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

M. Prank[1], M. Sofiev[1], H. A. C. Denier van der Gon[2], M. Kaasik[3], T. M. Ruuskanen[4], and J. Kukkonen[1]

[1]Finnish Meteorological Institute, P.O. Box 503, 00101 Helsinki, Finland
[2]TNO, Princetonlaan 6, 3584 CB Utrecht, The Netherlands
[3]University of Tartu, Ülikooli 18, 50090 Tartu, Estonia
[4]University of Helsinki, Department of Physics, P.O. Box 68, 00014 Helsinki, Finland

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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 metallurgy plant was missing or strongly under-estimated in the major European emission inventories, such as EMEP, EDGAR, TNO-GEMS, and PAREST-MEGAPOLI. In some cases it was 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 smaller under-estimation of SO_2 concentrations at 8 Finnish and Norwegian observational stations. For the nearest site to the plant the 10-fold underestimation turned to a 1.5-fold over-prediction. Temporal correlation improved more moderately (up to 45% for concentrations, up to 3 times for deposition). The study demonstrates the value of a combined usage of forward and inverse ensemble modelling for source apportionment in case of limited observational data.

Correspondence to: M. Prank
(marje.prank@fmi.fi)

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 anthropogenic emissions and some natural sources (volcanoes in Italy and DMS marine fluxes), with yearly time step and ca. 50 km spatial resolution. The emission inventory is based on the reports of the European countries and the estimations of 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.

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.

These databases contain comprehensive information about European emissions but in some cases additional efforts are needed to improve the quality. In particular, several 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., 2002, 2004, 2006; Zlatev et al., 2001; Sofiev et al., 1994, 2003; Sofiev, 2000; BACC, 2008, and also the EMEP own simulations, e.g. EMEP, 2007, 2008, 2009). As shown below, one of the reasons for that is the deficiency of emission information for the Kola Peninsula.

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An emerging approach to refine emission data is 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 measurement data are available from sufficiently dense network with sufficiently high time resolution, 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 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.

In Lapland the source apportionment problem 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 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).

The goals of the current paper are to (i) demonstrate the methodology of source apportionment suitable for the case of limited observational information and highly variable pollution patterns; (ii) refine the estimates of emission and distribution of sulphur oxides (SO_2 and SO_4^{2-}) and particulate matter (PM) 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 emission sector based 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 the impact of the emission refinement 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 plants are by far the largest in the region, roughly twice larger than that of the whole Finland (Ahonen et al., 1997). The Monchegorsk and Murmansk city emissions are also rich in

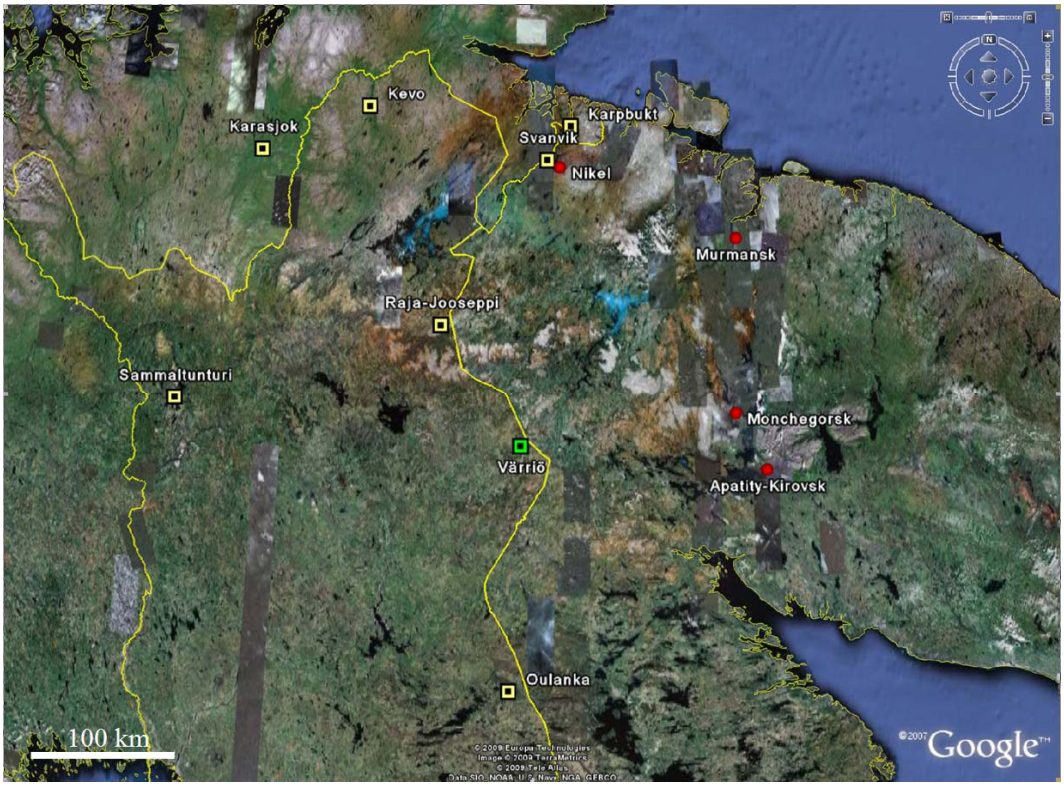

Fig. 1. Location of the major pollution sources of Kola Peninsula (red circles), the Varrio measurement station (green rectangle) and the other measurement sites (yellow rectangles).

NO_x, contrary to those of the Nikel plant, 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 PM non-industrial contributions are dispersed and originate from very different sources: road dust, sea salt, production of secondary aerosols, etc. The natural NO_x emissions around Lapland are very small and NO_x background concentration is caused by long range transport from Central and Eastern Europe (Ruuskanen et al., 2003; Virkkula et al., 2003). The natural SO_x in Lapland originate from marine DMS production, which forms a generally low background level (Ruuskanen et al., 2003; Tarrasson et al., 1995).

The best-articulated tracer for the industrial emission distribution in Lapland and Kola region is SO_2, which is also monitored by most of the observational stations of the region. The available information on other species is scarcer. Therefore, below we concentrate on the SO_x emission and project its emission sector specific modifications to particulate emission.

2.1 Evaluation of EMEP SO_2 emission data

The currently available EMEP data for Kola Peninsula reports strongly varying emission amounts and patterns for different years (see Table 1 and Fig. 2). These inconsistencies can be traced back to the evolution of the database. 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 while the patterns are assumed to be fixed and just scaled appropriately. 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 the Nikel plant belonged to two neighbouring grid-cells. For the year 1992 (the last available with 150 km resolution), over 250 kTon yr^{-1} of SO_2 emission was reported in the grid-cell covering the Nikel plant and about 30 kTon yr^{-1} attributed to Murmansk grid-cell (Sofiev, 2000).

In mid-1990s the default resolution of EMEP emission database was changed to 50×50 km and the emissions were recomputed retrospectively. That resulted in abrupt rearrangement of the emission pattern of Kola Peninsula. A strong source of SO_x was shown for 1980 in the grid-cell (48, 91) neighbouring Murmansk and for 1985 in the grid-cell

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

Gridcell		Lon	Lat	1980	1985	1990	1992	1995	2000	2003	2005	2007	original	corrected	1992 (150 × 150)
46	90	30.3	69.5	1606	588	86	66	37	29	21	7009	5551	438	110 085	
47	90	30.8	69.1	3113	480 238	162	125	69	54	41	35	42	849	849	
48	91	32.6	68.9	421 398	1072	27 815	22 347	14 146	10 179	9013	1619	781	114 916	39 121	
48	92	33.8	69.1	125 016	17 422	6214	4787	2646	2043	1560	1370	1118	34 092	240	
48	93	35.0	69.3	18 638	535	861	662	363	274	218	195	97	5083	5083	
49	91	33.1	68.5	3455	88 479	187	145	81	64	47	40	52	942	942	
50	88	30.4	67.4	5186	19 252	269	208	115	90	67	59	71	1414	1414	
50	90	32.5	67.9	16 811	1072	869	670	372	290	217	189	225	4584	4584	
50	91	33.7	68.0	12 280	17 107	652	503	280	220	163	141	180	3349	3349	
51	89	32.0	67.2	17 562	20 773	1130	902	560	410	352	331	286	4789	4789	
51	90	33.1	67.4	299 846	196 543	23 148	18 829	12 350	8846	7943	7660	6163	81 769	81 769	
51	91	34.2	67.6	88 199	1072	6747	5482	3586	2572	2303	2218	1794	24 052	24 052	
Total of Peninsula				1 070 305	863 490	71 009	56 937	35 829	26 019	22 667	21 496	17 007	291 875	291 875	507 800
150 km grid cells, aggregate from 50 km and the old dataset															
46–48	88–90	29.7	68.9	17 900	486 608	925	714	396	310	232	7192	5765	4881	114 529	253 100
46–48	91–93	33.2	69.5	675 634	21 273	35 411	28 197	17 376	12 665	10 922	3299	2096	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	98 480	98 480	193 500
49–51	91–93	34.9	68.2	115 647	113 091	8128	6547	4175	3029	2650	2522	2522	2104	31 537	31 400
ontribution of the main sources, %															
Nikel %				2	56	1	1	1	1	1	33	34	2	39	50
Murmansk %				54	2	50	50	48	49	48	15	12	54	16	6
Monchegorsk etc %				45	41	49	49	50	50	51	51	54	45	45	44

Presented data in:

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. Highlithed grid cells: green – Nikel, red – Murmansk, yellow – Monchegorsk and its surroundings.

The values from 1990–2007 mark the data recomputed after 2006.

(47, 90) neighbouring the Nikel plant location (Table 1 and Fig. 2). However, the plant itself was not represented as a source. The total emission of the Kola Peninsula stayed at similar level as in the 150 × 150 km resolving dataset. Until 2006, emission data with similar regional totals and patterns were available from EMEP for 1990s and beginning of 2000s.

In 2006 all emissions of Kola Peninsula starting from 1990 were recomputed following the latest reported data (EMEP, 2006) and appear more than an order of magnitude lower than the previous estimate and with a new distribution pattern (Table 1 and Fig. 2). For 2005, for the first time for the 50 km resolving dataset, somewhat higher emissions (compared to surrounding background level) show up in the grid cell (46, 90) containing the Nikel plant. However, the emission of that grid cell is still too low.

The changes of 2006 have not affected the projections for 2010 and 2020, which thus stayed at the previously reported levels and patterns.

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

For the following analysis we consider the problems of regional totals and the distribution patterns separately.

Considering the sharp changes since 1980s reported by Russia in the presently available version of the EMEP database, 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) and Berglen et al. (2008), the SO_2 emissions of the Nikel plant were reported around 250–300 kTon yr^{-1} until mid-1980s and reduced to ∼175 kTon 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 kTon yr^{-1} in 1992 to 380 kTon yr^{-1} in 1994.

The SO_2 concentration measurements in surrounding stations also do not support the changes shown by present EMEP data. The Svanvik measurement station in Norway reports about 2 times reduction in SO_2 annual mean concentrations from the late 1980's to beginning of 1990s (Hagen et al., 2002; Berglen et al., 2008). No significant change in SO_2 has been observed at Svanvik, Maajavri, Nikel, Viksjøfjell

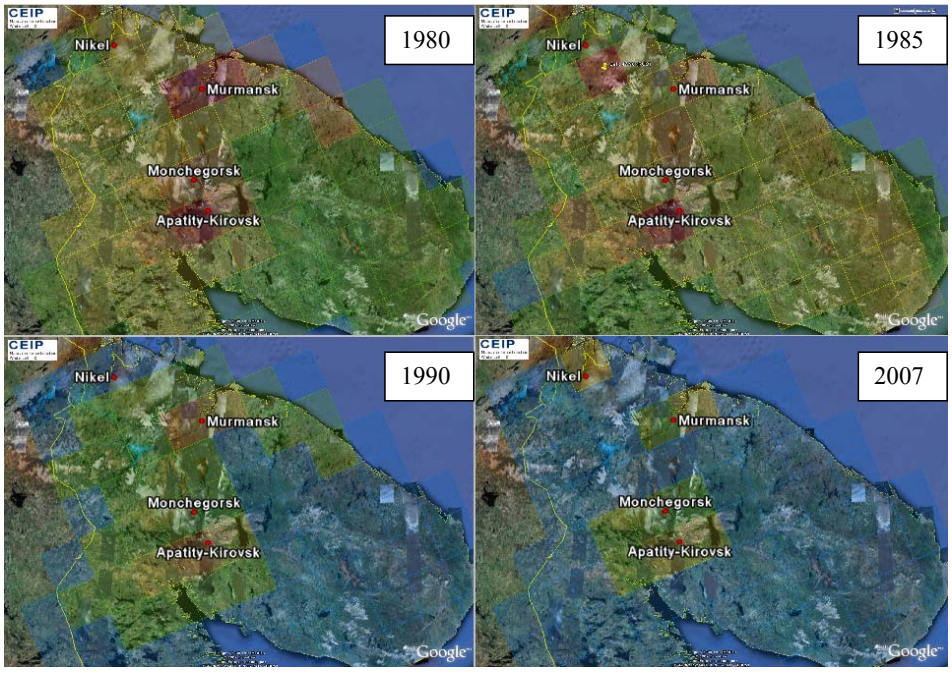

Fig. 2. The original EMEP emission for 1980–2007, WebDab status 2010. The 50km grid cells are shown with the colours reflecting the SO_x emission: blue $<=0.1$ kTon yr^{-1} SO_2, green $<=1$ kTon yr^{-1} SO_2, yellow $<=10$ kTon yr^{-1} SO_2, orange $<=100$ kTon yr^{-1} SO_2, red >100 kTon yr^{-1} SO_2.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

or Varrio stations in 1990s and 2000s (Hagen et al., 2002; Berglen et al., 2008; Ruuskanen et al., 2003; Virkkula et al., 2003). EMEP stations in Lapland also reported only gradual trends without any drastic changes during the last 20 years. 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.

2.2 Comparison of the emission inventories

There are numerous inventories of anthropogenic emission, 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 are EMEP, TNO-GEMS and PAREST-MEGAPOLI, and RETRO. The global databases, such as GEIA, EDGAR and CGEIC usually have low ($1 \times 1°$) resolution, which is insufficient for regional model applications. However, they can still be considered for comparison when it comes to regional totals.

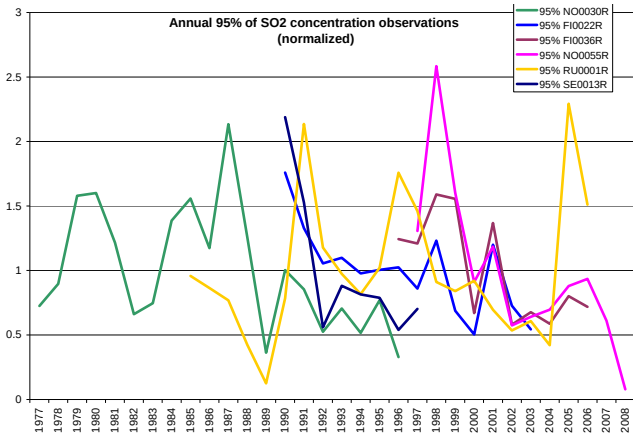

Fig. 3. Time series of the 95th percentile of the measurements of SO_2 concentration in air by the stations near Nikel, normalised to unit average.

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 kTon yr^{-1} of SO_2, 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 plant emission. However, it attributes about 80% of the emissions of the Peninsula to the Monchegorsk area and only about 15% (22 kTon yr^{-1} of SO_2) to the Nikel plant region, which is doubtful. For instance, Boyd et al. (2009) mentioned 300 kTon yr^{-1} (with a reference to Zientek et al., 1994) as a total-Kola industrial SO_2 emission with ∼70% attributed to the Nikel plant region.

The step from TNO-GEMS to PAREST-MEGAPOLI 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 kTon yr^{-1} of SO_2 (Fig. 4). However, the emission distribution still attributes only 19% of it (52.5 kTon yr^{-1}) 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 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 by a factor of 1.7 between these years. However, the emission pattern still does not show any significant emissions at the Nikel plant 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 simultaneously have (i) sufficient resolution, (ii) correct distribution of the major sources, (iii) 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 prior to 2006, 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 kTon yr^{-1} 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.

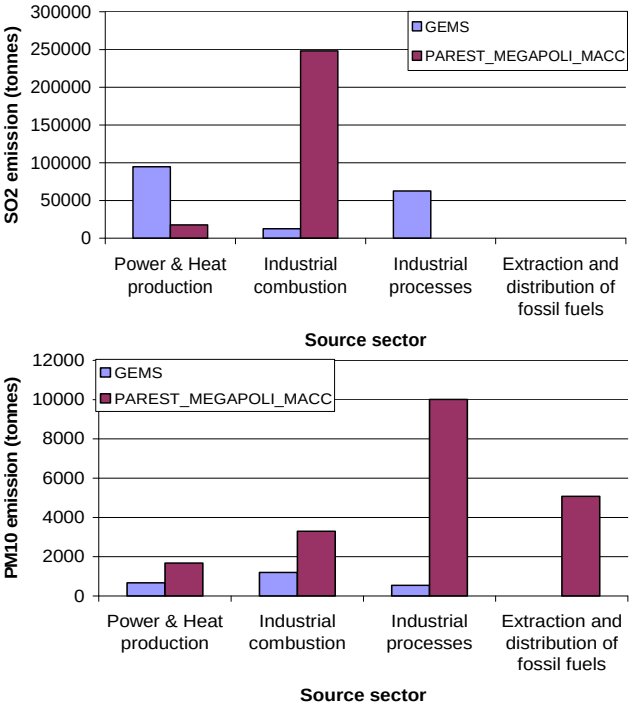

Fig. 4. Emissions of SO_2 (upper panel) and PM_{10} (lower panel) for the Kola domain in TNO-GEMS and PAREST-MEGAPOLI databases.

The datasets with the Kola emission totals close to 300 kTon yr^{-1} 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 plant emission. The next task of this work is, therefore, to correct this error. The emission data for year 2003, downloaded before 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 kTon yr^{-1} of SO_2), 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

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}, $PM_{2.5}$, PMcoarse, CO, POPs, HMs,	$50 \times 50\,km^2$ $150 \times 150\,km^2$	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	$1° \times 1°$	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	$1° \times 1°$	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	$1° \times 1°$	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.	$0.5° \times 0.5°$	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 Assessment Model (TEAM)
TNO-GEMS and PAREST-MEGAPOLI	NO_x, SO_2, CO, NMVOC, CH_4, NH_3, PM_{10}, $PM_{2.5}$	$0.25° \times 0.125°$	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).

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 of year 1990								
lat/lon	28	59	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	61 300	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 ton SO_2/yr								
Emissions for year 1995								
lat/lon	28	59	30	31	32	33	34	35
70	719	749	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 ton SO_2/yr								

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 plant was misplaced to near-Murmansk.

Since the time trends of the emission in 2000s are uncertain and probably not significant, we used the 2003 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, which must be relocated to Nikel plant place. 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 the Nikel plant, 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.

The above correction does not reposition the Nikel town emission, neither it reflects the details of the infrastructure,

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

Species, sector	EMEP 2000		Modified dataset	
	Nikel	Murmansk	Nikel	Murmansk
SO_x, S1	0	31 588	31 58	0
SO_x, S2	17	3020	1509	1528
SO_x, S3	418	114 164	76 989	37 596
$PM_{2.5}$, 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 Atmospheric 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.

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 the Nikel plant region, 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 an indication of industrial aerosols.

Apart from the Varrio campaign, the long-term analysis has been performed using the information from national networks of Norway (Aas et al., 2008) and Finland. For the purpose of the study, we used seven stations located close to the Nikel plant. Six of them monitor SO_2 concentrations in air, one reports SO_4^{2-} in aerosol, and three report SO_4^{2-} in precipitation, which were converted to wet deposition. The deposition was chosen as a target quantity of the study because it is the cause of acidification, the primary impact of SO_x in Lapland. None of the 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 transport (Sofiev et al., 2006b) incorporates a high-precision iterative 3-D 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. The verification of the model has been performed within the scope of EU-GEMS project (http://www.ecmwf.int/gems) and is continued on a routine basis within the EU-MACC (http://www.gmes-atmosphere.eu). According to the outcome, an overall bias of SILAM in Europe for SO_2 is within the limit of $\sim 1\,\mu g\,S\,m^{-3}$.

All simulations were performed with 0.1° horizontal and 6 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 contributions of Central and North-Western Europe were taken into account by nesting the domain into the SILAM European simulations, which cover the area 17° W–38° E and 33° N–72° N.

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° horizontal resolution. Simulations for 2006 were driven by the fields of the regional HIRLAM RCR system with 0.2° horizontal resolution. Both datasets have 3-hour time steps.

For the long term simulations the Eulerian kernel of SILAM was used. Simulations for the period of the Varrio 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.

The input emission, depending on the specific run, was either the EMEP-original dataset for 2003 (downloaded before the 2006 change) or the same dataset with the above described corrections. For the long-term analyses the computations were made for only sulphur compounds. For the Varrio campaign, total PM concentrations were computed, consisting of primary PM, sea-salt and secondary inorganic particles (sulphates, nitrates, and ammonium).

The SO_2/SO_4^{2-} split of the SO_x emission was assumed to be 95%/5% by volume for all the runs. All emission was considered in the model grid (no point sources). As insufficient amount of information is known about the Nikel stacks, no dynamic plume-rise computations were made and the emission was vertically distributed generally following the EMEP-recommended profile (Simpson et al., 2003). However, the Kola stacks are quite low: the highest one in Nikel region is about 160 m, the tallest one in Monchegorsk is about 200 m (Tuovinen et al., 1993). Therefore, the EMEP vertical emission profiles for SNAP sectors S1 and S3 were lowered by 150 m so that in average about half of emission was injected within 200 m up from the stack top. The related uncertainty is discussed in Sect. 5.

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 the Nikel plant 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

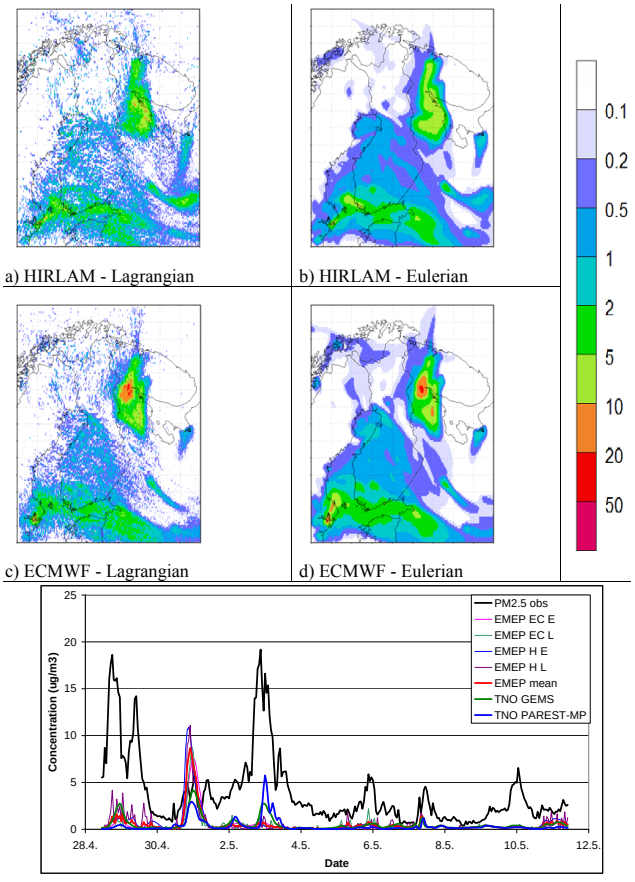

Fig. 5. Surface-level concentrations of $PM_{2.5}$, 00:00 at 3 May 2003, calculated using the original EMEP emissions. Panels present the 4 members of the ensemble: **(a)** Lagrangian SILAM, HIRLAM meteo, **(b)** Eulerian SILAM, HIRLAM meteo, **(c)** Lagrangian SILAM, ECMWF meteo, **(d)** Eulerian SILAM,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

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 for SO_x, NO_x, NH_x and primary PM, 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-MEGAPOLI emission data reproduced the peak time (Fig. 5, lowest panel) but showed strong underestimation of its value compared to the observations.

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).

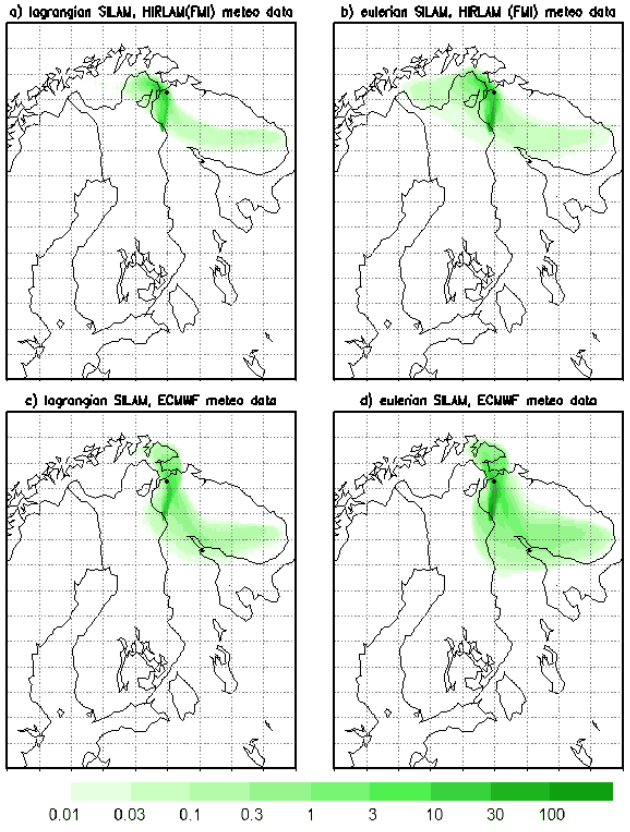

Fig. 6. A footprint of the highest peak of $PM_{2.5}$ concentration 2–3 May 2003. Panels: **(a)** Lagrangian SILAM, HIRLAM meteo, **(b)** Eulerian SILAM, HIRLAM meteo, **(c)** Lagrangian SILAM, ECMWF meteo, **(d)** Eulerian SILAM, ECMWF meteo. Location of the Nikel plant is marked by a dot.

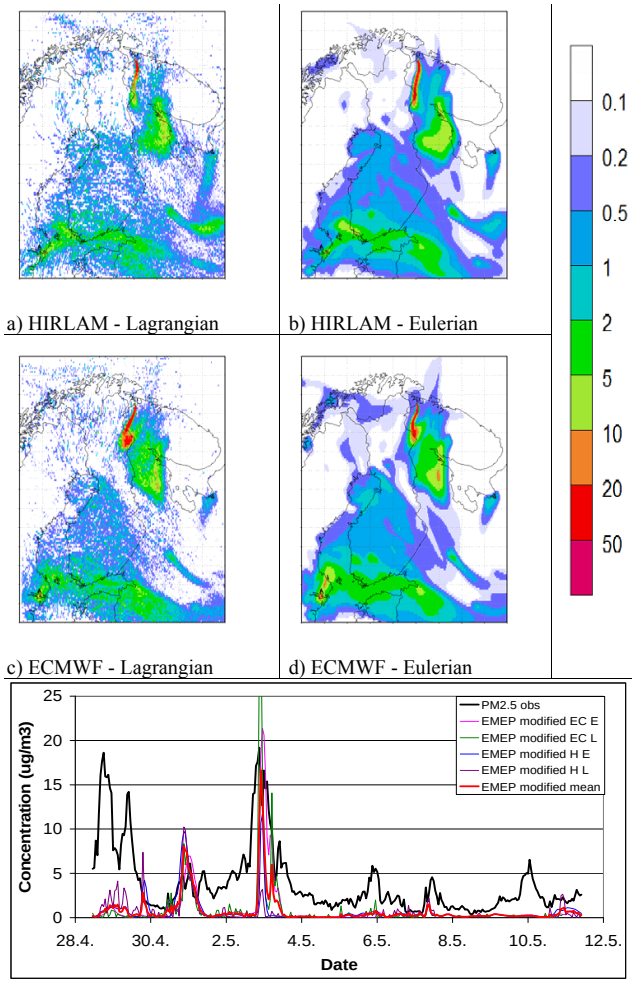

Fig. 7. Surface-level concentrations of $PM_{2.5}$, 00:00 at 3 May 2003, calculated with revised EMEP emissions. Panels: **(a)** Lagrangian SILAM, HIRLAM meteo, **(b)** Eulerian SILAM, HIRLAM meteo, **(c)** Lagrangian SILAM, ECMWF meteo, **(d)** Eulerian SILAM, ECMWF meteo, **(e)** time series for all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

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

The SILAM simulations with the revised PM and SO_x emissions (NO_x and NH_x emission was not changed) produced significantly different results. In all 4 ensemble runs the high $PM_{2.5}$ concentrations reached Varrio 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 total $PM_{2.5}$ 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 times 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 emission of the Nikel plant: long-term evaluation

The above described 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 emission correction to the model-measurement comparison, (ii) to re-check the suggested regional totals, (iii) to estimate how close the new distribution is to the real emission pattern in the region, (iv) to estimate

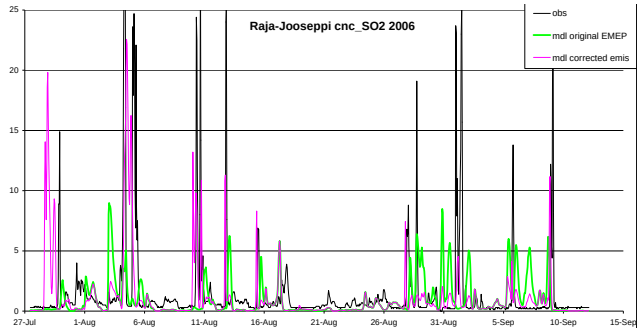

Fig. 8. Extraction of time series of modelled and measured SO_2 concentrations at Raja-Jooseppi station, 2006, [$\mu g\, SO_2\, m^{-3}$].

the impact of the correction onto the modelled acid deposition in the region. In order to stress the contribution of strong sources, only SO_2 concentrations higher than 1 $\mu g\, m^{-3}$ were taken into account in the model-measurement comparison.

In general, the new emission distribution leads to a significant improvement of the model-measurement agreement (Table 5). However, the impact is not homogeneous over the region. The influence on the predicted mean values and variability quickly decreases with the distance from the plant and depends on the site location with respect to both Murmansk and Nikel: from the 16-fold increase of the mean values (Svanvik, 9.6 km away from the Nikel plant) down to practically no impact at Oulanka (345 km from the Nikel plant, 334 km from Murmansk). The concentrations are still under-estimated at all the sites, apart from the closest site to the plant (Svanvik), where some 30% of over-estimation is reported. Improvement of the temporal correlation is moderate for the concentrations (up to ∼45%) but strong for wet deposition (up to 3 times). This is 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 modelled concentrations with the revised emissions. The over-estimation of the average concentrations at Svanvik apparently comes from the moderate concentrations (from 1 $\mu g\, S\, m^{-3}$ up to 100 $\mu g\, m^{-3}$). The frequency of episodes with 30–70 $\mu g\, S\, m^{-3}$ is over-stated, while the cases with concentrations 100–250 $\mu g\, S\, m^{-3}$ are under-estimated.

Quantile analysis for wet deposition is more uncertain due to weekly resolution of the observations. However, the tendency is that new source generates somewhat too high deposition near Nikel – up to 1.5 times. The apparent understatement of Svanvik deposition is due to just two extremely high observed episodes not fully reproduced by the model. It is only the farthest located site – Karasjok – where the wet deposition is still under-estimated.

To investigate whether the SO_2 emission of the Nikel plant is still underestimated in the revised data, we computed the

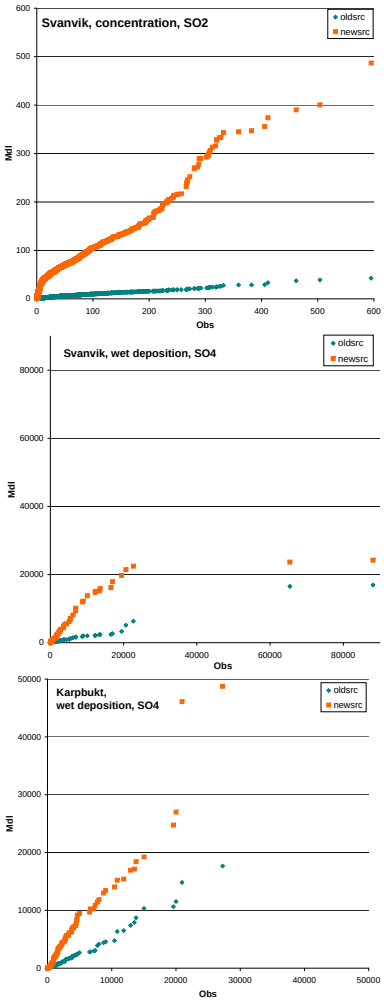

Fig. 9. Quantile charts for SILAM simulations vs. observations at the nearest sites to the Nikel plant. First panel: hourly concentration values in Svanvik [mug $SO_2\, m^{-3}$], second and third panel: weekly wet deposition in Svanvik and Karpbukt [$\mu g\, S\, m^{-2}\, week^{-1}$].

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.

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	
			Observed	Modelled Original source	Modelled Corrected source	Observed	Modelled Original source	Modelled Corrected source	Original source	Corrected source
Svanvik	cnc_so2	hour	6.34	0.67	10.93	25.60	2.31	26.77	0.33	0.26
Sammaltunturi	cnc_so2	hour	0.59	0.12	0.18	2.10	0.57	0.81	0.35	0.41
Raja-jooseppi	cnc_so2	hour	1.26	0.37	0.57	3.80	1.27	2.40	0.24	0.34
Oulanka	cnc_so2	hour	0.72	0.46	0.45	2.11	1.46	1.35	0.30	0.33
Kevo	cnc_so2	hour	0.98	0.25	0.57	3.80	1.01	2.36	0.49	0.50
Karasjok	cnc_so2	day	0.78	0.12	0.20	2.55	0.51	0.73	0.35	0.40
Karasjok	cnc_so4	day	2.18	0.18	0.22	2.81	0.63	0.66	0.39	0.42
Karasjok	wd_so4	day	189.50	134.50	144.00	556.49	590.74	617.52	0.40	0.46
Svanvik	wd_so4	week	8875.00	1874.00	7572.00	16 022.00	3458.00	7364.20	0.09	0.19
Karpbukt	wd_so4	week	3907.00	2091.00	6169.00	5138.60	3196.40	8248.70	0.21	0.65

Notations:

cnc_SO2 and cnc_SO4 – concentrations of SO_2 and SO_4 in air or in aerosol [$\mu g\,S\,m^{-3}$]

wd_SO4 – wet deposition of sulphates [$\mu g\,S\,m^{-2}\,day^{-1}$] or [$\mu g\,S\,m^{-2}\,week^{-1}$]

The comparison is provided for the periods with concentrations of SO_2 exceeding 1 $\mu g\,SO_2\,m^{-3}$.

The overlap of the yearly-mean footprints of the significant differences ($c_{model} - c_{observed}$) 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 analysis of the recent changes of the EMEP emission database are based on indirect considerations, such as the model-based source apportionment, land use analysis and 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 estimate for Kola Peninsula presented in the EMEP datasets generated before 2006 is close to the actual emission. Indeed, 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 emissions of 1990s can be used as estimates for 2000s. The changes during that period were not more than a factor of 2. It was also supported by the limited 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.

There are, however, uncertainties embedded in the 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 ∼20%. Finally, the specific uncertainty due to sparse station network in the region located upwind from the sources can be roughly estimated from the number of episodes N_{epi} when a particular site registered the plume from the plant. The standard deviation (StDev) of the mean over these episodes is proportional to $1/\sqrt{N_{epi}}$. With typical N_{epi} ∼30–40 per year, relative StDev ∼15%. This value is the lower estimate of the corresponding uncertainty. 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.

Uncertainties of the revised emission pattern can be summarised as follows.

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

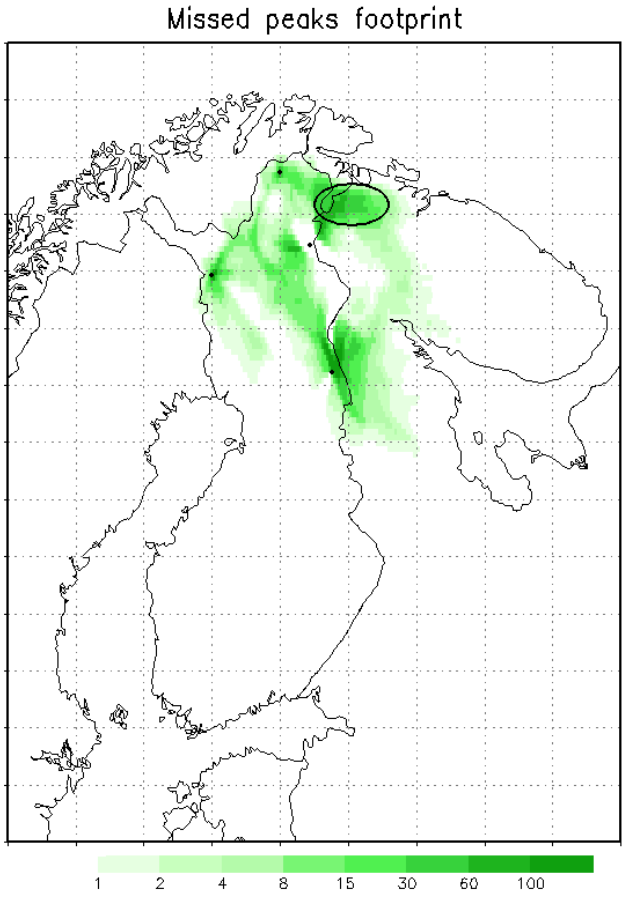

Fig. 10. Footprint of the major model-measurement differences ($c_{\mathrm{mdl}} - c_{\mathrm{obs}}$) of SO_2 concentration at monitoring sites (black dots), mean over 2006. The drawn quantity is the likelihood of the revised emission to be under-estimated. Presence of hot-spots around individual stations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

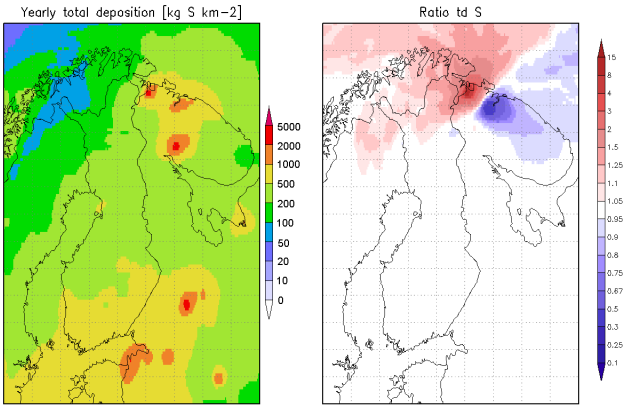

Fig. 11. Left panel: Total annual sulphur deposition after emission correction, [mg S m^{-2}], right panel: ratio of total sulphur depositions D before and after emission correction $D_{\mathrm{revised}}/D_{\mathrm{original}}$, [relative units], mean over the years 2003 and 2006.

the new input. Therefore, the emission in the Murmansk area is still probably over-estimated.

Some sources of background concentrations (<1 µg S m^{-3}) are not included in the computations (seen as missing background in Fig. 8). These are mainly the Arctic DMS marine and ship traffic emissions outside the SILAM European modelling domain. However, DMS emission is low in the Arctic seas (Tarrasson et al., 1995; Korhonen et al., 2008), as well as the ship traffic not covered by the computation grid (http://www.ceip.at/emission-data-webdab).

The model still underestimates the SO_2 peak concentrations at all stations except Svanvik by about a factor of 2 but variation is large. Wet deposition near the plant is over-stated up to 1.5 times but not farther away (Karasjok site) where it is still under-stated. Two model parameters of importance in this regard are the vertical diffusivity (K_z) and the scavenging ratio. Computations of K_z in stable stratification are challenging for models and recent evaluation of the SILAM diagnostic module confirmed it (Sofiev et al., 2010). The corresponding uncertainty is inter-connected with that of the vertical dilution of the emitted plumes and affects the surface concentrations. The relocated emission of the Nikel plant is represented as a point source with the injection height approximated by the adapted EMEP profiles. However, some part of the emission probably comes from the surrounding area and infrastructure of the Nikel town. The sensitivity study based on the model computations with high (EMEP-standard profile) and low (from stack top up to 150 m) injection showed that the near-source concentrations can be changed up to a few times between these two extremes, turning them e.g. at Svanvik from a factor of 2 under- to a factor of 2 over-estimation.

The SO_x scavenging ratio, after Galperin (1989), decreases for high SO_x concentration due to saturation of the rain droplets. However, the specific parameterization was obtained for average European conditions and may have higher uncertainties in Lapland.

Combined effect of the uncertainties in the vertical SO_x distribution, scavenging efficiency, and the emission total can probably explain the above-reported differences in the model scores for surface concentrations and wet depositions. It also shows that the refinement of the Kola emission pattern via source apportionment based on generally available data has reached its limit. Further refinement has to be based on different methodology, e.g. the bottom-up inventory.

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 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 (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 3-fold only.

5 Conclusions

An analysis of the different emission inventories, the observational campaigns and regular AQ monitoring in Northern Lapland, land-use, and sectoral emission split, allowed detecting problems with the total emission of Kola Peninsula and its distribution in the EMEP and other existing inventories.

A sudden 15–20-fold drop of the emission totals of SO_x and PM in Kola Peninsula in 1990s, reported to EMEP since 2006, is not supported by the long term observations, which rather suggest fairly constant emission throughout 1990s and 2000s – as in the previous versions of the database. Thus, the Kola Peninsula stays as the largest source of SO_x in Northern Europe being second only to Norilsk industrial region in Northern Eurasia.

In the prior-2006 EMEP data the emission of the Nikel metallurgy plant was found to be mis-allocated to the Murmansk city region.

A refined emission for Kola Peninsula is suggested, keeping the totals at the level of pre-2006 EMEP estimates and redistributing the industrial part of the emission from the city of Murmansk to the location of the Nikel metallurgy plant.

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 long-term model-measurement comparison showed sharp reduction of the model under-estimation (up to slight over-estimation in the nearest vicinity to the plant) and improvement of the temporal correlation coefficient (up to 3 times).

The impact of the emission redistribution on the deposition of sulphur compounds can reach an order of magnitude but becomes small when the distance from the sources exceeds the spatial scale of the emission redistribution, i.e., the distance between the Nikel plant and Murmansk.

Further refinement of the Kola Peninsula emissions with activity-based emission assessment methods could be recommended.

It is demonstrated that a combination of several types of analyses of emission and observational data with forward and adjoint ensemble modelling allows addressing the source apportionment problems even in case of strongly limited observational data.

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