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A refinement of the emission data for Kola Peninsula based on inverse dispersion modelling

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Abstract

The study reviews the emission estimates of sulphur oxides (SO_x) and primary particulate matter (PM) from the major industrial sources of Kola Peninsula. Analysis of the disagreements between the existing emission inventories for the Kola region combined with forward and inverse ensemble dispersion modelling, analysis of observation time-series and model-measurement comparison showed that the emission of the Nikel non-ferrous metallurgy plant was missing from the EMEP inventory, as well as from some others, being in some cases misplaced or mis-attributed to other sources of the region. A more consistent inventory of the anthropogenic emissions of SO_x and PM has been compiled for the peninsula, compared with the existing estimates and verified by means of dispersion modelling. In particular, the SILAM model simulations for 2003 and 2006 with the revised emission data showed much lower bias – up to 6 times for the most-affected sites – for SO_2 with regard to the measured concentrations of 8 Finnish and Norwegian observational stations in the region. Temporal correlation improved moderately (10–20%) but homogeneously over Lapland. The study demonstrates the value of a combined usage of forward and inverse ensemble modelling for source apportionment in case of limited observational data.

1 Introduction

The emission database of EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe, http://www.emep.int) (UNECE, 2009) includes both anthropogenic and natural emissions, with yearly time step and 50 km spatial resolution. The emission inventory is based on the reports of the European countries and the estimations of the EMEP experts. The EMEP database is one of the main sources of information for atmospheric dispersion modelling in Europe and contains one of the best-verified datasets.

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Other emission inventories covering Europe, such as GEIA (http://www.geiacenter.org), CGEIC (http://www.ortech.ca/cgeic), RETRO (http://retro.enes.org), EDGAR (http://www.mnp.nl/edgar), TNO-GEMS (Visschedijk et al., 2007) and PAREST_MEGAPOLI (Denier van der Gon et al., 2010), are partly independent from the EMEP database but still maintain some of its features. For example, the TNO-GEMS inventory distinguishes between the area and point sources and significantly rearranges the emission distributions but for most countries it keeps the EMEP-based national totals. Some other inventories, such as CGEIC, incorporate EMEP as the European component of the global patterns (http://www.ortech.ca/cgeic/poster.html). The EDGAR inventory is mostly independent but for some major sources it uses the same activity data as the ones underlying the EMEP emissions and, consequently, has some common features with EMEP.

An emerging approach to refine emission data is the inverse atmospheric dispersion modelling. It has become a useful tool in model-based analysis of observations and source apportionment studies (e.g. Kuparinen et al., 2007; Rannik et al., 2003; Bergamaschi et al., 2005; Saarikoski et al., 2007; Sofiev et al., 2006a, Elbern et al., 1997, 1999, 2007). The method can be used for both correcting the emission rates of known sources and delineating the origins of observed concentration peaks. Source apportionment using dispersion models is a corner stone of the nuclear emergency preparedness activity (Bocquet, 2005a, b; Issartel, 2005; Issartel and Baverel, 2003; Thomson et al., 2007; Loosmore et al., 2007; Chang et al., 1997 etc.).

The specific approach to the source apportionment depends on abundance and coverage of available observational information, modelling tools and a-priori information on the sources. If high quality frequent measurement data are available from sufficiently dense network, a full-scale data assimilation problem can be solved with the emission rate and/or its distribution being the assimilated quantities. However, the requirements to the observational data are very stringent in this case. Additionally, only advanced and expensive methodologies, such as the four-dimensional variational assimilation (4-D-VAR) or the ensemble Kalman filtering allow explicit emission treatment (Elbern et

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al., 2007).

When the source pattern is simple and the observational data are scarce, certain reductions of the methodology are possible or even inevitable. In an extreme case, a crude analysis can be based on trivial backward trajectories. Interpretation of such results is usually qualitative (e.g., Barletta et al., 2009; Skjøth et al., 2007), but sometimes quantitative analysis can be undertaken (Kulmala, 2000; Sogacheva et al., 2005, 2007; Heo et al., 2009). For quantitative and comparatively accurate assessment in case of limited observational information, the so-called "footprint" computations can be used (Rannik et al., 2003; Kuparinen et al., 2007; Saarikoski et al., 2007). This approach is based on solving the adjoint dispersion equation for e.g. an isolated episode registered by a single measurement device. The result of the adjoint computations describes the sensitivity distribution of that particular measurement. The observed values are sensitive to the emission fluxes from the area where the sensitivity is non-zero. This area is referred as the measurement footprint.

The source apportionment problem in Lapland has to be based on a limited set of stations, but fortunately the region has just a few almost-point sources dominating the emission pattern. Such distribution simplifies the source location problem, but also leads to a high sensitivity of the refined emission estimates to the uncertainties of the meteorological and dispersion models. For instance, a limited deviation of the predicted wind direction from the actual one may result in the model plume missing the particular station, thus jeopardizing the model-measurement comparison.

The uncertainties of the individual simulations can be reduced by constructing a modelling ensemble. This tool has proven to be useful for various tasks, including air quality analysis and forecasting (http://gems.ecmwf.int; http://www.gmes-atmosphere.eu; Sofiev et al., 1996; http://www.gse-promote.org; Delle Monache and Stull, 2003; Mallet and Sportisse, 2006; Pagowski and Grell, 2006) and also emergency modelling with point-type sources (Galmarini et al., 2004a, b; Potempski et al., 2008), i.e. for the emission distributions similar to the current study. It has been shown that even a simple arithmetical average, or the median as its robust analogy, of the individual ensemble

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members (air-quality models or specific simulations) usually shows better scores in the model-measurement comparisons than any single participating model (Galmarini et al., 2004c; Riccio et al., 2007; Potempski et al., 2008). The spread between the individual models then indicates the predictability of the episode, its stochastic features, and the potential range of the uncertainties in the results of the simulations. More sophisticated approaches are under construction, aiming at the optimal selection and combination of the ensemble members and at softening or lifting some of the underlying assumptions concerning the relation between the ensemble and the actual probability distribution (Galmarini et al., 2004c; Mallet and Sportisse, 2006; Riccio et al., 2007; Delle Monache et al., 2006; Potempski and Galmarini 2010).

The goal of the current study is to refine the estimates of sulphur oxides (SO_2 and SO_4^{2-}) and aerosol particulate matter (PM) pollution in Northern Lapland caused by the industrial sources of Kola Peninsula. The study includes the following steps: (i) the analysis of the emission patterns of Kola Peninsula in the existing emission inventories, (ii) the refinement of the emission data taking the EMEP inventory as a starting point, (iii) the verification of the proposed adjustments using ensemble forward and adjoint dispersion simulations with the SILAM modelling system, and (iv) the evaluation of their impact on the predicted air pollution of the region.

2 Analyses and refinement of the emission distribution of Kola Peninsula

The industrial pattern of Kola Peninsula is heavily dominated by three major centres of activity (Fig. 1): the Nikel (69°20′ N, 30°04′ E) and Monchegorsk (67°55′ N, 32°57′ E) non-ferrous metallurgy plants and mines, and the city of Murmansk (68°57′ N, 33°06′ E) with the nearby harbour. There is very limited anthropogenic activity outside these centres.

The SO_x emissions from Nikel and Monchegorsk are by far the largest in the region, also exceeding the SO_x emission of the whole Finland by a factor of times (Ahonen et al., 1997). The Monchegorsk and Murmansk emissions are also rich of NO_x, contrary

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to Nikel, which has high SO_x but low NO_x fraction (Ruuskanen et al., 2003; Virkkula et al., 2003). The PM emissions from these three sources are comparable but uncertainties are large, also due to relatively high contribution of other sources.

The natural SO_x in Lapland originate from marine DMS production, which forms a generally low background level. The natural NO_x emissions around Lapland are very small and their background concentration is caused by long range transport of PANs from Europe. The PM non-industrial contributions are disperse and originate from very different sources: road dust, sea salt, production of secondary aerosols, etc.

Summarising, the best-articulated tracer for the industrial emission distribution in Lapland and Kola region is SO_2. Uncertainties in the Nikel NO_x emission seem to be below the sensitivity of our methods. There was little observational information readily available on PM concentrations, apart from Varrio campaign. Therefore, below we concentrate on the SO_x emission and "project" its activity-specific modifications to particulate emission.

2.1 Evaluation of EMEP SO_2 emission data

According to the EMEP rules, every five years the emission distributions must be updated and reported afresh to the database by the member states. For intermittent years only the national totals are reported and the patterns are interpolated linearly in time. Upon the decision of the member states, the data can be revised retrospectively.

Until the early 1990s, the EMEP standard grid resolution was 150 km. In this grid the locations of Murmansk and Nikel belonged to two neighbouring grid-cells. For the year 1992 (the last available with 150 km resolution), over 250 kT yr^{-1} of SO_2 emission was reported in the grid-cell covering Nikel and about 30 kT attributed to Murmansk grid-cell (Sofiev, 2000).

The current EMEP database, with 50×50 km resolution, reports strongly varying emission amounts and patterns for different years for both SO_x and PM (see Table 1 and Fig. 2). Moreover, it differs considerably from the 1992 150×150 km distribution.

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A strong source of SO_x has been shown for 1980 in the grid-cell (48, 91) neighbouring Murmansk and for 1985 in the grid-cell (47, 90) neighbouring the Nikel location (Fig. 2). However, the plant itself is not represented as a source. Starting from 1990, all the reported emissions in the area suddenly fall by more than an order of magnitude, though in 2005 somewhat higher emissions (compared to surrounding background level) show up in the Nikel-containing grid cell (46, 90). The 2010 and 2020 emission projections again show a strong source near Murmansk but nothing around Nikel. Totals of these years return to the level of 1980s.

The same problems are evident for other substances, such as NO_x (to a less extent and somewhat different temporal pattern) and PM. The totals for other regions of Russia located within the EMEP domain do not exhibit such jumps.

Considering the above sharp changes since 1980s, one should take into account that the decline of the economy of the region in 1980s–1990s may indeed result in some decrease of the emission. However, we are not aware of any dedicated large-scale emission-reduction measures at the plant. Boyd et al. (2009) cautiously mentioned ~33% reduction during 1990s with a reference to the official values and assumed no modernisation of the plant. According to Hagen et al. (2002), Berglen et al. (2008), Nikel SO_2 emissions were reported around 250–300 $kT\,yr^{-1}$ until mid-1980s and reduced to ~175 $kT\,yr^{-1}$ by the beginning of 1990s. After that no significant long-term trend is reported but the data are scarce after 1993. Ahonen et al. (1997), referring to Baklanov (1994) and to Committee (1995) report, suggests the SO_2 emissions of the whole Kola Peninsula to fall by ~25% from 517 $kT\,yr^{-1}$ in 1992 to 380 $kT\,yr^{-1}$ in 1994.

The SO_2 concentration measurements in surrounding stations also do not support the changes shown by EMEP data (Hagen et al., 2002; Berglen et al., 2008; Ruuskanen et al., 2003; Virkkula et al., 2003). The Svanvik measurement station in Norway reports about 2 times reduction in SO_2 annual mean concentrations from the late 1980s to beginning of 1990s. No significant change in SO_2 has been observed at Svanvik, Maajavri, Nikel, Viksjøfjell or Varrio stations in 1990s and 2000s. EMEP stations in Lapland also reported only gradual trends without any drastic changes during the last

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20 yr. In particular, there was no dramatic decrease of the upper percentiles of the daily mean concentrations observed by any EMEP station of the region (Fig. 3).

Therefore, the sharp fall of all emissions over Kola Peninsula and large random changes in the emission distribution do not seem justified. Since the period of the fastest economical decline had ended by the mid-90s, the reported total emission of 1992 should not be too far from the emissions of later years, at least until 2008, when the current crisis started.

Finally, the 50 km resolving datasets obtained from EMEP before 2006 (used with some modifications by Hongisto et al., 2003; Sofiev et al., 2003; BACC, 2008; Bartnicki et al., 2002; EMEP, 2000; the EMEP assessment reports, see http://www.emep.int, etc.) differ drastically from the currently available EMEP information. In the old reports dated before 2006, the total SO_x emission for Kola Peninsula differed only slightly from the total of the 150 km resolution dataset for 1992. The source distribution pattern was again somewhat different, though still missing all emissions in the Nikel location. It is noteworthy that previous atmospheric dispersion simulations have shown that pollutant concentrations in Lapland are usually underestimated with respect to measurements at the monitoring stations in Finland, Sweden and Norway, unless extra information is included (Hongisto et al., 2003; Bartnicki et al., 2004, 2006; Zlatev et al., 2001; Sofiev et al., 1994; Sofiev, 2000).

2.2 Comparison of the emission inventories

There are numerous inventories of anthropogenic emission available, covering various regions and time periods with different spatial and temporal resolutions and containing different sets of pollutant species (Table 2). For Europe, the most extensive databases, with the largest number of pollutants and the highest spatial and temporal resolutions seem to be EMEP, TNO- GEMS and PAREST_MEGAPOLI, and RETRO. The global databases, such as GEIA, EDGAR and CGEIC usually have low (1×1 degree) resolution, which is insufficient for regional model applications. However, they can still be considered for comparison when it comes to regional totals.

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In the TNO-GEMS inventory for 2003, the initial EMEP emission distributions have been significantly rearranged but the national totals for most countries are based on values reported to EMEP. Independent bottom-up assessment from activity data and emission factors were used only if the reported data were missing or suspected to be erroneous. In particular, new emissions were generated for Russian Federation, including Kola Peninsula (Visschedijk et al., 2007). The total SO_2 emission of the region is assessed to be around 140 kT of SO_2 per year, which is of the same order of magnitude, though lower than the regional total of EMEP 1992 with 150 km grid resolution (Table 1). The emission distribution for SO_2 in the TNO-GEMS inventory differs considerably from that of EMEP and explicitly shows Nikel emission. However, it attributes about 80% of the emissions of the peninsula to the Monchegorsk area and only about 15% (22 kT of SO_2 per year) to the Nikel area, which is doubtful. For instance, Boyd et al. (2009) mentioned 300 kT (with a reference to Zientek et al., 1994) as a total-Kola industrial SO_2 emission with ~70% attributed to the Nikel plant area.

The step from TNO- GEMS to PAREST-MEGAPOLI (Denier van der Gon et al., 2010) included a complete overhaul of the European point source database including removal of the closed installations and expansion with all new point sources accessible through source-sector specific databases or statistics. There were two major reasons for this. Firstly, it improved the completeness of the list of European point sources. Secondly, for Russia the assessment relied on the estimates of the national sector total emissions by the IIASA RAINS/GAINS model (http://gains.iiasa.ac.at) which was adjusted significantly after releasing the TNO-GEMS database. The reconsideration of the point sources and Russian emission totals resulted in almost doubling the total SO_2 emission of the point sources in Kola Peninsula: from 170 to 266 kT of SO_2 per year (Table 5). However, the emission distribution still attributes only 19% of it (52.5 kT) to the Nikel plant.

The RETRO database does not provide anthropogenic SO_x emissions. For other pollutants, the RETRO emission assessments are independent from EMEP but still based on a similar set of activity data (energy statistics) and share most of its features

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concerning, in particular, the spatial distribution.

The EDGAR emission data are available only for years 1990 and 1995. The total levels are comparable with EMEP 1992 150 km resolution emissions, dropping moderately (by 1.7 times) between these years. However, the emission pattern still does not show any significant emissions at the Nikel location, and has an unrealistically large source in the Murmansk area (Table 3).

GEIA and CGEIC emissions for Europe are based on either EMEP or EDGAR assessments.

Concluding the analysis, none of the considered inventories contains information which would have (i) a sufficient resolution, (ii) a correct distribution of the major sources, (iii) a reasonable absolute emission level. Below we have compiled a dataset which seems to be matching these criteria better than the existing inventories.

2.3 Starting point for the emission correction

Selecting the initial dataset for modifications, we took into account that the previous modelling activities (Saarikoski et al., 2007; Galperin et al., 1994a, b, Sofiev et al., 1995; Galperin and Sofiev, 1998; Sofiev 2000; EMEP assessment reports from http://www.emep.int) have not shown significant over-estimation of SO_x and PM concentrations in 1990s and 2000s, when the data with absolute levels similar to those of the EMEP 150 km emissions for 1992 are used. Secondly, the EMEP datasets until mid-2000s reported ~40% reduction from these levels (e.g. EMEP, 1999, 2000), which is similar to the reduction reported by Ahonen et al. (1997), Boyd et al. (2009) and Zientek et al. (1994). Therefore, we assumed that the total emission for the peninsula in 1990s and first half of 2000s is close to 300 kT of SO_2. The unexplained sharp fall of the absolute level of emissions (by a factor of 15–20) in the later EMEP reports was considered to be unjustified and disregarded.

The datasets with the Kola emission totals close to 300 kT of SO_2 and 50 km resolution could be downloaded from the EMEP WebDab portal before 2006. They have only one evident error in the distribution: entirely missing Nikel emission. The next task

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of this work is, therefore, to correct this error. The emission data for year 2000, downloaded in 2006 was chosen as the reference point for the correction (Table 1). The dataset misses the Nikel plant emissions, while an extremely strong source of SO_2 (about 150 kT of SO_2 per year), NO_x, CO and PM is placed around Murmansk. A large fraction of the emission there is reported for the SNAP sector 1, (large combustion in energy and transformation industry), sector 2 (non-industrial combustion plants) and sector 3 (combustion in manufacturing industry) (SNAP=System Nomenclature of Air Pollutants, http://www.emep.int). As there are no known major sources in that area, apart from the city itself and the harbour, both reporting mainly into different SNAP sectors, such as 7 (transport) we assumed that in this reference dataset the emission of the Nikel metallurgy plant was misplaced to near-Murmansk.

Since the time trends of the emission after 2000 are uncertain and probably not significant, we used the 2000 emissions for all the modelling simulations described below.

2.4 Modification of the emission distribution

The correction of the emission database started from estimating the fraction of the emission attributed to Murmansk, that must be relocated to Nikel. The consideration can be based on individual SNAP sectors. Assuming that the emission of SNAP sector S1 (large combustion in energy and transformation industry) is dominated by Nikel, the S1 emissions in Murmansk area were moved to the Nikel plant location, leaving in the original grid cells only a small fraction, corresponding to the S1 level in the neighbouring cells. Similar logic was applied to other sectors and species that contribute to the infrastructure of a large factory (Table 4).

The new estimates are probably representative for 1990s and the first half of 2000s. With the limited amount of observational data, no trend analysis seems to be feasible but the trends suggested by Boyd et al. (2009) or reported by EMEP for other parts of Russia can still be applied.

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The above correction does not reposition the Nikel town emission, neither it reflects the details of the infrastructure, first of all, roads in the Nikel region. It is the plant emission only that has been repositioned. However, this is the biggest emission source in the Kola region.

Comparison of the emission fractions attributed to different sources (Table 1) shows, that the rearrangement of the emission pattern can be considered quite conservative, as only ~40% of the SO_2 emissions of the peninsula were moved to Nikel area compared to ~50% in 150 km resolving EMEP 1992 dataset and ~70% reported by Boyd et al. (2009).

3 Source apportionment via dispersion modelling

In this section we present the modelling-based evidence of the problems of the present emission distribution in Kola peninsula, demonstrate the improvements due to the above described changes and the need for further emission refinement.

3.1 Input data and SILAM system

3.1.1 Observational LAPBIAT-campaign at Varrio in 2003 and other datasets

An unequivocal indication of the missing emission source in the original EMEP inventory was obtained from the high-resolution atmospheric aerosol measurement campaign LAPBIAT carried out at Varrio, Finnish Lapland, 67°46′ N, 29°35′ E, from 28 April to 11 May 2003 (Ruuskanen et al., 2007). For the current study, we used the measurements of $PM_{2.5}$ (particulate matter smaller than 2.5 µm) as a prominent indication of industrial aerosols.

Apart from the Varrio campaign, the long-term analysis has been performed using the information from national networks of Finland and Norway. For the purpose of the study, we used seven stations located close to the Nikel site. Six of them monitor SO_2 concentrations in air, one reports SO_4^{2-} in aerosol, and three report SO_4^{2-} in

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precipitation. The latter were converted to wet deposition for this comparison. None of these stations reported PM over sufficiently long periods, so the long-term analysis was performed for sulphur oxides.

3.1.2 SILAM modelling system and setup

Limited observational information, unfavourable positions of most of the stations upwind of the main emission sources (regarding the prevailing synoptic wind pattern), and contradicting input emission data preclude a direct estimation of the emission in the Nikel and Murmansk areas via full-scale data assimilation and source apportionment techniques. Alternative analyses have therefore been used.

The pollution transport simulations and simplified source apportionment have been performed with the air quality modelling system SILAM version 4, which has two – Eulerian and Lagrangian – advection-diffusion cores. The Lagrangian 3-D transport (Sofiev et al., 2006b) incorporates a high-precision iterative advection algorithm after Eerola (1990) and a Monte-Carlo random-walk representation of atmospheric diffusion. The Eulerian core, also used in the current experiment, is based on the non-diffusive advection scheme of Galperin (2000) and the adaptive vertical diffusion algorithm of Sofiev (2002). For a more detailed description we refer to Sofiev et al. (2008) and http://silam.fmi.fi.

All simulations were performed with 0.1 degree horizontal and 10 min temporal resolution. The model vertical consisted of 11 layers up to about 9 km above the surface. The modelling domain covered the area of 15° E–42° E and 58° N–72° N. The boundary values were taken from SILAM European simulations. The modelling was performed for 2003 and 2006 – two arbitrarily selected years for which the observational data were available. For 2003, the meteorological data were taken from the operational forecasts of the global model of European Centre of Medium Range Weather Forecast (ECMWF). The data have 0.4 degrees horizontal resolution. Simulations for 2006 were driven by the fields of the regional HIRLAM RCR system with 0.2 degrees horizontal resolution. Both datasets have 3-h time steps. Simulations for the period of the Varrio

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campaign in 2003 were performed with both Lagrangian and Eulerian kernels, each driven by both ECMWF and HIRLAM meteo input. This 4-member modelling ensemble allowed more robust estimation of the dispersion patterns (compared to individual simulations) and also indicated the level of uncertainty of the results. For the long term simulations only one configuration based on the Eulerian kernel of SILAM was used to limit the computational costs.

The input emission, depending on the specific run, was either the EMEP-original dataset for 2000 (downloaded before 2006) or the same dataset with the above described corrections.

The SO_2/SO_4^{2-} split of the SO_x emission was assumed to be 80%/20% for all the runs. All emission was considered in the model grid (no point sources) with prescribed SNAP-sector-dependent initial vertical distribution after Simpson et al. (2003). Consequently, no dynamic plume-rise computations were made.

3.2 Modelling results

3.2.1 Is Nikel plant an active source in 2000s?

The LAPBIAT-campaign at Varrio in 2003 provided a direct confirmation that during that time Nikel was still an active source of airborne pollution. During this campaign, a few pollution episodes were observed over a generally low aerosol background of Arctic spring. The modelling attempts to reproduce some of the strongest ones (more than 10-fold from the background level), such as the peak of 2–3 May, using the original EMEP emission data, were unsuccessful – all 4 ensemble members showed neither significant concentrations near Varrio (Fig. 5) nor any probability for it: all high-concentration plumes were predicted far from the observational site. The dispersion simulations made using the TNO-GEMS and PAREST-MP emission data reproduced the peak time better (Fig. 5, lowest panel) but showed strong underestimation of its value compared to the observations.

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Adjoint computations performed for the time period of the peak pointed at a small area centred around the Nikel plant (Fig. 6). Therefore, it was confirmed that at least up to 2003 the plant was an active source of anthropogenic pollution (with no indication of the reduction seen up to 2006 – see Fig. 3), which is in agreement with, e.g., Boyd et al. (2009).

3.2.2 Revised Nikel emission data: re-analysis of the Varrio campaign

The SILAM simulations with the revised emissions produced significantly different results. In all 4 cases high $PM_{2.5}$ concentrations reached Värrio at the right time (Fig. 7). Both simulations with ECMWF meteorological input even overestimated the peak, whereas both HIRLAM-driven runs underestimated it, especially when using the Lagrangian dynamic kernel. However, the mean of the ensemble reproduces the measured peak value of $PM_{2.5}$ concentration with less than 10% error.

Analysis of Fig. 7 shows the value of the ensemble-type simulations when compared to the single-simulation assessments. Prediction of the position of narrow plumes originating from point-type sources is always uncertain and so are the absolute concentrations in the plumes. In this particular case, the variations between the model-runs exceed an order of magnitude (from less than 3 µg $PM_{2.5}$ m^{-3} up to 35 µg $PM_{2.5}$ m^{-3}, depending on the model setup and the input meteorological data). The time moments when the polluted masses arrive and leave the observation site are within 1–2 h for all the simulations. As a result the ensemble both reproduces the observed peak values and points out the high uncertainty and low predictability of the case.

3.2.3 New Nikel emission: long-term evaluation

The above correction of the emission distribution was used in two year-long simulations of the SO_x distribution over the area. The goals of the computations were: (i) to evaluate the impact of the source location correction to the model-measurement comparison, (ii) to re-check the suggested values, (iii) to preliminarily estimate how close

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the new distribution is to the real emission pattern in the region, (iv) to estimate the impact of the correction onto the modelled acid deposition in the region.

In general, the comparison of predicted concentrations and depositions with the observations (Table 5) shows that the new emission distribution leads to a significant improvement of the model-measurement agreement. However, the impact is not homogeneous over the region and varies between species. The influence on the predicted mean values and variability quickly fades out with the distance from the plant and depends on the site location with respect to both Murmansk and Nikel: from the 5.5-fold increase of the for mean values (Svanvik, 9.6 km away from Nikel) down to practically no impact at Oulanka (345 km from Nikel, 334 km from Murmansk). Improvement of the temporal correlation and root mean square error (RMSE) are much more moderate (up to ~20%) but also more homogeneous around the region. These are related to more accurate positioning of the plume from the plant, which leads to reduction of the "false alerts" and catching up the "missed peaks" in the predicted time series – see examples in Fig. 8.

From the quantile charts (Fig. 9) we can also see a substantial improvement in both modelled concentrations and wet deposition with the revised emissions, though the new Nikel emission estimate still seems to be low, leading to some 25–30% of underestimation. The almost linear character of the charts shows that SILAM reproduces all ranges of the concentrations with about the same accuracy, except for very low background levels. For Svanvik (the nearest site to the plant) the slope of the chart is the same for all concentrations, while Karpbukt (the site nearest to the coast and second-nearest to the plant) shows somewhat stronger under-estimation of the background, which can possibly be attributed to the missing marine dimethyl sulphide (DMS) emission.

Both concentrations of SO_2 and wet deposition of SO_4 show almost the same underestimation, meaning that the near-surface values and column loads are reproduced with the same moderate underestimation. In turn, it may indicate that there are no severe deficiencies in modelled vertical profile of concentration, but rather the emission

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pattern still has some room for improvement. This similarity also supports the estimation of the source injection height, evaluated using the stack height of Nikel plant, because concentration at Svanvik is strongly affected by the initial injection profile.

To directly confirm that Nikel SO_2 emission is still underestimated in the revised data, we computed the footprint of the differences between modelled and observed concentration peaks. The corresponding adjoint SILAM run covered the year 2006. The input for the run was compiled as a deviation of the model from the hourly concentrations reported by four monitoring sites close to Nikel. The closest site – Svanvik – was not included, as the distance from this site to the plant was less than a model grid cell size, which made its observations not representative for the current grid. For the other sites, a two-steps filtration procedure was applied to highlight only the significant problems in the model – measurement comparison. Firstly, the background concentrations in both modelled and observed time series were eliminated. Secondly, time periods with the model error less than 50% were excluded. The remaining time periods were analysed via the adjoint SILAM run.

The overlap of the yearly-mean footprints of the significant differences ($c_{\text{model}} - c_{\text{observed}}$) for the four sites (Fig. 10) shows that, apart from the areas near the sites, the footprints have a common highlighted area around the Nikel plant (circled in the map). This overlap suggests a common reason for the model under-estimation at all sites: the under-estimated emission from the Nikel plant and/or surrounding infrastructure.

4 Discussion

4.1 Reliability of the revised emission pattern

The suggested correction of the Kola emission distribution and suspected problems in the recent changes of the EMEP emission database are based on indirect considerations, such as the model-based source apportionment, land use analysis and

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heuristic analysis of the available data. All these considerations are prone to uncertainties, which in many cases are difficult to estimate. Locations of the sources are well known and easy to correct, but the actual emission rates of each of them are not. The most objective information comes from the observational sites, but in Lapland they are all located upwind from the major sources and thus require careful processing and combining with modelling for the source apportionment tasks.

The main assumption accepted as the starting point of the analysis was that the total SO_x emission from Kola Peninsula, as presented in the 1992 and the original 2000 datasets of EMEP, is approximately correct. It was supported by the small mean bias of the SILAM model and other CTMs including the EMEP model with regard to observations when run with this emission – also after 2000. According to J. M. Pacyna (personal communication), the uncertainty of the European SO_x emission in 1990s was ~30%. From the trend analysis of the observations (Fig. 3), it followed that there were no drastic changes in the emission during last two decades and the estimates of 1990s are valid up to 2006 within a factor of 1.5 or better. There are, however, uncertainties embedded in this approach: the model internal errors, limited representativeness of the monitoring sites, and a limited number of episodes when the impact of each of the major sources could be identified. Their crude assessment is as follows. According to Sofiev et al. (2006b), the SILAM-induced uncertainty of the mean concentrations inside the individual plumes from point sources is about 50%. Following Galperin and Sofiev (1994), the representativeness-related uncertainty of the observed annual mean value is at least 20%. Finally, the extra specific uncertainty due to sparse station network in the region located upwind from the sources is difficult to estimate but it is hardly lower than 10–15%. Indeed, let the number of episodes when a particular site registered the plume from the plant be N_{epi}. The standard deviation of the mean over these episodes is then proportional to $1/\sqrt{N_{epi}}$. With typical N_{epi}~30–40 per yr, the relative uncertainty will be ~15%. Summarising, a factor of 2 as an uncertainty of the above suggested total emission of SO_x in Kola Peninsula in 2000s may be a reasonable estimate.

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Uncertainties caused by rearrangement of the emission pattern can be summarised as follows.

Firstly, the relocated emission amount was chosen to some extent arbitrarily, with only moderate justification based on SNAP sectors and surrounding background emissions. As visible from the simulation results in Fig. 8, several false SO_2 concentration peaks remained in the time series modelled with the new input. Therefore, the emission in the Murmansk area is still probably over-estimated. Conversely, the model still underestimates the SO_2 concentrations at all stations to the west of the Nikel plant by a factor of 2 to 3, which only partly comes from missing background values due to missing marine DMS emissions. The peak concentrations are also underestimated, which indicates that the Nikel emissions are still too low.

Secondly, in this study, the relocated Nikel emission is represented as a point source with the corresponding stack height, while some part of the emission probably comes from the surrounding area and infrastructure of the Nikel town. The lack of low-level emissions can also be a reason for underestimating the peak concentrations at nearby stations.

Thirdly, the SO_2/SO_4^{2-} split of the SO_x emission as 80%/20% might not be exactly correct, as indicated by the measurements at Karasjok site. That station is located 197 km from Nikel and 318 km from Murmansk and is the only site reporting both SO_2 and SO_4^{2-} concentrations. There, the observed ratio is about 50/50, which, assuming the mean $SO_2 \rightarrow SO_4^{2-}$ conversion rate of 4–5% h^{-1} and travel time as 5–7 h suggests the actual ratio at the source place about 70–30%. Due to different removal intensities of SO_2 and sulphates, such change might lead to ~10% of additional uncertainty of the concentration and deposition fields.

4.2 Long-term impact of the Kola source onto Northern Lapland

The relocation of the Nikel plant emission, as shown in Fig. 11, has spatially limited and inhomogeneous but very substantial impact on the predicted sulphur deposition

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in Northern Lapland. These changes are particularly important due to the high sensitivity of the ecosystems in the region to acidifying deposition. Strong increase of the deposition (more than an order of magnitude) is predicted within ~20 km from the new source location, over an almost circular area. Since substantial emission is still present in the Murmansk region, the deposition in this region reduces about 5-fold only. The uncertainty of such deposition redistribution is moderate. Indeed, the normalised standard deviation of the deposition redistribution (Fig. 11b) is smaller than its mean value (Fig. 11a) almost everywhere and is less than a half of it over the areas with significant changes (more than a factor 1.5).

5 Conclusions

An analysis of the disagreements between different emission inventories, the results of the observational campaigns and regular AQ monitoring in Northern Lapland, land-use, and anthropogenic activity data, allowed detecting a mis-location of a significant part of the anthropogenic emission in Kola Peninsula, as well as problems with the regional totals currently available from EMEP. Firstly, a sudden 15–20-fold drop of the emission totals of SO_x and PM in Kola Peninsula in 1990s, reported by EMEP for the region after 2006, is not supported by long term observations. Secondly, the emission of the Nikel metallurgy plant is probably falsely allocated to the Murmansk city region in several available datasets.

The starting point for the modifications was the EMEP dataset for 2000 downloaded from WebDab before 2006. Following this dataset, the total emission of Nikel and Murmansk area is suggested to stay fairly constant at ~150 kT of SO_2 per year or more throughout both 1990s and 2000s.

Using forward and adjoint simulations of the SILAM system the suggested emission correction has been verified against two years of regular SO_2 monitoring data in Northern Lapland and the PM measurement campaign at Varrio in 2003. The ensemble simulations of Varrio campaign period reduced the dependence on specific input dataset

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and the model dynamics and increased the confidence in the suggested emission redistribution. The long-term simulations showed substantial reduction of the model bias (up to a factor of times) in the Northern Lapland and 10–20% increase of the temporal correlation coefficient compared to the measurements.

The impact of the discovered error in the database on the deposition of sulphur compounds is significant (over an order of magnitude) but limited in space. It becomes insignificant when the distance from the modified sources exceeds the spatial scale of the emission redistribution, i.e., the distance between Nikel and Murmansk.

The new Nikel emission estimate still seems to be low, leading to some 25–30% of under-estimation in modelled concentrations. Further refinement of the Kola Peninsula emissions with activity-based emission assessment methods could thus be recommended.

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Table 1. The EMEP SO_x emission data for Kola Peninsula (Unit: tons of SO_2 per grid cell per year).

Largest emitting 50 km grid cells																	
Gridcell		Lon	Lat	1980	1985	1990	1992	1995	2000	2003	2005	2007	2010	2020	original	corrected	1992 (150×150)
46	90	30.3	69.5	1606	588	86	66	37	29	21	7009	5551	616	736	438	110 085	
47	90	30.8	69.1	3113	480 238	162	125	69	54	41	35	42	1193	1426	849	849	
48	91	32.6	68.9	421 398	1072	27 815	22 347	14 146	10 179	9013	1619	781	148 558	150 334	114 916	39 121	
48	92	33.8	69.1	125 016	17 422	6214	4787	2646	2043	1560	1370	1118	47 916	57 286	34 092	240	
48	93	35,0	69.3	18 638	535	861	662	363	274	218	195	97	7143	8541	5083	5083	
49	91	33.1	68.5	3455	88 479	187	145	81	64	47	40	52	1324	1583	942	942	
50	88	30.4	67.4	5186	19 252	269	208	115	90	67	59	71	1988	2376	1414	1414	
50	90	32.5	67.9	16 811	1072	869	670	372	290	217	189	225	6443	7703	4584	4584	
50	91	33.7	68.0	12 280	17 107	652	503	280	220	163	141	180	4707	5627	3349	3349	
51	89	32.0	67.2	17 562	20 773	1130	902	560	410	352	331	286	6294	6603	4789	4789	
51	90	33,1	67,4	299 846	196 543	23 148	18 829	12 350	8846	7943	7660	6163	101 173	91 988	81 769	81 769	
51	91	34.2	67.6	88 199	1072	6747	5482	3586	2572	2303	2218	1794	29 875	27 437	24 052	24 052	
Total for Peninsula				1 070 305	863 490	71 009	56 937	35 829	26 019	22 667	21 496	17 007	379 152	387 849	291 875	291 875	507 800
150 km grid cells, aggregated from 50 km and the old dataset																	
46–48[1]	88–90	29.7	68.9	17 900	486 608	925	714	396	310	232	7192	5765	6861	8202	4881	114 529	253 100
46–48	91–93	33.2	69.5	575 634	21 273	35 411	28 197	17 376	12 665	10 922	3299	2096	207 673	221 010	156 977	47 329	29 800
49–51	88–90	31.5	67.7	361 125	242 517	26 545	21 479	13 881	10 015	8862	8484	7041	124 223	118 623	98 480	98 480	193 500
49–51	91–93	34.9	68.2	115 647	113 091	8128	6547	4175	3029	2650	2522	2104	40 395	40 014	31 537	31 537	31 400
Contribution of the main sources, %																	
Nikel %				2	56	1	1	1	1	1	33	34	2	2	2	39	50
Murmansk %				54	2	50	50	48	49	48	15	12	55	57	54	16	6
Monchegorsk etc. %				45	41	49	49	50	50	51	51	54	43	41	45	45	44

Presented data in columns: individual years: EMEP web emission portal WebDab, status 2010, original: the prior-2006 WebDab download for 2000, corrected: the outcome of this work, 1992 150 km × 150 km: the old 150 km dataset.
Highlighted grid cells: green – Nikel, red – Murmansk, yellow – Monchegorsk and its surroundings.

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Table 2. The summary of the databases for anthropogenic emissions in Europe.

Database	Anthropogenic emission species	Resolution	Time resolution; Available times	Data source for European emissions
EMEP WebDaB	SO_x, NO_x, NMVOC, NH_3, PM_{10}, $PM_{2.5}$, PM_{coarse}, CO, POPs, HMs,	$50\times50\,km^2$ $150\times150\,km^2$	Annual, with diurnal, weekly and monthly variations 1980, 1985, 1990, 1995–2007 and 2010, 2020	http://www.emep.int http://www.ceip.at/emission-data-webdab/ Emissions reported by the countries
EDGAR	NO_x, NMVOC, SO_2, HCs, CO_2, CH_4, N_2O, CO, halocarbons	1°×1°	Annual 1990, 1995	http://www.mnp.nl/edgar/documentation/methodology/ bottom-up inventory based on activity data and emission factors
GEIA	NH_3, Black Carbon, NO_x, SO_2, NMVOC, CO_2, CO, CFCs, HCFC-22, MCF, Pb, Hg, CH_4, N_2O, Pesticides, Reactive Cl	1°×1°	Annual, Seasonal 1985	http://www.geiacenter.org/ Emissions for western Europe taken from CORINAIR; the EMEP inventories for European areas not covered by CORINAIR.
CGEIC	SO_2, NO_x, Pb, HCH, Hg	1°×1°	Annual, seasonal 1985	http://www.ortech.ca/cgeic/poster.html GEIA, EDGAR GEIA 1A, annual sulphur and nitrogen global emission inventory
RETRO	NO_x, VOCs, CO	0.5°×0.5°	Annual, monthly mean 1960–2000?	http://retro.enes.org/reports/D1-6_final.pdf Bottom-up inventory based on activity data and emission factors of TNO Emission Assesment Model (TEAM)
TNO-GEMS and PAREST-MEGAPOLI	NO_x, SO_2, CO, NMVOC, CH_4, NH_3, PM_{10}, $PM_{2.5}$	0.25°×0.125°	Monthly 2003 2005	National and sector totals reported by the countries. IIASA RAINS/GAINS if reported values not available or suspicious (e.g. for Russian territory).

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Table 3. EDGAR SO_2 emissions (Unit: tons of $SO_2\,yr^{-1}$). Grid cells containing the largest sources have been highlighted (green – Nikel, red – Murmansk, yellow – Monchegorsk).

Emissions for year 1990								
lat\lon	28	29	30	31	32	33	34	35
70	890	983	0	0	0	0	0	0
69	151	1850	4440	377 000	0	0	0	0
68	179	0	0	39	1260	63 100	42	65
67	1430	647	2470	39	13 400	11 300	196	0
66	1030	301	7	52	209	196	120	0
						Total	481 396	t SO_2/yr
Emissions for year 1995								
lat\lon	28	29	30	31	32	33	34	35
70	719	794	0	0	0	0	0	0
69	169	1040	2480	227 000	0	0	0	0
68	188	0	0	40	716	33 700	41	66
67	1190	556	1380	40	7300	6180	199	0
66	870	339	7	53	212	199	122	0
						Total	285 600	t SO_2/yr

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Table 4. EMEP data for 2000 (WebDab before 2006) and corrected emission data for Nikel and Murmansk (unit: T yr^{-1}). Modified dataset is suggested as reference values for 1990s-mid-2000s.

	EMEP 2000		Modified dataset	
Species, sector	Nikel	Murmansk	Nikel	Murmansk
SO_x, S1	0	31 588	31 588	0
SO_x, S2	17	3020	1509	1528
SO_x, S3	418	114 164	76 989	37 593
$PM_{2.5}$, S1	0	343	343	0
$PM_{2.5}$, S2	12	2068	1121	960
$PM_{2.5}$, S3	10	2189	1383	816
$PM_{2.5}$, S4	0	9386	9386	0
$PM_{2.5}$, S7	6	435	116	324
$PM_{2.5}$, S8	3	194	51	146
$PM_{2.5}$, S9	3	118	65	57
PM_coarse, S1	0	398	398	0
PM_coarse, S2	8	1450	810	647
PM_coarse, S3	1	186	117	70
PM_coarse, S4	0	3604	3604	0
PM_coarse, S5	8	610	167	451

Note:
SNAP: System Nomenclature of Atmopsheric Pollutants
SNAP S1: Combustion in energy and transformation industries
SNAP S2: Non-industrial combustion plants
SNAP S3: Combustion in manufacturing industry
SNAP S4: Production processes
SNAP S5: Extraction and distribution of fossil fuels and geothermal energy
SNAP S6: Solvents and other product use
SNAP S7: Road transport
SNAP S8: Other mobile sources and machinery
SNAP S9: Waste treatment and disposal
SNAP S10: Agriculture

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Table 5. Statistical scores of SILAM two-years computations at the monitoring sites. Mean over 2003 and 2006.

Station	Quantity	Temporal resolution	Average value			Standard deviation			Temporal correlation		RMSE	
			Observed	Modelled Original source	Modelled Corrected source	Observed	Modelled Original source	Modelled Corrected source	Original source	Corrected source	Original source	Corrected source
Svanvik	cnc_SO_2	hour	6,35	0,53	3,60	25,53	1,48	11,12	0,31	0,31	25,80	24,70
Kevo	cnc_SO_2	hour	1,17	0,26	0,42	3,75	0,77	1,34	0,40	0,43	3,62	3,48
Raja-Jooseppi	cnc_SO_2	hour	1,57	0,37	0,45	3,71	0,88	1,24	0,19	0,28	3,84	3,74
Sammaltunturi	cnc_SO_2	hour	0,87	0,19	0,23	2,03	0,52	0,61	0,34	0,40	2,04	1,98
Oulanka	cnc_SO_2	hour	0,96	0,48	0,48	2,04	0,98	0,91	0,26	0,31	2,08	2,02
Karasjok	cnc_SO_2	day	0,25	0,17	0,24	0,62	0,43	0,57	0,40	0,45	0,60	0,62
Karasjok	cnc_SO_4	day	0,27	0,35	0,42	0,29	0,82	0,91	0,39	0,43	0,76	0,84
Karasjok	wd_xSO_4	day	188	59	63	555	229	229	0,33	0,37	542	532
Svanvik	wd_xSO_4	week	3044	709	3960	4526	1151	6739	0,45	0,49	4680	6220
Karpbukt	wd_xSO_4	week	3907	665	2639	5139	1043	3496	0,15	0,30	5990	5410

Notations:
cnc_SO_2 and cnc_SO_4 – concentrations of SO_2 and SO_4 in air or in aerosol [$\mu g\,S\,m^{-3}$]
wd_SO_4 – wet deposition of sulphates [$\mu g\,S\,m^{-2}\,d^{-1}$] or [$\mu g\,S\,m^{-2}\,week^{-1}$]

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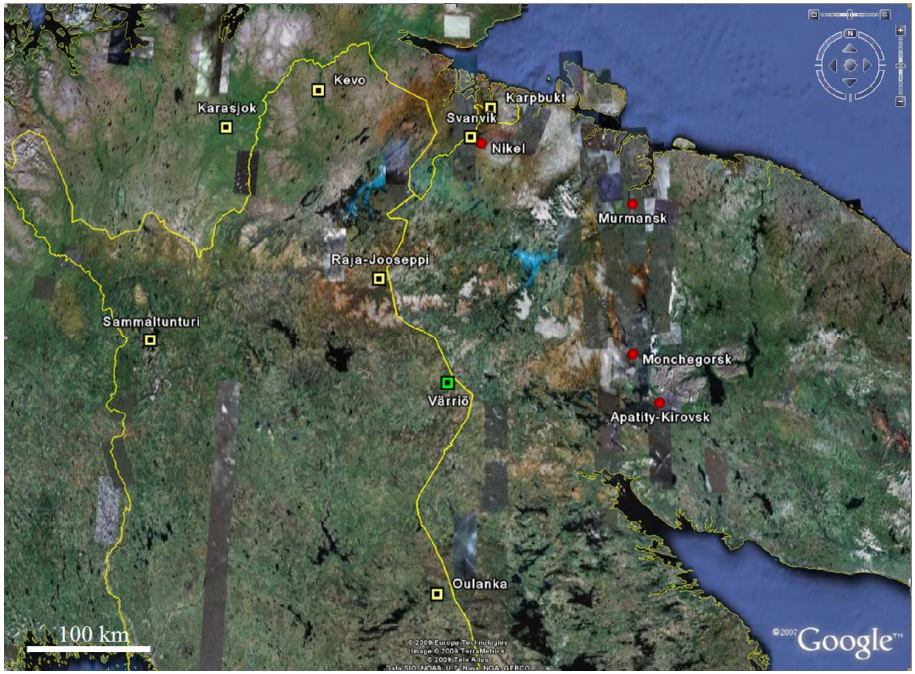

Fig. 1. Location of the Varrio measurement station (green rectangle), the other measurement sites (yellow rectangles) and the major pollution sources of Kola Peninsula (red circles).

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Fig. 2. The original EMEP emission for 1980–2007, WebDab status 2010. The 50 km grid cells are shown with the colours reflecting the SO_x emission: blue $\leq$ 0.1 kT SO_2 yr^{-1}, green$\leq$1 kT SO_2 yr^{-1}, yellow$\leq$10 kT SO_2 yr^{-1}, orange$\leq$100 kT SO_2 yr^{-1}, red$>$100 kT SO_2 yr^{-1}. Pictures from http://www.emep.int.

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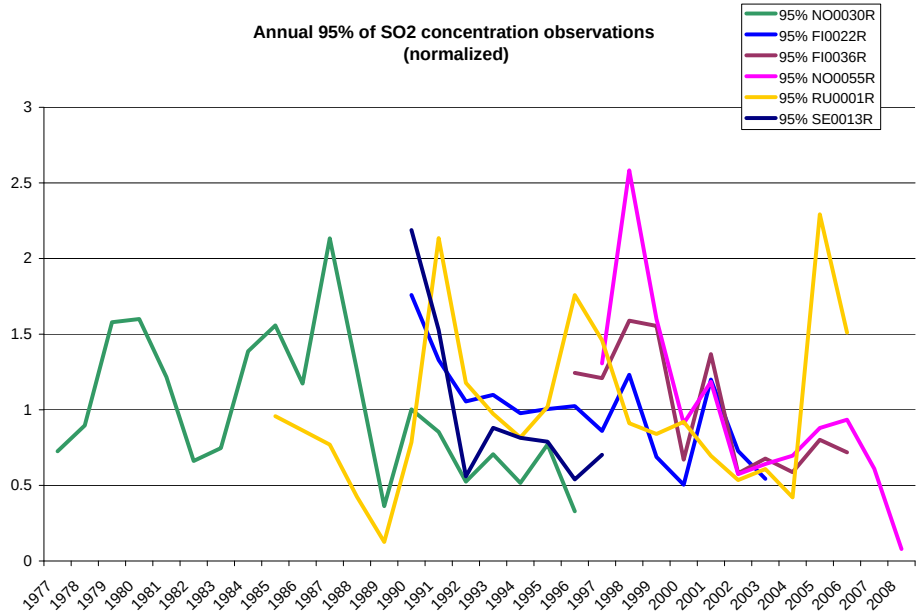

Fig. 3. Time series of the 95th percentile of the measurements of SO_2 concentration in air by the stations near Nikel, normalised to unit average.

Note:

Station Code	Name	Latitude	Longitude
NO0030R	Jergul	69°27′00″ N	24°36′00″ E
FI0022R	Oulanka	66°19′13″ N	29°24′06″ E
FI0036R	Pallas	68°00′00″ N	24°14′23″ E
NO0055R	Karasjok	69°28′00″ N	25°13′00″ E
RU0001R	Janiskoski	68°56′00″ N	28°51′00″ E
SE0013R	Esrange	67°53′00″ N	21°04′00″ E

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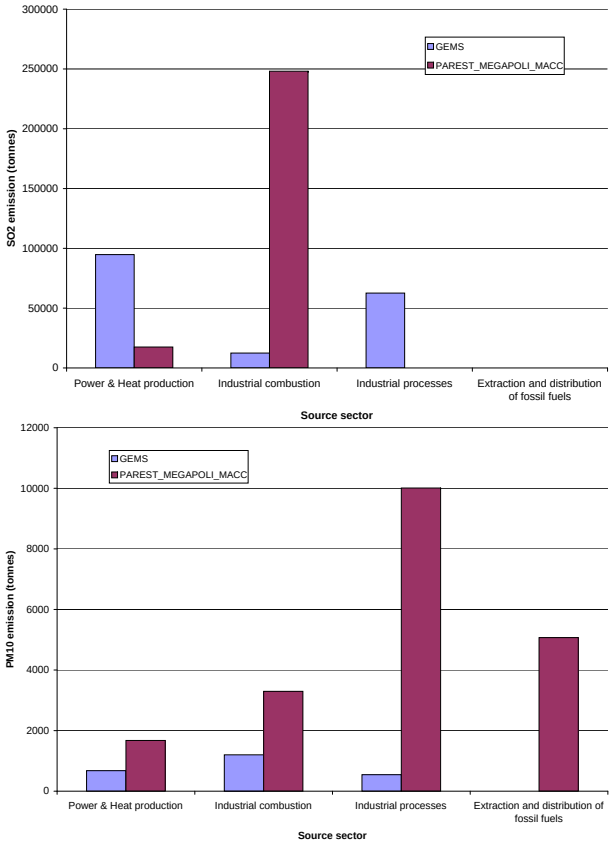

Fig. 4. Emissions of SO_2 (upper panel) and PM_{10} (lower panel) for the Kola domain in TNO-GEMS and PAREST_MEGAPOLI databases.

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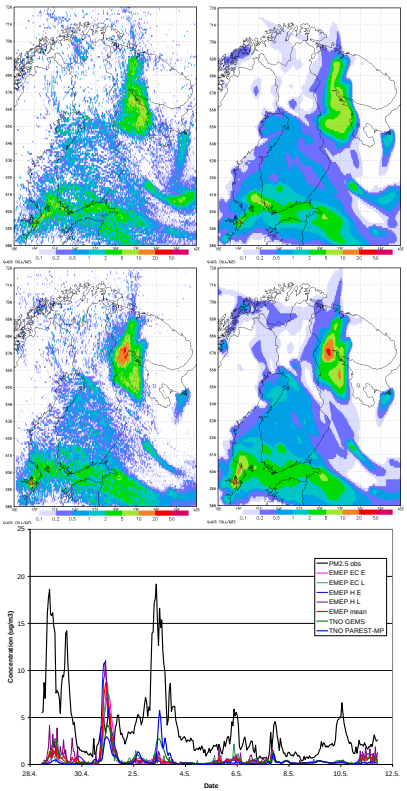

Fig. 5. Surface-level concentrations of $PM_{2.5}$, 00:00 UTC at 3 May 2003, calculated using the original EMEP emissions. Panels present the 4 members of the ensemble: **(a)** Lagrangian SILAM, HIRLAM meteo, **(b)** Eulerian SILAM, HIRLAM meteo, **(c)** Lagrangian SILAM, ECMWF meteo, **(d)** Eulerian SILAM, HIRLAM meteo, **(e)** time series for all four computations plus Eulerian SILAM with TNO-GEMS and PAREST_MEGAPOLI emissions, and Varrio $PM_{2.5}$ observations.

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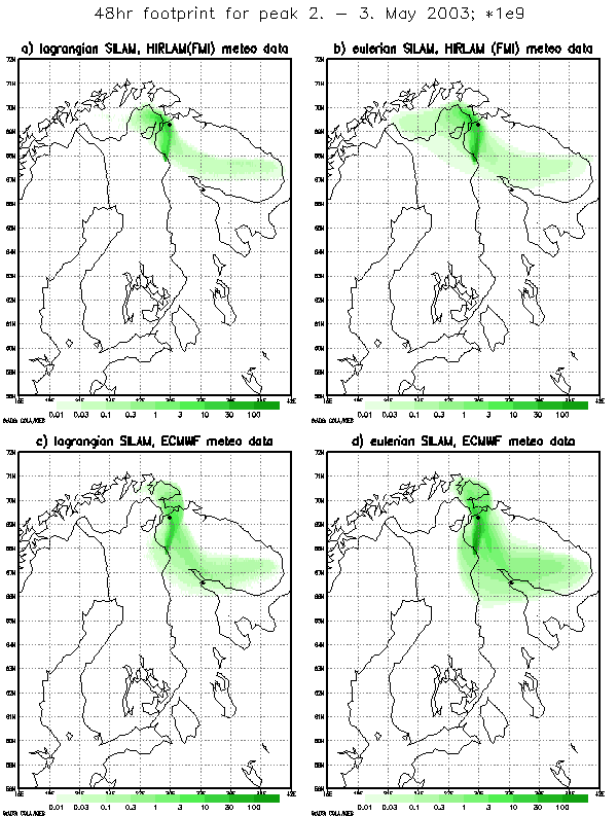

Fig. 6. A footprint of the highest peak of $PM_{2.5}$ concentration 2–3 May, 2003. Panels: **(a)** lagrangian SILAM, HIRLAM meteo, **(b)** eulerian SILAM, HIRLAM meteo, **(c)** lagrangian SILAM, ECMWF meteo, **(d)** eulerian SILAM, HIRLAM meteo. Location of the Nikel city with the metallurgy plant is marked by a dot.

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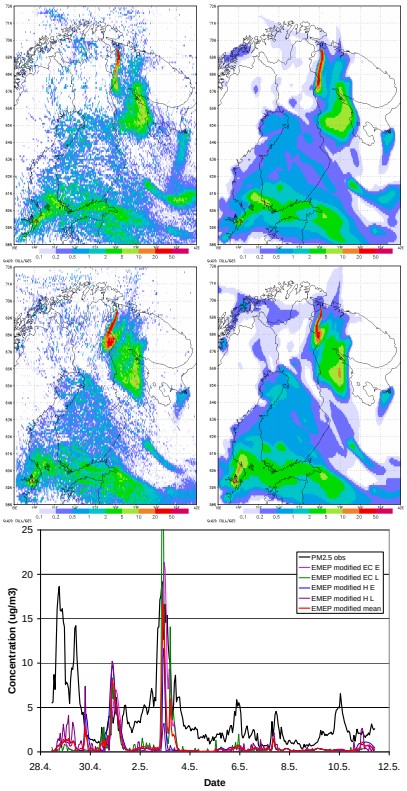

Fig. 7. Surface-level concentrations of $PM_{2.5}$, 00:00 UTC at 3 May 2003, calculated with revised EMEP emissions (a) Panels: **(a)** lagrangian SILAM, HIRLAM meteo, **(b)** eulerian SILAM, HIRLAM meteo, **(c)** lagrangian SILAM, ECMWF meteo, **(d)** eulerian SILAM, HIRLAM meteo, **(e)** time series for all four computations and Varrio $PM_{2.5}$ observations.

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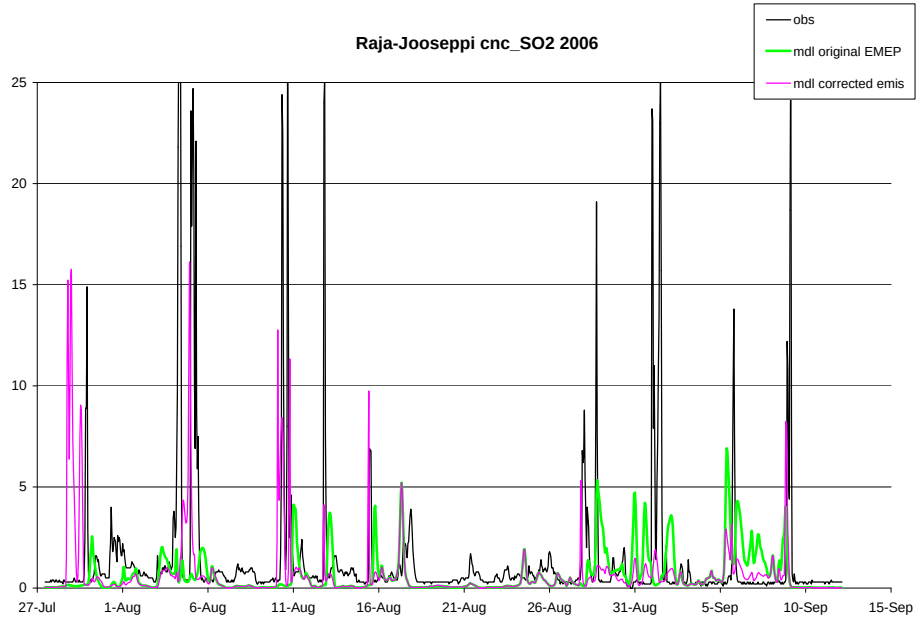

Fig. 8. Extraction of time series of modelled and measured SO_2 concentrations at Raja-Jooseppi station, 2006, unit $\mu g\,S\,m^{-3}$.

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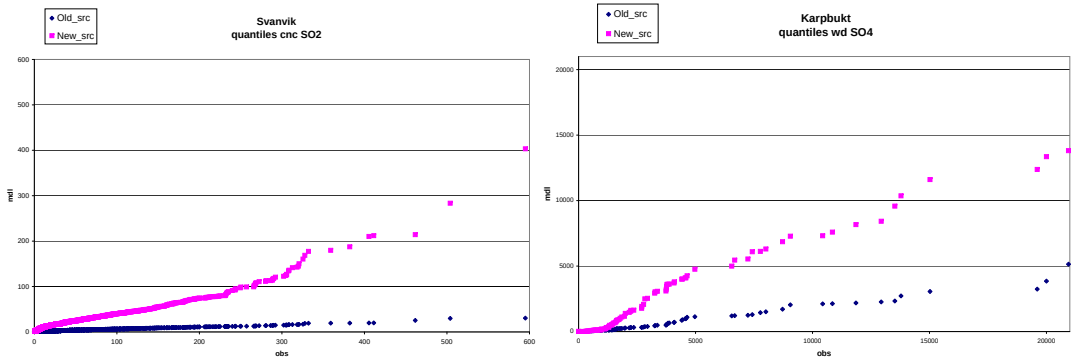

Fig. 9. Quantile charts for SILAM simulations vs. observations at the nearest sites to the Nikel plant. Unit: [μg S m^{-3}] for hourly concentration values in **(a)**; [μg S m^{-2} d^{-1}] for daily wet deposition in **(b)**.

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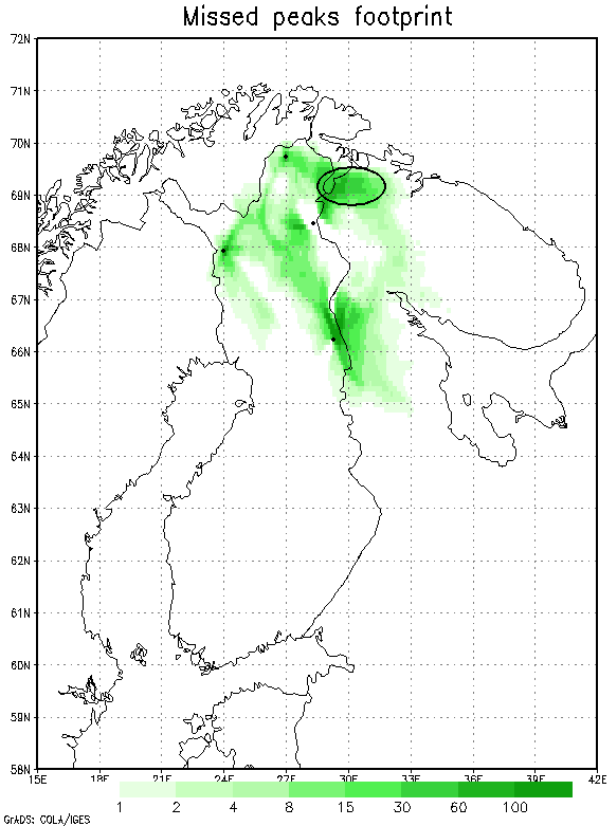

Fig. 10. Footprint of the major model-measurement differences ($c_{\mathrm{mdl}} - c_{\mathrm{obs}}$) of SO_2 concentration at monitoring sites (black dots), mean over 2006. The drawn quantity is the likelihood of the revised emission to be under-estimated. Presence of hot-spots around individual stations is an artefact originating from the low density of the observational network.

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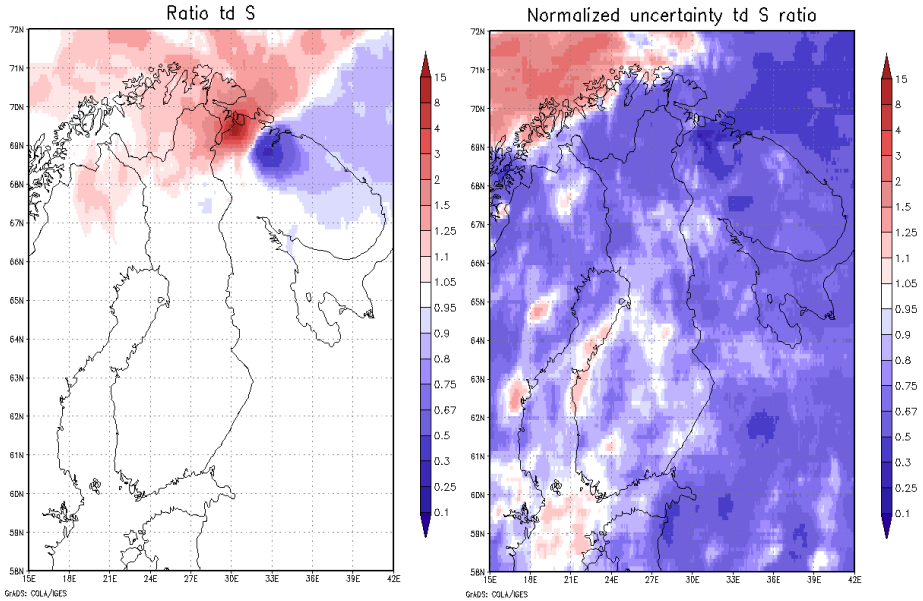

Fig. 11. Ratio of total sulphur depositions D before and after emission correction: **(a)** $D_{revised}/D_{original}$, relative units. **(b)** standard deviation of the ratio, relative to its mean. Averaged over years 2003 and 2006.

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