence need actions on a greater scale to affect the brownification and eutrophication, however for the purpose of this report the regional differences within Sweden were studied.

## 2.4 Water color

The color of water is an important parameter to regard. The water color is affected by the increased organic matter, increase in temperature, changes in the ground pH and intensified land use, primarily through agriculture (Worrall et al. 2003; Fransner 2018). The phenomena of increased water color is called ‘brownification’. There is a consensus that the brownification is directly connected to the increase of terrestrially derived organic matter in the water (Kritzberg and Ekström 2012). Another factor influencing the brownification is the amount of iron in the water (Kritzberg and Ekström 2012), however in this report the iron is not considered and only the absorbance of the water color investigated and discussed.

## 3 Materials and methods

### 3.1 Monitoring data

#### 3.1.1 Methods of measuring total organic nitrogen

The data used for this study was taken from Miljödata MVM (run by the Swedish University of Agricultural Sciences (SLU) as a cooperation between the Swedish Environmental Protection Agent as a commission by the Swedish Agency for Marine and Water Agency). The data is free and available at <http://miljodata.slu.se/mvm/>. The data selected were from the water chemistry parameter and the chosen components were: Tot-N\_ps (total nitrogen persulfate), Tot-N\_TNB (total nitrogen catalytic oxidation), Kjeld. -N (Kjeldahl nitrogen),  $\text{NH}_4^+$  (ammonium),  $\text{NO}_2^- + \text{NO}_3^-$  (nitrite and nitrate), TOC (total organic carbon) and Abs.\_F420 (filtered absorbance\_420).

The collected data resulted in monthly data from the years between 1965-2017 dependent on location. The data was adjusted according to recommendations in “Totalkväveanalyser vid Institutionen för vatten och miljö -En genomgång av olika analysmetoder och deras betydelse för tidsserierna” (Wallman et al. 2009). The Tot-N\_ps was multiplied with 110 % to modify for the average lower amount of nitrogen compared to the Tot\_sum (described in 3.1.1.1). The Tot-N\_TNB was adjusted by multiplication of 115 % for the same reason of lower average compared to the Tot\_sum (described in 3.1.1.3). This was done to be able to compare the results due to that the methods of analyzing nitrogen have been changed during the years.

Below follow the three different methods for collecting nitrogen through time, each one described as a method, and the equations to calculate the total organic nitrogen.

#### 3.1.1.1 Method 1, Total\_sum

The method that has been used for the longest period (1969/1972 until 2007) is the Total\_sum method, which is the method that sums the Kjeld. -N (Kjeldahl nitrogen) with the nitrate and nitrite nitrogen. The Kjeldahl nitrogen is the organically bound nitrogen combined with the ammonium (Wallman et al. 2009). The method of analyzing the Kjeldahl nitrogen is done by transferring the organically bound nitrogen to ammonium. This occurs at 387°C. The ammonium is colorimetrically determined by the indophenol method (SIS 028134) (Wallman et al. 2009). The Total\_sum is the method most used, internationally, to establish the nitrogen loading on the sea. The Total\_sum method is a more efficient at breaking down the organic material and may be one of the reasons for a higher nitrogen amount compared to the next two methods.

#### 3.1.1.2 Method 2, Tot-N\_ps

The Tot-N\_ps is the method of analyzing total nitrogen spectrophotometrically with a TrAAcs instrument. The instrument draws the sample reagents before the colorimetric mixing (Burton and Pitt 2001). The ps stands for persulfate digestion and this method was used from 1987 until 2006 in Sweden (Wallman et al. 2009).

#### 3.1.1.3 Method 3, Tot-N\_TNB

Another method of determining the total nitrogen is by determining the total amount of nitrogen after catalytic oxidation to nitrogen oxides after chemiluminescence. This method is abbreviated Tot-N\_TNB and is the standard method used in Sweden since 2007 until present day. The results are of high accuracy and precision (Wallman et al. 2009).

#### 3.1.1.4 Total organic nitrogen (TON)

The TON is determined by subtracting  $\text{NO}_3^-$ ,  $\text{NO}_2^-$  and  $\text{NH}_4$  from the Tot-N (Eq. 1). In the case of the Tot-N\_ps and the Tot-N\_TNB the total nitrogen is given, however to determine the organic nitrogen from the Kjeldahl nitrogen only the ammonium is subtracted (Eq. 2). This is if the Tot-N sum has not been calculated by adding the  $\text{NO}_3^-$ ,  $\text{NO}_2^-$ , in that case the equation would remain unchanged from equation 1.

$$\text{TON} = (\text{Tot-N}) - ((\text{NH}_4) + (\text{NO}_2^-) + (\text{NO}_3^-)) \quad (1)$$

$$\text{ON} = (\text{Kjeld. -N}) - (\text{NH}_4) \quad (2)$$

The few values in the  $\text{NH}_4\text{-N}$  ( $\mu\text{g L}^{-1}$ ),  $\text{NO}_2^-$  ( $\mu\text{g L}^{-1}$ ) and  $\text{NO}_3^-$  ( $\mu\text{g L}^{-1}$ ) that were showing  $< 1 \mu\text{g L}^{-1}$  and  $< 3 \mu\text{g L}^{-1}$  were changed to  $1 \mu\text{g L}^{-1}$  and  $3 \mu\text{g L}^{-1}$  respectively to be able to calculate the equations as well as the medians of the different nitrogen methods.

### 3.1.2 Total organic carbon (TOC)

TOC has been collected by SLU from 1987 and forward. Due to this the focus of this report began with that year. The methods of analyzing and collecting TOC has changed three times since the commencement. The method in -87 was a Swedish standard SS 02 81 99 with a Shimadzu TOC 500 with a test injector ASI-502. In 1999 the method maintained the same but the water tests were not conserved and they were pretreated with hydrochloric acid (SLU 2017a). In 2004 the method continued but the test was initially pretreated with hydrochloric acid as well as measured with a NDIR-detector (SLU 2017a). The method used today has remained the same since 2007. The method consists of the test being pretreated with hydrochloric acid to evaporate the inorganic carbon. The test is passed over a catalyzer to drive away the carbonic acid, with a non-carbon dioxide gas, and then quantified. The instrument used is a Shimadzu TOC-VCPH with a THM-1 model and a sample changer, ASI-V (SLU 2017a).

### 3.1.3 Water color

The water color is measured either by filtered or unfiltered tests. The filtered test measures primarily the dissolved nutrients in the water by measuring the light absorption (SLU 2017b), which mathematically represents the inverse logarithm of light transmittance through water. In Sweden the standardized method is in a 5 cm cuvette at 420 nm. Today the absorbance is measured with a spectrophotometer (SLU 2017b). Water in nature contains dissolved nutrients as well as humus substances, and particles which effects the color of the test. The more iron and humus particles in the water the higher the absorbance (Kritzberg and Ekström 2012; SLU 2017b).

There has only been two ways of measuring water color in Sweden effecting the time series. In 1965 to 1994 a method by Chalupa (1963) was used. In 1995 the SS-EN ISO 7887, part B, with a spectrophotometer was used. There have been two types of spectrophotometers; UNICAM 8625 used until September -09 and PerkinElmer Lambda 35 that has been used since October -09 and is used today (SLU 2017b).

## 3.2 Statistical method

In the cases of missing values, the average of the previous and the next values was chosen for replacement. To detect the distribution of the time series, Shapiro-Wilk normality test was used. To determine the long-term trends in the data set the non-parametric Seasonal Mann-Kendall trend test was used. The test was selected since it is a non-parametric test. The significant level was 0.05 and the period was 12, referring to months. The long-term trends were also tested with the Ljung-Box test to test the lack of fit by the model and the seasonality. The confidence interval was 95%.

To examine the time series of the three different methods of collecting total nitrogen, the linear regression of the yearly median values for each of the adjusted total organic nitrogen calculations (Tot\_sum, Tot-N\_ps and Tot-N\_TNB) were established.

The correlation between the parameters of TON, TOC and water color were tested with Pearson correlation of the yearly median values of each location. The Pearson correlation was chosen to determine the variance without classification of which parameter that were dependent

or independent. The median values were calculated without replacement for missing values, thus some years experienced less values than 12, however the median was still considered representative in these cases. The median values were chosen instead of the average value, since the data did not follow a normal distribution and the data showed large variation between high and low values. The significance level for the Pearson correlation tests were 0.01.

To test if there was any spatial variation between the Swedish catchment areas the rivers were divided according to the sub-basin in the sea that it runs into (Bothnian Bay, Bothnian Sea, Bothnian Proper, Kattegatt and Skagerrak). This was also carried out by Kritzberg and Ekström (2012). However, since the west coast only has six rivers and is divided into two separate sub-basins they were added together as one sub-basin to have even representation in each region.

Four rivers with the highest median over the period in each of the different sub-basins were chosen to establish if the different methods of measuring total organic nitrogen, are comparable and whether or not there seems to be an increase. The rivers were: Rickleån, Öre älv, Nyköpingsån and Lagan (Figure 2-5).

## 4 Results

### 4.1 Regional spatial variation

The median values for TON, TOC and the water color, throughout the time of 1987-2017 varied noticeably between the rivers (Table 2). The TON varied within 122.0 - 874.6  $\mu\text{g L}^{-1}$ , the TOC 2.6 – 16.8  $\text{mg L}^{-1}$  and the water color spanned 0.03 – 0.3  $5\text{cm}^{-1}$ . The highest values in all three components were in the most central and most southern rivers of the country. The rivers in this description included and followed Nyköpingsån (#17) which drains into Baltic Proper and Kattegatt and Skagerrak. The exception of high values was Rickleån (#8), in the north part of the country, which showed high nutrient values as well as an increase in color, considerably strong colored. There were seven rivers in total that showed high absorption coefficients ( $\text{Abs}_{420} 5\text{ cm}^{-1} > 0.20$ ). These were mostly located in the south, with Rickleån and Öre älv being exceptions. There were 11 weakly colored rivers ( $\text{Abs}_{420} 5\text{ cm}^{-1} < 0.10$ ) only located in the north. The north-south gradient displayed a clear difference in  $\text{Abs}_{420}$  values.

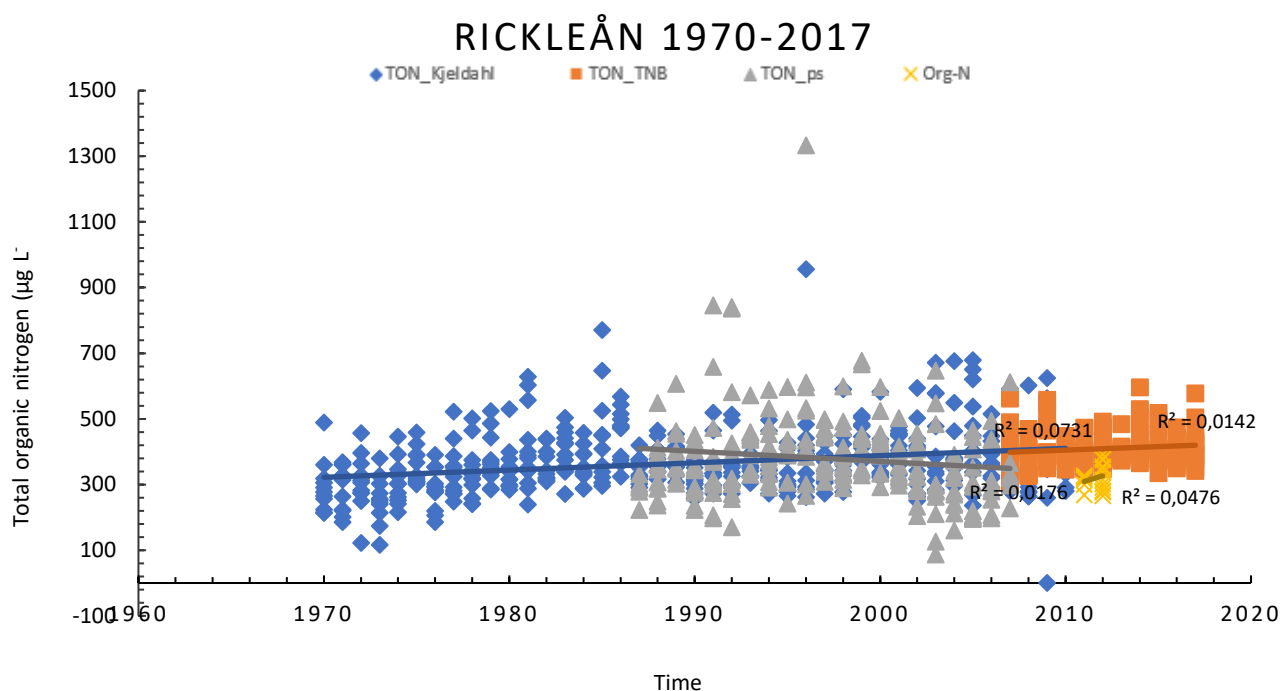
**Table 2.** Median values of total organic nitrogen (TON) (calculated by using Kjeldahl nitrogen for the years that existed and continued the data by the nitrogen measured by method 3, Tot-N\_TNB), total organic carbon (TOC) and water color (Abs\_F<sub>420</sub>) between 1987-2017.

River	Sub-basin	TON ( $\mu\text{g L}^{-1}$ )	TOC ( $\text{mg L}^{-1}$ )	Abs_F <sub>420</sub> ( $5\text{cm}^{-1}$ )
1 Torne älv	Bothnian Bay	255	5.7	0.11
2 Kalix älv	–II–	212	4.5	0.09
3 Råne älv	–II–	285	7.5	0.16
4 Lule älv	–II–	122	2.6	0.03
5 Pite älv	–II–	171	3.4	0.05
6 Kvistforsen	–II–	175	3.7	0.04
7 Rickleån	–II–	376	11.5	0.20
8 Ume älv	Bothnian Sea	156	3.8	0.05
9 Öre älv	–II–	316	10.2	0.24
10 Gideälv	–II–	312	9.5	0.19
11 Ångermanälven	–II–	184	4.9	0.08
12 Indalsälven	–II–	176	4.1	0.05
13 Ljungan	–II–	240	5.9	0.07
14 Delångersån	–II–	274	6.2	0.06
15 Ljusne strömmar	–II–	273	6.8	0.11
16 Dalälven	Baltic Proper	304	7.5	0.11
17 Nyköpingsån	–II–	875	11.1	0.07
18 Motala ström	–II–	525	7.5	0.06
19 Botorpsström	–II–	604	11.6	0.13
20 Emån	–II–	607	12.2	0.16
21 Ljungbyån	–II–	806	14.5	0.20
22 Lyckebyån	–II–	763	16.8	0.30
23 Mörrumsån	–II–	601	11.6	0.11
24 Helgeån	–II–	849	15.2	0.27
25 Lagan	Skagerrak+	550	11.7	0.21
26 Nissan	Kattegatt	542	12.7	0.27
27 Ätran	–II–	496	9.0	0.16
28 Bäveån	–II–	541	10.4	0.19
29 Örekilsälven	–II–	528	9.9	0.17
30 Enningsdalsälven	–II–	348	7.9	0.12

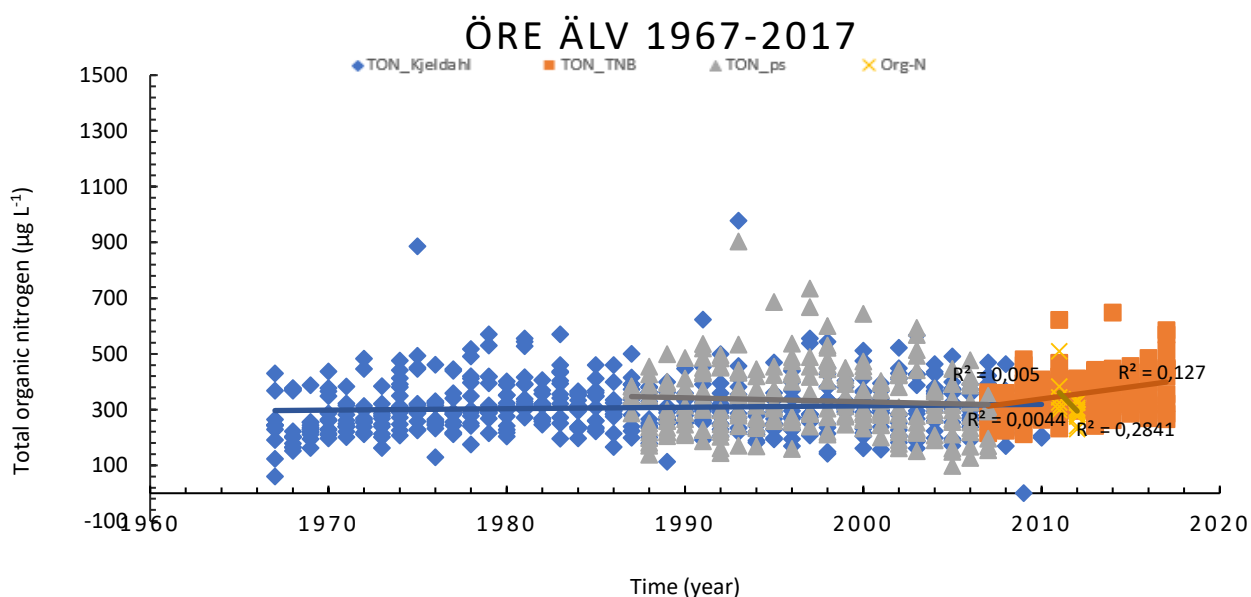


## 4.2 Trends in the representative rivers of each sub-basin

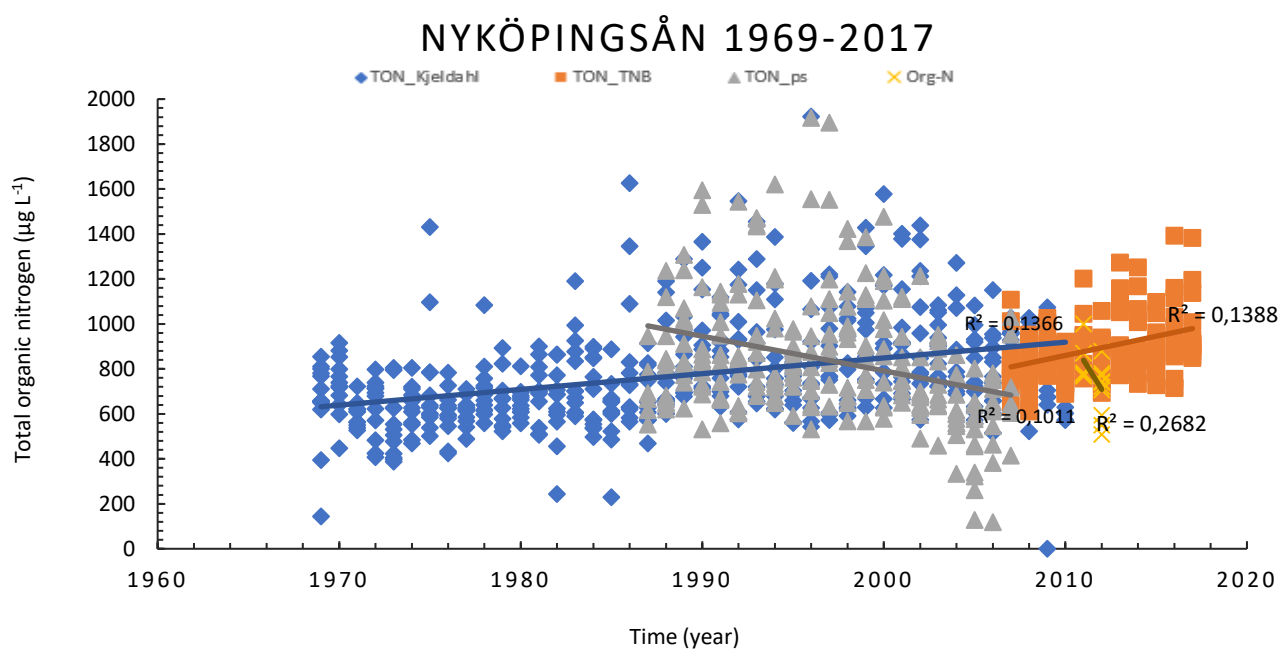
The TON estimated by the summation of the Kjeldahl nitrogen showed the longest time series with a low significance (Rickleån  $r^2 = 0.0731$ , Öre älv  $r^2 = 0.0044$ , Nyköpingsån  $r^2 = 0.1366$ , Lagan  $r^2 = 0.1204$ ). The results are shown in Figure 2-5. The graphs do however show the trends within all four methods. The line of regression was calculated for the summation of Kjeldahl summation nitrogen and the TNB nitrogen (Rickleån  $r^2 = 0.1017$ , Öre älv  $r^2 = 0.0438$ , Nyköpingsån  $r^2 = 0.1833$ , Lagan  $r^2 = 0.2229$ ).



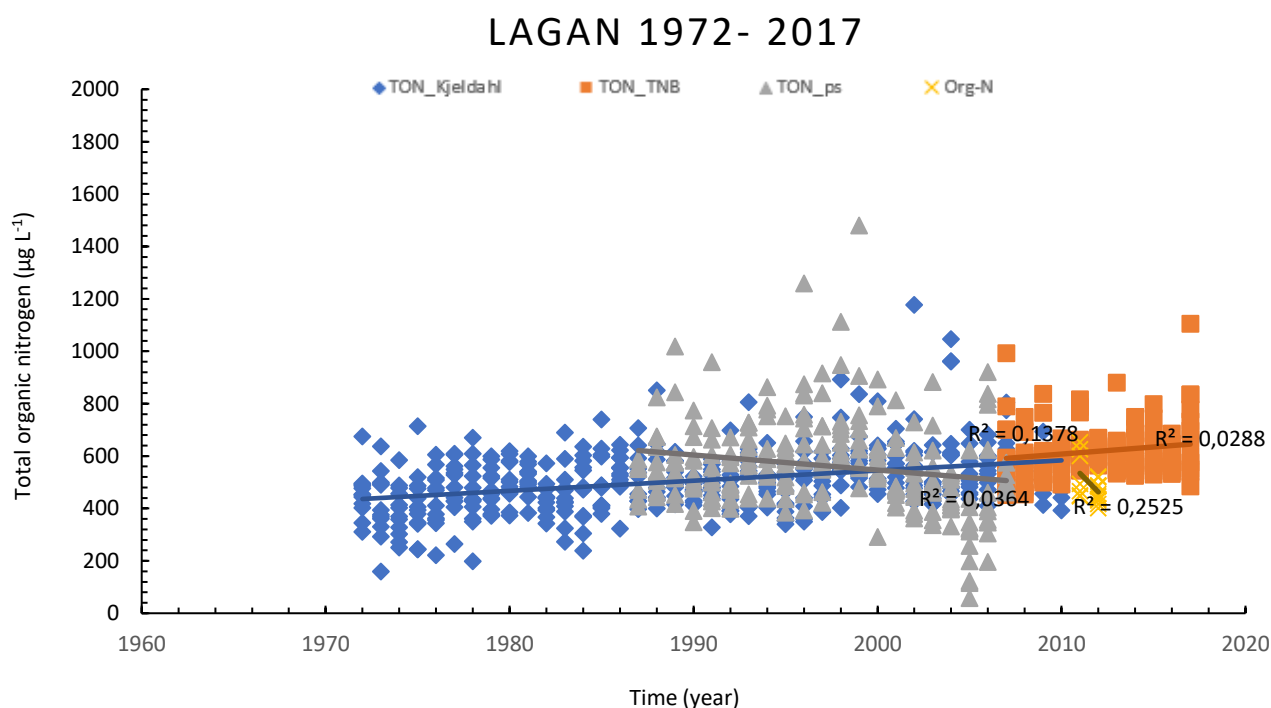
**Figure 2.** Total organic nitrogen in Rickleån 1970- 2017. The total organic nitrogen by the Kjeldahl method (◆), total organic nitrogen by the TNB method (■), total organic nitrogen by the ps method (▲) and organic nitrogen (×). Trendlines indicate each method individual trend and the  $r^2$  represents the coefficient of determination.



**Figure 3.** Total organic nitrogen in Öre älv through 1967–2017. The total organic nitrogen by the Kjeldahl method (◆), total organic nitrogen by the TNB method (■), total organic nitrogen by the ps method (▲) and organic nitrogen (×). Trendlines indicate each method individual trend and the  $r^2$  represents the coefficient of determination.



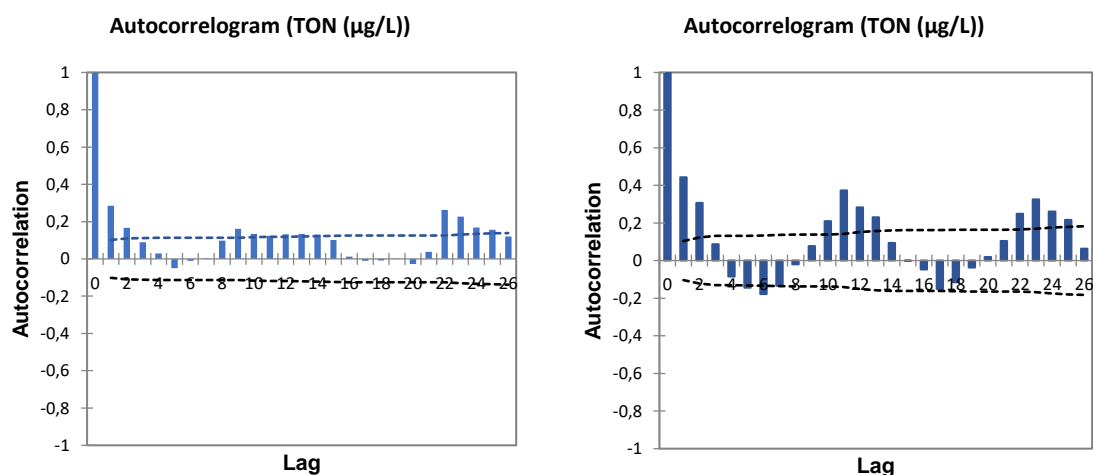
**Figure 4.** Total organic nitrogen in Nyköpingsån 1969–2017. The total organic nitrogen by the Kjeldahl method (◆), total organic nitrogen by the TNB method (■), total organic nitrogen by the ps method (▲) and organic nitrogen (×). Trendlines indicate each method individual trend and the  $r^2$  represents the coefficient of determination.



**Figure 5.** Total organic nitrogen in Lagan 1972 -2017. The total organic nitrogen by the Kjeldahl method (◆), total organic nitrogen by the TNB method (■), total organic nitrogen by the ps method (▲) and organic nitrogen (×). Trendlines indicate each method individual trend and the  $r^2$  represents the coefficient of determination.

### 4.3 Distribution and serial autocorrelation

The non-parametric Seasonal Mann-Kendall trends showed trend for the components in all the rivers, for Båveån ( $p = 0.0002$ ,  $\alpha = 0.05$ , Kendall  $\tau_{av} = 0.2664$ ) and for Botorpsström ( $p < 0.0001$ ,  $\alpha = 0.05$ , Kendall  $\tau_{av} = 0.2648$ ). The test indicates a serial autocorrelation, which is shown in the Ljung-Box test with a seasonality of 12 months (Figure 6). Botorpsström ( $df = 6$ , value = 45.158,  $p < 0.0001$ ).

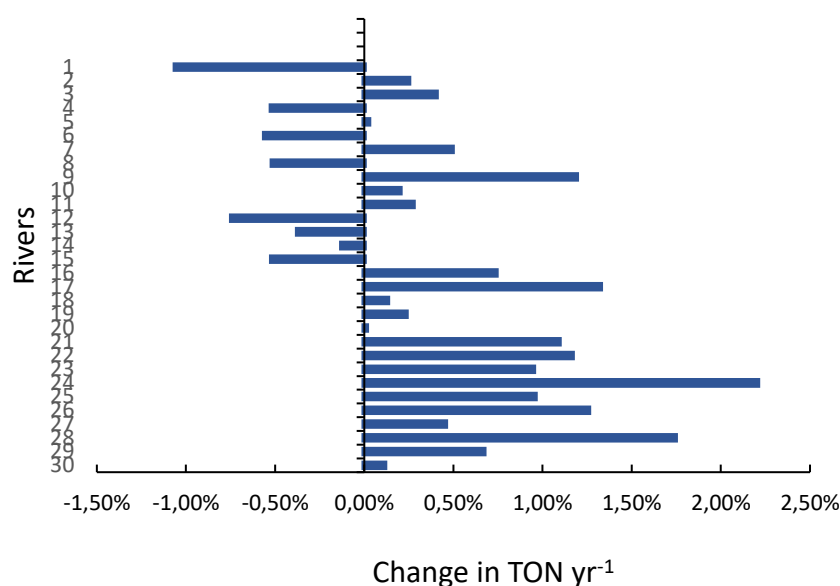


**Figure 6.** Seasonality Box-Ljung trend test in Botorpsström (left) and Båveån (right). The 95% confidence interval are displayed as the dotted line.

To avoid serial autocorrelation for the correlation result, the median values for each year and component was used for further analysis.

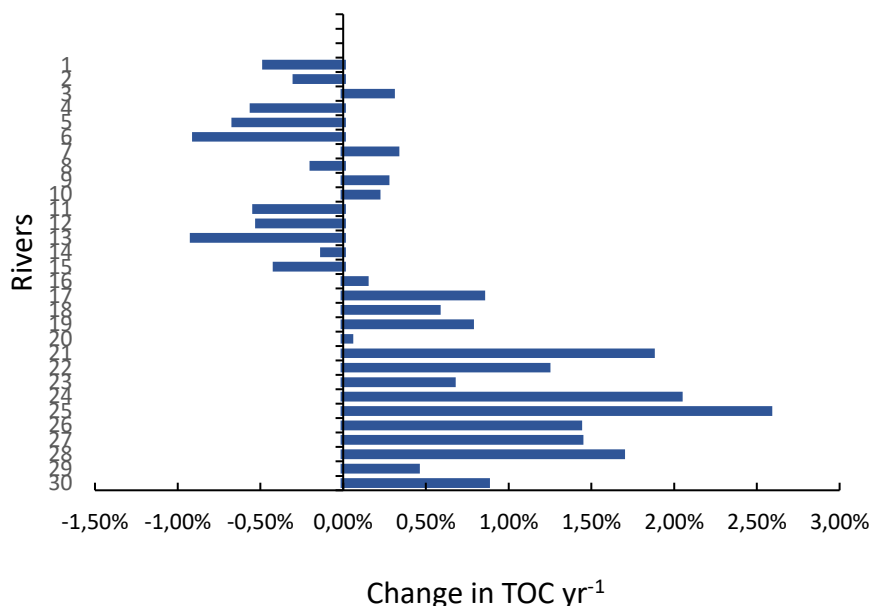
### 4.4 Rate of change

The total organic nitrogen increased in 22 of 30 rivers throughout the time (Figure 7). The increase is the highest in the most southern rivers, with an exception in Öre älv (#9). The increase varies from 0.40 to 66.00 % throughout the time which corresponds to 0.01- 2.20% per year. There are however eight rivers that show a decrease from 1987-2017. These rivers are located north of Dalälven (#16). The decrease is the highest in the most northern river, Torne älv (#1). The decrease over time varies from -3.80 to -31.80% which corresponds to -0.13 to -0.60% per year.



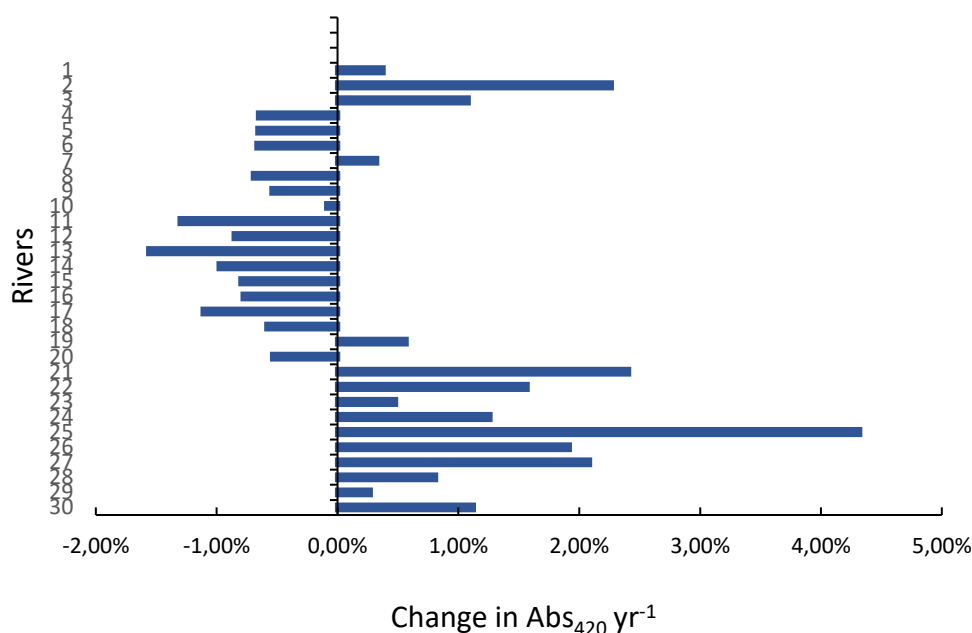
**Figure 7.** The rate of change, per year, of total organic nitrogen in 30 rivers in Sweden. The time change is from 1987-2017. The negative values indicate a decrease.

The total organic carbon increased in 19 out of 30 rivers south of Ljusne strömmar (#15) with four exceptions (Figure 8). The increase varies from 1.36- 77.33% over the time which corresponds to 0.05 – 2.58% per year. The decrease varied from -3.73 to -27.34 over time which corresponds to -0.12 to -0.91% per year.



**Figure 8.** The rate of change, per year, of the total organic carbon in 30 rivers in Sweden. The time change is from 1987-2017. The negative values indicate a decrease.

The absorbance, water color, increased in 14 of 30 rivers and decreased in the remaining 16 (Figure 9). The increase is most prominent south of Ljungbyån (#21) and on the west coast, and in the three most northern rivers, Torne-, Kalix- and Råne älv. The increase over time in all the 14 rivers varies from 8.00 to 130.00 % over time, corresponding to 0.30-4.30% per year.



**Figure 9.** The rate of change, per year, of water color in 30 rivers in Sweden. The time change is from 1987-2017. The negative values indicate a decrease.

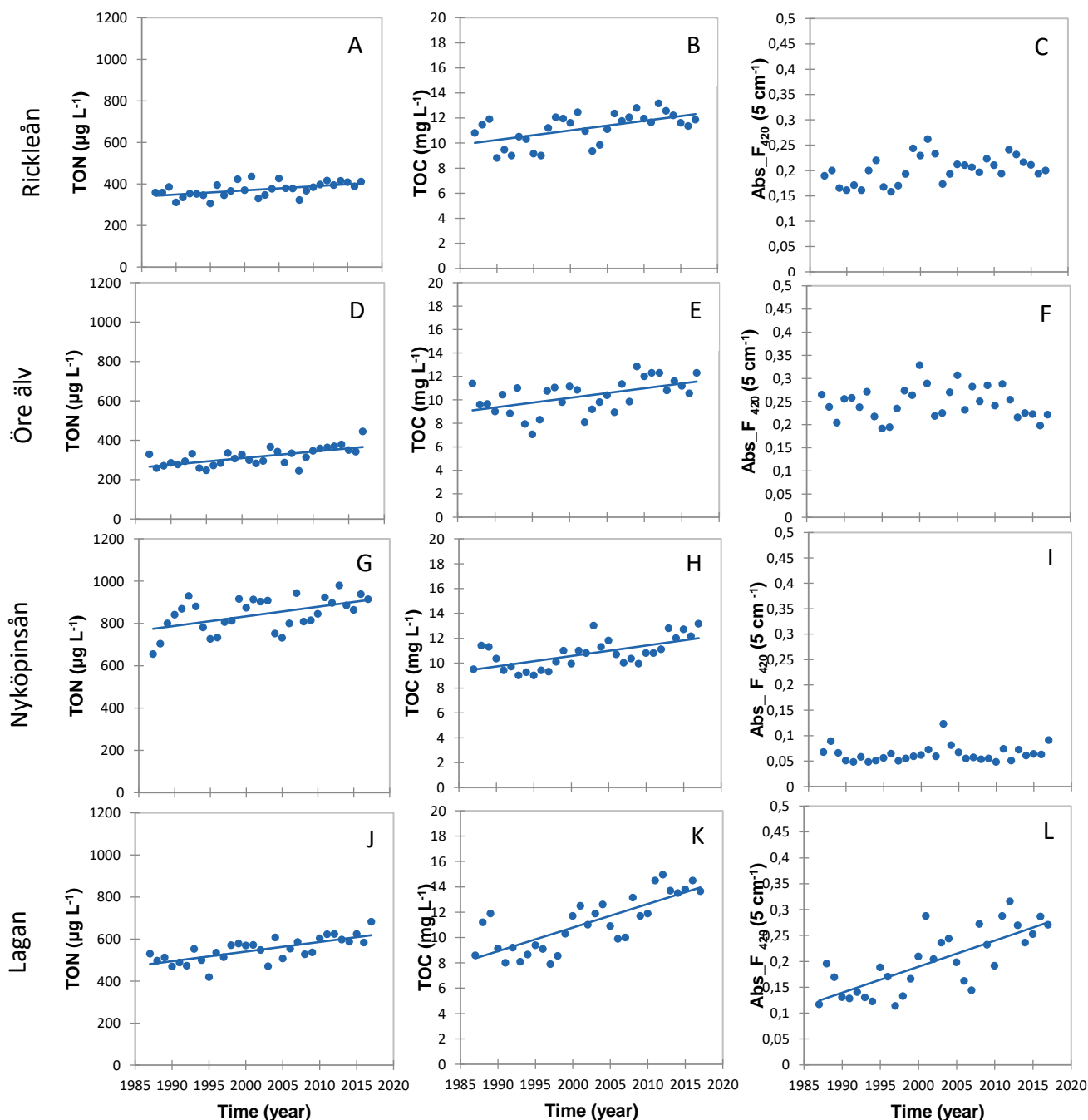
#### 4.5 Correlation between TON, TOC and water color

The TON increased in the majority of the rivers, however only 11 of them showed significance ( $r^2 = 0.434 \pm 0.121$ ,  $p < 0.0001$ ). Among the eleven were the regional representation rivers (Rickleån, Öre älv, Nyköpingsån and Lagan) included (Figure 10. A, D, G, J) ( $r^2 = 0.377 \pm 0.130$ ,  $p < 0.0001$ ).

The TOC showed significant positive change in 18 rivers ( $r^2 = 0.456 \pm 0.148$ ,  $p < 0.0001$ ). Rickleån, Öre älv, Nyköpingsån and Lagan ( $r^2 = 0.398 \pm 0.153$ ,  $p < 0.0001$ ) were included in the 18 rivers (Figure 10. B, E, H, K).

The significant change over time for water color was almost exclusively in the far south, started from Emån, and on the west coast, all the way to Enningsdalsälven (#30). It resulted in a total of 14 rivers with significant increase ( $r^2 = 0.394 \pm 0.154$ ,  $p < 0.0001$ ). There were only three rivers in the far north, that were also included in the previous statement, that showed significance ( $r^2 = 0.230 \pm 0.010$ ,  $p < 0.0001$ ). Out of the four regional representation rivers (Figure 10. CFIL) only, Lagan, on the west coast showed an increase with a strong significance ( $r^2 = 0.570$ ,  $p < 0.0001$ ) (Figure 10. L).

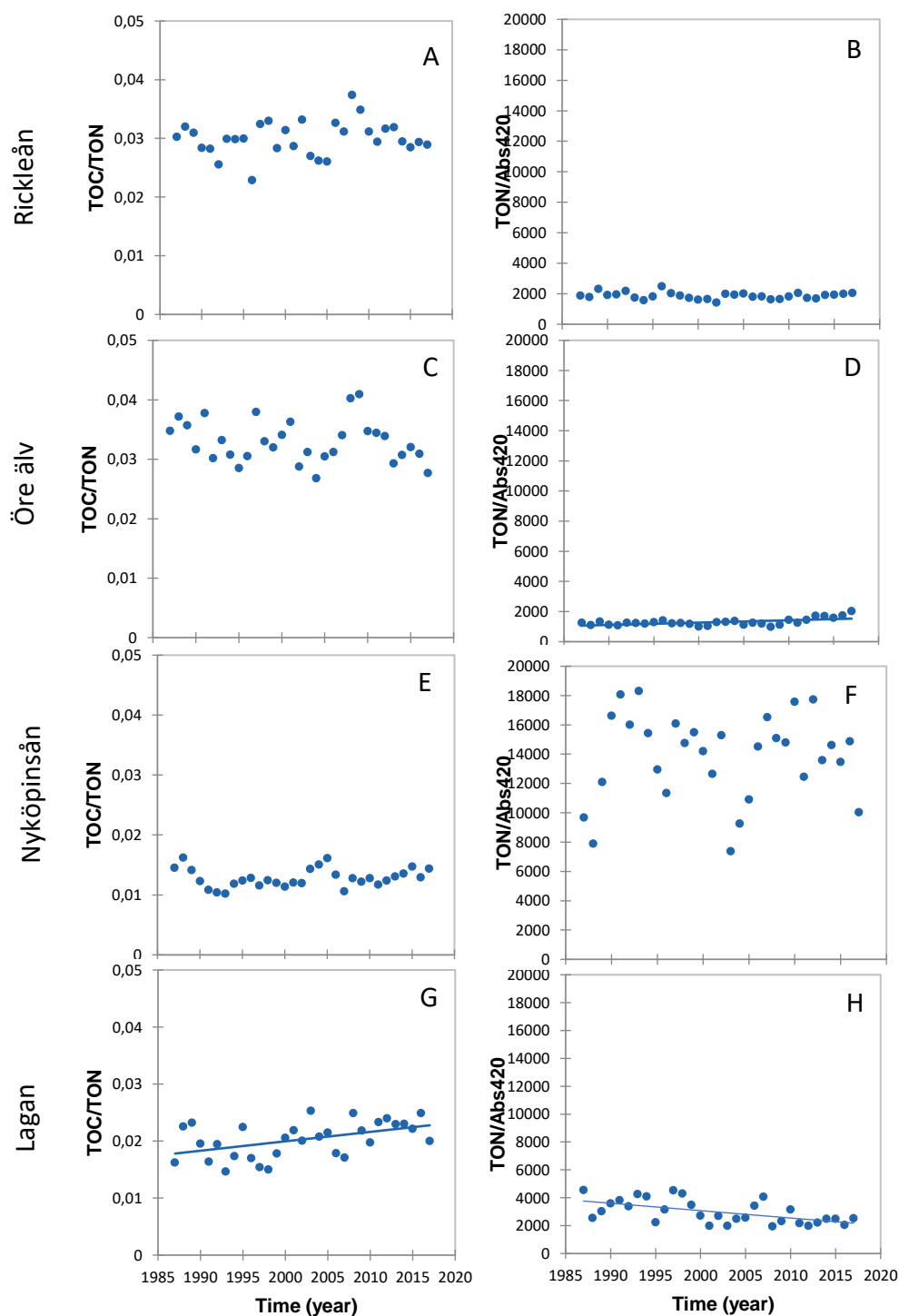
The Pearson correlation matrix and the Pearson coefficients of determination for each river can be viewed in Appendix 8.1.



**Figure 10.** Mean annual values of total organic nitrogen (TON) in four rivers draining into in the different regions of the Baltic Sea. Where there is no trend line, the correlation is not significant. Trend lines show significant change.

The relationship between the total organic nitrogen and water color (TON/Abs420) showed a significance with a negative trend in the case of ten rivers, located south of Öre älv (#9) with a continued consistency on the west coast ( $r^2 = 0.321 \pm 0.111$ ,  $p < 0.0001$ ) (see Appendix 8.1). The four rivers that represented the regional differences, showed no significant result at Rickleån and Nyköpingsån while there was a considerable significance in Öre älv ( $r^2 = 0.359$ ,  $p = 0.000$ ) and a negative significance in Lagan ( $r^2 = 0.332$ ,  $p < 0.0001$ ) (Figure 11. B, D, F, H).

Nine rivers showed a positive increase with significance with the relationship between total organic carbon through total organic nitrogen ( $r^2 = 0.313 \pm 0.093$ ,  $p < 0.0001$ ) (see Appendix 8.2). However, only Lagan (#25) ( $r^2 = 0.232$ ,  $p < 0.0001$ ) out of the regional representation lakes was included in these nine rivers and the three remaining showed a variation in result with no significance (Figure 11. A, C, E, G).



**Figure 11.** Trends in TOC/TON and TON/Abs420 in four rivers with different sub-basins in Sweden for the period 1987-2017. Trend lines show significant change.

## 5 Discussion

The discussion question regarding whether phosphorus or nitrogen is the primarily culprit for eutrophication is an ongoing discussion. The differences in salinity, temperature, water turnover and ecosystem influence these factors and makes it a difficult subject to tackle on a general scale. The results found in this report shows that there is a significant increase in total organic nitrogen, total organic carbon and in some cases a significant change in the water color in Swedish river systems. The effects that total organic nitrogen is increasing can be a reason for the increase of eutrophication.

### 5.1 Changes in composition

The correlation between TOC and TON indicates that the total organic carbon increase more for the same time period compared to the total organic nitrogen. This rejects  $H_1$ , since the correlation is significant however the quota is not. The composition of C: N can be explained by an increase of humic matter in the water, which would increase the carbon to a greater extent than the nitrogen (Mcknight DM and GR 1988).

### 5.2 Eutrophication at the respective sites

The country is showing extreme variation in these components through time and the south-north gradient which makes this both a regional and a national problem. The result also indicates that  $H_2$  should be accepted, however only on the regard the spatiotemporal differences within the country, since the correlation between the organic nitrogen and organic carbon was not proportional.

Since this study was carried out in the river mouths, away from direct coastline to not be affected by the Baltic Sea, the measurements cannot argue for the eutrophication that occurs in the Baltic Sea but are sources for it. On the other hand, one can argue that because of the different salinities that the water has along Sweden's coast, the eutrophication would be impacted by the salinity (Blomqvist et al. 2004).

### 5.3 Rate of change

A misconception regarding rate of change is unavoidable since the first and the last values were used to calculate the rate of change. This can lead to deception in the general trend over time due to that the last year can have a lower value than the previous years which will result in a negative trend over the time period. However, in this study the values that displayed a decrease in the rate of change, did not show any significant correlation and only a few showed a negative correlation without significance.

### 5.4 Sources of error

#### 5.4.1 Seasonality variation

A factor that was not considered during this study, is the seasonal variation, referring to the differences between spring flood and the pronounced storms during the fall/early winter season. The primary reason for this was the data collected. All measured years did not have



measurements for all twelve months which would have excluded years from the collected time frame of 1987-2017. By measuring the median values for each year, the extreme values were disregarded, however it would be interesting to examine whether or not the higher median values show matching values to when there have been extreme rain periods, followed by floods in Sweden.

A report by the Civil Protection and Preparedness Authority (Myndigheten för samhällsskydd och beredskap (MSB)) was written in 2012, as a consequence to the directive by the European Union, in 2007, to govern the flood management risks. The report include floods in the past that have had serious impact on the environment, human health, economic activity and cultural heritage (MSB 2012).

Since the result of this study showed high median values and high correlation for all three parameters in the southern rivers, following and including #17 Nyköpingsån, the interest regarding past flood occurrences in this area is unavoidable. In 2007 the area defined as of Götaland in Sweden, which includes rivers #18-30, received heavy precipitation during a few days, with values up to 100 mm daily. This caused a consistent flood for a period of four weeks during the summer of 2007 (MSB 2012). An area that was particularly affected was the drainage area of Emån. The consequences of the flood caused traffic accidents, canceled trains and damage to private and industrial properties (MSB 2012). The TON value measured for Emån in June 2007 is the lowest during that entire year,  $552 \mu\text{g L}^{-1}$ . In May, the month prior, measured  $664 \mu\text{g L}^{-1}$  and in July after the flood measured  $692 \mu\text{g L}^{-1}$ .

In the beginning of August in 2002, an area on the west coast called Orust, not far from river #28-29 Bäveån and Örekilsälven, was hit by a severe thunderstorm with precipitation measured to 200 mm the first night and between 40-90 mm the second night (MSB 2012). The flood caused damage on private and public property and 20 000 residents were without telephony and 6000 residents were without electricity (MSB 2012). Bäveån, located closer to Orust than Örekilsälven, registered the second highest values of TON of the year 2002 in August, a measurement of  $884 \mu\text{g L}^{-1}$ , followed by  $508 \mu\text{g L}^{-1}$ . In July it was measured to  $566 \mu\text{g L}^{-1}$ . Örekilsälven on the other hand measured  $427 \mu\text{g L}^{-1}$  in July,  $621 \mu\text{g L}^{-1}$  in August and  $544 \mu\text{g L}^{-1}$  in September causing the August to measure one of the higher values that year.

Since the median, and not weighted average of the water flow of each of the years were included in this report, it would be, given the flood incidents at these respective locations an interesting hypothesis for future studies.

## **6 Conclusion and implications**

This thesis set out to answer how the concentrations of TOC and TON in river discharge have changed over the last 30 years in Sweden. The results show that the null hypothesis, which assumes that there has been no increase of TON, TOC or water color during the period between 1987-2017, should be rejected. The results show a significant increase in the three components, as well as a spatiotemporal significant relevance. The northern part of Sweden shows less of a rate of change trend and not a significant change in the correlation between the total organic carbon through total organic nitrogen.

The question to ask for further research is the underlying cause for the spatiotemporal variations within the country. How is the land use affecting these results? To get a more accurate result, further studies with a weighted average and/or where the flow of the water is considered, could provide interesting results.

The importance of understanding how our river mouths are affected by the chemical components is the gate to implementing restrictions or adaptations to minimize the eutrophication problems that are occurring in the Baltic Sea. The results found were based on measurement within Sweden, however similar patterns in the Northern Hemisphere are to be expected and hence the problem is beyond countries borders. The results of this report contribute to the scientific discussion regarding increased nitrogen and carbon over the last 30 years.

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## 8 Appendices

### 8.1 Correlation matrix of TON, TOC and Abs\_F<sub>420</sub> for each river

The significance level for both the appendices show a significance level of 0.01. The bold represent the significance.

RIVER		COEFFICIENTS OF DETERMINATION (PEARSON):				P-VALUES (PEARSON):				CORRELATION MATRIX (PEARSON):					
		Variables	Time (year)	TON (µg L <sup>-1</sup> )	TOC (mg L <sup>-1</sup> )	Abs_F <sub>420</sub> (5 cm <sup>-1</sup> )	Time (year)	TON (µg L <sup>-1</sup> )	TOC (mg L <sup>-1</sup> )	Abs_F <sub>420</sub> (5 cm <sup>-1</sup> )	Variabl es	Time (year)	TON (µg L <sup>-1</sup> )	TOC (mg L <sup>-1</sup> )	Abs_F <sub>420</sub> (5 cm <sup>-1</sup> )
1	Torne älv	Time (year)	1				0				Time (year)	1			
		TON (µg L <sup>-1</sup> )	0.023	1			0.411	0			TON (µg L <sup>-1</sup> )	0.153	1		
		TOC (mg <sup>-1</sup> )	0.158	0.476	1		0.027	< 0.0001	0		TOC (mg <sup>-1</sup> )	0.397	0.690	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.236	0.347	0.588	1	0.006	0.000	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.486	0.589	0.767	1
2	Kalix älv	Time (year)	1				0				Time (year)	1			
		TON (µg L <sup>-1</sup> )	0.067	1			0.161	0			TON (µg L <sup>-1</sup> )	0.258	1		
		TOC (mg <sup>-1</sup> )	0.087	0.570	1		0.106	< 0.0001	0		TOC (mg <sup>-1</sup> )	0.296	0.755	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.218	0.445	0.475	1	0.008	< 0.0001	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.467	0.667	0.689	1

3	Råne älv	Time (year)	1			0			Time (year)	1								
		TON (µg L <sup>-1</sup> )	0.105	1			0.075	0			TON (µg L <sup>-1</sup> )	0.324	1					
		TOC (mg <sup>-1</sup> )	0.225	0.301	1			0.007	0.001	0			TOC (mg <sup>-1</sup> )	0.475	0.548	1		
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.345	0.164	0.480	1			0.001	0.024	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.587	0.405	0.693	1	
4	Lule älv	Time (year)	1			0			Time (year)	1								
		TON (µg L <sup>-1</sup> )	0.017	1			0.480	0			TON (µg L <sup>-1</sup> )	-0.132	1					
		TOC (mg <sup>-1</sup> )	0.021	0.539	1			0.442	< 0.0001	0			TOC (mg <sup>-1</sup> )	0.143	0.734	1		
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.000	0.398	0.490	1			0.963	0.000	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.009	0.631	0.700	1	
5	Pite älv	Time (year)	1			0			Time (year)	1								
		TON (µg L <sup>-1</sup> )	0.007	1			0.646	0			TON (µg L <sup>-1</sup> )	-0.086	1					
		TOC (mg <sup>-1</sup> )	0.009	0.060	1			0.613	0.186	0			TOC (mg <sup>-1</sup> )	0.095	0.244	1		
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.008	0.083	0.413	1			0.639	0.116	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.088	0.288	0.642	1	
6	Kvistforsen	Time (year)	1			0			Time (year)	1								
		TON (µg L <sup>-1</sup> )	0.018	1			0.476	0			TON (µg L <sup>-1</sup> )	0.133	1					
		TOC (mg <sup>-1</sup> )	0.012	0.383	1			0.554	0.000	0			TOC (mg <sup>-1</sup> )	0.110	0.619	1		
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.000	0.299	0.588	1			0.999	0.001	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.000	0.546	0.767	1	

7	Rickleån	Time (year)	1		0		Time (year)			1				
		TON (µg L <sup>-1</sup> )	0.277	1	0.002	0	TON			0.526	1			
		TOC (mg <sup>-1</sup> )	0.314	0.332	1	0.001	0.001	0	TOC			0.560	0.576	1
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.197	0.295	0.508	1	0.012	0.002	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.444	0.544	0.712
8	Ume älv	Time (year)	1		0		Time (year)			1				
		TON (µg L <sup>-1</sup> )	0.063	1	0.174	0	TON			0.251	1			
		TOC (mg <sup>-1</sup> )	0.055	0.381	1	0.202	0.000	0	TOC			0.236	0.617	1
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.004	0.239	0.387	1	0.748	0.005	0.000	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.06	0.488	0.622
9	Öre älv	Time (year)	1		0		Time (year)			1				
		TON (µg L <sup>-1</sup> )	0.446	1	< 0.0001	0	TON			0.668	1			
		TOC (mg <sup>-1</sup> )	0.266	0.514	1	0.003	< 0.0001	0	TOC			0.516	0.717	1
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.001	0.043	0.239	1	0.864	0.260	0.005	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	-0.032	0.209	0.489
10	Gideälven	Time (year)	1		0		Time (year)			1				
		TON (µg L <sup>-1</sup> )	0.081	1	0.120	0	0.002	TON			0.285	1		
		TOC (mg <sup>-1</sup> )	0.248	0.292	1	0.004	0.002	0	TOC			0.498	0.540	1
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.077	0.169	0.548	1	0.130	0.022	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.278	0.411	0.740



11	Ångermanälven	Time (year)	1			0			Time (year)	1			
		TON (µg L <sup>-1</sup> )	0.083	1		0.116	0		< 0.0001	TON (µg L <sup>-1</sup> )	0.288	1	
		TOC (mg <sup>-1</sup> )	0.042	0.563	1	0.271	< 0.0001	0	TOC (mg <sup>-1</sup> )	0.204	0.750	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.048	0.287	0.473	1	0.234	0.002	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	-0.220	0.535
12	Indalsälven	Time (year)	1			0			Time (year)	1			
		TON (µg L <sup>-1</sup> )	0.001	1		0.853	0		TON (µg L <sup>-1</sup> )	-0.035	1		
		TOC (mg <sup>-1</sup> )	0.044	0.198	1	0.257	0.012	0	TOC (mg <sup>-1</sup> )	0.210	0.445	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.007	0.029	0.657	1	0.645	0.357	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.086	0.171
13	Ljungan	Time (year)	1			0			Time (year)	1			
		TON (µg L <sup>-1</sup> )	0.069	1		0.153	0		TON (µg L <sup>-1</sup> )	-0.263	1		
		TOC (mg <sup>-1</sup> )	0.002	0.100	1	0.796	0.083	0	TOC (mg <sup>-1</sup> )	-0.048	0.316	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.020	0.059	0.530	1	0.449	0.186	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	-0.141	0.244
14	Delångersån	Time (year)	1			0			Time (year)	1			
		TON (µg L <sup>-1</sup> )	0.126	1		0.050	0		TON (µg L <sup>-1</sup> )	0.355	1		
		TOC (mg <sup>-1</sup> )	0.125	0.159	1	0.051	0.026	0	TOC (mg <sup>-1</sup> )	0.353	0.399	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.000	0.130	0.228	1	0.984	0.046	0.007	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	-0.004	0.361

15	Ljusne strömmar	Time (year)	1			0			Time (year)	1					
		TON (µg L <sup>-1</sup> )	0.169	1			0.022	0			TON	-0.411	1		
		TOC (mg <sup>-1</sup> )	0.069	0.063	1		0.153	0.174	0		TOC	0.263	0.251	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.096	0.045	0.743	1		0.090	0.250	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.310	0.213	0.862
16	Dalälven	Time (year)	1			0			Time (year)	1					
		TON (µg L <sup>-1</sup> )	0.130	1			0.047	0			TON	0.360	1		
		TOC (mg <sup>-1</sup> )	0.378	0.116	1		0.000	0.061	0		TOC	0.615	0.340	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.179	0.007	0.541	1		0.018	0.666	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.423	0.081	0.735
17	Nyköpingsån	Time (year)	1			0			Time (year)	1					
		TON (µg L <sup>-1</sup> )	0.260	1			0.003	0			TON	0.510	1		
		TOC (mg <sup>-1</sup> )	0.398	0.142	1		0.000	0.036	0		TOC	0.631	0.377	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.017	0.002	0.415	1		0.484	0.810	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.131	0.045	0.644
18	Motala ström	Time (year)	1			0			Time (year)	1					
		TON (µg L <sup>-1</sup> )	0.033	1			0.327	0			TON	-0.182	1		
		TOC (mg <sup>-1</sup> )	0.187	0.017	1		0.015	0.480	0		TOC	0.432	0.132	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.175	0.065	0.422	1		0.019	0.165	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.419	0.256	0.650

19	Botorpsströ m	Time (year)	1		0				Time (year)	1					
		TON (µg L <sup>-1</sup> )	0.372	1	0.000		0		TON	0.610	1				
		TOC (mg <sup>-1</sup> )	0.509	0.606	1	< 0.0001		< 0.0001		0	TOC	0.713	0.778	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.024	0.326	0.378	1	0.515	0.001	0.000	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.155	0.035	0.156	1
20	Emån	Time (year)	1		0				Time (year)	1					
		TON (µg L <sup>-1</sup> )	0.362	1	0.000		0		TON	0.602	1				
		TOC (mg <sup>-1</sup> )	0.343	0.560	1	0.001		< 0.0001		0	TOC	0.586	0.749	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.236	0.432	0.771	1	0.006	< 0.0001	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.486	0.657	0.878	1
21	Ljungbyån	Time (year)	1		0				Time (year)	1					
		TON (µg L <sup>-1</sup> )	0.186	1	0.015		0		TON	0.432	1				
		TOC (mg <sup>-1</sup> )	0.399	0.437	1	0.000		< 0.0001		0	TOC	0.632	0.661	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.340	0.396	0.919	1	0.001	0.000	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.583	0.630	0.959	1
22	Lyckebyån	Time (year)	1		0				Time (year)	1					
		TON (µg L <sup>-1</sup> )	0.578	1	< 0.0001		0		TON	0.760	1				
		TOC (mg <sup>-1</sup> )	0.494	0.826	1	< 0.0001		< 0.0001		0	TOC	0.703	0.909	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.375	0.706	0.872	1	0.000	< 0.0001	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.612	0.840	0.934	1

23	Mörrumsån	Time (year)	1		0				Time (year)	1				
		TON (µg L <sup>-1</sup> )	0.678	1	< 0.0001		0		TON (µg L <sup>-1</sup> )	0.824	1			
		TOC (mg <sup>-1</sup> )	0.495	0.639	1	< 0.0001		< 0.0001	0	TOC (mg <sup>-1</sup> )	0.703	0.799	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.265	0.544	0.779	1	0.003	< 0.0001	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.515	0.738	0.883
24	Helgeån	Time (year)	1		0				Time (year)	1				
		TON (µg L <sup>-1</sup> )	0.476	1	< 0.0001		0		TON (µg L <sup>-1</sup> )	0.690	1			
		TOC (mg <sup>-1</sup> )	0.492	0.464	1	< 0.0001		< 0.0001	0	TOC (mg <sup>-1</sup> )	0.701	0.681	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.374	0.295	0.803	1	0.000	0.002	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.612	0.543	0.896
25	Lagan	Time (year)	1		0				Time (year)	1				
		TON (µg L <sup>-1</sup> )	0.525	1	< 0.0001		0		TON (µg L <sup>-1</sup> )	0.724	1			
		TOC (mg <sup>-1</sup> )	0.613	0.391	1	< 0.0001		0.000	0	TOC (mg <sup>-1</sup> )	0.783	0.625	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.570	0.296	0.892	1	< 0.0001	0.002	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.755	0.544	0.944
26	Nissan	Time (year)	1		0				Time (year)	1				
		TON (µg L <sup>-1</sup> )	0.398	1	0.000		0		TON (µg L <sup>-1</sup> )	0.631	1			
		TOC (mg <sup>-1</sup> )	0.514	0.456	1	< 0.0001		< 0.0001	0	TOC (mg <sup>-1</sup> )	0.717	0.675	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.498	0.368	0.836	1	< 0.0001	0.000	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.706	0.607	0.914

27	Ätran	Time (year)	1		0				Time (year)	1				
		TON (µg L <sup>-1</sup> )	0.144	1	0.035		0		TON (µg L <sup>-1</sup> )	0.379	1			
		TOC (mg <sup>-1</sup> )	0.602	0.217	1	< 0.0001		0.008	0	TOC (mg <sup>-1</sup> )	0.776	0.466	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.498	0.147	0.788	1	< 0.0001		0.033	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.706	0.383
28	Bäveån	Time (year)	1		0				Time (year)	1				
		TON (µg L <sup>-1</sup> )	0.369	1	0.000		0		TON (µg L <sup>-1</sup> )	0.608	1			
		TOC (mg <sup>-1</sup> )	0.591	0.516	1	< 0.0001		< 0.0001	0	TOC (mg <sup>-1</sup> )	0.769	0.719	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.429	0.360	0.703	1	< 0.0001		0.000	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.655	0.600
29	Örekilsälven	Time (year)	1		0				Time (year)	1				
		TON (µg L <sup>-1</sup> )	0.028	1	0.372		0		TON (µg L <sup>-1</sup> )	0.166	1			
		TOC (mg <sup>-1</sup> )	0.547	0.048	1	< 0.0001		0.234	0	TOC (mg <sup>-1</sup> )	0.740	0.220	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.353	0.001	0.541	1	0.000		0.866	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.594	-0.032
30	Enningsdalsälven	Time (year)	1		0				Time (year)	1				
		TON (µg L <sup>-1</sup> )	0.462	1	< 0.0001		0		TON (µg L <sup>-1</sup> )	0.680	1			
		TOC (mg <sup>-1</sup> )	0.787	0.561	1	< 0.0001		< 0.0001	0	TOC (mg <sup>-1</sup> )	0.887	0.749	1	
		Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.779	0.477	0.912	1	< 0.0001		< 0.0001	< 0.0001	0	Abs_F <sub>420</sub> (5cm <sup>-1</sup> )	0.883	0.690

## 8.2 Correlation matrix over TOC/TON and TON/Abs\_F420

RIVER		COEFFICIENTS OF DETERMINATION (PEARSON):				P-VALUES (PEARSON):			CORRELATION MATRIX (PEARSON):			
		Variables	Time (year)	TOC/TON	TON/Abs <sub>420</sub>	Time (year)	TOC/TON	TON/Abs <sub>420</sub>	Variables	Time (year)	TOC/TON	TON/Abs <sub>420</sub>
1	Torne älv	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.154	1		0.029	0		TOC/TON	0.392	1	
		TON/Abs <sub>420</sub>	0.184	0.377	1	0.016	0.000	0	TON/Abs <sub>420</sub>	-0.429	-0.614	1
2	Kalix älv	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.032	1		0.338	0		TOC/TON	0.178	1	
		TON/Abs <sub>420</sub>	0.103	0.183	1	0.078	0.016	0	TON/Abs <sub>420</sub>	-0.321	-0.428	1
3	Råne älv	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.066	1		0.162	0		TOC/TON	0.257	1	
		TON/Abs <sub>420</sub>	0.138	0.424	1	0.040	< 0.0001	0	TON/Abs <sub>420</sub>	-0.371	-0.651	1
4	Lule älv	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.145	1		0.034	0		TOC/TON	0.381	1	
		TON/Abs <sub>420</sub>	0.010	0.218	1	0.602	0.008	0	TON/Abs <sub>420</sub>	-0.097	-0.466	1
5	Pite älv	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.010	1		0.594	0		TOC/TON	0.099	1	
		TON/Abs <sub>420</sub>	0.012	0.451	1	0.561	< 0.0001	0	TON/Abs <sub>420</sub>	-0.109	-0.672	1
6	Kvistforsen	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.001	1		0.854	0		TOC/TON	0.034	1	
		TON/Abs <sub>420</sub>	0.001	0.448	1	0.851	< 0.0001	0	TON/Abs <sub>420</sub>	0.035	-0.669	1
7	Rickleån	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.025	1		0.394	0		TOC/TON	0.159	1	
		TON/Abs <sub>420</sub>	0.020	0.385	1	0.447	0.000	0	TON/Abs <sub>420</sub>	-0.142	-0.620	1
8	Ume älv	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.000	1		0.955	0		TOC/TON	0.010	1	
		TON/Abs <sub>420</sub>	0.005	0.331	1	0.707	0.001	0	TON/Abs <sub>420</sub>	0.070	-0.575	1
9	Öre älv	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.032	1		0.336	0		TOC/TON	-0.179	1	
		TON/Abs <sub>420</sub>	0.359	0.310	1	0.000	0.001	0	TON/Abs <sub>420</sub>	0.599	-0.557	1

<b>10</b>	Gideälven	Time (year)	<b>1</b>		<b>0</b>			Time (year)	<b>1</b>		
		TOC/TON	0.069	<b>1</b>		0.154	<b>0</b>	TOC/TON	0.262	<b>1</b>	
		TON/Abs <sub>420</sub>	0.007	0.526	<b>1</b>	0.665	<b>&lt; 0.0001</b>	<b>0</b> TON/Abs <sub>420</sub>	-0.081	<b>-0.725</b>	<b>1</b>
<b>11</b>	Ångermanälven	Time (year)	<b>1</b>		<b>0</b>			Time (year)	<b>1</b>		
		TOC/TON	0.001	<b>1</b>		0.851	<b>0</b>	TOC/TON	-0.035	<b>1</b>	
		TON/Abs <sub>420</sub>	0.260	0.227	<b>1</b>	<b>0.003</b>	<b>0.007</b>	<b>0</b> TON/Abs <sub>420</sub>	<b>0.510</b>	<b>-0.476</b>	<b>1</b>
<b>12</b>	Indalsälven	Time (year)	<b>1</b>		<b>0</b>			Time (year)	<b>1</b>		
		TOC/TON	0.079	<b>1</b>		0.126	<b>0</b>	TOC/TON	0.281	<b>1</b>	
		TON/Abs <sub>420</sub>	0.008	0.685	<b>1</b>	0.622	<b>&lt; 0.0001</b>	<b>0</b> TON/Abs <sub>420</sub>	-0.092	<b>-0.827</b>	<b>1</b>
<b>13</b>	Ljungan	Time (year)	<b>1</b>		<b>0</b>			Time (year)	<b>1</b>		
		TOC/TON	0.028	<b>1</b>		0.369	<b>0</b>	TOC/TON	0.167	<b>1</b>	
		TON/Abs <sub>420</sub>	0.001	0.574	<b>1</b>	0.870	<b>&lt; 0.0001</b>	<b>0</b> TON/Abs <sub>420</sub>	-0.031	<b>-0.758</b>	<b>1</b>
<b>14</b>	Delångersån	Time (year)	<b>1</b>		<b>0</b>			Time (year)	<b>1</b>		
		TOC/TON	0.000	<b>1</b>		0.994	<b>0</b>	TOC/TON	0.001	<b>1</b>	
		TON/Abs <sub>420</sub>	0.041	0.196	<b>1</b>	0.275	0.013	<b>0</b> TON/Abs <sub>420</sub>	0.202	-0.443	<b>1</b>
<b>15</b>	Ljusne strömmar	Time (year)	<b>1</b>		<b>0</b>			Time (year)	<b>1</b>		
		TOC/TON	0.264	<b>1</b>		<b>0.003</b>	<b>0</b>	TOC/TON	<b>0.514</b>	<b>1</b>	
		TON/Abs <sub>420</sub>	0.297	0.728	<b>1</b>	<b>0.002</b>	<b>&lt; 0.0001</b>	<b>0</b> TON/Abs <sub>420</sub>	<b>-0.545</b>	<b>-0.853</b>	<b>1</b>
<b>16</b>	Dalälven	Time (year)	<b>1</b>		<b>0</b>			Time (year)	<b>1</b>		
		TOC/TON	0.068	<b>1</b>		0.158	<b>0</b>	TOC/TON	0.260	<b>1</b>	
		TON/Abs <sub>420</sub>	0.050	0.648	<b>1</b>	0.226	<b>&lt; 0.0001</b>	<b>0</b> TON/Abs <sub>420</sub>	-0.224	<b>-0.805</b>	<b>1</b>
<b>17</b>	Nyköpingsån	Time (year)	<b>1</b>		<b>0</b>			Time (year)	<b>1</b>		
		TOC/TON	0.020	<b>1</b>		0.449	<b>0</b>	TOC/TON	0.141	<b>1</b>	
		TON/Abs <sub>420</sub>	0.002	0.597	<b>1</b>	0.574	<b>&lt; 0.0001</b>	<b>0</b> TON/Abs <sub>420</sub>	-0,224	<b>-0,805</b>	<b>1</b>
<b>18</b>	Motala ström	Time (year)	<b>1</b>		<b>0</b>			Time (year)	<b>1</b>		
		TOC/TON	0.239	<b>1</b>		<b>0.005</b>	<b>0</b>	TOC/TON	<b>0.489</b>	<b>1</b>	
		TON/Abs <sub>420</sub>	0.230	0.414	<b>1</b>	<b>0.006</b>	<b>&lt; 0.0001</b>	<b>0</b> TON/Abs <sub>420</sub>	<b>-0.479</b>	<b>-0.643</b>	<b>1</b>



19	Botorpsström	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.251	1		0.004	0		TOC/TON	0.501	1	
		TON/Abs <sub>420</sub>	0.011	0.140	1	0.555	0.038	0	TON/Abs <sub>420</sub>	0.104	-0.375	1
20	Emån	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.071	1		0.148	0		TOC/TON	0.266	1	
		TON/Abs <sub>420</sub>	0.137	0.496	1	0.041	< 0.0001	0	TON/Abs <sub>420</sub>	-0.370	-0.705	1
21	Ljungbyån	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.278	1		0.002	0		TOC/TON	0.527	1	
		TON/Abs <sub>420</sub>	0.268	0.632	1	0.003	< 0.0001	0	TON/Abs <sub>420</sub>	-0.517	-0.795	1
22	Lyckebyån	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.065	1		0.167	0		TOC/TON	0.254	1	
		TON/Abs <sub>420</sub>	0.177	0.561	1	0.018	< 0.0001	0	TON/Abs <sub>420</sub>	-0.421	-0.749	1
23	Mörrumsån	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.101	1		0.082	0		TOC/TON	0.317	1	
		TON/Abs <sub>420</sub>	0.163	0.492	1	0.024	< 0.0001	0	TON/Abs <sub>420</sub>	-0.404	-0.702	1
24	Helgeån	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.129	1		0.047	0		TOC/TON	0.360	1	
		TON/Abs <sub>420</sub>	0.159	0.589	1	0.026	< 0.0001	0	TON/Abs <sub>420</sub>	-0.399	-0.768	1
25	Lagan	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.232	1		0.006	0		TOC/TON	0.481	1	
		TON/Abs <sub>420</sub>	0.332	0.847	1	0.001	< 0.0001	0	TON/Abs <sub>420</sub>	-0.576	-0.920	1
26	Nissan	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.113	1		0.064	0		TOC/TON	0.337	1	
		TON/Abs <sub>420</sub>	0.258	0.654	1	0.004	< 0.0001	0	TON/Abs <sub>420</sub>	-0.508	-0.809	1
27	Ätran	Time (year)	1			0			Time (year)	1		
		TOC/TON	0.462	1		< 0.0001	0		TOC/TON	0.680	1	
		TON/Abs <sub>420</sub>	0.403	0.719	1	0.000	< 0.0001	0	TON/Abs <sub>420</sub>	-0.635	-0.848	1

<b>28</b>	Bäveån	Time (year)	<b>1</b>	<b>0</b>				Time (year)	<b>1</b>			
		TOC/TON	0.253	<b>1</b>	<b>0.005</b>				<b>0</b>	TOC/TON	<b>0.503</b>	<b>1</b>
		TON/Abs <sub>420</sub>	0.172	0.420	<b>1</b>	0.023	<b>0.000</b>	<b>0</b>	TON/Abs <sub>420</sub>	-0.415	<b>-0.648</b>	<b>1</b>
<b>29</b>	Örekilsälven	Time (year)	<b>1</b>	<b>0</b>				Time (year)	<b>1</b>			
		TOC/TON	0.339	<b>1</b>	<b>0.001</b>				<b>0</b>	TOC/TON	<b>0.583</b>	<b>1</b>
		TON/Abs <sub>420</sub>	0.213	0.615	<b>1</b>	<b>0.009</b>	<b>&lt; 0.0001</b>	<b>0</b>	TON/Abs <sub>420</sub>	<b>-0.462</b>	<b>-0.784</b>	<b>1</b>
<b>30</b>	Enningsdalsälven	Time (year)	<b>1</b>	<b>0</b>				Time (year)	<b>1</b>			
		TOC/TON	0.443	<b>1</b>	<b>&lt; 0.0001</b>				<b>0</b>	TOC/TON	<b>0.665</b>	<b>1</b>
		TON/Abs <sub>420</sub>	0.590	0.774	<b>1</b>	<b>&lt; 0.0001</b>	<b>&lt; 0.0001</b>	<b>0</b>	TON/Abs <sub>420</sub>	<b>-0.768</b>	<b>-0.880</b>	<b>1</b>