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TEOTIL METALS: DEVELOPMENT REPORT



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Summary - sammendrag

The model TEOTIL has been widely used in Norway for simulating nutrient fluxes at national scale. This report describes extending TEOTIL to incorporate metals. A review of metal behaviours is presented and the requirements and constraints for extending TEOTIL described. Key datasets for the metals of interest are identified and statistical analyses undertaken to explore relevant relationships. A new model is constructed using metal concentrations from the 2019 "1000 Lakes" survey to represent spatial variability, and regionally aggregated data from Elveovervåkingsprogrammet to infer temporal patterns. Performance of the model is evaluated by comparison to observed datasets and shown to be better than the current method used to estimate metal fluxes for annual reporting to the OSPAR Commission.

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Front page photo

View towards the Norwegian Border from the nickel smelter at Nikel, Russia. The smelter is a major source of copper and nickel pollution in Pasvikelva. Image © [Ninara](#) under [CC2.0 Generic](#) licence.

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Summary

The model TEOTIL has been widely used in Norway for simulating nutrient fluxes at national scale. This report describes extending TEOTIL to incorporate metals. A review of metal behaviours is presented and the requirements and constraints for extending TEOTIL described. Key datasets for the metals of interest are identified and statistical analyses undertaken to explore relevant relationships. A new model is constructed using metal concentrations from the 2019 “1000 Lakes” survey to represent spatial variability, and regionally aggregated data from Elveovervåkingsprogrammet to infer temporal patterns. Performance of the model is evaluated by comparison to observed datasets and shown to be better than the current method used to estimate metal fluxes for annual reporting to the OSPAR Commission.

Sammendrag

Modellen TEOTIL har blitt mye brukt i Norge for å simulere transport av næringsstoffer på nasjonal skala. Denne rapporten vurderer å utvide TEOTIL til å inkludere metaller. En gjennomgang av viktige datasett presenteres og forutsetningene og begrensningene for utvidelse av TEOTIL er beskrevet. Nøkkeldata for de aktuelle metallene er identifisert og en statistisk analyse er gjennomført for å utforske relevante sammenhenger. En ny modell er laget ved hjelp av metalkonsentrasjoner fra «1000-sjøers undersøkelsen» i 2019 for å representere romlig variasjon, samt regionalt aggregerte data fra Elveovervåkingsprogrammet for å utlede tidsmønstre. Modellens egnethet har blitt evaluert ved å sammenligne predikerte/modellerte data med observerte datasett. Den nye modellen viste seg å være bedre enn den gjeldende metoden som brukes til å estimere metalltilførsler for årlig rapportering til OSPAR-kommisjonen.

1. Introduction

As part of a commitment to protect the marine environment of the Northeast Atlantic, Norway estimates annual terrestrial loads for selected pollutants and submits them to the OSPAR Commission (<https://www.ospar.org/>). Riverine fluxes are estimated using long-term monitoring datasets where possible, supplemented by model simulations in regions where direct observations are not available. Since the mid-1990s, the model TEOTIL ("TEOretiske TILførselsberegninger"; Tjomsland *et al.*, 2010) has been used to simulate fluxes of nitrogen (N) and phosphorus (P) in unmonitored areas. This report describes work to extend TEOTIL to include metals: arsenic (As; strictly a metalloid), lead (Pb), cadmium (Cd), copper (Cu), chromium (Cr), mercury (Hg), nickel (Ni) and zinc (Zn).

2. Review of metal behaviours

According to Skjelkvåle *et al.* (2006), the main (non-point) sources of metals influencing surface water concentrations in Norway are:

- **Geological.** Metals are derived directly from the weathering of bedrock and surficial deposits
- **Atmospheric.** Metals may be transported from elsewhere in Europe as "long-range, transboundary air pollution" (LRTAP), or from local sources within or near Norwegian borders. LRTAP is particularly important in Southern Norway

The relative importance of these sources is different for different metals. Historically, As, Cd, Hg, Pb and Zn have been primarily determined by LRTAP, whereas Cr, Cu and Ni have been mostly influenced by geology (except close to the Russian border, where Cu and Ni are strongly affected by local industry). Furthermore, the mobility of metals derived from these sources is influenced by the following factors:

- **Water chemistry.** Some metals are more easily mobilised at lower pH, and many metals form complexes with organic matter, leading to higher concentrations in areas with high total organic carbon (TOC; Groenenberg *et al.*, 2015)
- **Hydrology.** Short residence times and/or high water fluxes mean reduced contact time between water and soil, leading to lower metal concentrations. Sedimentation rates are also important, as a high proportion of the total metal load is transported in solid phases (Vink and Peters, 2003)

Key factors believed to determine surface water concentrations for the eight metals of interest are summarised in *Table 1*.

Element	Key factors in Norway
As	Strongly influenced by LRTAP
Cd	Strongly influenced by LRTAP. Concentrations higher in acidified regions
Cu	Primarily determined by bedrock geology, except close to the Russian border, where local industrial sources dominate. LRTAP not a dominant factor. Some old mining regions exhibit high concentrations
Cr	Concentrations primarily determined by bedrock geology and TOC. Not strongly influenced by atmospheric deposition
Hg	Primarily influenced by LRTAP. Strong affinity with organic matter
Ni	Primarily determined by bedrock geology, except close to the Russian border, where local industrial sources dominate. LRTAP not a dominant factor
Pb	Strongly influenced by LRTAP. Strong affinity with organic matter. Concentrations higher in acidified regions
Zn	Strongly influenced by LRTAP, but with local geological effects too. Some clear point sources in mining areas. Concentrations higher in acidified regions

Table 1: Key factors influencing metal concentrations in Norwegian surface waters. LRTAP, Long-range, transboundary air pollution. Summarised from Skjelkvåle *et al.* (2006)

3. TEOTIL

3.1 Development history

The original TEOTIL model was created in 1992 (Ibrekk and Tjomslund, 1992). In 2001, it was recoded into Visual Basic and, in 2017, the code was refactored again into Python – a language that is increasingly popular for scientific applications. The Python implementation of the model currently lacks a graphical user interface, but it is more flexible and efficient than the previous version, making it feasible to extend the model to other parameters while also reducing runtimes.

3.2 Nitrogen and phosphorus

The purpose of the existing TEOTIL model is to integrate nutrient inputs from a variety of sources and accumulate them downstream. Key datasets for simulating nitrogen and phosphorus include: diffuse background inputs from forests & uplands; diffuse human inputs based on population density & agricultural activities; and point discharges from industry, sewage treatment & aquaculture. The model uses the “export coefficient” concept to estimate diffuse fluxes, and it also incorporates a simple representation of catchment-level nutrient cycling and retention. For national scale applications, the model typically uses the “regime” catchment network, which comprises roughly 20 000 sub-catchments across Norway, ranging in size between 0.01 and 10 000 km².

3.3 Model structure and constraints

Extending TEOTIL to incorporate additional parameters requires understanding the model's structural framework, which places constraints upon further development. To simulate fluxes for any given parameter, X , the model must perform the following calculations:

1. **Sum “local” inputs of X for each sub-catchment.** In the case of N and P, diffuse inputs are estimated by applying export coefficients to datasets of land use and agricultural activities in each catchment. These are then added to the sum of all point discharges located within the catchment boundary to give the total local input of X to catchment i , denoted L_i^X
2. **Accumulate loads downstream.** Each sub-catchment, i , is assigned a parameter-specific retention factor, α_i^X , between 0 and 1. This indicates the proportion of the load of X entering the catchment that is retained due to internal cycling.

The model begins by identifying the uppermost sub-catchment(s) in each river system. For these headwater catchments, the amount of X transmitted to the next catchment downstream is

$$T_i^X = (1 - \alpha_i^X)L_i^X \quad (1)$$

For the next catchment downstream, j , the total input of X is equal to the sum of local sources, L_j^X , plus any inputs from catchments upstream (numbered 1 to n in equation [2], below). The output from catchment j is therefore

$$T_j^X = (1 - \alpha_j^X) \left(L_j^X + \sum_{p=1}^n T_p^X \right) \quad (2)$$

These calculations are illustrated schematically in *Fig. 1*. Adding additional chemical species to TEOTIL therefore requires estimating two parameters per species for each sub-catchment: (i) the **total local inputs**, L_i^X and (ii) the **retention factors**, α_i^X .

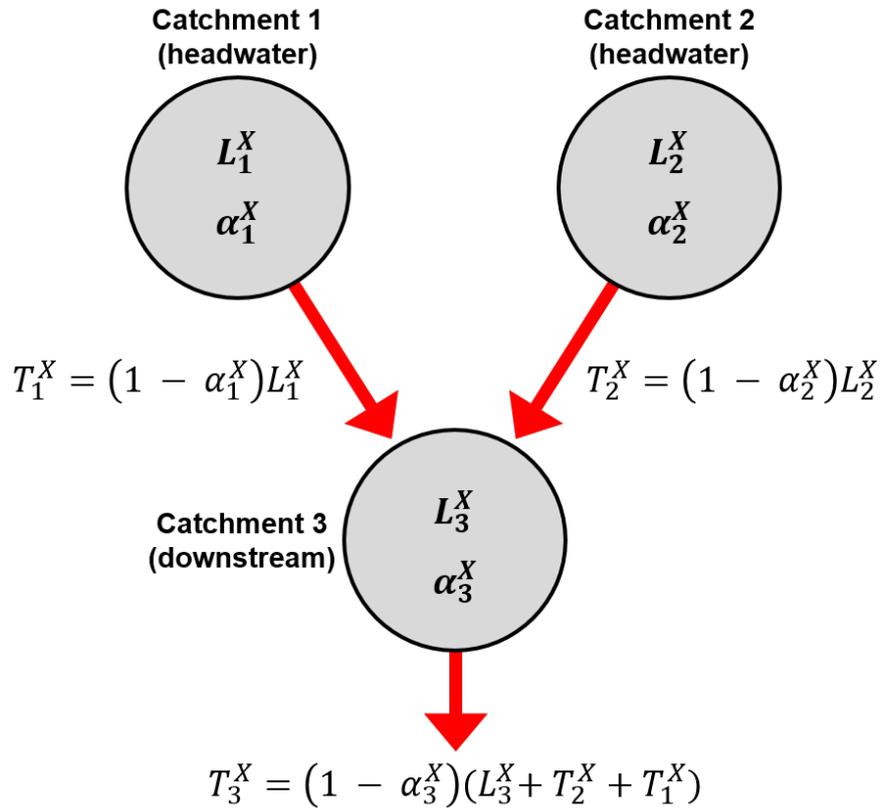


Fig. 1: Schematic illustration of the flux accumulation procedure in TEOTIL. L_i^X , sum of local inputs of parameter X to sub-catchment i ; α_i^X , retention factor for parameter X in sub-catchment i ; T_i^X , load of X transmitted downstream from sub-catchment i

4. TEOTIL Metals

The methodology currently used in Norway to estimate metal fluxes for OSPAR reporting is simplistic: the inputs from all point discharges in each main catchment are added together, and this total is assumed to reach the sea. This approach ignores key processes such as internal cycling and catchment-level retention, as well as failing to account for diffuse inputs, which are significant.

The eight metals of interest in this report exhibit a range of behaviours in the natural environment, some of which are quite different to N and P. For this reason, development of TEOTIL Metals requires identifying key datasets and relationships capable of summarising sub-catchment-level metal inputs and retention.

4.1 Estimating local inputs

Estimating annual local inputs of metals to each regime catchment requires information describing both point and diffuse sources. Data for point sources can be obtained from the Norwegian Environment Agency's discharge licensing database (as for nutrients), but diffuse inputs must be inferred from other datasets. Based on the discussion in *Section 2*, key datasets

are those describing geology, atmospheric deposition, water chemistry and hydrology. To be of practical use, candidate datasets must meet the following criteria:

- Be available for most or all of the metals of interest
- Be available at national scale
- Be updated with a frequency that reflects the variable's natural rate of change

4.1.1 Geology

The most comprehensive geological datasets available at national scale come from the Geochemical Atlas compiled by the Norwegian Geological Survey (NGU; Ottesen *et al.*, 2000). Raw point data for six of the eight metals of interest (As, Cr, Cu, Ni, Pb and Zn) were supplied by Tor Erik Finne at NGU. Data were collected during 1984-85 and represent metal concentrations measured in flood sediments at 690 locations across Norway. For the purposes of this report, they are assumed to be representative of concentrations in locally eroded material transported by rivers. It is further assumed that these concentrations remain constant over time, so the local geological inputs to each catchment are proportional to the amount of weathering and erosion, which is linked to hydrology (*Section 4.1.3*).

The raw point data for geochemistry has been spatially interpolated onto a common grid using Inverse Distance Weighting (IDW). An example for zinc is shown in *Fig. 2*.

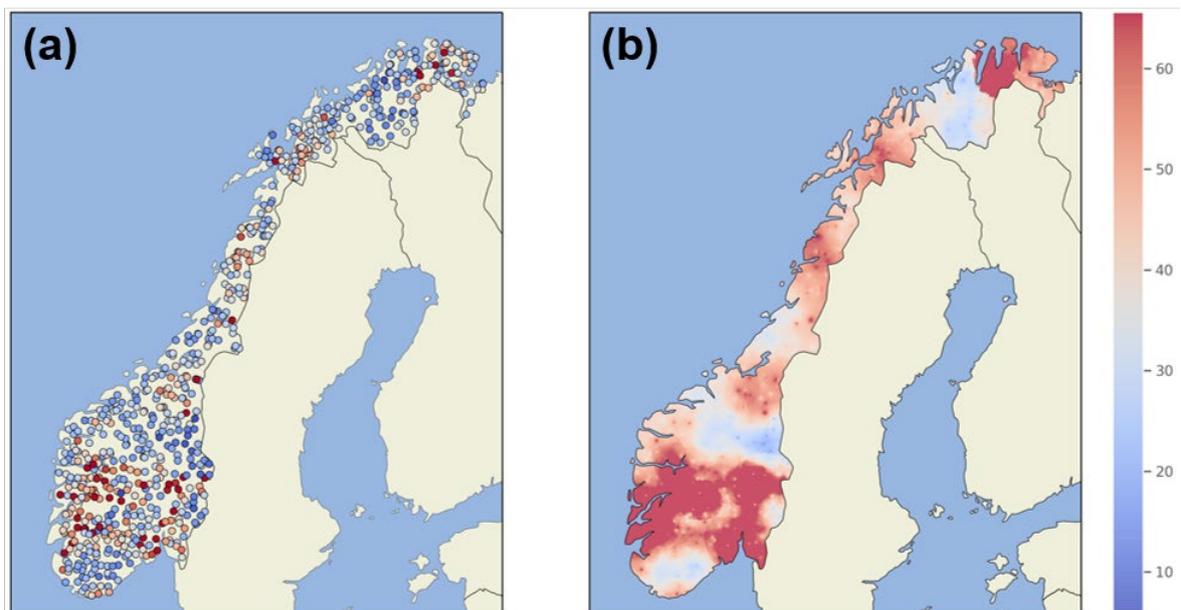


Fig. 2: Geochemical data from flood sediments for zinc (mg/kg). (a) Raw point data from NGU; (b) Spatially interpolated using Inverse Distance Weighting

4.1.2 Atmospheric deposition

Estimates of atmospheric deposition based on dynamic modelling are available from EMEP (<http://www.emep.int/>), but only for Cd, Hg and Pb. For the other metals of interest (As, Cr, Cu, Ni and Zn), the best available data come from measurements of concentrations in mosses, which aggregate deposition inputs over periods of about a year (Skjelkvåle *et al.*, 2006).

Model results from EMEP are produced every year, while the moss surveys have been conducted every five years since 1985. Lab methods and limits of detection have improved substantially in the more recent moss surveys, but the number of sites sampled has also reduced. The dataset from 2005 seems to provide the most complete picture in terms of spatial coverage and measurement accuracy.

Raw point datasets from the moss surveys were made available by Eiliv Steinnes at NTNU. The same IDW interpolation procedure used for geochemistry was applied to translate values onto a common grid (Fig. 3).

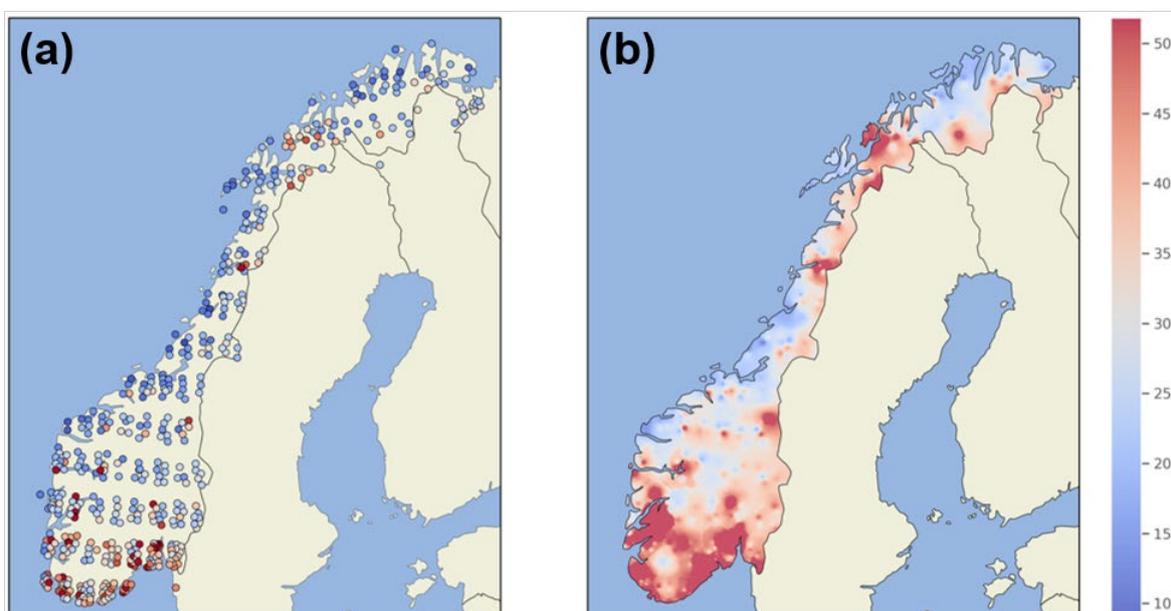


Fig. 3: Zinc concentrations measured in mosses in 1990 (mg/kg). (a) Raw point data; (b) Spatially interpolated using Inverse Distance Weighting

4.1.3 Water chemistry

National surveys of Norwegian water chemistry took place in 1986, 1995 and 2019. The 2019 “1000 Lakes” survey considered all the metals of interest in this report, while the 1995 survey included all except Hg. pH and TOC were also measured, making it possible to explore effects such as acidification and organic matter complexation.

Analytical methods used in the 2019 survey have improved considerably since 1995; a key limitation of the earlier survey datasets is that measured concentrations are frequently low compared to laboratory limits of detection (LODs). Table 2 shows the proportion of samples from the 1995 survey that were at or below the LOD for seven metals, plus TOC.

Element	Number of samples	Number \leq LOD	Percent \leq LOD
As	504	328	65
Cd	970	610	63
Cr	970	648	67
Cu	971	261	27
Ni	971	180	19
Pb	969	37	4
Zn	970	87	9
TOC	1007	81	8

Table 2: Proportion of limit of detection (LOD) values from the 1995 lake chemistry survey. TOC, total organic carbon

To develop a robust implementation of TEOTIL Metals, it is necessary to estimate inputs of metals to each regine catchment. Obvious ways of doing this are either (i) deriving statistical relationships linking geological and atmospheric datasets to water chemistry, or (ii) using the water chemistry datasets directly to infer catchment concentrations. However, for either approach, deriving meaningful quantitative relationships is difficult when a significant proportion of the data are censored (and this is especially true when more than 50% of the data are censored, because even simple descriptive statistics – like the median – become difficult to estimate). *Table 2* makes it clear that, from the national survey data available, only the 2019 survey will be useful in the context of developing TEOTIL Metals.

The best available datasets describing metal concentrations through time are those collected as part of Elveovervåkingsprogrammet (e.g. Gundersen *et al.*, 2019). Since 1990, 155 major Norwegian rivers have been monitored within this programme, and since 2017 a subset of 20 has been sampled and analysed four times per year for heavy metals.

Raw point datasets from the 2019 1000 Lakes survey were interpolated using the same procedure as for mosses and geochemistry. The interpolated grids for each metal are shown in *Fig. 4*.

4.1.4 Hydrology

As part of Elveovervåkingsprogrammet, national scale hydrology data are supplied annually by the Norwegian Water Resources and Energy Directorate (NVE) and incorporated into the existing TEOTIL model for N and P. For TEOTIL Metals, hydrology plays an important role in determining weathering and mobilisation, and in making it possible to translate metal concentrations in the geochemical, moss or water chemistry datasets into riverine fluxes. Hydrological parameters such as specific runoff and catchment residence time are also required to derive estimates of retention (*Section 4.2*).

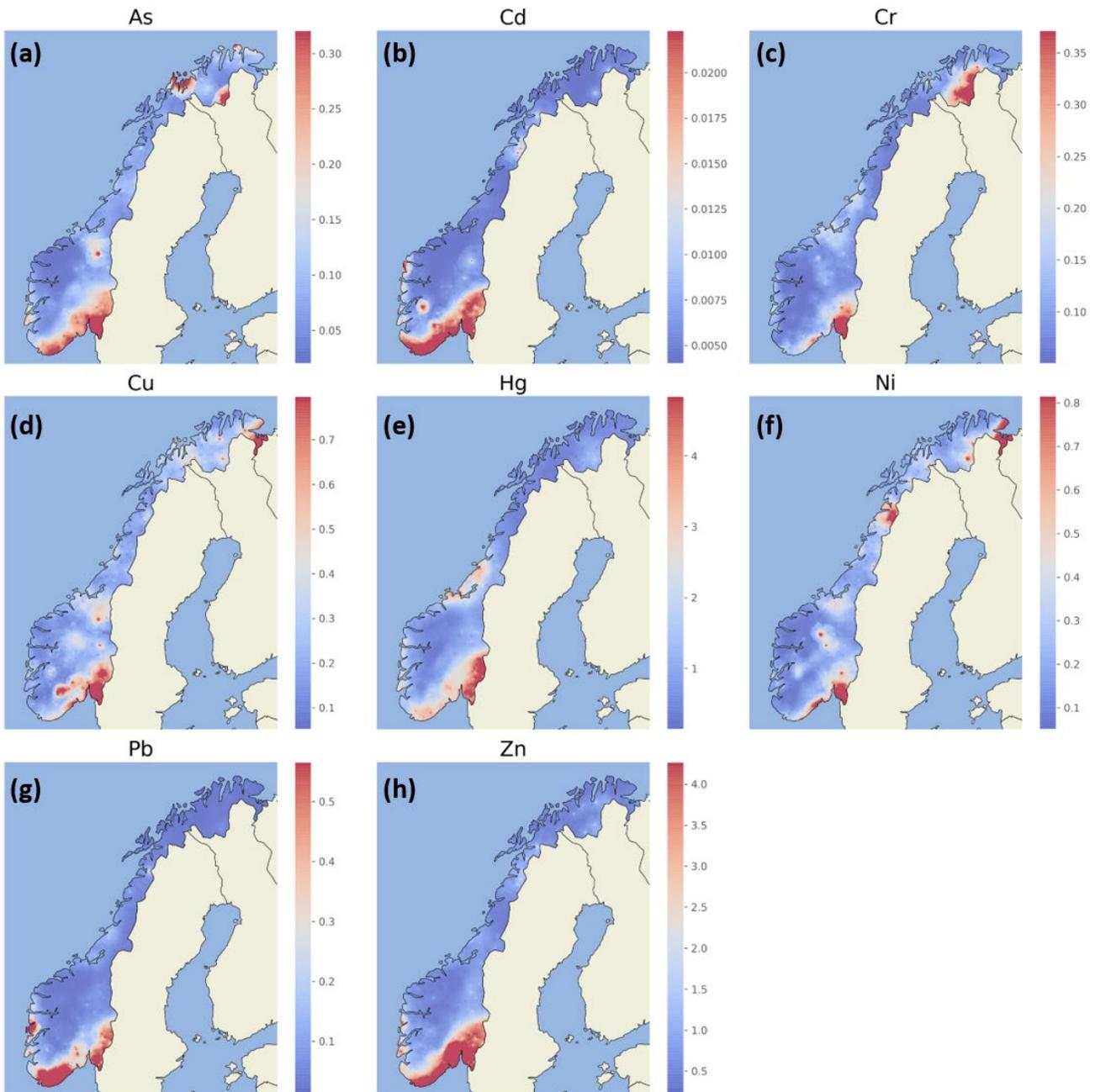


Fig. 4: Water chemistry from the 2019 1000 Lakes survey, spatially interpolated using Inverse Distance Weighting. All units are µg/l except Hg, which is ng/l.

4.2 Estimating retention

4.2.1 Theory

The theoretical principles for estimating retention in lakes are described by Vollenweider (1975). The mass balance for a non-volatile substance in a lake or reservoir is given by

$$\frac{dM}{dt} = m_i - m_o - S \quad (3)$$

where M is the mass of substance in the lake; m_i is the sum of all inflow fluxes; m_o is the flux from the lake outflow; and S represents all internal processes that remove the substance from the lake. S is assumed to be proportional to M , such that the greater the amount of substance in the lake, the faster it is removed by internal processing

$$S = \sigma M \quad (4)$$

Rewriting the input and output fluxes as the product of discharge and concentration, and combining equations [3] and [4] gives

$$\frac{dM}{dt} = q_i[m_i] - q_o[m_o] - \sigma M \quad (5)$$

where q_i and q_o are the inflow and outflow discharges, respectively, and $[m_i]$ and $[m_o]$ are the inflow and outflow concentrations. For a perfectly mixed lake, $[m_o] = M/V$, where V is the lake volume. Assuming steady state, $\frac{dM}{dt} = 0$ and V is constant, which implies $q_i = q_o = q$

$$q[m_i] - q[m_o] - \sigma V[m_o] = 0 \quad (6)$$

Defining the water renewal rate as $\rho = q/V$ (i.e. the reciprocal of the residence time) and rearranging gives

$$\frac{[m_o]}{[m_i]} = \frac{\rho}{\sigma + \rho} \quad (7)$$

Because q is constant, the ratio of concentrations, $[m_o]/[m_i]$, is also the ratio of fluxes, m_o/m_i . The retention factor, R , is defined as

$$R = 1 - \frac{m_o}{m_i} = 1 - \frac{\rho}{\sigma + \rho} = \frac{\sigma}{\sigma + \rho} \quad (8)$$

In this formulation, the water renewal rate, ρ , is hydrologically determined, and σ is a parameter-specific constant representing the rate of internal cycling and removal. Although equation [8] has a sound scientific basis, several authors (e.g. Larsen and Mercier, 1976; Vink and Peters, 2003) have found empirically that a modified equation of the form

$$R = \frac{\sigma}{\sigma + \rho^n} \quad (9)$$

provides a better fit to observations. Note, however, that there is no mechanistic theory behind equation [9]; unless $n = 1$, the units in the denominator are not physically compatible.

4.2.2 Datasets

For some metals, the literature provides suggested values for the parameters σ and n in equation [9] (e.g. Vink and Peters, 2003). However, these values depend on catchment characteristics and are therefore specific to the hydrological, geochemical and climatic conditions under which they were derived. Studies focusing on the Norwegian context are rare, so for TEOTIL Metals it will likely be necessary to estimate relationships directly from Norwegian data.

An alternative is to use existing water chemistry data and/or statistical relationships to estimate surface water metal concentrations directly for each regime catchment. This approach implicitly accounts for retention by estimating $[m_o]$, instead of starting with an estimate for m_i and assuming steady state. In this case, retention for all catchments in the model can be assumed to be zero, and fluxes for each substance calculated simply as the product of outflow discharge and concentration, $q_o[m_o]$.

4.3 Statistical relationships

TEOTIL Metals is underpinned by the idea that significant relationships exist between key sources/drivers of metal inputs (geology, atmospheric deposition, hydrology and point discharges) and the concentrations observed in surface waters. To test this, statistical models were developed linking the 2019 national water chemistry dataset to a range of possible explanatory variables (*Section 7*). The analysis considered only the metals As, Cr, Cu, Ni, Pb and Zn; Cd and Hg were not considered, as they are missing from one or more of the required input datasets. The following variables were incorporated:

- **Water chemistry.** Concentrations of metals [$\mu\text{g/l}$], plus TOC [mgC/l] and pH [-] were extracted from the database for the 2019 lake survey. Catchment boundaries representing the area upstream of each sampling location were also calculated
- **Geology.** The mean metal concentration [mg/kg] measured in river sediments was calculated for each catchment by averaging IDW interpolated values within each catchment boundary (*Fig. 2b*)
- **Atmospheric deposition.** The IDW interpolated moss dataset from 2005 was averaged over each catchment to obtain values proportional to deposition inputs [mg/kg]. The 2005 dataset was chosen because later moss surveys contain fewer sampling points (see *Section 4.1.2*)

4.3.1 Exploratory data analysis

Scatter plots with lowess smoothing were used to summarise relationships between variables and to highlight general trends (*Section 7*). Although there is a lot of noise, the smoothing picks out plausible relationships: most metal concentrations are higher in organic-rich or acidic

conditions, and there are clear positive relationships with deposition (mosses) and geology (river sediments). Furthermore, in some cases the relationship with pH approximates exponential decay, which can be linearised by converting from pH units to the concentration of H^+ ions

$$[H^+] = 10^{-pH} \quad (10)$$

When this is done, relationships on the scatter plots are all approximately linear, suggesting multiple linear regression as an appropriate tool for further analysis.

4.3.2 Multiple linear regression

As an initial test of relevant relationships, a multiple linear regression model of the following form was fitted for each metal, X

$$C_i^X = \beta_0^X + \beta_1^X M_i^X + \beta_2^X G_i^X + \beta_3^X C_i^{TOC} + \beta_4^X C_i^{H^+} \quad (11)$$

Where C_i^X is the concentration of metal X in lake i ; M_i^X is the average concentration of X in mosses in catchment i ; G_i^X is the average concentration of X in river sediments in catchment i ; C_i^{TOC} is the concentration of TOC in lake i ; $C_i^{H^+}$ is the concentration of H^+ ions in lake i (estimated using equation [10]) and the β_j^X are the regression parameters for each metal.

All variables were standardised prior to regression and a “best subsets” approach applied to find the model with the lowest Akaike information criterion (AIC) from all possible combinations of the four explanatory variables. Note that, in general, best subsets is prone to overfitting the data (regularised techniques such as lasso or ridge regression are usually preferable), but it does provide a useful comparison to the “full” model, which is of interest here.

Fig. 5a shows the fitted versus observed values for Pb for the “best” model identified. This model explains roughly 70% of the data variance ($R^2 = 0.70$) and the residuals (*Fig. 5b*) are approximately normally distributed. All variables are highly significant ($p \ll 0.05$) and the model coefficients are plausible, indicating that Pb has positive relationships with both geological and atmospheric inputs, as well as with concentrations of organic matter and H^+ . Looking at the standardised coefficients (*Table 3*), the concentration of H^+ has the biggest effect size, followed by concentration of TOC, geochemistry and, finally, mosses (i.e. deposition). These results are broadly consistent with the literature survey in *Section 2*, which states that Pb concentrations in Norway are affected by LRTAP, but strongly modified by pH and complexation with organic matter (*Table 1*).

Variable	Slope	p-value
Mosses	0.0267	0
Geochemistry	0.0674	0
TOC	0.0770	0
H^+	0.1433	0

Table 3: Standardised regression coefficients and p-values for the regression model shown in *Fig. 5*

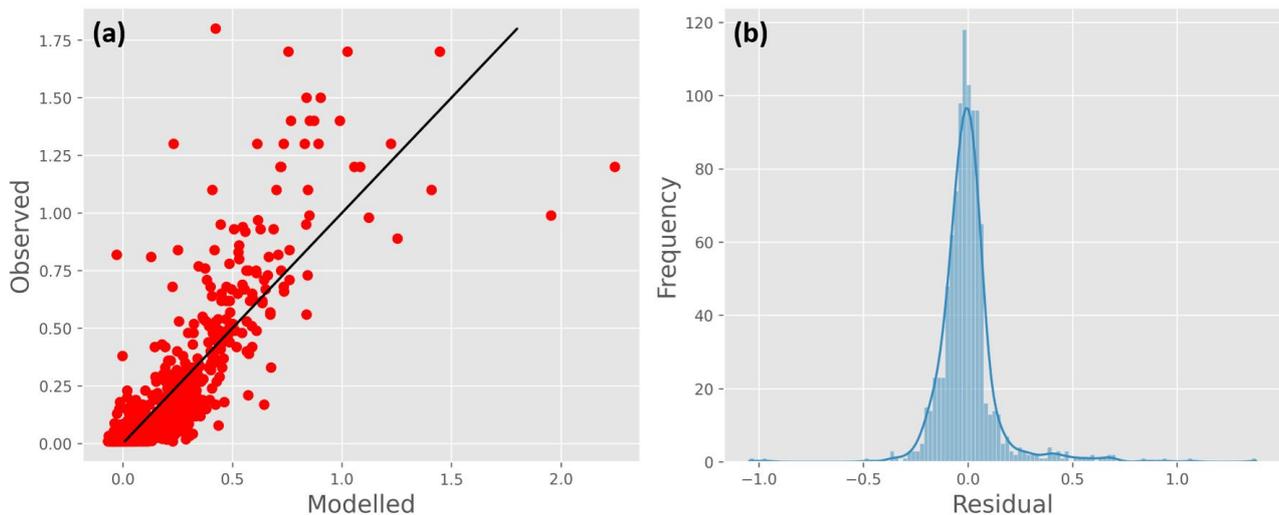


Fig. 5: Multiple linear regression results for Pb. (a) Observed versus modelled values (note the black line is the 1:1 relationship, not the line of best fit); (b) distribution of residuals

Detailed regression results for all metals can be found in the Supplementary Information (Section 7, [notebook 01](#)). Although all relationships are highly statistically significant, and most are physically plausible, the explanatory power for several of the metals is poor (i.e. the effect size is small/negligible). Furthermore, in most cases, the effects of other water chemistry parameters (especially TOC concentration) are more important than either the moss or geochemistry variables. This is problematic, because TEOTIL enforces a simple conceptual framework that is incapable of representing such “process-based” or “mechanistic” effects¹.

Restricting the statistical models to only consider the moss and geochemistry datasets yields poor relationships and, since we do not have national scale datasets of pH and TOC through time (the “1000 Lakes” surveys are 25 years apart), it is not possible to use these relationships to effectively estimate regime-scale catchment inputs from the moss and geochemistry data. The alternative approach of directly estimating catchment metal concentrations from the 2019 1000 Lakes dataset must therefore be considered in more detail.

4.4 Direct estimates of metal concentrations

Poor statistical relationships (except when incorporating pH and TOC) and incomplete historic datasets make it difficult to use the moss and geochemistry surveys to derive export coefficients and retention factors for TEOTIL2 Metals. However, the 2019 1000 Lakes survey provides a detailed picture of the spatial distribution of metal concentrations in Norwegian freshwaters. To some extent, this dataset can be used to bypass challenges with the moss and geochemistry data: since we already know the concentrations in surface waters, it is not necessary to allow for catchment level retention (i.e. retention can be assumed to be 0; Section 4.2.2); it is also

¹ Similar problems have been documented with the original TEOTIL model when simulating phosphorus: because a significant fraction of the P-load is typically bound to particulates, the model performs poorly when compared to nitrogen (where most of the flux is dissolved).

no longer necessary to derive export coefficients from the “geology” and “deposition” datasets, because direct estimates of concentrations are available instead.

The downside of this approach is that it cannot distinguish between inputs from bedrock weathering versus atmospheric deposition. This is unfortunate, as one of the main benefits of models such as TEOTIL is the ability to perform “source apportionment” i.e. to separate total fluxes into several contributing components. However, in the case of TEOTIL Metals, we have no choice but to assume that geochemical concentrations are constant, because there is only one geochemical survey (from 1984/5). Rather than attempting to separate the bedrock weathering and atmospheric deposition components using poor statistical relationships, it may therefore be better to estimate the combined (weathering + deposition) component directly, and then assume that changes in this component are driven primarily by atmospheric deposition. This approach simultaneously solves the problems of estimating retention factors and export coefficients and, although conceptually different to the way nutrients are handled in TEOTIL, it is similar to how the model estimates catchment-level water fluxes.

4.4.1 2019 concentrations from the “1000 Lakes” survey

Metal concentrations measured during the 2019 1000 Lakes survey are assumed to reflect a combination of point discharges (e.g. from industry) and diffuse inputs from bedrock weathering and atmospheric deposition. To isolate the diffuse component, 947 stations with no upstream point discharges were identified from the 1000 Lakes dataset using point discharge data supplied by the Norwegian Environment Agency. Data from just these lakes was interpolated using inverse distance weighting, and mean concentrations calculated for each metal in each regine catchment using zonal statistics.

A prototype TEOTIL2 Metals model for 2019 was then constructed by combining the estimated “diffuse” inputs with the data on point discharges from the discharge licensing database. This model was used to simulate fluxes for the 20 “main rivers” monitored as part of Elveovervåkingsprogrammet during 2019. *Fig. 6* compares output from this experimental TEOTIL2 model with observed fluxes (left column), and also shows results obtained by simply summing all point discharges, which is the current method used for e.g. OSPAR reporting (right column).

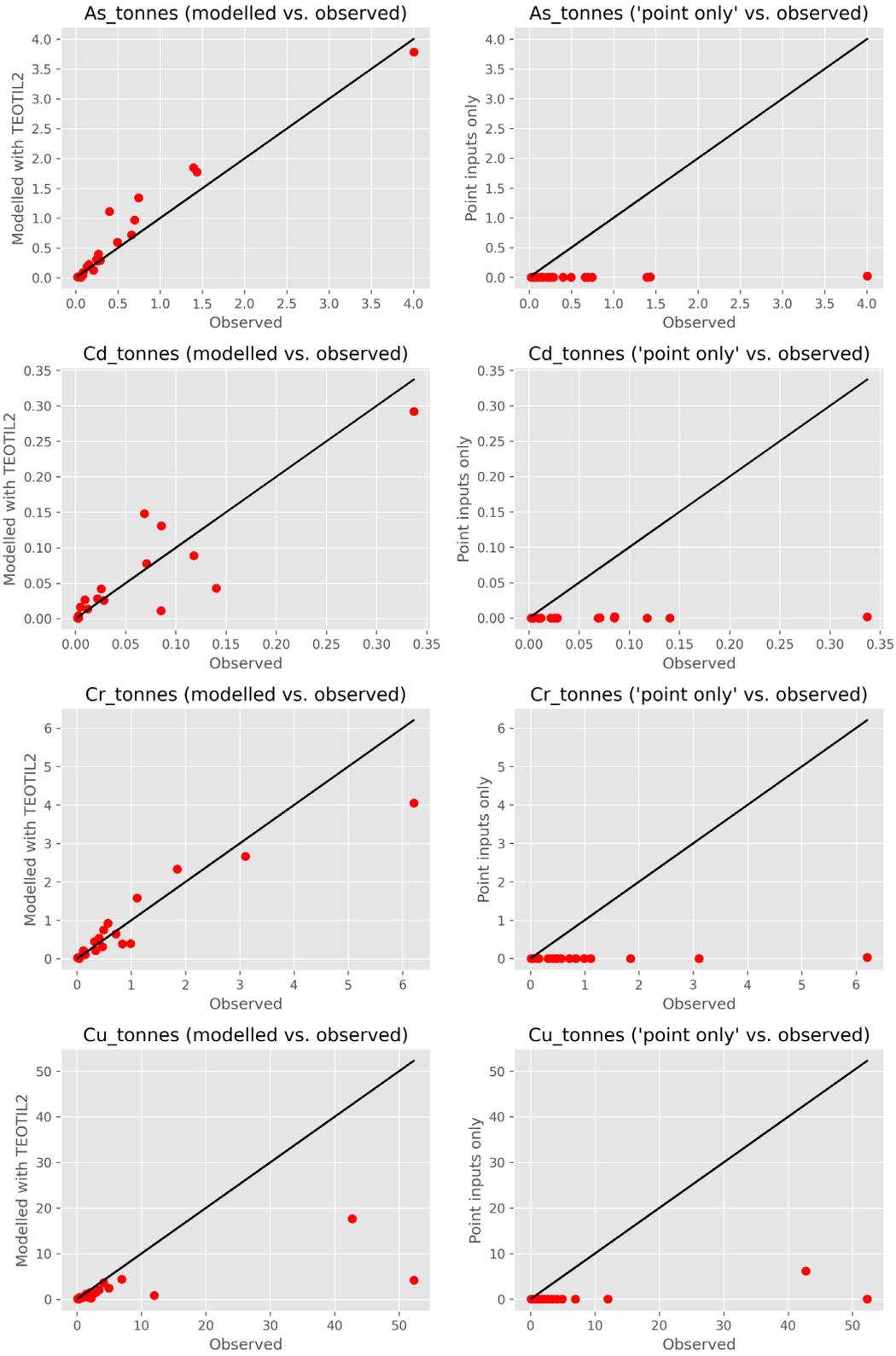


Fig. 6: Continued next page.

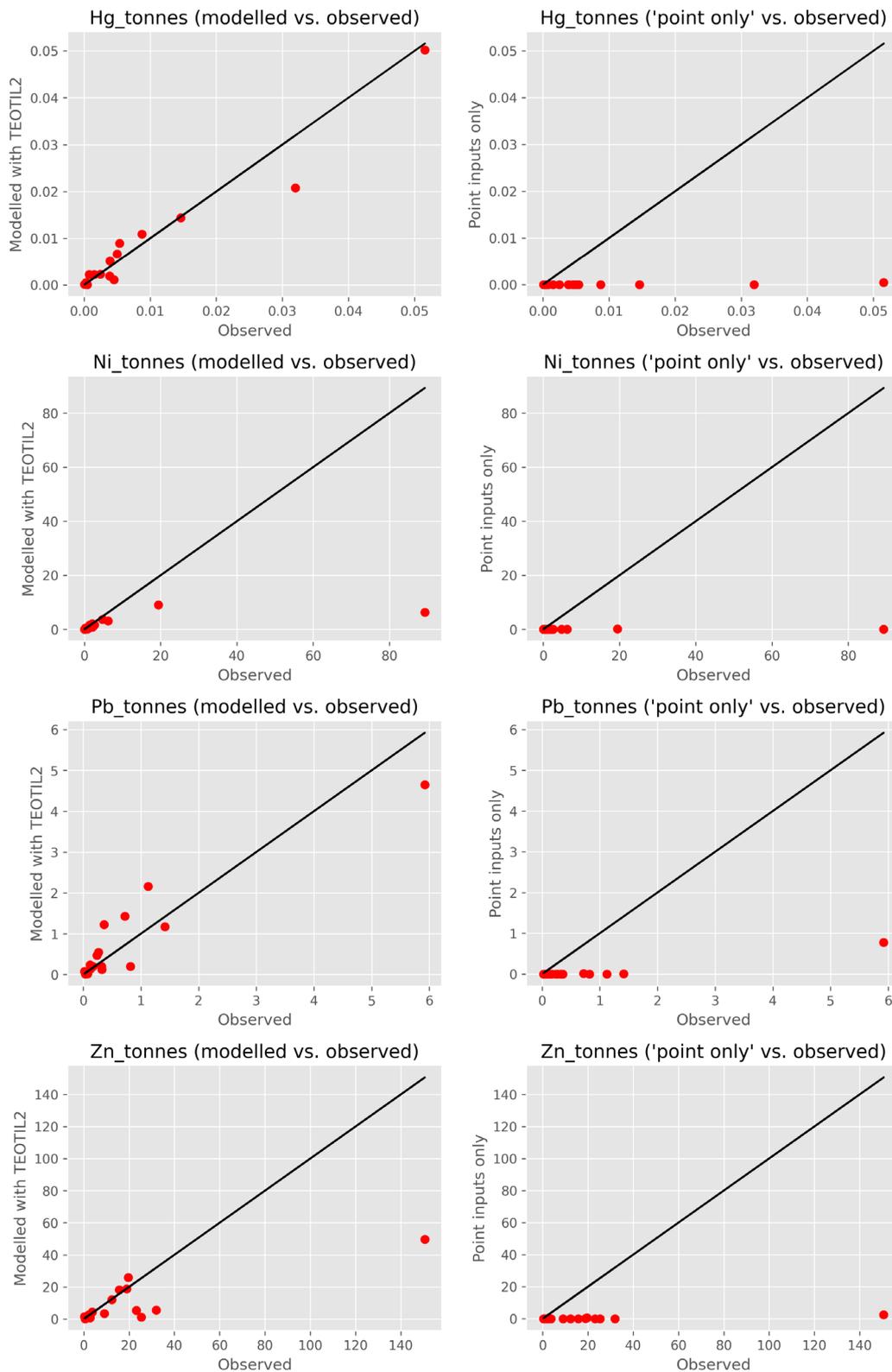


Fig. 6: The left column of plots shows output from TEOTIL2 Metals for 2019; the right column is the sum of point discharges only (i.e. the current “default” method for OSPAR reporting). The x-axis in all plots shows observed fluxes calculated as part of Elveovervåkingsprogrammet; black lines show 1:1 relationships for comparison.

Fig. 7 shows the same data but this time as a set of “target plots”. These provide a convenient way of summarising and comparing the performance of different models: the y-axis shows normalised bias between simulated and observed values; the x-axis is the unbiased, normalised root mean square difference (RMSD); and the distance between any point and the origin is the normalised total RMSD. The solid circle highlights $RMSD = 1$, and any point within this has positively correlated simulated and observed data series and positive Nash-Sutcliffe model efficiency. The dashed circle marks $RMSD = 0.7$, and is chosen because the area within this is approximately equal to the area of the outer “donut”. Better models will plot closer to the centre of the target.

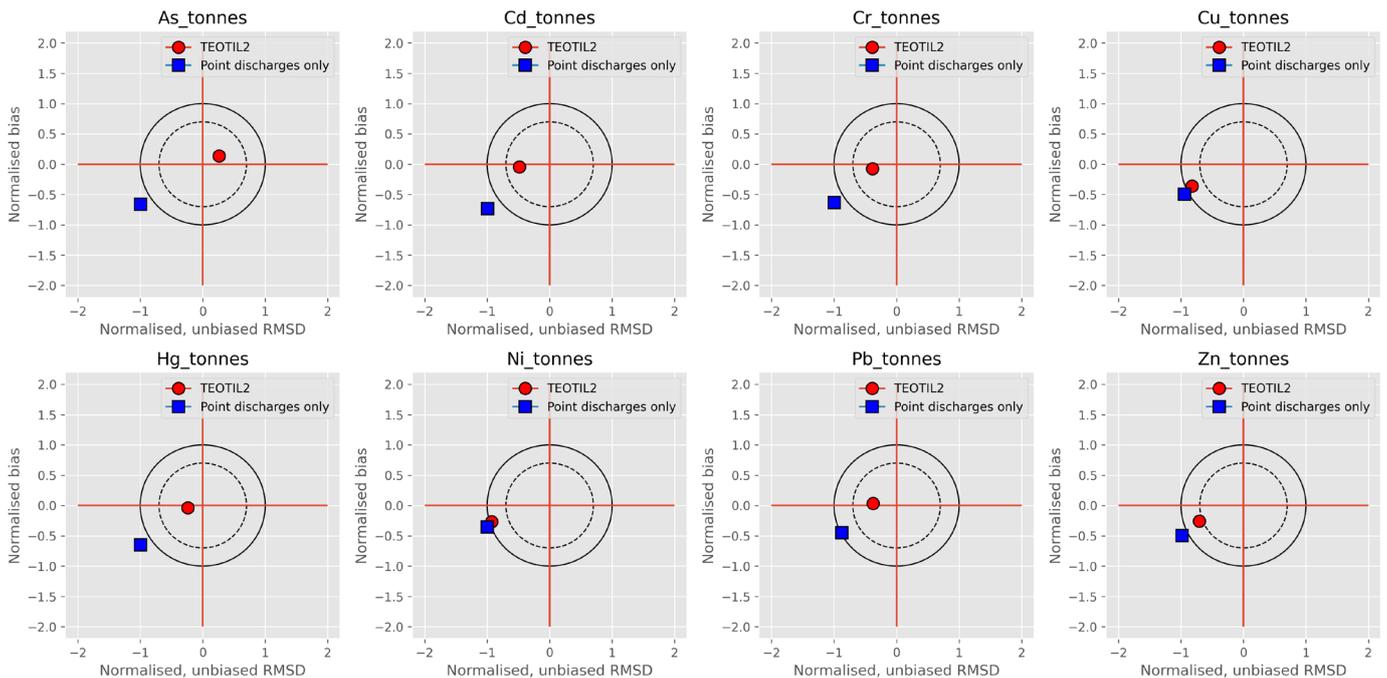


Fig. 7: Target plots comparing the performance of the new model (red circles) to the “default” approach of summing point discharges (blue squares)

It is clear from these plots that TEOTIL2 Metals implemented using the 1000 Lakes dataset is substantially better than the current “default” workflow, at least for 2019: scatter plots for the new model show clear positive correlations for most metals, with slopes that are close to 1 in five out of eight cases. By contrast, using only point discharges dramatically underestimates observed fluxes (which is not surprising). The target plots also clearly illustrate the improved performance of the new model, which has normalised total $RMSD < 1$ for all metals.

Both sets of plots show the new model consistently underestimates metal fluxes for Ni and Cu. These two metals exhibit similar behaviour in the natural environment, with fluxes usually dominated by geological inputs (Table 1). Further investigation reveals that most of the “outlier” points on plots with high observed fluxes of Cu and Ni are actually from a single river, Pasvikelva (regine 246.A5), which is located in the far north of Norway close to the Russian border. The majority of the Pasvikelva catchment is situated in Finland & Russia and there are extensive nickel mining activities in this region, as well as a large nickel smelter just across the Russian border (less than 15 km upstream of the water sampling location; see cover page). Cu-

and Ni-rich runoff from the mines and smelting works are known to pose a significant environmental problem in this area, with discharges transported via Koloselva to Kuetsjärvi across the Norwegian border into Pasvikelva (Pasvik-Programme, 2020).

Since the database used to generate input files for TEOTIL Metals only includes Norwegian data, it is not surprising that fluxes of Cu and Ni are underestimated in this region. If Pasvikelva is removed from the model comparison, performance for Cu and Ni improves substantially: both target plots show total normalised *RMSD* < 0.7 (i.e. within the inner circle) and *R*² values increase to around 90%. Given the current scope of TEOTIL Metals and the difficulty of obtaining point discharge data for non-Norwegian regions, these performance metrics seem acceptable.

4.4.2 Changing concentrations through time

The 1000 Lakes dataset shows the *spatial* distribution of metal concentrations during 2019, but one of the main uses of TEOTIL2 is to simulate riverine fluxes *over time*. It is therefore necessary to also consider temporal - as well as spatial - changes.

The best available monitoring data showing changing metal concentrations over time is collected as part of Elveovervåkingsprogrammet (e.g. Gundersen *et al.*, 2019). 11 rivers have been consistently monitored since the 1990s, and less complete data exist from 144 other rivers. For the purposes of OSPAR reporting, data from these rivers are grouped into four hydrologically determined “maritime regions”, illustrated in *Fig. 8*. *Table 4* shows how these areas relate to the 262 “vassdragsområder” defined by NVE.

OSPAR region	OSPAR vassdragsområder	Additional vassdragsområder
Skagerrak	1 to 23	309 to 315
North Sea	24 to 90	
Norwegian Sea	91 to 170	303 to 308
Lofoten-Barents Sea	171 to 247	301 to 302

Table 4: Vassdragsområder associated with the four maritime regions defined for OSPAR reporting. Note that the official OSPAR regions do not include 15 catchments (301 to 315) that drain to Sweden (*Fig. 8*), but for TEOTIL Metals these catchments have been assigned to the nearest OSPAR area

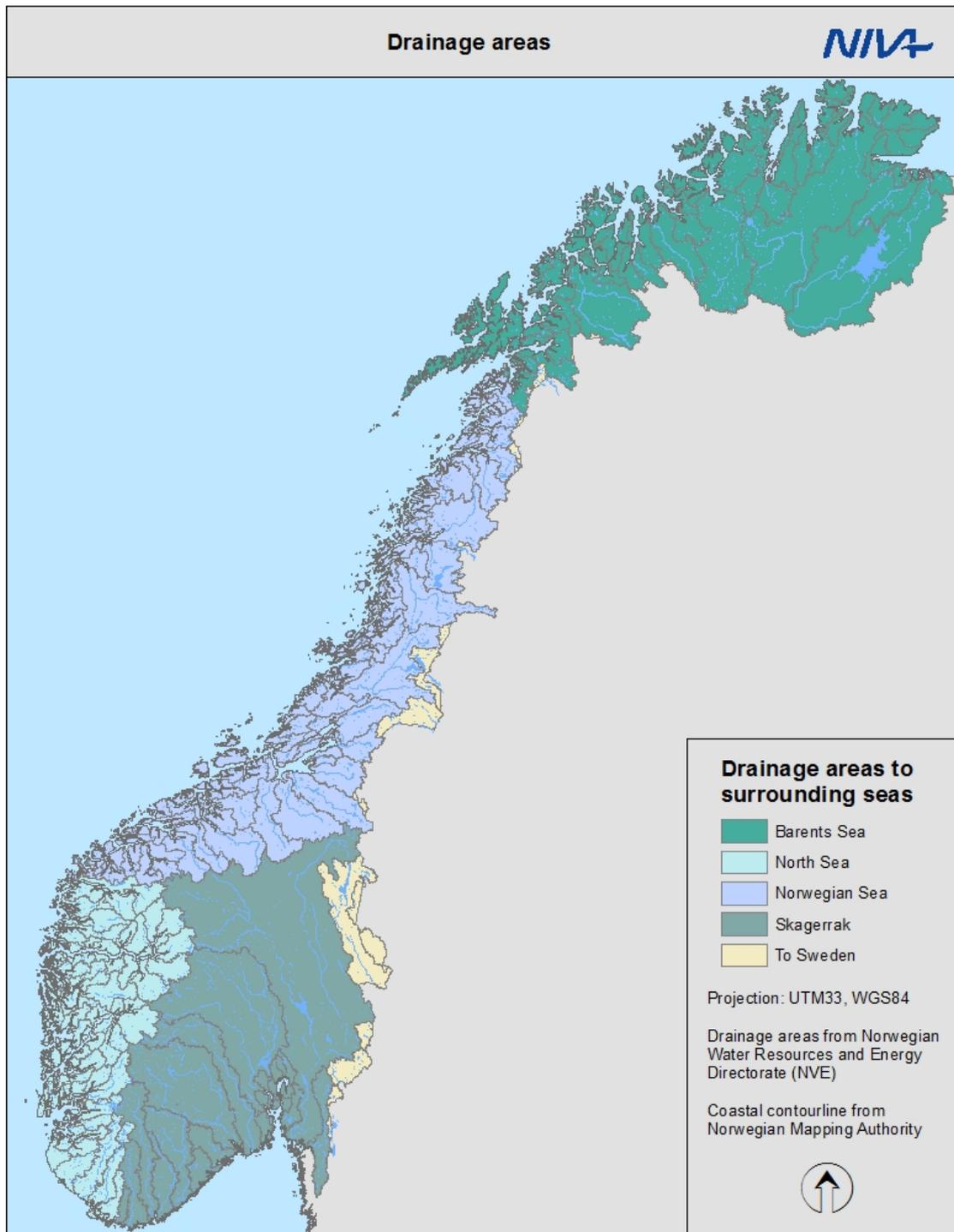


Fig. 8: Map of the four Norwegian maritime areas defined for OSPAR reporting: Skagerrak, North Sea, Norwegian Sea and Lofoten-Barents Sea

In order to model temporal changes using TEOTIL Metals, the observed data for each metal and each maritime area were aggregated to produce a single time series for each metal representative of each region. Data gaps were filled by linear interpolation, and a moving median filter with a window width of three years was applied to each aggregated series to remove extreme spikes (which appear to be due to issues with the monitoring data in some years). The resulting annual time series for each metal and region were expressed relative to

the observed value in 2019, giving a series of “change factors” for each region that could be applied to the spatially interpolated dataset from the 1000 Lakes survey. In this way, the detailed spatial patterns from the 2019 survey are preserved, but modified through time based on regional historic trends. Full details of the data processing are provided as Supplementary Information (Section 7, [notebook 02](#)) and time series of change factors derived for each parameter in each region are shown in **Fig. 9**.

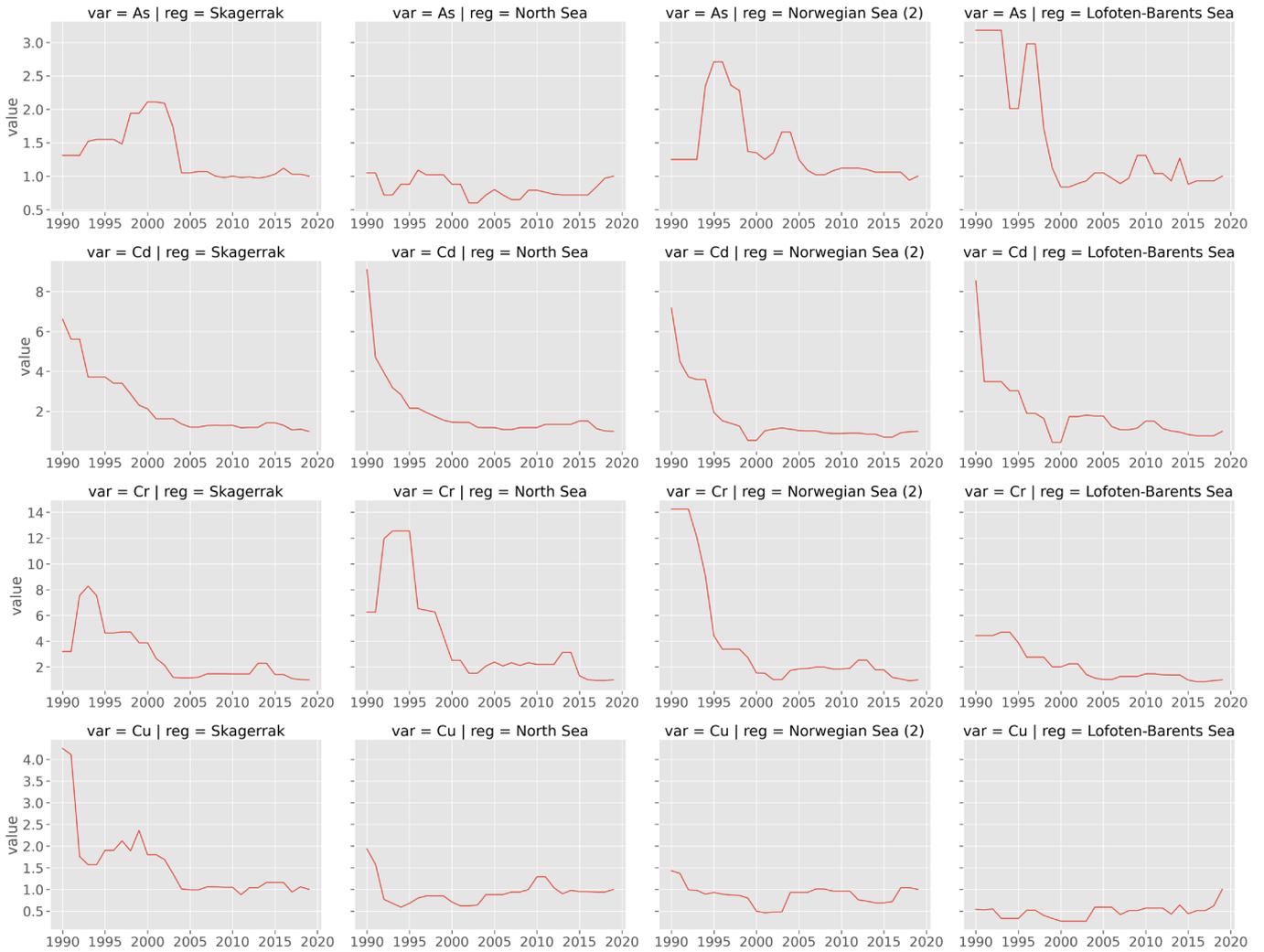


Fig. 9: Continued next page.

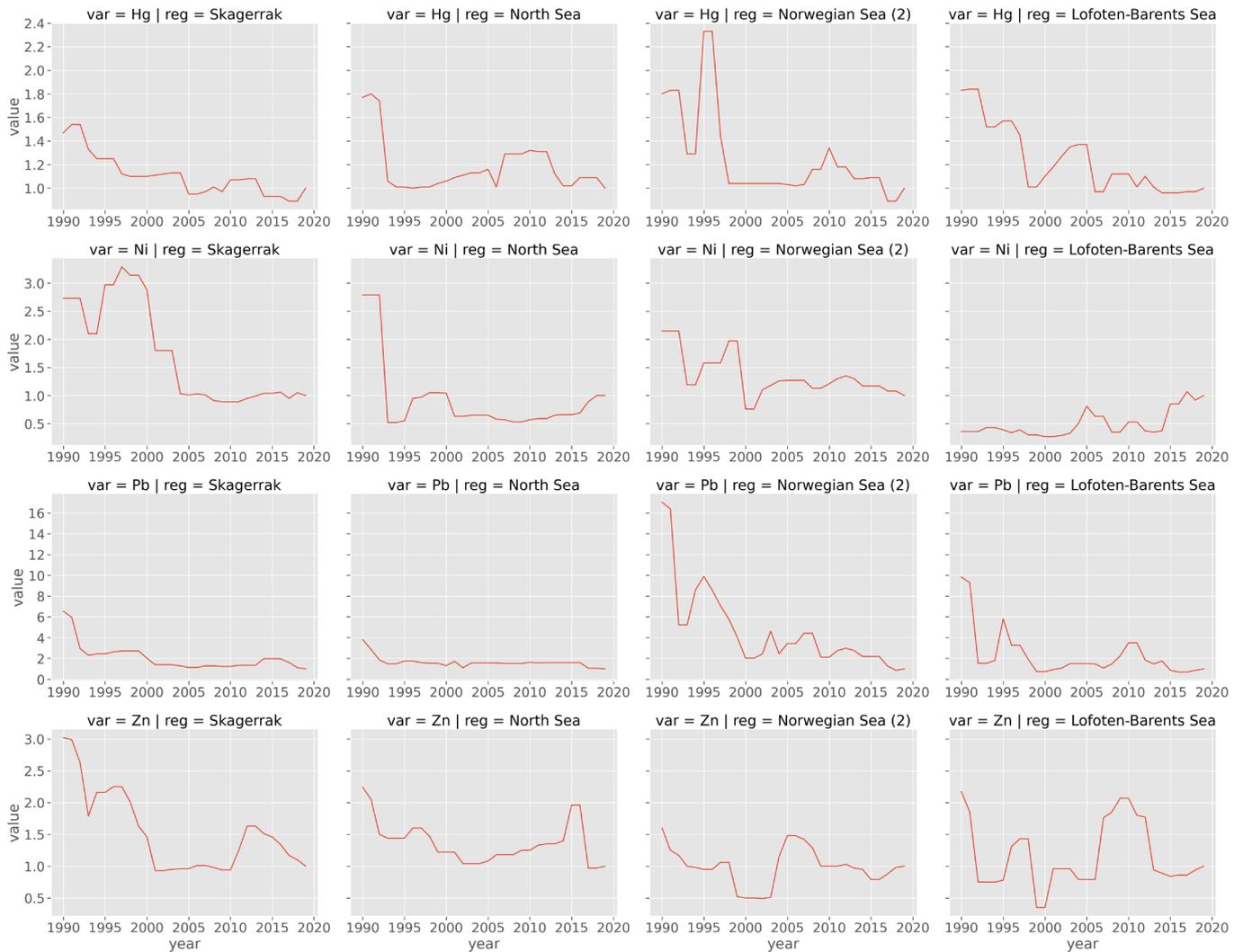


Fig. 9: Smoothed, average annual time series for each parameter in each OSPAR region, relative to 2019 values. Based on data from Elveovervåkingsprogrammet

Using these change factors, the spatio-temporal version of TEOTIL Metals was used to simulate metal fluxes for the 20 “main rivers” currently monitored under Elveovervåkingsprogrammet. These rivers have been sampled quarterly for metals since 2017 and therefore have reasonably consistent estimates of measured fluxes. *Table 5* shows a summary of model skill based on comparison of simulated and observed values for the period from 2017 to 2019 inclusive. Further details, including scatter plots and target plots, are provided as Supplementary Information (*Section 7, [notebook 04](#)*).

Metal	Regression slope	R ²	p-value
As	0.95	0.93	7.86e-36
Cd	0.82	0.77	1.35e-18
Cr	1.15	0.91	6.78e-33
Cu	2.44	0.32	1.88e-06
Hg	0.89	0.91	5.74e-20
Ni	4.59	0.40	4.99e-08
Pb	0.77	0.83	3.45e-24
Zn	1.51	0.72	3.77e-18

Table 5: Ordinary least squares linear regression results (without intercept term) for observed versus modelled values for the 20 “main rivers” monitored under Elveovervåkingsprogrammet (2017 to 2019). Good models have regression slopes and R² values close to 1

For six out of eight metals, the performance of TEOTIL2 seems promising: all relationships are highly significant ($p \ll 0.05$); the slopes of the regressions are close to 1 with zero intercept; and R² is in the range from 72 to 93%. Target and scatter plots (*Section 7, [notebook 04](#)*) also demonstrate the new model is substantially better than the default approach of simply summing point discharges.

As described in *Section 4.4.1*, model performance is poor for Cu and Ni, with regression slopes larger than 1 and R² values less than or equal to 40%. However, as noted above, this is mostly due to “outlier” data points from Pasvikelva, which receives large point inputs of Cu and Ni from industrial sites across the border in Russia. If this river is excluded from the skill assessment, the model’s performance for these metals improves substantially (normalised total RMSD < 0.7).

4.4.3 National scale simulations for 1990 to 2019

The new TEOTIL2 Metals model has been used to simulate fluxes for the entire regine catchment network for the period from 1990 to 2019. Results were evaluated by comparing modelled versus observed fluxes for the 11 stations within Elveovervåkingsprogrammet with the most detailed monitoring data. *Fig. 10* shows modelled and observed annual time series for five of these stations selected to be widely distributed around the Norwegian coastline. Results for all 11 stations are provided in Supplementary Information (*Section 7, [notebook 05](#)*).

Overall, the model is capable of capturing the main spatial and temporal patterns observed in most rivers, although it fails to reproduce much of the temporal detail. This is to be expected, as the only complete datasets representing temporal changes are available at regional - rather than local - scale. However, in several cases where there are large discrepancies between observed and simulated values, the most likely source of error seems to be the observed data rather than the simulations. For example, some of the observed (blue) curves on *Fig. 10* show isolated high spikes (e.g. Pb in regine 002.A51; Glomma), which are unlikely to be genuine. Similarly, high spikes in the simulated data (red curves on *Fig. 10*), such as those for Hg in regine 002.A51 (Glomma) during 2004 and 2006, are caused by dramatic but isolated increases in point discharges from industrial sites upstream (as recorded by Miljødirektoratet’s discharge

licensing database). Since these increases are both large and short-lived, and because they are not reflected in the river monitoring data, it seems likely these spikes are caused by problems with the reporting of industrial discharges (e.g. unit conversion errors). One option for improving the model is therefore to undertake a more thorough review of the point discharges used to generate input files.

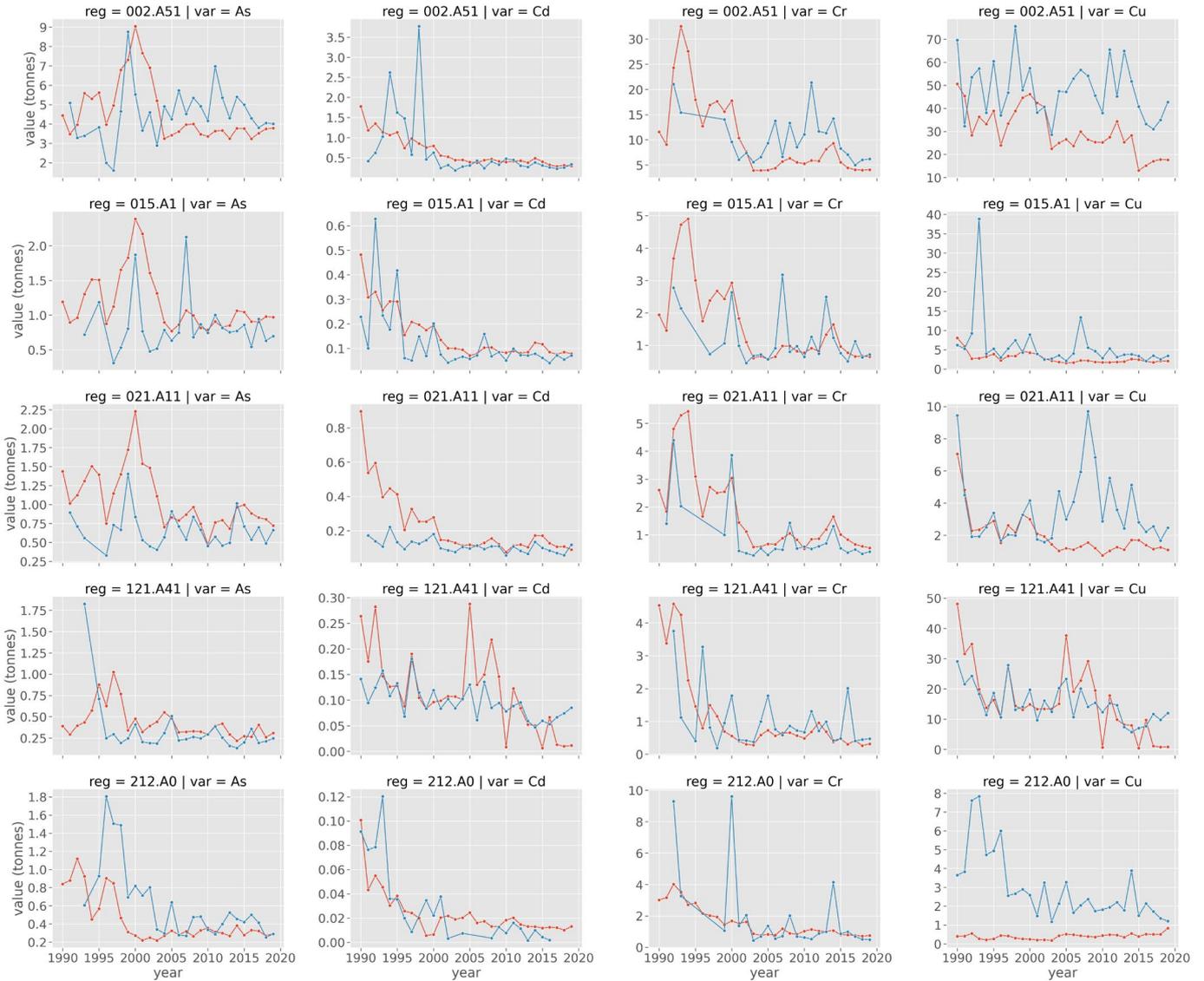


Fig. 10: Continued next page.

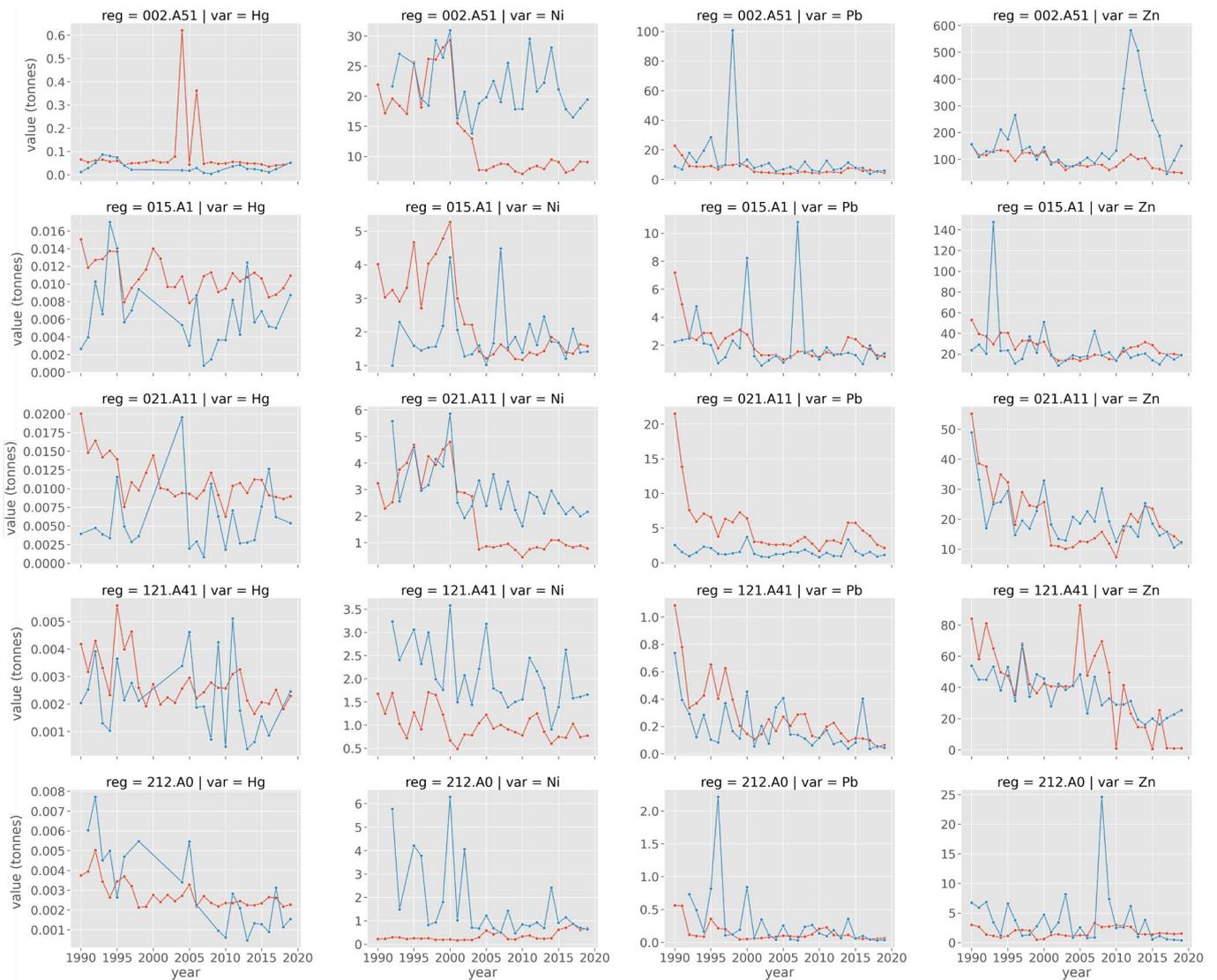


Fig. 10: Simulated (red) and observed (blue) annual time series (1990 to 2019) for each metal for five rivers within Elveovervåkingsprogrammet. The y-axis on all plots shows the annual flux in tonnes. Regime codes as follows: 002.A51, Glomma; 015.A1, Numedalslågen; 021.A11, Otra; 121.A41, Orkla; 212.A0, Altaelva.

Finally, although the model performs adequately for most rivers, there are some locations, most notably vassdragsområder 006 (Alna) and 028 (Orreelva), where the model dramatically underestimates both the mean and variability of observed fluxes (see Section 7, [notebook 05](#)). For Alna this is unsurprising, as the model does not explicitly include urban inputs and these are likely to be substantial from a large city such as Oslo. For Orreelva, monitoring data from Elveovervåkingsprogrammet indicates the catchment is strongly influenced by agriculture, so it is possible that additional inputs of metals come from e.g. pesticides or herbicides. These are not currently accounted for by the model.

5. Conclusion

The TEOTIL model has been extended to simulate fluxes of seven metals (lead, cadmium, copper, chromium, mercury, nickel and zinc) and one metalloid (arsenic) at the scale of Norway's "regine" catchment network. The spatial distribution of "diffuse source" metal concentrations in Norwegian surface waters has been derived from the 2019 "1000 Lakes" dataset, while temporal changes at regional scale are inferred from long-term monitoring in major rivers collected as part of Elveovervåkingsprogrammet. Direct point discharges (e.g. from industry) are also included, based on data extracted from Miljødirektoratet's discharge licensing database.

For most metals and most locations, the model adequately reproduces patterns in observed metal fluxes. Assessment of the model's performance demonstrates it is superior to the current "default" approach of considering only point discharges, as done e.g. for OSPAR reporting. There is some evidence that the point discharge datasets used to drive the model may contain significant errors for some locations and years, leading to sudden implausible spikes in the model output. With further refinement of the input datasets, it may be possible to improve these aspects of performance.

Despite these limitations, TEOTIL Metals seems capable of producing plausible simulations of metal fluxes for the period from 1990 to 2019. The model is simple and fast (run times are typically less than 10 seconds) and it will be straightforward to use the model to simulate fluxes for future years by assimilating new water chemistry data as it becomes available.

The entire TEOTIL2 modelling framework (including TEOTIL2 Metals) is Open Source and available to download from GitHub ([here](#)).

6. References

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7. Supplementary information

The TEOTIL model is Open Source and available on GitHub [here](#). The repository includes source code, installation instructions, further documentation and a series of short tutorials. Supporting material (including code) for the development of TEOTIL Metals is also available as a series of Jupyter notebooks, which form appendices to this report:

- [Metals development 01](#): Interpolation of key datasets and preliminary statistical analysis
- [Metals development 02](#): Incorporating the 2019 “1000 Lakes” dataset and exploring spatial patterns
- [Metals development 03](#): Simulating changes through time
- [Metals development 04](#): Evaluating the model by comparing to observed data for the period from 2017 to 2019
- [Metals development 05](#): Using TEOTIL Metals to simulate national scale fluxes for the period from 1990 to 2019

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