A Statistical Method for
Estimation of European Sulphur Emissions
Using EMEP Monitoring Data

by

GUDMUND HØST[1]

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[1]Norwegian Computing Center, Box 114 Blindern, 0314 Oslo, Norway.

Preface and Acknowledgements

This report was prepared for the twentieth session of the Steering Body of EMEP (Co-operative Programme for Monitoring and Evaluation of the Long Range Transmission of Air Pollutants in Europe). The purpose of this report is to present a statistical method that may prove useful in future compliance monitoring of emissions. The method uses the EMEP-model to match official emissions data from European countries and measured depositions from the EMEP monitoring network. An application to 1990 sulphate data demonstrates the method. The problem of designing an optimal monitoring network may also be assessed within the proposed statistical framework.

Contents

1 Introduction

The Convention on Long-Range Transboundary Air Pollution was adopted in Geneva in 1979. It is an international agreement with the objective of protecting man and his environment against air pollution including long-range transboundary air pollution. Various protocols to the Convention have put explicit obligations on the contracting parties pertaining to control and reduction of emissions. A protocol on the reduction of sulphur emissions or their transboundary fluxes by at least 30% was adopted at Helsinki in 1985. Related protocols on nitrogen oxides and volatile organic compounds were adopted in 1988 and 1991, respectively. In addition, a second sulphur protocol on further reduction of sulphur emissions was adopted in Oslo in 1994. Rather than proportional reduction of sulphur emissions, this protocol calls for cost-effective reductions to minimize the effects on the environment. Of particular relevance to this article is the emphasis of the Oslo Protocol on review of the information supplied by the parties. Indeed, the Oslo Protocol establishes a committee to review compliance by the parties with their obligations.

The purpose of this article is to introduce a statistical framework that may prove useful for monitoring compliance by European countries with their obligations to reduce air pollutant emissions. The method is illustrated by an application to 1990 data.

In our example, we use average concentrations of sulphate in precipitation from the EMEP monitoring network in 1990 (Pedersen, Schaug and Skjelmoen 1992). We also use national sulphur emissions as reported to the Geneva Convention. The reported sulphur emissions used in this article are given in Barrett et al (1995). In our approach, the reported sulphur emission from each country characterizes a probability distribution and the true emission is a random variable drawn from this distribution. The reported emissions are linked to the monitored depositions by using the acid deposition model developed by the Meteorological Synthesizing Centre – West (MSC–W) of EMEP, located at Norwegian Meteorological Institute. This model is described in various publications, its latest implementation and further references are given in Barrett et al (1995). Coefficients from the acid depo-

sition model describe the transfer of emitted sulphur dioxide from countries to wet deposited sulphate at monitoring stations. These transfer coefficients are used as regressors in a spatial linear regression model with the measured depositions as response and the emissions as regression coefficients. Within an empirical Bayes framework (Berger 1985), we obtain a mathematical consistent method for combining the various sources of information.

The results given in this article pertaining to sulphur emissions may be improved upon by including measured sulphur dioxide and particulate sulphate in the statistical model. For future assessment of national reductions of sulphur emissions, we also recommend combining data from subsequent years.

The statistical model presented here may have wide applications to other fields of environmental modeling, such as water pollution. Although the focus here is on estimation of input variables (emissions), our model may also be used for enhancing the prediction of output variables (depositions). Thus, our results are also relevant to prediction of critical load exceedances.

Our article is structured as follows. The data are presented in Section 2. In Section 3, the statistical framework is introduced and some special cases are discussed. The application to European sulphur emissions is described in Section 4. A discussion of our results and suggestions for further work are given in Section 5.

2 Data and Available Information

The emissions information used in the analysis were 48 national and regional emissions from 1990, as presented in Barrett et al (1995). These sulphur dioxide emissions will be referred to as *prior emissions*, and the values are given in Table 1 (left column). Of the prior emissions, only 30 values are officially submitted, the remaining 18 values are estimated by MSC–W.

The deposition data used in this study are yearly averages of sulphate in precipitation measured at 42 locations in Europe, obtained by averaging daily values from the EMEP monitoring network (Pedersen, Schaug and Skjelmoen 1992) throughout 1990. For each location, the data value is a weighted average of sulphate concentrations, with the weights taken as the proportion of daily precipitation to the total 1990 precipitation at the location. Stations with more than 10% missing data in any month have been excluded from the analysis. The sulphate data are further described in Pedersen, Schaug, Skjelmoen and Kvalvågnes (1992).

The acid deposition model of MSC–W (Barrett et al 1995) was used to obtain transfer coefficients from emitted sulphur dioxide to wet deposited sulphate. We will refer to this acid deposition model as the *EMEP-model*. Each transfer coefficient we use describe wet deposited sulphate contribution at a specific monitoring station from a unit emission in a specific country or region. The transfer coefficients quantifies current knowledge of atmospheric transport, chemical transformations and deposition processes, and it is the explanatory tool linking emissions to monitoring data. While the monitoring data are spatial point values, the sulphate predictions from the EMEP-model are spatial averages over 150 km × 150 km grid blocks. Particular attention must be given to monitoring stations near large emission sources. This is because the acid deposition model is not designed to describe local deposition phenomena. Therefore, including monitoring stations near large sources may amplify the variance in emission estimates. On the other hand, the monitoring data are often not sensitive to local emissions, because monitoring locations are selected to measure background concentrations. Consequently, a monitoring station located in a grid block with large local emissions is likely to measure lower sulphate concentrations than the true grid-block average. This

effect may introduce systematic errors and would give biased emission estimates. Therefore, to take proper care of such data, we would need a much more elaborate statistical model than we have used in this study. Since our present model is Gaussian, we have instead applied some reasonable data screening procedure. The data screening criterion used in this analysis was to delete stations in grid blocks with yearly emissions exceeding 10 tons of sulphur per square km. One British station and 5 stations in central Europe were removed by this criterion. In addition, a station on Faeroe Islands was removed due to very strong marine influence. The resulting set of 42 monitoring stations used in the analysis is shown in Figure 1.

Figure 2 shows the EMEP-predicted sulphate concentrations using reported emissions versus the measured concentrations for 1990 after the data screening described above. We see that the EMEP-predicted concentrations are mostly smaller than the measured concentrations. The predicted concentrations from the EMEP-model are also shown in the leftmost column of Table 2.

The method also requires a prior estimate for the precision of the reported emissions. We have used a prior coefficient of variation $\gamma_0 = 0.3$, based on recommendations from MSC–W. This reflects the view that reported emissions have a standard deviation of 0.3 times the reported value. A discussion on the sensitivity of the results to this parameter is given in Sections 4 and 5.

Code	Region	Prior	Posterior	Increase	Rel. Change	CV
AL	Albania	120	120	0	1.00	0.30
AT	Austria	90	91	1	1.01	0.30
BE	Belgium	317	317	0	1.00	0.30
BG	Bulgaria	2020	2030	10	1.01	0.30
DK	Denmark	180	187	7	1.04	0.29
FI	Finland	260	273	13	1.05	0.28
FR	France	1298	1281	-17	0.99	0.28
DD	German Dem. Rep.	4755	4545	-210	0.96	0.28
DE	Germany,Fed. Rep.	878	885	7	1.01	0.29
GR	Greece	510	511	1	1.00	0.30
HU	Hungary	1010	1079	69	1.07	0.28
IS	Iceland	6	6	0	1.00	0.30
IE	Ireland	178	178	0	1.00	0.30
IT	Italy	2251	2715	464	1.21	0.24
LU	Luxembourg	16	16	0	0.99	0.30
NL	Netherlands	201	205	4	1.02	0.29
NO	Norway	54	54	0	1.00	0.30
PL	Poland	3210	3111	-99	0.97	0.21
PT	Portugal	282	282	0	1.00	0.30
RO	Romania	1504	1523	19	1.01	0.30
ES	Spain	2316	2336	20	1.01	0.30
SE	Sweden	130	132	2	1.02	0.29
CH	Switzerland	62	63	1	1.01	0.30
TR	Turkey	354	354	0	1.00	0.30
GB	United Kingdom	3760	4206	446	1.12	0.21
REM	Remaining Areas	813	817	4	1.00	0.30
BAS	Baltic Sea	72	73	1	1.01	0.30
NOS	North Sea	174	176	2	1.01	0.30
ATL	North East Atlantic Ocean	316	317	1	1.01	0.30
MED	Mediterranean Sea	12	12	0	1.00	0.30
NAT	Natural Oceanic	721	733	12	1.02	0.29
RU1	Kola/Karelia	759	766	7	1.01	0.30
RU2	Leningrad/Novgorod-Pskov	317	321	4	1.01	0.30
RU3	Kaliningrad	44	45	1	1.01	0.30
BY	Belarus	710	726	16	1.02	0.29
UA	Ukraine	3850	3904	54	1.01	0.30
MD	Reublic of Moldova	91	91	0	1.00	0.30
RU4	Rest of Russia	3339	3362	23	1.01	0.30
EE	Estonia	240	244	4	1.02	0.29
LV	Latvia	82	82	0	1.00	0.30
LT	Lithuania	136	138	2	1.02	0.30
CS*	Czech Republic	1876	1822	-54	0.97	0.30
SK	Slovakia	543	557	14	1.03	0.29
SI	Slovenia	195	218	23	1.12	0.26
HR	Croatia	180	184	4	1.02	0.29
BA	Bosnia-Hercegovina	480	529	49	1.10	0.27
YU*1	F.Yugoslavia (-SI,HR,BA,YU*1)	508	513	5	1.01	0.30
YU*2	FYR Macedonia	10	10	0	1.00	0.30
	Total	41228	42140	912	1.02	-

Table 1: *Results of emissions estimation. Prior Emission, Posterior Estimated Emission and Increase are in units of 1000 tons of sulphur dioxide.*
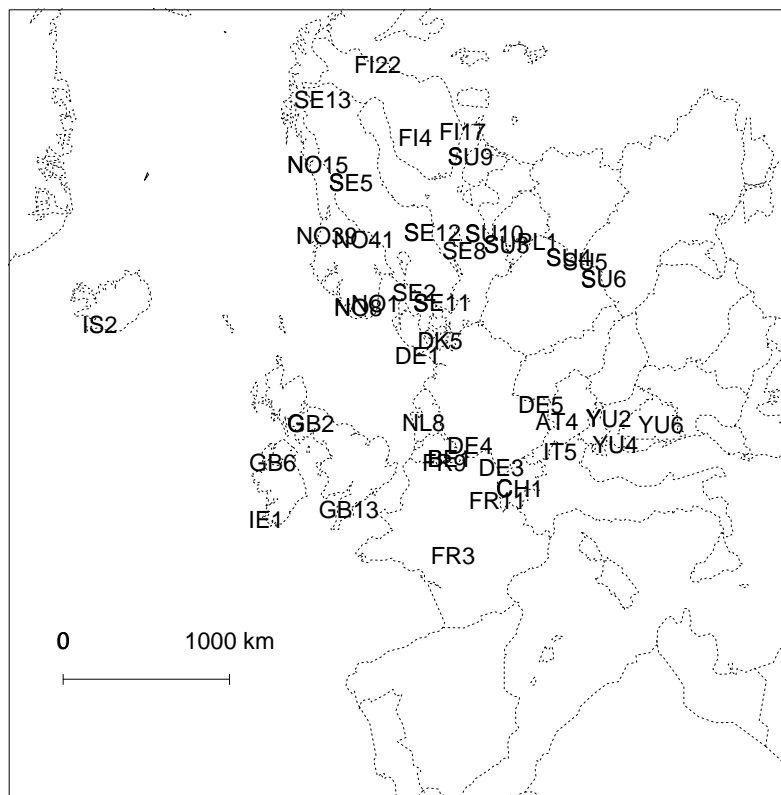
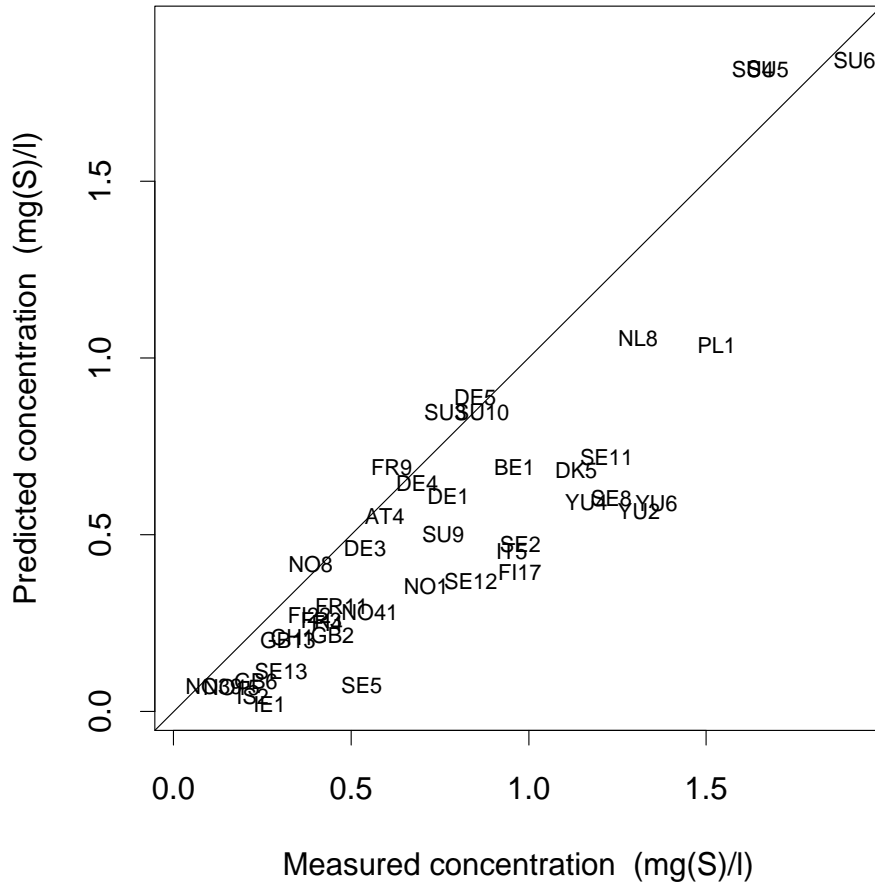Figure 1: *Study area and data locations. Concentrations at the monitoring stations are given in Table 2.*

Figure 2: *Average concentration of sulphate in precipitation in 1990 as measured by monitoring stations and corresponding prior prediction from the EMEP-model*

Grid block ID	Prior	Posterior	Increase	Rel. Change	Measured
AT4	0.55	0.56	0.004	1.01	0.59
BE1	0.69	0.69	0.003	1.00	0.96
CH1	0.21	0.22	0.007	1.03	0.34
DE1	0.61	0.66	0.050	1.08	0.77
DE3	0.46	0.46	-0.004	0.99	0.54
DE4	0.65	0.65	0.001	1.00	0.69
DE5	0.89	0.88	-0.011	0.99	0.85
DK5	0.68	0.72	0.034	1.05	1.13
FR3	0.26	0.26	0.002	1.01	0.42
FR9	0.69	0.69	0.003	1.00	0.61
FR11	0.30	0.30	0.000	1.00	0.47
IT5	0.46	0.48	0.026	1.06	0.95
IE1	0.02	0.02	0.000	1.02	0.27
IS2	0.04	0.04	0.002	1.04	0.22
NO1	0.35	0.38	0.027	1.08	0.71
NO8	0.42	0.46	0.039	1.09	0.39
NO15	0.07	0.07	0.004	1.05	0.16
NO39	0.07	0.08	0.005	1.06	0.11
NO41	0.28	0.29	0.010	1.04	0.55
NL8	1.06	1.14	0.081	1.08	1.31
PL1	1.04	1.03	-0.011	0.99	1.53
SE2	0.47	0.50	0.022	1.05	0.98
SE5	0.08	0.08	0.004	1.05	0.53
SE8	0.60	0.61	0.004	1.01	1.23
SE11	0.72	0.74	0.023	1.03	1.22
SE12	0.37	0.38	0.009	1.02	0.84
SE13	0.11	0.12	0.001	1.01	0.30
FI4	0.25	0.26	0.005	1.02	0.43
FI17	0.40	0.40	0.007	1.02	0.98
FI22	0.27	0.28	0.003	1.01	0.38
SU3	0.85	0.85	0.002	1.00	0.77
SU4	1.82	1.78	-0.036	0.98	1.63
SU5	1.82	1.78	-0.036	0.98	1.67
SU6	1.84	1.81	-0.033	0.98	1.92
SU9	0.50	0.51	0.008	1.02	0.76
SU10	0.85	0.85	0.002	1.00	0.87
GB2	0.22	0.24	0.021	1.09	0.45
GB6	0.09	0.09	0.002	1.02	0.23
GB13	0.20	0.22	0.013	1.06	0.32
YU2	0.57	0.63	0.062	1.11	1.31
YU4	0.59	0.64	0.042	1.07	1.16
YU6	0.59	0.64	0.046	1.08	1.36

Table 2: *Predicted concentrations at the monitoring stations (See Figure 1) included in the study. Prior predicted concentrations, Posterior (based on estimated emissions) predicted concentrations and Increase are in units of mg(S)/l.*

3   Statistical Model

Consider a random field $y(\boldsymbol{x})$, where $\boldsymbol{x}$ is the location within some geographic region. Let $y(\boldsymbol{x})$ have the decomposition

$$y(\boldsymbol{x}) = \boldsymbol{b}'(\boldsymbol{x})\boldsymbol{\beta} + \epsilon(\boldsymbol{x}). \tag{1}$$

Here, $\boldsymbol{b}(\boldsymbol{x})$ is an M-vector of known functions and $\boldsymbol{\beta}$ is an M-vector of unknown constants. Furthermore, $\epsilon(\boldsymbol{x})$ is a Gaussian random field with zero mean and covariance $\text{Cov } (\boldsymbol{x}_1, \boldsymbol{x}_2) = c(\|\boldsymbol{x}_1 - \boldsymbol{x}_2\|; \boldsymbol{\theta})$, where $c(\cdot; \boldsymbol{\theta})$ is a covariance function to be specified in Section 4.1. The focus of this article is on estimation of $\boldsymbol{\beta}$, but the unknown covariance parameters $\boldsymbol{\theta} = (\sigma^2, a)'$ must also be estimated. Here, σ^2 is the variance of the residual process and a is the correlation range.

In our application, $y(\boldsymbol{x})$ represents the (true) sulphate concentration at location $\boldsymbol{x}$. Furthermore, $b_j(\boldsymbol{x})$ represents the contribution to this concentration as predicted by the EMEP-model for a unit emission of sulphur dioxide from country j; $j = 1, \ldots, M$. The unknown emission from country j is β_j, so $\boldsymbol{b}'(\boldsymbol{x})\,\boldsymbol{\beta}$ is the sulphate concentration predicted by the EMEP-model. Deviations between the true sulphate concentration and the concentration predicted by the EMEP-model are absorbed into the residual field $\epsilon(\boldsymbol{x})$.

We introduce an *empirical Bayes* framework (Berger 1985, pp. 167–), and use *prior* and *posterior* to denote knowledge of $\boldsymbol{\beta}$ without or with observations of the y–field. A priori, we take each β_j to be a normal distributed random variable with prior mean $\beta_0 j$ and prior coefficient of variation γ_0. Then the probability density of $\boldsymbol{\beta}$ is

$$\begin{aligned}
p(\boldsymbol{\beta}; \boldsymbol{\beta}_0, \gamma_0) &= |2\pi\gamma_0^2 \operatorname{diag}(\boldsymbol{\beta}_0\boldsymbol{\beta}_0')|^{-1/2} \\
&\times \exp\left\{ -\frac{1}{2}(\boldsymbol{\beta} - \boldsymbol{\beta}_0)'[\gamma_0^2 \operatorname{diag}(\boldsymbol{\beta}_0\boldsymbol{\beta}_0')]^{-1}(\boldsymbol{\beta} - \boldsymbol{\beta}_0) \right\}.
\end{aligned}$$

Here, we take the prior mean $\boldsymbol{\beta}_0$ to be the vector of reported emissions from each country. The model for prior variance signifies that large reported emissions are likely to be more uncertain than small emissions. Without observations of the y-field, the natural "estimate" of $\boldsymbol{\beta}$ is $\boldsymbol{\beta}_0$, i.e. use the reported emissions. The motivation for regarding $\boldsymbol{b}(\boldsymbol{x})$ as fixed and $\boldsymbol{\beta}$ as random is the assumption that the reported emissions are much more uncertain than the EMEP-model. Thus, possible errors in the EMEP-model would be interpreted as errors in the reported emissions.

Since we have no a priori knowledge about the variance parameter σ^2, we use the common non-informative prior $p(\sigma^2) \propto \sigma^{-2}$. For the range parameter a, we use the prior $p(a) \propto (1 + a)^{-2}$, as suggested by Handcock and Wallis (1994). The prior chosen for a reflects that the residual field ϵ should capture high-frequency fluctuations, hence a small range is more likely a priori than a long range. The interested reader is referred to Berger (1985) for an introduction to Bayesian statistics and discussions on how to choose prior densities.

Let $\boldsymbol{y}$ be the data vector of N measurements of sulphate, $\boldsymbol{y} = (y(\boldsymbol{x}_1), \ldots, y(\boldsymbol{x}_N))'$. The information in the data are contained in the likelihood

$$L(\boldsymbol{\beta}, \boldsymbol{\theta}; \boldsymbol{y}) = |2\pi\boldsymbol{\Sigma}(\boldsymbol{\theta})|^{-1/2} \exp\left\{-\frac{1}{2}[\boldsymbol{y} - \boldsymbol{B}\boldsymbol{\beta}]'\boldsymbol{\Sigma}^{-1}(\boldsymbol{\theta})[\boldsymbol{y} - \boldsymbol{B}\boldsymbol{\beta}]\right\}. \quad (2)$$

Here, the j'th column of $\boldsymbol{B}$ is $b_j(\cdot)$ evaluated at the data locations $\{\boldsymbol{x}_1, \ldots, \boldsymbol{x}_N\}$, and the (i, j)'th element of the covariance matrix $\boldsymbol{\Sigma}(\boldsymbol{\theta})$ is $c(\|\boldsymbol{x}_i - \boldsymbol{x}_j\|; \boldsymbol{\theta})$.

By Bayes' theorem, the posterior density of $(\boldsymbol{\beta}', \boldsymbol{\theta}')$ is

$$p(\boldsymbol{\beta}, \boldsymbol{\theta}|\boldsymbol{y}) \quad \propto \quad p(\sigma^2)\, p(a)\, p(\boldsymbol{\beta}; \boldsymbol{\beta}_0, \gamma_0^2)\, L(\boldsymbol{\beta}, \boldsymbol{\theta}; \boldsymbol{y}).$$

Inserting the appropriate expressions for the prior and the likelihood, we get

$$p(\boldsymbol{\beta}, \boldsymbol{\theta}|\boldsymbol{y}) \;\;\propto\;\; \frac{1}{\sigma^2}\frac{1}{(1+a)^2}\,|\gamma_0^2\,\mathrm{diag}(\boldsymbol{\beta}_0\boldsymbol{\beta}_0')|^{-1/2}$$

$$\times \exp\left\{-\frac{1}{2}(\boldsymbol{\beta}-\boldsymbol{\beta}_0)'[\gamma_0^2\,\mathrm{diag}(\boldsymbol{\beta}_0\boldsymbol{\beta}_0')]^{-1}(\boldsymbol{\beta}-\boldsymbol{\beta}_0)\right\} \qquad (3)$$

$$\times |\boldsymbol{\Sigma}(\boldsymbol{\theta})|^{-1/2}\exp\left\{-\frac{1}{2}[\boldsymbol{y}-\boldsymbol{B}\boldsymbol{\beta}]'\boldsymbol{\Sigma}^{-1}(\boldsymbol{\theta})[\boldsymbol{y}-\boldsymbol{B}\boldsymbol{\beta}]\right\}.$$

According to the Bayesian paradigm, inference about $\boldsymbol{\beta}$ and $\boldsymbol{\theta}$ should be based on the posterior density (3). In the present application, we have $M+2 = 50$ parameters and $N = 42$ observations. Therefore, the parameters can not be estimated by standard methods. In contrast, the Bayesian framework allows for coherent parameter estimation by including prior information. Since the number of parameters is large, it is computationally expensive to integrate the posterior density (3). To avoid integration, we instead maximize (3), and use the *posterior mode* as the parameter estimate. The a posteriori density is a penalized likelihood function, and we may apply a method of numerical maximum likelihood estimation. We use the scoring method proposed for spatial data by Mardia and Marshall (1984). Some caution should be exercised when selecting the covariance function, see Warnes and Ripley (1987), and the rejoinder of Mardia and Watkins (1989). Precision estimates are obtained from the estimated parameter covariance matrix,

$$\mathrm{Var}\left\{(\widehat{\boldsymbol{\beta}}', \widehat{\boldsymbol{\theta}}')\right\} = -\left[\mathrm{E}\frac{\partial^2 \log p(\boldsymbol{\beta},\boldsymbol{\theta}|\boldsymbol{y})}{\partial(\boldsymbol{\beta}',\boldsymbol{\theta}')^2}\right]^{-1}_{(\boldsymbol{\beta}',\boldsymbol{\theta}')=(\widehat{\boldsymbol{\beta}}',\widehat{\boldsymbol{\theta}}')}. \qquad (4)$$

Some special cases will be illustrated by fixing σ^2 and a. Denote by $\widehat{\boldsymbol{\Sigma}}$ the covariance matrix based on the parameter estimates $\widehat{\sigma^2}$ and $\widehat{a}$. Conditional on $\widehat{\sigma^2}$ and $\widehat{a}$, the expression for the estimated emissions $\widehat{\boldsymbol{\beta}}$ is found by differentiating the right hand side of (3). We get

$$\widehat{\boldsymbol{\beta}} = (\boldsymbol{\Sigma}_0^{-1} + \boldsymbol{B}'\widehat{\boldsymbol{\Sigma}}^{-1}\boldsymbol{B})^{-1}(\boldsymbol{\Sigma}_0^{-1}\boldsymbol{\beta}_0 + \boldsymbol{B}'\widehat{\boldsymbol{\Sigma}}^{-1}\boldsymbol{y}), \qquad (5)$$

where $\boldsymbol{\Sigma}_0 = \gamma_0^2\,\mathrm{diag}(\boldsymbol{\beta}_0\boldsymbol{\beta}_0')$. From (5), the variance of $\widehat{\boldsymbol{\beta}}$ is

$$\text{Var } \widehat{\boldsymbol{\beta}} = (\boldsymbol{\Sigma}_0^{-1} + \boldsymbol{B}'\widehat{\boldsymbol{\Sigma}}^{-1}\boldsymbol{B})^{-1}. \tag{6}$$

Formulae (5)–(6) are discussed in the following remarks.

1. Let $\gamma_0 \rightarrow \infty$ and $N \geq M$. This represents a situation with many monitoring stations and no prior information on emissions. Then

$$\begin{aligned}
\widehat{\boldsymbol{\beta}} &\rightarrow (\boldsymbol{B}'\widehat{\boldsymbol{\Sigma}}^{-1}\boldsymbol{B})^{-1}\boldsymbol{B}'\widehat{\boldsymbol{\Sigma}}^{-1}\boldsymbol{y} \\
\text{Var } \widehat{\boldsymbol{\beta}} &\rightarrow (\boldsymbol{B}'\widehat{\boldsymbol{\Sigma}}^{-1}\boldsymbol{B})^{-1},
\end{aligned}$$

which is the maximum likelihood estimate. When $\widehat{\boldsymbol{\Sigma}}$ is known, this is equivalent to the generalized least squares estimate. Furthermore, inserting $\widehat{\boldsymbol{\Sigma}} = \sigma^2\boldsymbol{I}$, we get the ordinary least squares estimate.

2. Let $\gamma_0 \rightarrow 0$. Then $\widehat{\boldsymbol{\beta}} \rightarrow \boldsymbol{\beta}_0$ and Var $\widehat{\boldsymbol{\beta}} \rightarrow \boldsymbol{\Sigma}_0$. This is just the mean and variance of the prior density, which is data-independent. Thus, with exact emissions reports, there is no need to consider the monitoring data.

3. Formula (5) represents the general situation with γ_0 finite and positive. Then we may estimate $\boldsymbol{\beta}$ also for $N < M$, and the emission estimates will be a weighted combination of the prior emission and the maximum likelihood estimate.

4. A measure of the total variance in estimating $\boldsymbol{\beta}$ is tr(Var $\widehat{\boldsymbol{\beta}}$). Since $\boldsymbol{B}'\widehat{\boldsymbol{\Sigma}}^{-1}\boldsymbol{B}$ is a positive definite matrix, we see from (6) that for $\widehat{\boldsymbol{\Sigma}}$ fixed, the total variance is minimized for some positive finite value of γ_0. Thus, better emission estimates (smaller total variance) are obtained by using both prior information and data than by using either of these sources of information separately.

4 Results

4.1 Main Results

The model was fitted using an exponential covariance function $c(h; \sigma^2, a) = \sigma^2 \exp(-3h/a)$. Here, the correlation range a is the distance beyond which correlation is less than 0.05. The maximum posterior density algorithm gave residual variance estimate $\widehat{\sigma^2} = 0.085 \ [mg(S)/l]^2$ and correlation range estimate $\hat{a} = 851 \ km$. This suggests that sulphate residuals will be virtually uncorrelated for lags greater than 851 km. The estimated standard deviation of the estimated residual variance and correlation range were $0.022 \ [mg(S)/l]^2$ and $300 \ km$, respectively. Figure 3 shows the empirical correlogram and the estimated correlation function. The *correlogram* is the average covariance between station pairs at a specified lag divided by the overall variance. Since both the correlogram and the fitted correlation function tend to one as the lag tends to zero, there are no indications of measurement error effects in the sulphate data. This is partly because the sulphate data used in this analysis are averages over many values (i.e. one year of daily observations), but also because removal of stations near large emission sources may reduce residual short-range variability.

The results for the estimated emissions are described in Figure 4 and in Table 1. In Figure 4, the reported emissions and the estimated emissions are shown. The upper and lower boundaries of the boxes indicate 63% "confidence" intervals for the emissions, and the central bars are the reported (broken line) or estimated (full line) emissions. Figure 4 shows that the estimation method gives larger emissions for Italy and Great Britain than reported. For Italy, United Kingdom and Poland, the heights of the full boxes are smaller than the heights of the broken boxes. A shrinkage of the full box compared to the broken box in Figure 4 (for example Italy, United Kingdom and Poland) signifies that the estimated emission has smaller estimated posterior uncertainty than the prior uncertainty. Figure 4 also shows smaller estimated emissions than reported for former German Democratic Republic and Poland. Table 1 shows that our method of including information from the EMEP model and sulphate measurements allocate an additional 464 000

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tons of sulphur dioxide to Italy (21% increase) and an additional of 446 000 tons to Great Britain (12% increase), as compared to the reported emissions. Similarly, our method indicates that emissions from former German Democratic Republic were 210 000 tons smaller than reported and emissions from Poland were 99 000 tons smaller than reported, corresponding to 3–4% of the respective reported emissions. The estimation method suggests that the total emissions of sulphur dioxide from all European sources are 912 000 tons greater (2%) than reported. A total of 32 estimated emissions exceeded the prior emissions, while 4 estimates are smaller than reported. For 12 regions the estimated and reported emissions are equal.

Our method also gives estimated correlation between posterior estimated emissions from different countries. All these estimated correlations were smaller than ±0.3.

Table 2 shows the sulphate concentrations at monitoring locations resulting from using the estimated emissions instead of the reported emissions. Only 6 locations get slightly smaller concentrations than the prior predictions. Some regions of largest increase seem to be former Yugoslavia and Norway. These results are only indicative for the updated deposition situation in Europe, because the density of monitoring stations vary geographically. To obtain the complete picture, we would have to evaluate the EMEP-model for all grid blocks in Europe, not only for the monitoring stations.

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4.2 Sensitivity Study

The sensitivity of the statistical model was checked through analysis of some runs under various model assumptions. We are particularly interested in the capability of the method to "detect" unusual prior emissions. In addition, we will investigate the sensitivity to prior uncertainty. We reestimate emissions for prior coefficient of variation $\gamma_0 = 0.2, 0.3, 0.5, 1.0$. Furthermore, we vary the prior emission of the greatest contributor, former German Democratic Republic (DD), and investigate the results. The hypothetical prior emissions chosen are the official emission (4 755 000 tons) multiplied by factors 0.5, 1.0 and 2.0.

The results are shown in Table 3 for German Democratic Republic (DD), United Kingdom (GB), Italy (IT) and Poland (PL). The three latter countries were selected because they are important contributors, whose emissions can be estimated with relatively good precision (small posterior coefficient of variation).

Table 3 (upper panel) shows that when the DD prior emission is unreasonable small, both the DD and PL posterior emissions are increased compared to the prior emissions. Correspondingly, Table 3 (lower panel) shows that when the DD prior emission is unreasonably large, both the DD and PL emissions are reduced a posteriori. Therefore, it seems that the DD and PL emissions may be confounded in our statistical model. Possibly, a different configuration of the monitoring network could give better separation of the emissions from these two regions. On the other hand, it is possible that we may need higher resolution in the EMEP-model to separate these regional emissions.

When interpreting the columns of Table 3, recall that the posterior variability is proportional to the hypothetical DD prior emission. Therefore, for a fixed prior coefficient of variation, the IT, PL and GB posterior emissions will tend to be closer to the reported emission when the hypothetical DD emission is small (upper panel) than when the hypothetical DD emission is large (lower panel). Across all three panels of Table 3, we see that the emissions from Italy (IT) and United Kingdom (GB) are increased a posteriori. In particular, Table 3 (middle panel, right column) gives the posterior emissions for

the reported DD prior emissions when the deposition data are given much weight. We see that the deposition data suggest a small posterior decrease of DD and PL emissions, and a substantial posterior increase of the GB and IT emissions. However, to obtain significant results, we would need more monitoring data.

Emitter	Prior coefficient of variation				
	0	0.2	0.3	0.5	1
DD	**2377**	2378	2391	2517	2632
IT	2251	2436	2671	3453	5011
PL	3210	3260	3308	3406	3295
GB	3760	3910	4063	4506	4680
DD	**4755**	4617	4545	4682	4332
IT	2251	2455	2715	3587	5300
PL	3210	3131	3111	3086	3009
GB	3760	3991	4206	4753	4898
DD	**9510**	7865	7115	6601	5519
IT	2251	2471	2755	3694	5433
PL	3210	2863	2793	2736	2801
GB	3760	4073	4345	4937	5166

Table 3: *Posterior estimated emissions for various prior specifications, in units of 1000 tons of sulphur dioxide. Boldface numbers indicate hypothetical reported emission from former German Democratic Republic (DD)*

4.3   Cross-validation of Sulphate Values

While we in previous sections described emissions estimation, we now focus on implications for prediction of sulphate concentrations. We will do this by introducing a statistical interpolator, and compare the predictive capability of the acid deposition model predictor with this interpolator.

For a given location $\boldsymbol{x}$, the sulphate value can be inferred from the (empirical Bayes) kriging interpolator

$$\widehat{y}(\boldsymbol{x}) = \boldsymbol{b}'(\boldsymbol{x})\widehat{\boldsymbol{\beta}} + \boldsymbol{c}'(\boldsymbol{x})\widehat{\Sigma}^{-1}(\boldsymbol{y} - \boldsymbol{B}\widehat{\boldsymbol{\beta}}). \tag{7}$$

Here, the elements of the N-vector $\boldsymbol{c}(\boldsymbol{x})$ are $(\boldsymbol{c}(\boldsymbol{x}))_i = c(\|\boldsymbol{x} - \boldsymbol{x}_i\|; \widehat{\sigma^2}, \widehat{a})$. The term $\boldsymbol{b}'(\boldsymbol{x})\widehat{\boldsymbol{\beta}}$ on the right hand side of (7) can be interpreted as the acid deposition model prediction based on estimated emissions. The second term on the right hand side of (7) interpolates the fitted residuals $(\boldsymbol{y} - \boldsymbol{B}\widehat{\boldsymbol{\beta}})$. This term represents deviations between the observed and predicted sulphate concentrations. The corresponding interpolation error estimate is the usual

$$\text{Var}\,\{y(\boldsymbol{x}) - \widehat{y}(\boldsymbol{x})\} = \boldsymbol{\alpha}'\widehat{\Sigma}\boldsymbol{\alpha} + 2\,\boldsymbol{\alpha}'\boldsymbol{c}(\boldsymbol{x}) + \sigma^2, \tag{8}$$

where the weights $\boldsymbol{\alpha}$ are

$$\boldsymbol{\alpha} \;=\; \widehat{\Sigma}^{-1}\boldsymbol{c}(\boldsymbol{x}) \;+\; \widehat{\Sigma}^{-1}\boldsymbol{B}(\Sigma_0^{-1} + \boldsymbol{B}'\widehat{\Sigma}^{-1}\boldsymbol{B})^{-1}[\boldsymbol{b}(\boldsymbol{x}) - \boldsymbol{B}'\widehat{\Sigma}^{-1}\boldsymbol{c}(\boldsymbol{x})]$$

After estimating the parameters, the following cross-validation exercise was carried out. One monitoring station at a time was omitted and that location was interpolated using all other data. The procedure was repeated for each of the 42 monitoring stations. Then the cross-validated (interpolated) sulphate values were compared with the observed sulphate values.

Figure 5 shows observed and cross-validated sulphate concentrations. Compared to Figure 2, there seems to be a smaller bias in the interpolated sulphate values. For each location, we form the squared difference between the

measured sulphate value and the interpolated (cross-validated) value. Denote the root mean of all such values by the *rms true interpolation error*. In this article, we have used the arithmetic mean, although one could alternatively use a spatially weighted mean. The cross-validation exercise also gives an estimate of the interpolation variance at each location, as given by (8). Denote the root mean of all such values by the *rms estimated interpolation error*. The rms true interpolation error is 0.227 and the rms estimated interpolation error is 0.235. This close correspondence between estimated and true interpolation errors indicates that the proposed statistical model is a reasonable method for assessing sulphate concentrations in Europe.

In comparison, we can also calculate the *rms prediction error* from the squared difference between measured sulphate and the value predicted by the acid deposition model (i.e., not interpolating the residual). The rms prior prediction error (using reported emissions only) is 0.334, while the rms posterior prediction error is 0.318. Thus, there is only a slight improvement on sulphate predictive capability by incorporating the monitoring data, as long as we do not interpolate the residuals. This is because much weight is given to the prior emissions in the present example. A larger prior coefficient of variation would give smaller posterior prediction errors. For example, using $\gamma_0 = 0.5$ reduces the rms posterior prediction error to 0.288. Prediction performance would also be improved by including more data, either from other years or from other monitored components. The results of this section indicate that prediction performance may be further improved by also interpolating the residual field. Although this may be preferable in terms of predicting actual sulphate concentrations, it will reduce interpretability, since unexplained variability would be introduced into the predictor. This unexplained variability represents sulphur that cannot be accounted for in the mass–balance formulation of the EMEP–model.

The results of this section should be interpreted with some care, since we are in fact assessing the quality of interpolation and prediction of sulphate, not emissions estimations. Also, there may be some dispute as to how one should average the cross-validated and predicted values. In particular, we may want to give greater weight to distant locations, since these locations "represent" a larger area than locations from densely sampled regions.
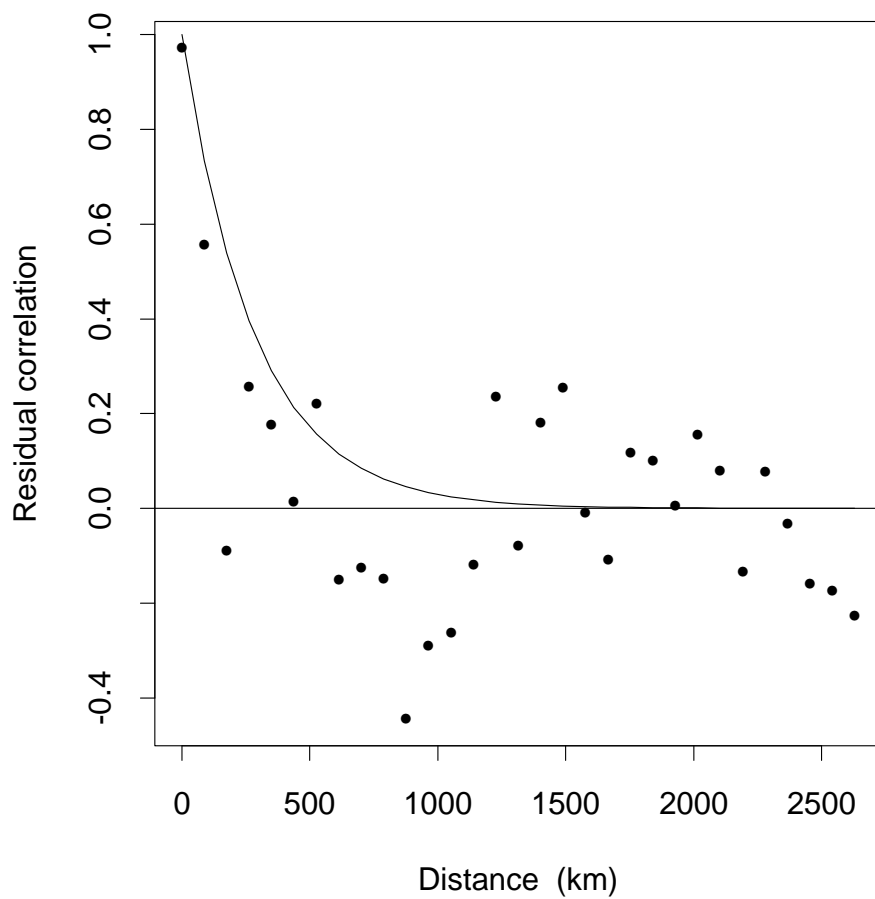
Figure 3: *Correlogram of fitted residuals (points) and estimated correlation function (line) with range=851 km*
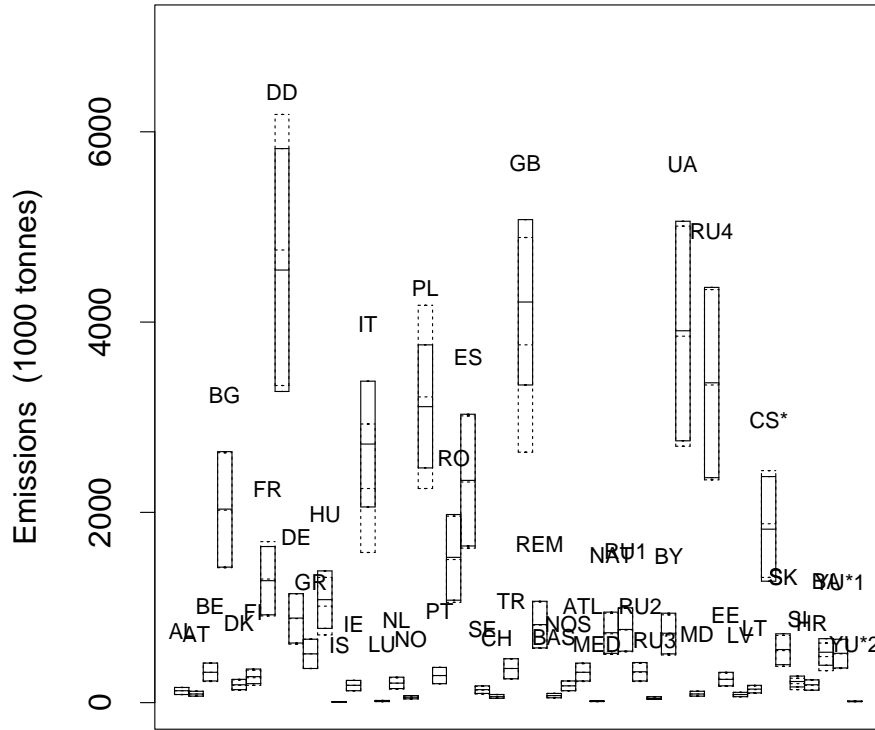
Figure 4: *Posterior estimated emissions (full boxes) and prior emissions (broken boxes) for each region. Predicted emission and ± one (estimated) standard deviation. Region codes are explained in Table 1*
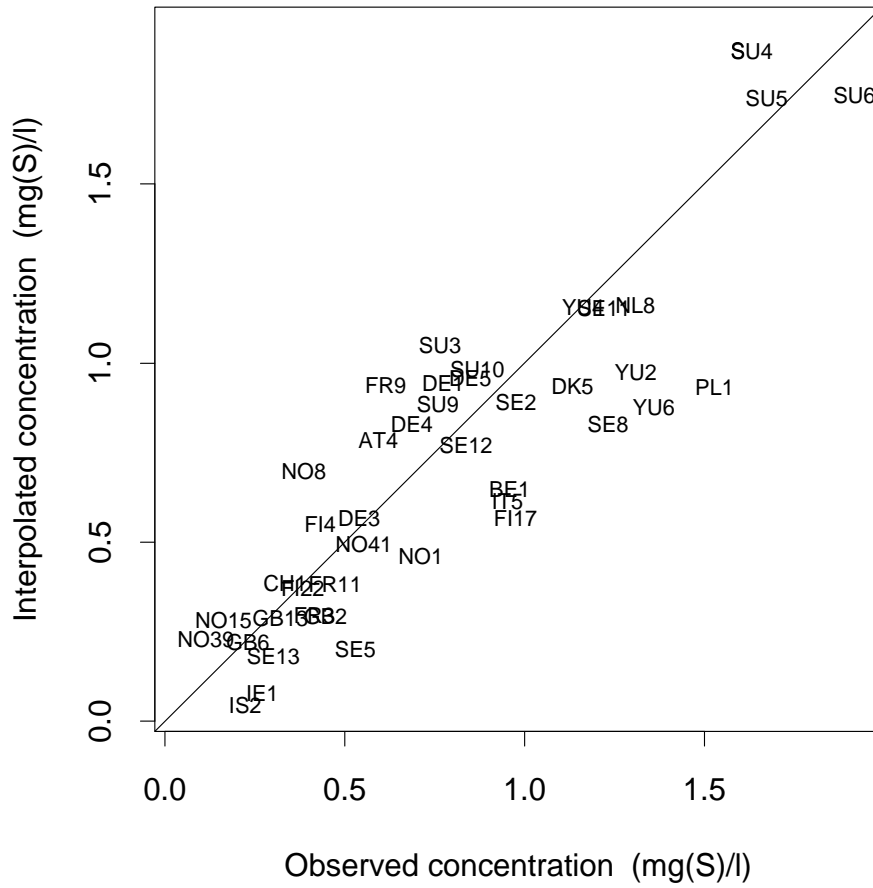
Figure 5: *Observed versus cross-validated sulphate concentrations at monitoring stations. See Figure 1 for location of monitoring stations*

5 Discussion

We have proposed a statistical framework for combining reported emissions, monitored deposition data and information from an acid deposition model. Our method was used to investigate sulphur emissions and sulphur wet depositions in Europe during 1990 with the view towards improving the emissions estimates.

Our main findings are that the 1990 sulphur emissions from Italy and United Kingdom seem to be larger than reported, while emissions from Poland and former German Democratic Republic probably were somewhat smaller than reported. Furthermore, there are indications that the total emissions from all sources are somewhat greater than reported in Barrett et al (1995).

The precision of our results depends on the prior uncertainty about reported emissions. The prior uncertainty is quantified by the hyperparameter γ_0, which has to be specified prior to the analysis. In the absence of exact knowledge of γ_0, the following considerations apply. Choosing γ_0 too small will overemphasize the confidence in reported emissions and give emissions estimates that are close to the reported emissions. On the other hand, choosing γ_0 too large will underemphasize emission reports and the resulting emissions estimates will be based mainly on sulphate data and the acid deposition model. This will give less biased emissions estimates, but greater posterior variance. Thus, a pragmatic point of view is to regard γ_0 as a smoothing parameter governing the bias-variance trade-off. If the main concern is to identify potentially unreliable emission reports, the emphasis should be on bias reduction. Consequently, we would be better off using a value for γ_0 which is somewhat too large rather than one which is too small. Alternatively, a topic for further research could be to estimate γ_0. Possibly, this problem may be addressed through cross-validation of the prior emissions $\boldsymbol{\beta}_0$.

The precision of our estimates also depends on the locations of monitoring stations. In general, increasing the number of monitoring stations will give better emissions estimates. In Section 2, we argued that locating monitoring stations near large emission sources may introduce bias in emissions

estimates. Furthermore, the detailed geometry of the monitoring network affects the variance of emissions estimates in a non-trivial manner through interaction with the acid deposition model coefficients. Although beyond the scope of this article, the problem of locating new monitoring stations can be addressed by the current framework. In particular, the possible improvement on emission estimates from a particular region resulting from adding or deleting monitoring stations can be quantified.

A further question is the sensitivity of the present results to possible systematic errors in the acid deposition model. Such errors should not be important if the emissions are the main source of uncertainty. Other questions on sensitivity include alternative regional subdivisions. Stochastic simulation may prove a useful tool for addressing these and related topics.

Our framework can also be used for enhancing the predictions of sulphate concentrations and depositions. In general, one should expect better correspondence between predicted and measured concentrations when using the proposed method than when using reported emissions only. This has bearing also on predicting exceedances of critical loads, and is another topic for further research.

Even though statistical interpolation of the residuals may further improve predictions, only posterior predicted concentrations can be attributed to regional sources, through the link provided by the EMEP-model. In this context, the residual field is a nuisance term representing unexplained variability, i.e. sulphate not accounted for by the EMEP–model. The proposed framework is a tool for reducing the importance of the unexplained variability relative to the explained variability. This would be preferable for decision making, since only the explained variability is subject to negotiations on emissions reductions.

Further work may include extending the method to include other monitored sulphur compounds, such as sulphur dioxide and particulate sulphate. This may enhance predictions of total depositions and would modify predicted exceedances of critical loads. Emissions estimates may be further improved by including data from several years, although a more elaborate statistical model would be required to incorporate possible temporal correlations. To

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this date, all data should be available for estimating compliance with the Helsinki protocol on 30% reduction of sulphur dioxide emissions. In the future, we hope to use the method for investigating compliance with the Oslo Protocol, and to extend the method to also include nitrogen and volatile organic compounds.

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