Trends in nutrients and metals in Norwegian rivers and point sources 1990–2009

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ABSTRACT

This assessment of nine river catchments in Norway covers 20 years (1990–2009) of water quality data on total phosphorus (TP), orthophosphate, total nitrogen (TN), ammonium, nitrate, copper, zinc, cadmium, lead and nickel. The nine catchments are located from the south to the north of the country and are included in the Riverine Inputs and Direct Discharges (RID) programme of the OSPAR Commission for Protection of the Marine Environment of the North-East Atlantic. The partial Mann–Kendall test was used to evaluate long-term monotonic trends. For both TP and TN, decreasing trends ($p < 5\%$) were found in three out of nine rivers under study. Downward trends in riverine metal loads were found in 23 of 45 tests. Only one significant increasing trend in nutrient loads was found, and there were no upwards trends in metal loads. To some extent, the trends in riverine loads could be explained by trends in discharges from point sources. Even after taking potential sources of error into consideration, these results indicate that mitigation measures implemented since 1990 to reduce pressures from point sources have had an impact on water quality in Norwegian rivers.

Key words | metals, nutrients, point sources, riverine loads, trends

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INTRODUCTION

Concern for the marine environment has led to several international agreements and directives with a common goal of reducing pollutant loads to the seas. Examples of these actions are the OSPAR (OSlo-PARis) Convention for the Protection of the Marine Environment of the North-East Atlantic (www.ospar.org), the Helsinki Commission (HELCOM) for the Protection of the Marine Environment of the Baltic Sea Area (http://www.helcom.fi) and the relatively recently established Marine Strategies Framework Directive of the European Union (http://ec.europa.eu/environment/marine). In turn, these measures have led to several international programmes for monitoring the seas, the air and the rivers draining to the seas. The OSPAR programme for monitoring land-based sources of pollution of the North Atlantic is entitled Riverine Inputs and Direct Discharges (RID). The objectives of RID were drawn up jointly by the contracting parties, and, among other things, state that all riverine inputs and direct discharges of specific pollutants to OSPAR Convention waters should be assessed on an annual basis and be reviewed periodically to determine temporal trends (Parcom 1998). The Norwegian RID programme was started in 1990, and it uses three methods to record loads from the mainland to the sea: monitoring of water quality in rivers; monitoring of direct discharges from point sources; and modelling/estimating loads from unmonitored areas. The data provided by such programmes can be valuable for many purposes, including the assessment of trends in pollution from both long-range and local land-based anthropogenic activities (Howarth *et al.* 1996; Skjelkvåle *et al.* 2001; Littlewood & Marsh 2005; Iital *et al.* 2010; Bouraoui & Grizzetti 2011). Long-term RID data can also be used as a basis for modelling aimed at tasks like predicting possible impacts of climate change on water quality in rivers and adjoining marine areas (Kaste *et al.* 2006; Wright *et al.* 2008). However, an evaluation of data from the British RID programme revealed that several constituents were less useful for trend analyses for various reasons, such as data missing for certain years, a large

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number of values below detection limits, low sampling frequencies and problems with extreme outliers (Littlewood *et al.* 1998). The need to ensure the quality of data before analysing for trends was also emphasized in the latest periodic assessment of the European RID data (OSPAR Commission 2009). Consequently, a comprehensive review of the Norwegian RID database was recently carried out in which the data were checked and corrected against the original records and anomalies investigated (Stålnacke *et al.* 2009). Parameters less suitable for trend analyses were identified using criteria similar to those reported by Littlewood & Marsh (2005), including several values at or below detection limits, changes in detection limits or analytical methods over time, and/or several years without records.

There are surprisingly few publications concerning water quality trends in the rivers of northern Europe. Räike *et al.* (2003) described nutrient trends in rivers in Finland, and Stålnacke *et al.* (1999b) investigated trends in nitrogen loads in rivers in Sweden. In Norway, studies regarding trends in river water quality have focussed primarily on smaller waterways, such as heathland brooks with limited local anthropogenic pressures (Kaste & Skjelkvåle 2002) or agricultural streams (Bechmann *et al.* 2008), and hence there is a lack of scientific literature on trends in pollutants in the larger Norwegian rivers.

The RID data have the advantage of also containing two decades of records on effluents from point sources such as industrial facilities and wastewater treatment plants (WWTPs), and this enables comparison of trends in rivers under pressure from pollution. However, it is not always a straightforward task to link trends in rivers with activities in the upstream catchments, where a number of simultaneous terrestrial and aquatic processes can conceal the direct connections between pressures and impacts (e.g., Moss 1999; Harris & Heathwaite 2012). Nonetheless, it is important that managers can show that expensive mitigation measures or changes in land use will actually have an effect on the water quality, not least to satisfy the requirements of the EU Water Framework Directive (EU 2000). Hence, during the last decade, many studies in the literature have concerned the water quality response to implemented measures and large-scale changes in land use, particularly with respect to nutrients. For example, Pastuszak *et al.* (2012) investigated the two largest rivers in Poland (the Vistula and the Oder) and found that the nitrogen load decreased by 20 and 25% and the phosphorus load by 15 and 65%, respectively, from 1988 to 2008. The fall of the Iron Curtain in the early 1990s should also be mentioned in this context, because it resulted in a dramatic reduction in the intensity of agriculture, as illustrated by a 70–90% decrease in the use of fertilizers (Stålnacke *et al.* 2004). Despite this, there was a certain time lag before it was possible to discern a significant downward trend, and this inertia has also been demonstrated and discussed by many authors (e.g., Grimvall *et al.* 2000; Sileika *et al.* 2006; Iital *et al.* 2010). For example, Bouraoui & Grizzetti (2011) pointed out that in large rivers like the Elbe and the Loire it would take 8 and 14 years, respectively, for the entire system to respond to changes in nutrient application. Grimvall *et al.* (2000) used examples from western Europe to demonstrate that riverine loads of phosphorus could be rapidly reduced from high to moderate levels, but that any further reduction, if achieved at all, could take decades.

The main objective of the present study was to employ quality assured data from two decades of RID monitoring in Norway to assess nutrient and metal trends in riverine loads and direct discharges from point sources. In addition, possible connections between the riverine loads and point sources of pollution were examined, while at the same time considering other pressures in the upstream catchments.

MATERIALS AND METHODS

River catchments

A total of 247 rivers drain to the North Atlantic from the mainland of Norway. However, for economic reasons, only 46 of the rivers have been monitored regularly in the Norwegian RID programme; 36 of these are sampled four times a year and the other 10 once a month. Those 10 rivers were selected for monthly sampling in the late 1980s based on criteria related to the geographical distribution, size and availability of adequate flow data. Furthermore, eight of the 10 were chosen because they were among the most load-bearing rivers in the country, whereas the remaining two (the Alta and the Suldalslågen) were to represent semi-natural catchments with little or no local pollution. Although most of the monitored rivers are affected by hydropower regulation, the flow in the Suldalslågen River is particularly affected by water abstractions. Accordingly, the Suldalslågen was removed from the monitoring programme and replaced with the Vosso River in 2008. Due to the disrupted time series for both of those rivers, they were excluded from our study, which consequently comprised a total of nine rivers (cf. Table 1 and Figure 1). The total catchment area for these nine rivers is about 94,000 km^2, which constitutes about 30% of the Norwegian mainland.

The Glomma, Drammenselva, Numedalslågen, Skienselva and Otra flow into the Skagerrak and represent the Norwegian rivers with the highest nutrient and metal loads within this marine area. The Glomma has the largest catchment area of all Norwegian rivers, and the Drammenselva has the third largest catchment area. The Orre River, which drains into the North Sea, is relatively small but is included in the RID programme because it drains one of the most intensive agricultural areas in Norway. More than 30% of the Orre catchment is used for farming, and eutrophication problems such as toxic algal blooms have been reported (Bechmann *et al.* 2005). The Orkla and Vefsna Rivers flow into the Norwegian Sea, and only 4 and 8%, respectively, of the land in their catchment areas is used for agricultural purposes. In other words, farming is less intensive in this part of the country compared to the region around the Orre River, but a more important issue in the Orkla–Vefsna area is the presence of abandoned mines and also some industrial activities. The last of the main rivers, the Alta, flows into the Barents Sea, and its catchment area has a population density of only 0.3 persons per km^2 and no industrial plants reporting discharges.

Water quality and flow

In each river, water is collected as grab samples in sections of the stream with good water mixing. The sampling stations are located as close to the river outlet as possible, but upstream of any salt water intrusion. Sampling frequency is once a month except in the Glomma and Drammenselva Rivers, where four additional samples are taken in the months of May and June to improve representation of the high water discharge during the snow melt season. Analyses include six fractions of nutrients (total phosphorus (TP), orthophosphate (PO_4-P), total nitrogen (TN), ammonium, nitrate and silicate), eight heavy metals (copper, zinc, cadmium, lead, chromium, nickel, mercury and arsenic), one pesticide (lindane), seven polychlorinated biphenyl (PCB) compounds (CB28, CB52, CB101, CB118, CB138, CB153 and CB180), and four other parameters (suspended particulate matter [SPM], total organic carbon, pH and conductivity). Analytical methods and detection limits for the entire period have been reported by Skarbøvik *et al.* (2011). During the course of the 20 yr of monitoring, some analytical results, especially for

Table 1 | Geographical characteristics of the nine studied Norwegian rivers. The rivers are listed from south (Glomma River) to north (Alta River)

					Location of water quality station	
No.	**Name of river**	**Marine waters discharge area**	**Size of catchment area (km^2)**	**Long-term average specific flow (l/s km^2)**	**Latitude**	**Longitude**
1	Glomma	Skagerrak	41,918	16.9	59.27800	11.13400
2	Drammenselva	Skagerrak	17,034	18.2	59.75399	10.00903
3	Numedalslågen	Skagerrak	5,577	21.1	59.08627	10.06962
4	Skienselva	Skagerrak	10,772	25.3	59.19900	9.61100
5	Otra	Skagerrak	3,738	39.8	58.18742	7.95411
6	Orre	North Sea	105	47.4	58.73143	5.52936
7	Orkla	Norwegian Sea	3,053	14.7	63.20100	9.77300
8	Vefsna	Norwegian Sea	4,122	40.0	65.74900	13.23900
9	Alta	Barents Sea	7,373	11.9	69.90100	23.28700

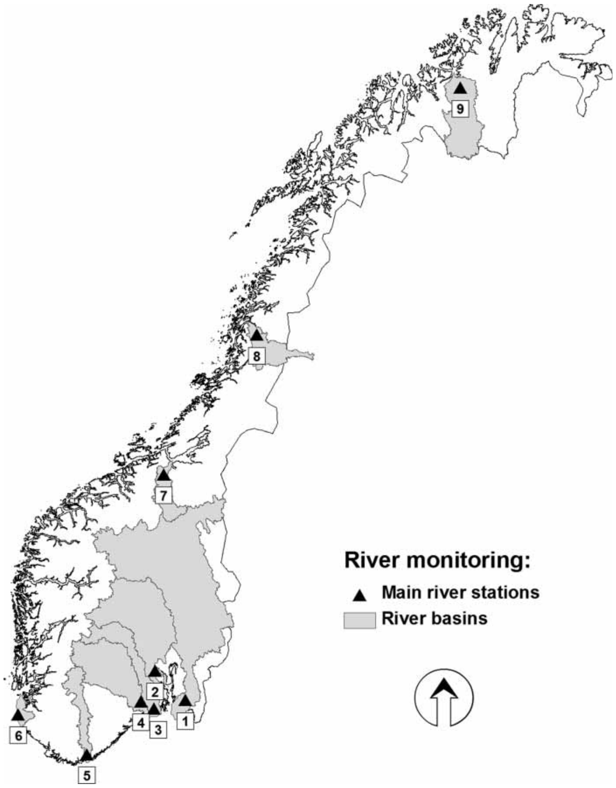

Figure 1 | Map showing the nine rivers included in the investigation. The numbers refer to those assigned to the rivers in Table 1.

orthophosphate, nitrate and cadmium were below the detection limit; in these cases, the concentrations were set to zero.

Daily water discharge measurements were used to calculate loads. In some rivers, the stations for water level/water discharge registration are not located at the same sites as the water quality stations, and therefore the discharge at the quality sampling sites was calculated by up- or downscaling based on the size of the respective drainage areas.

Load calculation

As outlined by Stålnacke *et al.* (2009), the load calculation formula applied to the Norwegian RID data has been slightly modified from the original formula recommended by the RID programme (Parcom 1998), and the following formula is now used:

$$\text{Load} = Q_r \frac{\sum_1^n Q_i C_i t_i}{\sum_1^n Q_i t_i}$$

where Q_i represents the water discharge on the day of sampling (day i); C_i is the concentration on day i; t_i is the time period from the midpoint between day i – 1 and day i to the midpoint between day i and day $i + 1$ (i.e., half the number of days between the previous and next sampling); and Q_r is the annual water volume. The main improvement in this modified formula compared to the original formula is that it can better handle irregular sampling frequency, including samples collected during flood conditions. This is particularly important when calculating loads in the Glomma and Drammenselva Rivers, where additional sampling is done during the snow melt period in spring.

Discharges from point sources

Three point sources of pollution are recorded in the Norwegian RID programme: WWTPs, industries and fish farms. Most fish farms are located in the sea, and consequently such facilities have little influence on riverine loads and were excluded from the trend analyses in our study. Data on nutrients from all WWTPs with a capacity of more than 50 person equivalents (PE) are registered annually in a national database. Municipal wastewater can also contain industrial effluents, but larger amounts of such discharges must be reported separately according to concession agreements.

Data quality assessment

As mentioned in the introduction, a basis for the present study was the quality assurance of the Norwegian RID data that was performed in the late 2000s (Stålnacke *et al.* 2009). That work revealed a pronounced alteration in TP concentrations during the period 1999–2003, which was due to a switch to a different laboratory for analysis of samples. This resulted in rather severe anomalies in TP concentrations, and thus the data from this period were replaced with interpolated data. Inter- and extrapolation of data have also been done for data on reported discharges from point sources. Not all industrial units or WWTPs report each year, and in order to ensure a more consistent dataset, interpolation, and to some degree also extrapolation, of data have been done in years where units still in operation had omitted to report their data. The criteria and methodology for this inter- and extrapolation are described in Stålnacke *et al.* (2009). Furthermore, assessments of changes in analytical methods and levels of detection over time, and in the length of the data series, resulted in a decision to limit the trend analyses to the following constituents: cadmium, copper, nickel, lead, zinc, ammonium nitrogen (NH_4-N), nitrate nitrogen (NO_3-N), TN, PO_4-P and TP. For the discharges from point sources, trend analyses were performed on the same constituents except for PO_4-P, NH_4-N and NO_3-N.

Methodology for trend analyses

The partial Mann–Kendall test (Libiseller & Grimvall 2002) was used to detect long-term monotonic trends in loads in the nine rivers and in discharges from WWTPs and industrial facilities. Here, monotonic is defined as an increase or decrease that is consistent over time and can be linear (the same slope over time) or non-linear. This approach has its methodological basis in the seasonal Mann–Kendall test (Hirsch & Slack 1984), with the difference being that water discharge is included as explanatory variable when analysing rivers. Accordingly, spurious trends caused by trends in water discharges and statistical relationships between the constituent load and water discharge are eliminated as far as possible. The partial Mann–Kendall test also includes a correction for serial correlation up to a user-defined time span (in our case 1 year), and it offers convenient handling of missing values. Trends were considered as statistically significant at the 5% level (double-sided test). All trend tests were also subjected to a visual inspection, because some apparent trends might have been influenced by the interpolated or extrapolated values calculated during the data assessment.

RESULTS AND DISCUSSION

Current pollutant loads to the sea

The main purpose of the RID monitoring is to assess annual pollutant loads to the marine areas and to detect any trends in those loads. Annual loads in the rivers vary significantly depending on water discharges, and thus the annual mean values for the period 2005–2009 were used to estimate current loads to the sea. During the indicated period, the total annual loads from the Norwegian mainland were calculated based on monitoring of 37 additional rivers, estimated diffuse inputs from unmonitored rivers, and discharges from sewage treatment plants and industries (excluding fish farms) (for details of the calculation methodology and results, see Skarbøvik *et al.* (2009)). As indicated in Table 2, the nine rivers contribute from 22 to 47% of the total inputs of the constituents in question from the Norwegian mainland to the sea. This reflects the importance of these rivers, particularly bearing in mind that Norway actually has 247 rivers that run to the North Atlantic.

Norwegian rivers drain into four different OSPAR marine areas (Table 1), two of which (i.e., the North Sea and the Skagerrak) also receive inputs from other countries (Sweden, Denmark, Germany, the Netherlands, Belgium and the United Kingdom). In 2009, Norway contributed the following proportions of the total loads of the indicated constituents (including pollutants from fish farms) to the North Sea and Skagerrak (Pengerud & Skarbøvik 2010): 8, 5, 16, 21, 6 and 14% of the nitrogen, phosphorus, cadmium, copper, lead and zinc, respectively. Nickel is not monitored in all of the mentioned countries, and therefore it was not possible to compare loads of that element from Norway with those from the other nations.

Area-specific pollutant loads varied significantly between the nine catchments studied (Table 3). Nutrient loads varied from 3 to 180 kg TP/km^2 and from 74 to 3,343 kg TN/km^2. By comparison, rivers flowing to the Baltic Sea have been reported to have area-specific loads ranging from 11 to 42 kg TP/km^2 and from 204 to 1,220 kg TN/km^2 (Stålnacke *et al.* 1999a). The area-specific loads of suspended sediments, nutrients, nickel and lead were highest in the smallest river, the Orre, whereas the Orkla River had the highest area-specific loads of copper and zinc. The Alta River represents relatively pristine conditions, and it had the lowest area-specific loads of all substances.

Discharges from point sources in the nine catchments

The relative importance of the discharges from point sources as compared to the riverine loads was assessed by

Table 2 | Annual loads (based on an average for the period 2005–2009) of metals and nutrients in the nine rivers, and as total for the Norwegian mainland (excluding fish farms)

River	TP tonnes	TN tonnes	SPM tonnes	Cu tonnes	Zn tonnes	Cd tonnes	Ni tonnes	Pb tonnes
Glomma	455	13,606	246,000	51	101	0.34	21	7.8
Drammenselva	78	4,978	37,000	12	45	0.11	7.3	2.2
Numedalslågen	71	1,734	55,000	6	23	0.08	2.0	2.9
Skienselva	45	2,772	11,000	5	24	0.10	2.3	0.7
Otra	19	1,154	7,000	6	22	0.10	2.8	1.5
Orre	19	351	4,000	0.3	1	0.00	0.2	0.2
Orkla	16	765	9,000	20	42	0.12	2.1	0.2
Vefsna	15	610	12,000	2	13	0.01	1.1	0.3
Alta	24	547	12,000	2	7	0.00	0.9	0.2
Sum 9 rivers	744	26,517	393,000	104	278	0.87	40	16
Totals to the sea[a]	3,377	113,129	828,000	240	621	2.4	163	41
	%	%	%	%	%	%	%	%
% of the 9 rivers	22	23	47	43	45	36	25	39

[a] Based on data from Skarbøvik *et al.* (2009).

Table 3 | Average annual area-specific loads of eight constituents in nine Norwegian rivers for the period 2005–2009

River	TP kg/km^2	TN kg/km^2	SPM kg/km^2	Cu kg/km^2	Zn kg/km^2	Cd kg/km^2	Ni kg/km^2	Pb kg/km^2
Glomma	11	325	5,869	1.22	2.41	0.008	0.50	0.187
Drammenselva	5	292	2,172	0.70	2.64	0.007	0.43	0.130
Numedalslågen	13	311	9,862	1.08	4.12	0.015	0.36	0.518
Skienselva	4	257	1,021	0.46	2.23	0.009	0.21	0.066
Otra	5	309	1,873	1.61	5.89	0.028	0.74	0.403
Orre	180	3,343	38,095	2.86	9.52	0.040	1.95	1.305
Orkla	5	251	2,948	6.55	13.76	0.039	0.68	0.052
Vefsna	4	148	2,911	0.49	3.15	0.001	0.27	0.087
Alta	3	74	1,628	0.27	0.95	0.000	0.12	0.022

simple tests that did not take retention into account. During the period 2005–2009, the average TP and TN discharges from the WWTPs located upstream of the sampling points in the nine rivers corresponded to 3–4% of the riverine loads. The Skienselva and Orkla Rivers had the highest proportions of TP from WWTPs (6–7%), whereas the highest proportion of TN from WWTPs (6–10%) was found in four of the rivers draining to the Skagerrak area (Glomma, Drammenselva, Numedalslågen and Skienselva Rivers). In all nine rivers, the discharges of metals from WWTPs comprised only a very small part (less than 1%) of the riverine loads.

Considering industrial nutrient discharges, the TP effluents constituted approximately 8% and TN about 1% of the loads in the nine rivers (again based on annual averages for 2005–2009 and without adjusting for retention). However, for the three northernmost rivers, no nutrients in industrial effluents had been reported. The metal discharges from industry varied considerably between the rivers. Metal-laden effluents to the Numedalslågen, Skienselva, Orre, Otra, Vefsna and Alta Rivers were very small compared to the riverine loads, whereas the industrial metal discharges in the other rivers were much larger. There is probably a certain degree of retention in the Orkla River catchment, because the average industrial discharges of copper, zinc and cadmium over the 5-year period were actually higher than the riverine loads: more specifically, 25% higher for copper and 50% higher for zinc and cadmium. In the Glomma River, copper discharged from industries constituted 29% of the riverine loads, whereas the corresponding figures for zinc and cadmium were 33 and 21%, respectively. In the Drammenselva River, industrial discharges of zinc and cadmium represented about 3 and 5% of the riverine loads, respectively. Industrial discharges of nickel and lead constituted only 0–1% of the riverine loads.

Nutrient trends

In general, loads of substances in rivers are flow dependent, and therefore it is useful to include analyses of variations in water discharges in the overall assessment. Our evaluation of annual water discharges showed the following: a significant upward trend in the Drammenselva River ($p < 0.03$); an upward, albeit not significant, trend in the Skienselva River ($p < 0.08$) (Table 4); and a tendency towards increased water discharges since 2004 in the Orre River.

Examining riverine loads of TP, statistically significant downward trends were detected in three rivers: the Otra, the Vefsna and the Altaelva (Table 4). Orthophosphate showed a statistically significant downward trend in the Vefsna River and weak downward trends in the Otra and Altaelva Rivers ($p < 0.08$).

Downward trends in phosphorus discharges from WWTPs were found for four of the five rivers flowing to the Skagerrak region, and the same was noted for the Altaelva River in the north (Table 4). There were also downward trends in the phosphorus content in industrial effluents released to the Otra River. However, significant increases in TP emitted from industries to the Glomma River were noted, but the proportion of effluents of TP from industries was low in comparison to total TP loads in this river.

Regarding nitrogen, statistically significant downward trends ($p < 0.05$) in loads of both TN and nitrate nitrogen were detected in three of the nine rivers: the Skienselva, the Vefsna and the Altaelva (Table 5). In addition, downward trends were found for ammonium nitrogen in the Glomma, Orkla, Vefsna and Altaelva Rivers and for nitrate nitrogen in the Otra River. For the Skienselva, Drammenselva and Glomma Rivers, where WWTP emissions contributed about 7–10% of the riverine TN loads, there were statistically significant reductions in TN effluents from WWTPs. Significant downward trends in nitrogen discharges from WWTPs were also noted for the Orkla and Alta Rivers, but the TN discharges to these rivers were

Table 4 | Trends (*p*-values) in water discharge (*Q*), phosphorus loads and direct discharges from WWTPs and industries shown for the nine studied rivers[a]

		Riverine loads		**WWTPs**	**Industries**
River	***Q***	**TP**	**PO_4-P**	**TP**	**TP**
Glomma	0.349	0.288	0.562	0.002 (–)	0.002 (+)
Drammenselva	0.026 (+)	0.830	0.640	<0.001 (–)	0.475
Numedalslågen	0.195	0.615	0.687	<0.001 (–)	0.183
Skienselva	0.071	0.278	0.942	0.005 (–)	0.733
Otra	0.747	0.039 (–)	0.070	0.144	0.002 (–)
Orre	0.182	0.218	0.879	NA	0.061
Orkla	0.662	0.499	0.314	0.454	NA
Vefsna	0.964	<0.001 (–)	0.024 (–)	0.845	NA
Altaelva	0.992	0.013 (–)	0.081	0.003 (–)	NA

[a] For statistically significant trends ($p < 0.05$), + denotes an upward trend and – signifies a downward trend.

NA: not applicable.

Table 5 | Trends (*p*-values) in water discharges (*Q*), nitrogen loads and direct discharges from WWTPs and industries shown for the nine studied rivers[a]

		Riverine loads			**WWTPs**	**Industries**
River	***Q***	**TN**	**NH_4-N**	**NO_3-N**	**TN**	**TN**
Glomma	0.349	0.544	0.001 (–)	0.469	0.018 (–)	0.032 (+)
Drammenselva	0.026 (+)	0.369	0.112	0.938	0.029 (–)	0.795
Numedalslågen	0.195	0.032 (+)	0.134	0.405	0.144	0.324
Skienselva	0.071	0.003 (–)	0.033 (–)	0.001 (–)	0.012 (–)	0.621
Otra	0.747	0.106	0.808	<0.001 (–)	0.051	0.001 (–)
Orre	0.182	0.476	0.007 (–)	0.393	NA	0.869
Orkla	0.662	0.200	0.041 (–)	0.269	0.037 (–)	NA
Vefsna	0.964	0.007 (–)	<0.001 (–)	<0.001 (–)	0.229	NA
Altaelva	0.992	0.007 (–)	0.027 (–)	0.011 (–)	0.023 (–)	NA

[a]For statistically significant trends ($p < 0.05$), + denotes an upward trend and – signifies a downward trend.
NA: not applicable.

small compared to the riverine loads (1–2%). For point sources, the only significant upwards trend in TN loads was found for industrial effluents in the Glomma River, but the proportion of effluents of TN from industries was low in comparison to total TN loads in this river.

The only significant upward trend in riverine nutrient loads was found for TN in the Numedalslågen River. In this region, there is evidence that concentrations of dissolved organic carbon (DOC) have increased since 1990 (de Wit *et al.* 2007; Monteith *et al.* 2007). It has been observed that DOC concentrations and concentrations of organically bound nitrogen follow similar patterns (Haaland *et al.* 2008).

Analysis of the concentration data for the same time period revealed good agreement between the patterns in the trends in loads and the trends in concentrations of the analysed nutrients (Skarbøvik *et al.* 2009). By comparison, Bouraoui & Grizzetti (2011) analysed data from 90 river stations in Europe (including four Norwegian RID stations) and, for levels of dissolved inorganic nitrogen, found increasing trends in 12% of the rivers and decreasing trends in 23%; results regarding TP in the same study showed increasing trends in 8% of the rivers and decreasing trends in 38%.

In addition to point sources, diffuse sources will contribute nutrients to the rivers. The environmental authorities in Norway have focussed on mitigation measures to decrease nutrient losses from agricultural areas, but this has mainly concerned phosphorus (Bechmann *et al.* 2008) because of the role this element plays as a limiting nutrient in most of our freshwater ecosystems. By comparison, few targeted mitigation efforts have been implemented to reduce nitrogen losses, although the official recommendations have stipulated lower use of fertilizers in recent years, which also apply to nitrogen. For the country as a whole, this has lowered the annual sales of fertilizers and thereby also reduced application of nutrients from about 12–14 kilotonnes (kt) of phosphorus and 100–120 kt of nitrogen during the period 1990–2007 to 6 kt of phosphorus and 80 kt of nitrogen in 2008–2009 (www.mattilsynet.no). However, this latter reduction in fertilizers is not likely detected in the riverine loads presented in this paper since many studies have shown that, due to the effects of retention processes, it can take several years or even decades for rivers to respond to reductions in fertilizer application (cf. Grimvall *et al.* 2000; Stålnacke *et al.* 2003, 2004; Bouraoui & Grizzetti 2011). Nevertheless, it is apparent that levels contributed from other sources have also been reduced, because national monitoring programmes have revealed that nitrate concentrations in precipitation at all monitoring stations in southern Norway have declined significantly since 1980 (Schartau *et al.* 2011). A significant decrease in ammonium has been observed at most of the stations as well.

An example of nutrient trends: the Skienselva River

Of the nine rivers studied here, the Skienselva had a relatively high proportion of discharges of both TP and TN from WWTPs (about 7%). The number of WWTPs in this catchment has been relatively stable around 32 units, which are distributed along the main river courses from 1 and up to 150 km upstream of the sampling point. There were statistically significant downward trends in effluents of TN and TP from WWTPs, and in riverine loads of TN, NH_4-N and NO_3-N (Table 5). The annual TN loads in this river (Figure 2(a)) correlated well with the water discharges during 1990–2009 ($R^2 = 0.7$), although there is a tendency of increased water discharges during the 20-year period ($p < 0.08$; Table 5). The decrease in TN is further illustrated in Figure 2(b), which shows the annual flow-weighted TN concentration in this river (i.e., load divided by water flow). This chart further illustrates that the reductions were most pronounced in the 1990s and that the levels have been more or less stable in the 2000s. This does, to a large extent, coincide with the change in the levels of TN effluents from WWTPs over the years (Figure 2(c)).

Trends in metals

Statistically significant downward trends in riverine loads of metals were found in 23 of the 45 trend tests performed (9 rivers × 5 metal constituents; Table 6). In four additional cases, there were indications of downward trends

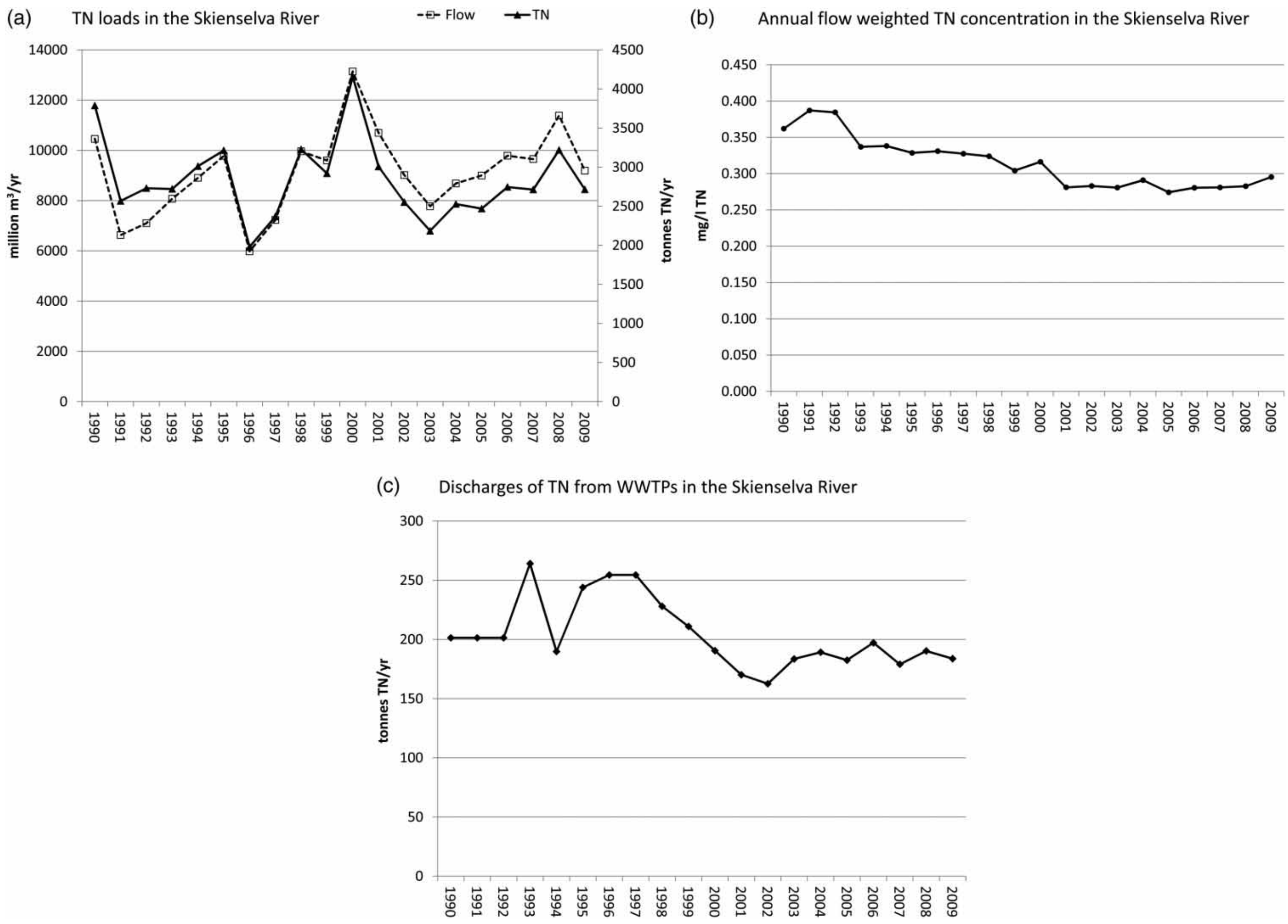

Figure 2 | Data on the Skienselva catchment showing riverine TN loads (tonnes) and water flow (mill m^3/yr) (a); annual flow-weighted TN concentrations (mg/L) (b); and TN effluents (tonnes) from WWTPs (c).

Table 6 | Long-term trends (*p*-values) in water discharge (*Q*) and metal loads in nine Norwegian rivers during the period 1990–2009[a]

		Riverine loads				
River	***Q***	**Cd**	**Cu**	**Ni**	**Pb**	**Zn**
Glomma	0.349	0.128	0.183	0.014 (–)	0.067	0.014 (–)
Drammenselva	0.026 (+)	<0.001 (–)	0.773	0.949	0.279	0.642
Numedalslågen	0.195	0.020 (–)	0.078	0.326	0.054	0.026 (–)
Skienselva	0.071	0.012 (–)	0.036 (–)	0.135	0.187	0.011 (–)
Otra	0.747	0.297	0.192	0.002 (–)	0.321	0.065
Orre	0.182	0.182	0.190	<0.001 (–)	0.514	0.794
Orkla	0.662	0.036 (–)	0.017 (–)	0.011 (–)	0.008 (–)	0.007 (–)
Vefsna	0.964	<0.001 (–)	0.001 (–)	<0.001 (–)	<0.001 (–)	0.017 (–)
Altaelva	0.992	0.001 (–)	0.001 (–)	0.005 (–)	0.581	0.379

[a] For statistically significant trends ($p < 0.05$); + denotes an upward trend and – signifies a downward trend.

($0.05 < p < 0.1$). No upward trends in riverine loads were detected for the investigated metals. In greater detail, the trends in riverine loads can be described as follows:

- For copper loads, statistically significant downward trends were detected in the Skienselva, Orkla, Vefsna and Altaelva Rivers, and tendencies towards decreases were found in the Numedalslågen River ($p < 0.08$).
- For zinc loads, five out of nine rivers showed statistically significant downward trends and one additional river (the Otra) showed a tendency towards declining trends ($p < 0.07$).
- For lead loads, only two statistically downward trends were found in the Orkla and Vefsna Rivers. In two additional rivers, the Glomma and the Numedalslågen, the levels of lead declined, but this was not statistically significant ($0.05 < p < 0.07$).
- For cadmium and nickel loads, there was a tendency towards decreases in a majority of the rivers (Table 6). However, the trends noted for this element must be interpreted with caution, because many values were at or below the limit of detection (LOD), and there were also changes in the LOD values over the monitoring period.

In terms of direct discharges of metals, there were relatively few significant trends, and some of those trends were observed in rivers where the direct discharges constituted less than 1% of the total riverine loads and were therefore less relevant. Some apparent trends were discarded upon closer scrutiny of the data, for example when a high proportion of the data had been interpolated or extrapolated. The metal loads from WWTPs were generally very low compared to the total riverine loads. In terms of industrial effluents, declining trends could be detected for cadmium and copper loads to the Otra River, lead loads to the Orkla River, and nickel loads to the Glomma, Skienselva, Otra and Orre Rivers.

In general, the data on metal trends in other Norwegian rivers are limited, but long-term series of heavy metal concentrations in precipitation are available for three stations in southern Norway and one in the northern part of the country (Aas *et al.* 2012). Levels of lead have declined over the last 20–30 years at all the stations in southern Norway, and the same pattern is apparent for cadmium and to a lesser extent also for zinc. Considering all heavy metals together, the decrease was more pronounced in the 1980s than in later years.

An example of trends in metals: the Orkla River

The Orkla River drains into the Norwegian Sea, and it has relatively high area-specific loads of copper, zinc and cadmium (Table 3). The sources of metals in the catchment area include two mining industries currently in operation, as well as probable seepage from old mining activities, but there is no available information on the latter that can be used in trend analyses. Of the two mining companies in

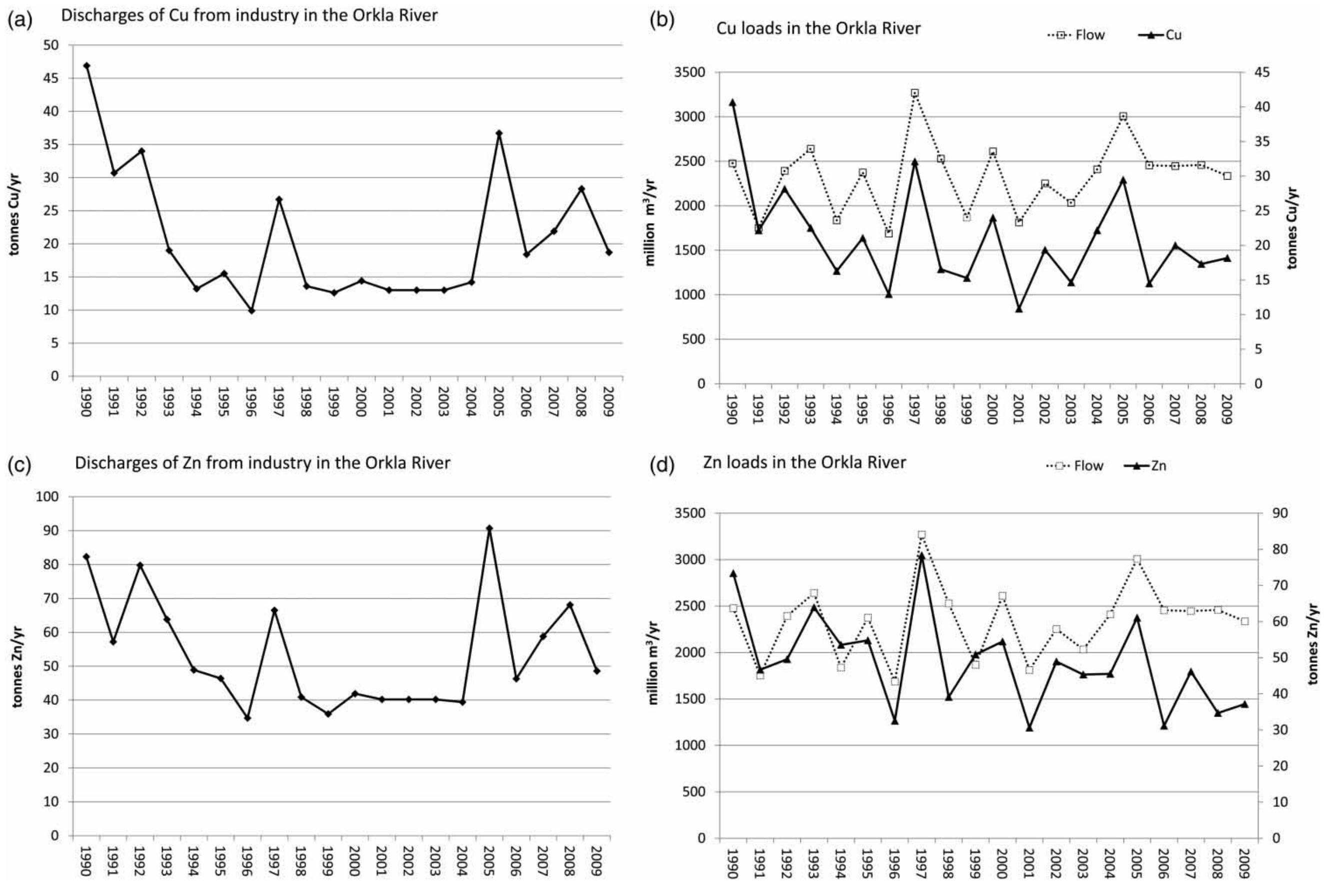

Figure 3 | Data on the Orkla catchment showing discharges of copper (a) and zinc (c) from industries and riverine loads of copper (b) and zinc (d), in tonnes/yr. Water flow (mill m^3/yr) is also indicated in the charts illustrating riverine loads.

operation, the one with the smallest discharges is located about 80 km from the sampling site and the one with the highest discharges is located about 10 km upstream of the sampling site, as the crow flies.

A significant decrease was noted in the riverine loads for all the metals investigated (Table 6), but only one downward trend in industrial discharges was found, and this was for lead, a metal for which industrial discharges only constituted 0.3% of the load in this river. However, a certain degree of correlation was observed in comparisons of riverine loads and industrial discharges of copper and zinc (Figure 3), both of which comprised around 150% of the riverine loads. For copper, the coefficient of determination (R^2) when comparing riverine loads and industrial effluents was 0.7. For zinc, the corresponding coefficient was low ($R^2 = 0.2$), but several peaks of riverine loads nonetheless corresponded with industrial discharges (e.g., in 1990, 1997 and 2005).

Uncertainty

As mentioned above, caution should be observed when analysing trends in long-term datasets on water quality, because a number of factors can add to the uncertainty. Accordingly, even though the dataset evaluated here was subjected to thorough inspection, there are still many weaknesses that should be considered.

One of the most important uncertainties is related to sampling frequency. In a previous study, Skarbøvik *et al.* (2012) used daily data on SPM from 5 years (2001–2005) in one of the nine rivers (i.e., the Numedalslågen) to assess the effects of sampling frequency on annual loads of particles. The results showed that the loads were seriously underestimated when using monthly datasets, as compared to both weekly sampling and biweekly sampling with additional flood samples. In an investigation of the Garonne

River, Coynel *et al.* (2004) concluded that in order to achieve an error rate of less than 20% in estimates of annual suspended sediment loads, it would be necessary to conduct sampling every 3rd day, and the most likely result of reduced sampling frequency would be underestimation of loads. Whereas suspended sediments are notorious for their variability, it should be noted that there are also other substances that can exhibit extensive variability in larger rivers. Moatar & Meybeck (2005) studied the Loire River in France and found that it would be necessary to use the following sampling frequencies to obtain 10% precision in the annual loads of both particulate and dissolved matter: every 10th day for TP, every 5th day for particulate phosphorus, every 15th day for nitrate, and every 10th day for orthophosphate.

On the other hand, a major strength of the dataset used in the present study is the length of the time series. Monthly samples over 20 years gave a total of 240 samples per river, which must be regarded as a good basis for statistical testing of long-term monotonic trends. In addition, the fact that we have previously performed all the statistical tests on the concentration data from the rivers (Skarbøvik *et al.* 2009), and the results of those tests confirmed the trends seen in this study on the loads, further strengthens the predictive power of the data analysis.

CONCLUSIONS

In this study of nine Norwegian rivers, geographically distributed from the River Alta in the north to the River Glomma in the south, statistically significant downward trends in loads of nutrients were detected in 17 of 45 tests (nine rivers and five compounds) for the period 1990–2009. Furthermore, significant downward trends in riverine loads of metals (copper, zinc, cadmium, lead and nickel) were detected in 23 of 45 tests. There was only one significant increasing trend in the riverine loads of nutrients (TN loads in one river) and no upwards trends in the riverine loads of metals. Efforts to explain the trends in the riverine loads in relation to point sources were partly successful, inasmuch as relatively good agreement between trends in some riverine loads and direct discharges of pollutants were found. Even after taking potential sources of errors into consideration, these results indicate that mitigation measures that have been implemented since 1990 to reduce pressures from point sources have indeed had an impact on the quality of water in the rivers. Obviously, there are also other changes in pressures from both land-based and airborne sources that have played a role in the observed riverine trends. Despite the evident decline in pollutant loads, the levels of these substances still remain high in some of the rivers investigated, in particular the Orre River. This demonstrates that additional measures must be taken to achieve acceptable conditions.

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