ATMOSPHERIC MODELLING OF HEAVY METAL POLLUTION IN EUROPE:
Further development and evaluation of the MSCE-HM model

DRAFT

I. Ilyin, O. Rozovskaya, V. Sokovykh, O. Travnikov
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INTRODUCTION

The MSCE-HM chemical transport model has been developed for assessment of heavy metal airborne pollution in Europe and support of the review, extension and implementation of the Protocol on Heavy Metals. The model performance have been reviewed at the EMEP Task Force on Measurements and Modelling meeting in Zagreb and TFMM Workshop on model review in Moscow in 2005. Along with the recognition that the model is “appropriate for the operational modelling of heavy metals at the regional scale”, a number of recommendations aimed at further model improvement have been formulated. Particularly, the recommendations included the following issues:

- Extensive evaluation of input meteorological data for regional heavy metal modelling;
- Movement to the ECMWF input data with 1°×1° spatial resolution for the meteorological fields pre-processing;
- Development of the model parameterization for heavy metal re-suspension from soil and seawater;
- Extension of the MSCE-HM model parameterisation for the second priority heavy metals (As, Cr, Ni, Cu, Zn and Se);
- Improvement of the model description of atmospheric removal processes;
- Extension of the heavy metal hemispheric model to the global scale;
- Inclusion of a shallow lowest model layer;
- Investigation of mercury dry deposition to forests;
- Further research and improvement of the model description of mercury chemical transformations in the atmosphere.

Besides, the Workshop concluded that official emission data for heavy metals suffered from significant uncertainties and are of limited value in terms of model applications. Therefore further improvement of official emission data was highly recommended.

First results of MSC-E activities on implementation of the above mentioned recommendations have been reflected in the EMEP/MSC-E technical report [Gusev at al., 2006]. Particularly, an approach to validate input meteorological fields prepared for atmospheric transport modelling has been elaborated and described. A tentative parameterization of wind re-suspension of particle-bound heavy metals from soil and seawater has been developed. Pilot results of the assessment of lead and cadmium re-suspension contribution to the pollution levels in Europe have been described. Extension of the MSCE-HM model parameterisation for the second priority heavy metals (As, Cr, Ni, Cu, Zn and Se) has been reported along with modelling results of their long-range atmospheric transport in Europe. The model performance has been evaluated using different emission inventories (official emissions data and ESPRIM expert estimates).

This report continues consideration of the MSCE-HM model development and evaluation in accordance with the recommendations of the Workshop on model review. Particular attention is paid to comparison and analysis of discrepancies of different anthropogenic emissions inventories. Three available datasets of heavy metals anthropogenic emissions in Europe in 2000 are taken into consideration: the EMEP official emissions data, TNO and ESPRIM non-official expert estimates. The analysis is focused on emissions data for lead and cadmium since these two heavy metals give the major concern from the modelling point of view [Ilyin and Travnikov, 2005]. The emission
inventories were analyzed by intercomparison of annual emission totals from individual European countries and contributions of key source categories to national emissions.

Further development of the quality control system of input meteorological data is described and illustrated. The main goal of the system is performing quantitative quality control and verification of meteorological fields prepared by the meteorological pre-processor (MM5) for atmospheric transport modelling on the routine basis. Statistical algorithms characterizing reliability of meteorological data through comparison of their spatial and temporal variation as well as frequency distributions with reference datasets are presented. Besides, progress of MSC-E activities aimed at moving to ECMWF meteorological fields with higher spatial resolution ($1^\circ \times 1^\circ$) as input information for the data preprocessing is reported. First results of comparison of meteorological parameters obtained using this dataset with those based on data from the NCEP/DOE reanalysis are considered.

The model parameterization of wind re-suspension of particle-bound heavy metals from soil and seawater is further developed. Particularly, the effect of soil characteristics (soil texture, size distribution of soil grains etc.) on dust production and suspension are taken into account. A global dataset of soil properties is used to characterize wind erosion and re-suspension of heavy metals from different soil types in Europe. Besides, some special effects influencing dust mobilization (such as wind drag partition, inhibition by soil moisture, the Owen effect) are introduced or revised in the model. Full description of the revised dust production scheme is presented along with estimates of heavy metal re-suspension in Europe. Plans for further research are discussed.

A special attention of this report is paid to evaluation of modelling results against measurements. One of the inferences of the Workshop on model review was the conclusion that MSCE-HM model tends to underestimate measured concentrations of heavy metals in air and precipitation when using the official emissions data for modelling. Similar underestimation was demonstrated by other models participated in the intercomparison studies. An improvement of the agreement was achieved by taking into consideration the wind re-suspension process, however, some underestimation still remains [Gusev et al., 2006]. To analyse the problem the model results are compared with results of the CMAQ model – one of well-developed contemporary chemical transport models. Besides, the underestimation is analysed using measurements data with high (daily or weekly) temporal resolution. It allows evaluation of the model performance to reproduce both short-term and long-term variation of measured values and reveal possible reasons of the underestimation.
Chapter 1

COMPARISON OF LEAD AND CADMIUM EUROPEAN EMISSION INVENTORIES

In this Chapter three available European emission inventories for lead and cadmium for the year 2000 are compared. The first inventory is the official emission data submitted to the UN ECE Secretariat by the Parties to the LRTAP convention by March 2006 (hereafter denoted as EMEP data). The officially reported emission data are available from the EMEP emission database WEBDAB (http://webdab.emep.int). The second inventory is emission estimates prepared by TNO (hereafter denoted as TNO data). The TNO data are mostly based on the EMEP official data but corrected and supplemented with non-official expert estimates if no country official data were available or data were qualified to be unrealistic [Denier van der Gon et al., 2005]. The third emission inventory developed within the framework of EU ESPREME project mostly consists of non-official expert estimates (hereafter denoted as ESPREME data). The ESPREME emission dataset is available at the ESPREME website (http://espreme.ier.uni-stuttgart.de). Brief description and general comparison of the mentioned above inventories can be found in [Vestreng et al., 2006]. In this study we perform quantitative comparison of different estimates of lead and cadmium emissions from individual European countries in order to reveal major differences and present data for national experts analysis. Comparison of key source categories of lead and cadmium emissions from most European countries is presented in Annex A.

1.1 Lead

National totals

Total annual emissions of lead from European countries in 2000 according to EMEP official data along with TNO and ESPREME estimates are presented in Table 1.1. Only countries that officially reported national emissions data were included. In addition, ratios of TNO and ESPREME estimates to EMEP data for each country are presented for facilitation of the analysis.
Table 1.1. Total annual emissions of lead from European countries in 2000, t/y

<table>
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<tr>
<th>Country</th>
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<th>ESPREME</th>
<th>TNO/EMEP</th>
<th>ESPREME/EMEP</th>
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<td>647.5</td>
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<td>192.8</td>
<td>571.1</td>
<td>1.18</td>
<td>3.50</td>
</tr>
</tbody>
</table>

* European part of the country

Figure 1.1 illustrates comparison of national emission totals for the above-mentioned countries from different emission inventories for lead. The largest deviation between the EMEP emission data and ESPREME and TNO estimates (about 37 and 23 times, respectively) is noted for Germany. There is also substantial deviation (more than 4 times) between national totals from EMEP and ESPREME estimates for Austria, Denmark, Ireland, Luxembourg, the Netherlands, Norway and Sweden. TNO data suggest higher emissions for Romania, Russian Federation and Spain as compared to the other inventories. For Italy, large absolute discrepancy (about 1000 t/y) between the EMEP official data and estimates by ESPREME is observed. Portugal reported significantly higher total emissions than both TNO and ESPREME estimates.
Deviations of the TNO and ESPREME estimates from the EMEP official data are clearly illustrated by scatter plots in Fig. 1.2. In general, TNO and ESPREME estimates demonstrate good correlation with the official data (the correlation coefficients are 0.94 and 0.92, respectively). As seen from Fig. 1.2a, the TNO estimates mostly coincide with the EMEP data. Significant deviations are observed only for a few countries (Germany, Switzerland, Luxemburg, Portugal and Latvia). On the other hand, ESPREME estimates of lead emissions significantly exceed EMEP data for many countries (Fig. 1.2b). For about 45% of countries the discrepancy exceeds a factor of two. However, it should be noted that the largest deviations are characteristics of countries with moderate and low emissions (e.g. the Netherlands, Sweden, Ireland, Austria, Denmark etc.). The exceptions are Germany, the United Kingdom and Portugal.

**Key source analysis for selected countries**

To provide insight into the reasons of discrepancies between different inventories of lead emission the simple comparison of emissions from various source categories was performed. In order to compare the official EMEP data and estimates by TNO and ESPREME, the Nomenclature for Reporting (NFR)
source categories adopted within EMEP [UN ECE, 2002] and the source categories utilized in TNO inventory [Denier van der Gon et al., 2005] have been aggregated to the key source categories defined within the ESPREME inventory. Results of the comparison of lead emissions from various key source categories for countries reported the official data are presented in Annex A. Examples of the key source categories analysis for countries with high absolute and/or relative discrepancies of total national emission estimates from different inventories are presented below.

Figure 1.3 demonstrates comparison of key source categories of lead emissions in Germany. As can be seen from the figure the EMEP data are dramatically lower than the data from TNO and ESPREME because of lack of lead emissions from industrial processes and gasoline combustion. Contribution of gasoline combustion to lead emissions is evaluated by the ESPREME inventory even for countries where consumption of leaded gasoline is banned. It is expected that even unleaded gasoline (i.e. gasoline without lead additives) contains lead as an impurity due to its content of crude oil [Vestreng et al., 2006]. Beside contribution of gasoline combustion ESPREME data expect more significant contribution of industrial/residential combustion and public power production to lead emission in Germany.

All three emission inventories expect gasoline combustion as prevailing source category of lead emissions in Italy (Fig. 1.4). However, the ESPREME estimates contain much larger contribution of industrial sources than two other inventories. It leads to approximately twice higher total emission estimate from this country in comparison with EMEP and TNO values.

An opposite situation with discrepancies between different inventories is presented in Figs. 1.5 and 1.6 for Portugal and Latvia, respectively. In both cases the TNO and ESPREME data significantly underestimate the EMEP official data. According to EMEP inventory industrial/residential combustion is the dominant source category of lead emissions in Portugal. On the other hand, this source category is negligible according to TNO and ESPREME estimates. Contribution of gasoline combustion is comparable in EMEP and ESPREME data whereas TNO almost neglect this category.

Lead emissions in Latvia mostly consist of emissions from industrial sources according to EMEP official data (Fig. 1.6). Two other emissions inventories assess considerably lower absolute contribution of this source category with some addition of gasoline combustion and industrial/residential combustion. All this leads to several times lower estimates of total lead emissions in Latvia by TNO and ESPREME in comparison with EMEP data.
TNO inventory demonstrates highest estimates of lead emissions in comparison with other datasets for such countries as Romania and Russian Federation (Figs. 1.7 and 1.8). All inventories agree that gasoline combustion is the prevailing source category in these countries. However, its absolute contribution is the largest in TNO estimates. Besides, TNO assessed much higher contribution of public power production to lead emissions in Russian Federation. The EMEP official data expect significantly smaller emissions from industrial processes than those from ESPREME and TNO inventories.
1.2. Cadmium

National totals

Total annual emissions of cadmium from European countries in 2000 according to EMEP official data along with TNO and ESPREME estimates are presented in Table 1.2. Only countries that officially reported national emissions data were included into the comparison. In addition, ratios of TNO and ESPREME estimates to EMEP data for each country are presented for facilitation of the analysis.

Table 1.2. Total annual emissions of cadmium from European countries in 2000, t/y

<table>
<thead>
<tr>
<th>Country</th>
<th>EMEP</th>
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<th>ESPREME</th>
<th>TNO/EMEP</th>
<th>ESPREME/EMEP</th>
</tr>
</thead>
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</table>

* European part of the country

The comparison of national totals for above-mentioned countries from different emission inventories for cadmium is illustrated in Fig. 1.9. Similarly to lead, the largest deviation between EMEP emission totals and estimates by ESPREME and TNO (about 22 and 7 times, respectively) is noted for Germany. Besides, there are significant discrepancies between different estimates of national totals of cadmium emissions in France, Italy, Russian Federation and the United Kingdom. Large relative deviations are also characteristics of Austria, Denmark, the Netherlands and Sweden. In most cases emissions estimates from the ESPREME inventory exceed both the EMEP official data and the TNO estimates. Nevertheless, for some countries, such as Poland, Romania, Slovakia, EMEP data and/or TNO estimates suggest higher emission values in comparison with ESPREME data.
Scatter plots in Fig. 1.10 illustrate relative deviations of the TNO and ESPREME estimates of cadmium emissions from the EMEP data. In general, TNO and ESPREME estimates demonstrate significant correlation with the official data (the correlation coefficients are 0.95 and 0.72, respectively). Good agreement of the TNO and EMEP values (Fig. 1.10a) can be explained by the fact that the TNO dataset is mostly based on the official data. Significant deviations are observed only for three countries (Germany, Romania, and Republic of Moldova). ESPREME estimates of cadmium emissions are significantly higher (more than twice) those from other inventories for 60% of countries. As it was mentioned above the deviations are characteristics of both countries with large total emission (Italy, France, Germany, the United Kingdom) and countries with smaller emissions.

Fig. 1.10. Scatter plots of comparison of different emission inventories for cadmium in 2000: (a) TNO vs. EMEP official data; (b) ESPREME vs. EMEP official data Red solid line delineates 1:1 ratio, green dashed lines show deviation by a factor of two

Key source analysis for selected countries

In order to analyze discrepancies between different inventories of cadmium emission the simple comparison of emissions from key source categories was performed using the same methodology as described above for lead. Examples of the analysis for some countries with high absolute and/or
relative discrepancies of cadmium emission estimates are presented below. Comparison of different estimates of the key source categories of cadmium emissions for all countries is given in Annex A.

Figure 1.11 demonstrates EMEP, TNO, and ESPREME estimates of cadmium key source categories in Germany. TNO and ESPREME inventories expect much higher total cadmium emission in Germany than that from the EMEP official data. The reason for that is the official data does not include cadmium emissions from industrial processes. Contribution of industrial/residential combustion is also negligible in comparison with the TNO and ESPREME data. Besides, the ESPREME estimates expect higher emissions from public power production.

Relative discrepancies between EMEP data and other estimates of cadmium emission in Russian Federation are lower but the absolute differences are very large: 30 t/y and 60 t/y for TNO and ESPREME data, respectively. The official data expect much lower contribution of industrial processes and public power production to cadmium emissions than that from two other inventories. According to the TNO estimates public power production is prevailing source category of cadmium emissions in this country. On the other hand, industrial processes and industrial/residential combustion dominate in the ESPREME inventory.

Figures 1.13 and 1.14 illustrate somewhat different ratio between cadmium emission estimates by various inventories for Portugal and Spain, respectively. In both cases the TNO estimates are lower than the EMEP official data, whereas the ESPREME ones exceed them considerably. Contribution of key source categories to the national totals also significantly varies between the inventories. Industrial and residential combustion is expected to be the dominant source category in Portugal cadmium emissions according to the EMEP data. The ESPREME inventory expects also significant contribution of industrial processes, whereas contribution of these two categories is much lower in the TNO inventory. All three datasets assess similar contribution of public power production to cadmium emissions in Spain. However, contributions of industrial/residential combustion and industrial processes vary considerably. Besides, the EMEP data expect some cadmium emissions from gasoline combustion.
The TNO estimates fairly agree with the EMEP official data for total cadmium emissions in Poland (Fig. 1.15). However, they deviate in the estimates of the key source categories contribution. Particularly, the EMEP data expect much higher contribution of industrial/residential combustion. The ESPREME inventory gives somewhat lower estimates of total cadmium emission in Poland expecting also significant contribution of public power production. The TNO estimates of total cadmium emission from Republic of Moldova twice exceed those from both the official data and the ESPREME estimates (Fig. 1.16). The reason for that is large contribution of gasoline combustion to cadmium emissions in this country expected in the TNO inventory, which is absent in other datasets.

To summarize the analysis presented above it can be concluded that the EMEP official data and TNO and ESPREME estimates of lead and cadmium emissions well correlates with each other. However, significant differences are observed for many countries, particularly, between the EMEP and ESPREME datasets. In general the ESPREME inventory presents more higher emission estimates than two other datasets. The deviation is more pronounced for cadmium emissions. Significant discrepancies take place also in contribution of source categories to national emissions of lead and cadmium estimated by different emission inventories. It could be partly explained by different definitions of emission sectors and source categories accepted in the inventories. Participation of national experts is highly required for in depth analysis of discrepancies of the available emission inventories and improvement of emissions data.
Chapter 2

SYSTEM OF QUALITY CONTROL OF METEOROLOGICAL DATA

Quality of atmospheric transport model results strongly depends on quality of input data, in particular, on gridded meteorological fields. Meteorological data for MSC-E regional-scale transport model are generated by MM5 modelling system, using NCEP Re-analysis or ECMWF analyses as input data. Technical description of this system is available in [Grell et al., 1995]. Parameterizations of physical processes and technical aspects of the use of MM5 in MSC-E are overviewed in [Travnikov and Ilyin, 2005]. To be sure that meteorological data used to run the transport model are correct, the data should be verified. Besides, the need of verification of meteorological data acquired by the models of MSC-E was reflected by TFMM [ECE/EB.AIR/GE.1/2006/4].

MSC-E has started developing a system of quality control of meteorological data. In the MSC-E technical report [Gusev et al., 2006] brief description of this system was presented, and some preliminary results for 2000 were overviewed. This report deals with more detailed description of the system. Besides, some results of the validation for entire period of 1990 – 2004 are presented.

The main idea of validation of meteorological data is based on comparison of the processed parameters with similar parameters from independent (“reference”) data set. The validation was focused on three aspects. First of all, we analyzed spatial variability of meteorological fields reproduced by MM5. Then we studied its ability to capture temporal variations. Finally, we compared statistical distributions of meteorological parameters produced by MM5 with the reference meteorological data. Similarity of spatial distribution of meteorological fields from MM5 and the reference data or of their temporal trends is characterized by the correlation coefficient:

$$R_{\text{corr}} = \frac{\sum_{i=1}^{N} (M_i - \overline{M})(O_i - \overline{O})}{\sqrt{\sum_{i=1}^{N} (M_i - \overline{M})^2 \sum_{i=1}^{N} (O_i - \overline{O})^2}}, \quad \overline{M} = \frac{1}{N} \sum_{i=1}^{N} M_i, \quad \overline{O} = \frac{1}{N} \sum_{i=1}^{N} O_i,$$

where $M$ stands for MM5 result, $O$ – for the reference data, $N$ – number of values, $\overline{O}$ and $\overline{M}$ are averaged reference and MM5-generated values.

In order to describe relative difference between MM5 and the reference data the relative bias was calculated:

$$B_R = \frac{\overline{M} - \overline{O}}{\overline{O}} \cdot 100\%$$

The absolute difference was characterized by the absolute bias:

$$B_A = \frac{1}{N} \sum_{i=1}^{N} (M_i - O_i) = \overline{M} - \overline{O}$$

However, in case of precipitation, $\overline{O}$ and $\overline{M}$ were replaced by more common accumulated values (e.g. annual or monthly sums).

The work of the verification system is demonstrated on the example of 2000, using 2.5°x 2.5° data of ECMWF re-analysis data (ERA-40) as reference information [http://www.ecmwf.int/research/era/]. Besides, some meteorological parameters modelled by MM5 were compared with results of
NCEP/DOE reanalysis-II project [Kanamitsu et al., 2002]. In future we are planning to use the data of observations carried out at synoptic stations for this task.

In addition to this, precipitation amounts generated by MM5 were compared with precipitation data obtained in the framework of GPCP project [http://cics.umd.edu/~yin/GPCP/, Rudolf et al., 1994]. Spatial resolution of these data is 1°x 1°, and temporal resolution is one day [Huffman et al., 2001]. GPCP precipitation data are based on rain gauge observations. In regions where measurement network is scarce (e.g., over oceans), information from satellites was used. Therefore, GPCP data seem to be especially relevant for purpose of evaluation of modelled precipitation, especially for regions with dense network of meteorological stations. These GPCP data are available since 1996.

Validation of meteorological data involved analysis of variety of input parameters, such as wind velocity magnitude and its components, precipitation, humidity, air and surface temperatures. In this report we focused on the two parameters crucial for modelling of atmospheric transport and deposition of metals and POPs – wind velocity and precipitation.

2.1. Spatial and temporal variability

Wind velocity

Spatial distribution of parameters characterizing absolute and relative difference (absolute and relative bias, respectively) between wind velocity magnitude generated by MM5 and ERA-40 are demonstrated in Fig. 2.1 and 2.2. The statistics were computed only for those EMEP grid cells which coincide with latitude-longitude grid points with step 2.5°x2.5°. This approach was used in order avoid interpolation of reference meteorological parameters from 2.5°x2.5° grid to finer EMEP 50x50 km grid, since the interpolation would give rise of additional uncertainties. As seen from the figures, over the Atlantic and East of Europe MM5 tends to generate wind velocities mostly 0.5 - 1.5 m/s (or 10-25%) lower compared to ERA-40. Over Scandinavian Peninsula the winds can be lower even by more than 25%. Over southern part of Europe, Turkey, and west of Scandinavia wind magnitudes computed by MM5 are higher than those of ERA-40 by 0.5 – 1.5 m/s (10-25% higher) compared to ERA-40.

![Fig. 2.1. Absolute bias (a), relative bias (b) between MM5 and ERA-40 near-surface wind velocities and temporal correlation coefficient (c, 6-hour time step) for near-surface wind velocity over EMEP domain in 2000. MM5 vs. ERA-40.](image-url)
Coefficient of correlation ($R_c$) was used to characterize similarity of temporal variability of meteorological parameters modelled by MM5 and provided by ERA-40. $R_c$ between annual 6-hour time series over every latitude-longitude point of EMEP domain with 2.5° step in 2000 are shown in Fig. 2.1. Over major part of EMEP modelling domain the coefficients are larger than 0.8. Lower $R_c$ was obtained over elevated terrain (e.g., the Balkans, the Caucasus, the Alps). This can be partly connected with coarser resolution of ERA-40 data, which leads to smoother and less detailed relief.

Similarity of spatial distributions of wind magnitudes of MM5 and ERA-40 is confirmed by significant spatial correlation coefficient between monthly mean wind magnitudes (Fig. 2.2). In all months of the considered period it was always above 0.75. In winter months the correlation is typically above 0.9, while in summer it is mostly around 0.80 – 0.85.

**Fig. 2.2.** Spatial coefficients of correlation between monthly-mean near-surface wind velocity magnitudes provided by MM5 and ERA-40

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**Precipitation amounts**

Comparison of precipitation annual sums from MM5 and ERA-40 reveals that over most of EMEP domain the difference between precipitation fits ±150 mm/year range (Fig. 2.3a). Over the Balkans, Iberian Peninsula and west of Scandinavia MM5 produced precipitation amounts 300 mm (25 – 50%) higher compared to ERA-40. Large relative bias between the two precipitation data sets can also be mentioned for Germany, east of the Ukraine, Spain, west of Kazakhstan and Caspian region. In these regions the relative difference can exceed 50%.

**Fig. 2.3.** Absolute bias (a) and relative bias (b) between MM5 and ERA-40 surface annual precipitation sum over EMEP domain in and 2000
Comparison with annual precipitation sums available in GPCP data set shows that over Norwegian Sea, central part of Europe, Scandinavia, southern part of Iberian Peninsula and the Black Sea region MM5 produces precipitation amounts lower than GPCP by 100 – 300 mm or even more (Fig. 2.4). Relative bias between MM5 precipitation and GPCP in these regions is less then ~50%. Over the Atlantic and the Arctic oceans MM5 precipitation significantly (100 – 300 mm or even more) exceeds GPCP precipitation. The Arctic is the region where relative bias between the two data sets is the highest and exceeds 50%. However, it also worth mentioning that precipitation measurement data over the Arctic is scarce, and thus GPCP precipitation there were basically retrieved from satellite data, and not estimated from real rain gauge measurements.

![Fig. 2.4. Absolute bias (a) and relative bias (b) between MM5 and GPCP surface annual precipitation sum over EMEP domain in 2000.](image)

Over major part of EMEP domain coefficient of correlation between 10-days sums of MM5 and ERA-40 precipitation is higher than 0.5 (Fig. 2.5). The highest coefficients (> 0.8) are seen over the Atlantic. Low temporal correlation is often takes place over regions with complex terrain (the Caucasus, the Balkans, the Alps etc.).

Spatial distribution of temporal correlation coefficients between GPCP and MM5 precipitation differs from that between ERA-40 and MM5 precipitation (Fig. 2.5). The lowest correlation was found for the oceanic part of the Arctic, which can be partly explained by the uncertainties of precipitation derived from satellite information. In the mid latitudes the coefficient is generally significant (> 0.5), while in the south of the EMEP domain it is relatively low (from < 0.3 to 0.5), especially over land areas.

![Fig. 2.5. Temporal coefficients of correlation between annual precipitation sums provided by MM5 and ERA-40 for entire 2000, with 10-day time step](image)
Spatial correlation coefficients between monthly sums of precipitation from MM5 and from reference data sets demonstrate strong seasonal variations (Fig. 2.6). Higher correlation (0.8 – 0.9) was obtained for winter seasons, while lower (0.55 – 0.7) – for summer. Lower correlation for summer can be explained by the fact that precipitation originating from convective cloudiness occur more often in summer then in winter. Compared to large-scale precipitation, modelling of convective precipitation is more challenging problem, because they are characterized by smaller spatial scales and shorter duration, and hence, not easily reproduced explicitly by models. That is why modelled summer-time precipitation are typically characterized by less accuracy than winter-time ones.

Fig. 2.6. Spatial coefficients of correlation between monthly-mean near-surface precipitation sums provided by MM5 and ERA-40 or GPCP

2.2. Frequency distribution functions

Another way to characterize ability of meteorological driver to reproduce meteorological fields is to compare frequency distributions of parameters generated by the driver and those available in reference data set.

Comparison of frequency distribution functions for near-surface wind velocity and air temperature in 2000 is shown in Fig. 2.7. The functions shown in Fig. 2.7 are probability distribution of the 6-hour parameter values over EMEP domain during a whole 2000 year. Thus, it characterizes both temporal and spatial variability of wind velocity and air temperature.

Frequency distribution functions for MM5 and ERA-40 wind velocity look are quite similar. Maximum of distribution for ERA-40 wind velocity took place at around 2.5 m/s, and for MM5 – at around 3 – 3.5 m/s. Near-surface wind velocities higher than 20 m/s are rare in both data sets. The curve for ERA-40 is slightly higher than that for MM5 near wings and lower near the maximum. This implies that wind ERA-40 velocities are characterized by somewhat higher variability.

ERA-40 contains more values with air temperature around 0 °C (Fig. 2.7). For both sets of data almost all near-surface temperature values range from - 50 to 40 °C. Frequency distribution function for MM5 is higher than that for ERA-40 in −20 - 0°C temperature range, and lower in 0 - 5°C range. This means that near-surface air described by MM5 tends to be slightly colder compared to ERA-40.
Fig. 2.7. Distribution functions for near-surface wind velocity (a) and air temperature (b) generated by MM5 and available in ERA-40 data.

2.3. Use of ECMWF analyses data

Following the recommendations of TFMM, MSC-E has started to move to utilization of the analysis data provided by ECMWF. Global analysis data with 1°x 1° spatial resolution were retrieved from ECMWF data storage system, and were processed by MM5. Detailed analysis of the meteorological fields derived on the base of ECMWF data has not been performed yet. In particular, it is necessary to compare meteorological parameters based on ECMWF and NCEP initial data. A few examples of this comparison are demonstrated below.

Precipitation sums in January 2005 based on ECMWF and NCEP meteorology and relative difference between them are shown in Fig. 2.8. As seen, the fields of precipitation look quite similar. Over most of Europe the relative difference between the two fields is within ±25%. However, over Mediterranean region precipitation based on ECMWF are significantly (more than 50%) lower than those based on NCEP data. The opposite situation takes place over north of Scandinavia, central Italy, Hungary, east of Turkey. As seen from Fig. 2.8a and b, the precipitation amounts based on ECMWF data are characterized by higher spatial variability compared to those based on Reanalysis-II. This can be partly connected with finer spatial resolution of ECMWF data compared to NCEP/DOE.

Fig. 2.8. Precipitation amounts in January, 2005 computed by MM5 on the base of ECMWF(a), NCEP/DOE Reanalysis-II (b), and relative difference between them (c), estimated as: (ECMWF-NCEP)/NCEPx100%.

Monthly mean wind magnitudes based on ECMWF and NCEP data and relative difference between them are shown in Fig. 2.9. Over major part of EMEP domain the difference in wind magnitudes lies within ±10% range. The exceptions are some regions in Mediterranean and western coast of Europe.
Changes in input meteorological data can lead to changes in modelled pollution levels. In order to explain their reasons, differences between meteorological data should be studied in more details. This task is planed as MSC-E future activity.

**Further activities**

The system of quality control of regional-scale meteorological input data has been developed and applied. The most important input meteorological parameters have been compared with those from the reference data set (ECMWF ERA-40 archive). The verification of meteorological data was focused on the analysis of spatial patterns, temporal variability and frequency distribution of meteorological parameters. Besides, first results of generating meteorological input data on the base of ECMWF analyses with 1°x1° resolution were overviewed. Further activities regarding the supply of MSC-E with regional-scale meteorological data include the following tasks:

1. Complete transition from NCEP/DOE Re-analyses to ECMWF analyses data
2. Carry out detailed comparison of pre-processed meteorological data based on NCEP/DOE Re-analysis and ECMWF analysis
3. Perform quality control of input meteorological information on the routine basis
4. Where possible, make use of observational data for the purposes of evaluation of meteorological data.
Chapter 3

RE-SUSPENSION OF PARTICLE-BOUND HEAVY METALS

Wind re-suspension of particle-bound heavy metals (like lead and cadmium) from soil and seawater appears to be important process affecting ambient concentration and deposition of these pollutants, particularly, in areas with low direct anthropogenic emissions. Recent data demonstrate significant decrease of lead and cadmium emissions in Europe during last decade [ECE/EB.AIR/WG.5/2006/2]. However, long-term historic emissions of these metals during previous century from industrial processes, fuel combustion, road transport etc. followed by atmospheric dispersion and deposition to the ground resulted in their accumulation in the surface soils all over Europe. Aeolian erosion of bare soils or human disturbed areas leads to suspension of dust particles containing heavy metals into the atmosphere. This process supplements current anthropogenic emissions and can partly explain discrepancy between sharp trends of emission reduction and moderate decrease of measured heavy metal concentrations in precipitation [Ilyin and Travnikov, 2005].

Pilot parameterization of heavy metal re-suspension developed for the MSCE-HM model along with tentative estimates of the re-suspension effect on pollution levels in Europe were described in [Gusev et al., 2006]. This section contains description of further development and improvement of the heavy metal re-suspension scheme. Particularly, effect of soil characteristics (soil texture, size distribution of soil grains etc.) on dust production and suspension are taken into account. Besides, some special effects influencing dust mobilization (such as wind drag partition, inhibition by soil moisture, the Owen effect) are introduced or revised in the model.

3.1. Saltation

In mineral dust production models the process of wind erosion and suspension of dust aerosol from the ground is commonly parameterized as combination of two major processes: saltation and sandblasting [e.g. Gomes et al., 2003; Zender et al., 2003; Gong et al., 2003]. The first process (saltation) presents horizontal movement of large soil aggregates driven by wind stress. These aggregates are too heavy to be directly suspended by wind in usual conditions. Instead, they are moved by wind stress close to the surface jumping from one place to another. When the saltating aggregates impact the ground they can eject much smaller particles (few micrometers), which can be easily suspended by wind and transported far away from the source region. This process is called the sandblasting.

The saltation process is characterized by the critical wind stress value, over which movement of soil particles can be initiated. This critical wind stress can be described by the threshold wind friction velocity, which depends on the soil particle size, soil wetness, and protection of the erodible soil by roughness elements (drag partitioning). In order to characterize this threshold friction velocity (\( U'_t \)) in the model we used a simplified empirically based parameterization proposed by Marticorena and Bergametti [1995]:

\[
U'_t = \begin{cases} 
\frac{0.129\kappa}{(1.928Re^{0.092} - 1)^{0.5}}, & \text{Re} \leq 10 \\
0.129\kappa(1 - 0.858\exp(-0.0617(\text{Re} - 10))), & \text{Re} > 10 
\end{cases}
\]

(3.1)

where \( \kappa = \frac{D_s}{\rho_a} \left( \frac{\rho_s g + 6 \cdot 10^{-7}}{D_s^{5/2}} \right) \), \( \text{Re} = 1.755D_s^{1.56} + 0.38 \).

Here \( D_s \) is the soil particle size, \( \rho_a \) and \( \rho_s \) are air and soil mass densities, respectively; \( g \) is the gravity acceleration.
The threshold value increases for very small and very large particles and has a minimum corresponding approximately to 75 μm.

The saltation threshold is affected by soil moisture content. Soil water retention (because of interparticle capillary and molecular adsorption forces) leads to cohesion of soil particles and increase of the wind erosion threshold. In the first version of the re-suspension scheme we used a simplified approach to account for the soil moisture effect suggested by Grini et al. [2005]. It is based on rainfall events and implements the following assumptions:

- The dust production is stopped if precipitation during the last 24 hours exceeds 0.5 mm.
- The period without the dust production (in days) is equal to precipitation amount (in mm) during the last 24 hours.
- The dust production is resumed if no rain has fallen in the last 5 days.

A more sound parameterization of the threshold friction velocity dependence on soil wetness was proposed by Fécan et al. [1999] based on empirical data. According to this work the threshold is a function of gravimetric soil moisture and clay content in soil.

\[
\frac{(U_t')}{{U_t^*}} = \begin{cases} 
1 & , w < w' \\
1 + 1.21(w - w')^{0.68} & , w > w'
\end{cases}
\]  

(3.2)

where \( w \) is gravimetric soil moisture; and the critical value \( w' \) is a function of soil clay content:

\[ w' = 0.0014(\% \text{clay})^2 + 0.17(\% \text{clay}). \]

Figure 3.1 illustrates dependence of the saltation threshold ratio \( ((U_t')_{\text{wet}} / U_t^* ) \) on soil moisture for different values of clay content in soil (0%, 20%, and 40%) corresponding roughly to sandy, loamy and clay soils as calculated using Eq. (3.2). As seen from the figure, the higher of clay content in soil the larger critical value of soil moisture (\( w' \)) from which the saltation threshold friction velocity becomes increasing. Besides, increase of the saltation threshold at fixed soil moisture is more significant for soils with lower clay content. It can be explained by balance between competitive cohesion forces in different soil types. Sand is characterized by prevailing interparticle capillary forces whereas contribution of much weaker water adsorption forces increases in more hydroscopic clay soils. Thus, wet soil cohesion is more effective for sandy soils.

Under natural conditions wind stress is commonly sharing between erodible and non-erodible surfaces. It leads to decrease of actual friction velocity describing wind momentum transfer to the erodible surface and initiation of the saltation process. Extend of the wind drag partition depends both on size and density of non-erodible roughness elements. In the current dust suspension scheme we apply parameterization based on the drag partition scheme developed by Shao and Yang [2005]. According to this scheme the total drag is partitioned into a pressure drag on the roughness elements \( \tau_p \), skin drag on the surface of the roughness elements \( \tau_b \), and skin drag on the underlying surface \( \tau_s \):

\[
\tau = \tau_p + \tau_b + \tau_s
\]

(3.3)
Only the drag on the underlying surface is responsible for wind erosion and dust suspension. Following Shao and Yang [2005] relative contribution of the surface drag can be presented as:

\[
\frac{r_s}{r} = \left(1 - \frac{\beta \lambda_{ef}}{1 + \beta \lambda_{ef}}\right) \exp(-5\eta),
\]

(3.4)

where \(\beta = C_d/C_0\) is the ratio of the drag coefficient for isolated roughness elements to that for bare surface; \(\lambda_{ef}\) is effective frontal area index representing the effect of mutual sheltering of roughness elements defined as:

\[
\lambda_{ef} = \frac{\lambda}{(1 - \eta)^{1.0}} \exp\left(-\frac{6\lambda}{(1 - \eta)^{1.0}}\right);
\]

(3.5)

\(\lambda\) and \(\eta\) are the frontal area index and skin area index characterizing density and form of roughness elements, respectively.

The frontal area index is generally defined as \(\lambda = nbh\); where \(b\) and \(h\) are the characteristic width and height of roughness elements, and \(n\) is the number of roughness elements per unit surface area. The value \(\sigma = \eta/\lambda\) (called as the aspect ratio of roughness elements) characterizes average ratio of basal area to profile area of the roughness elements. Marticorena et al. [2006] measured dimensions of different obstacles in arid and semi-arid areas and obtained average aspect ratios close to 2. Thus, we set \(\eta = 2\lambda\) in the dust suspension scheme.

Wind friction velocity is defined as \(U' = \sqrt{r_s/\rho_a}\), therefore ratio of friction velocity to erodible surface to the whole friction velocity can be calculated as follows:

\[
\frac{(U')_s}{U'} = \frac{r_s}{r} = \frac{1 - \beta \lambda_{ef}}{1 + \beta \lambda_{ef}} \exp(-5\lambda).
\]

(3.6)

Figure 3.2 shows dependence of the fraction of wind friction velocity related to smooth erodible surface on density of roughness elements (frontal area index) for several values of the drag coefficients ratio. The fraction of smooth friction velocity quickly decreases while roughness density increases. Values of the drag coefficients ratio typically vary from 100 to 200 for different types of rough surfaces [Shao and Yang, 2005]. As seen from the figure the fraction of smooth friction velocity only slightly depends on this parameter. Therefore a fixed value \(\beta = 150\) is accepted in the scheme. The frontal area index significantly varies for different rough surfaces and land cover types. According to field measurements and conditions of wind tunnel experiments [Marticorena et al., 2006; Shao and Yang, 2005] this parameter commonly ranges from 0.001 to 0.2. In tentative calculations we accepted the following values of the frontal area index: 0.01 for bare land and urban areas, 0.002 for arable land during cultivation period. However, more extensive analysis is required for attribution the frontal area index to different land cover categories and different locations.

Another process affecting wind erosion and saltation of soil particles is the Owen effect [Owen, 1964] responsible for positive feedback of saltation on wind friction velocity. Saltating soil aggregates interact with underlying surface and transfer part of wind momentum to the ground. It leads to increase of the wind shear stress, which can be expressed in increase of the wind friction velocity.
Gillette et al. [1998] derived a simple formula for this effect using field measurements at dry Owens Lake:

\[ \Delta U^* = 0.003(U_{10} - U_{10,t})^2, \]  

(3.7)

where \( \Delta U^* \) (in m/s) is increase of wind friction velocity due to the Owen effect; \( U_{10} \) and \( U_{10,t} \) (in m/s) are the wind speed and the threshold wind speed at 10 m height, respectively.

Once the wind friction velocity exceeds the threshold value, the vertically integrated size-resolved saltation flux is given in the following form [Gomes et al., 2003]:

\[ F_s(D_s) = \frac{K\rho_s}{g} \left( (U^*)^2 - U_1^2 \right) \left( (U^*)^2 + U_1^2 \right)^2. \]  

(3.8)

The constant \( K \) in this expression reflects possibility of the limitation of soil aggregates supply because of depletion of loose material on the surface. According to Gomes et al. [2003] this constant is close to unity for sandy soils but much lower (about 0.02) for soils with elevated clay content where wind erosion is inhibited by crust formation after rainfalls. Thus, we adopted \( K=1 \) for deserts, \( K=0.02 \) for other bare soils and urban areas and \( K=0.1 \) for all cultivated agricultural soils.

### 3.2. Soil properties data

In general the integral saltation flux strongly depends on size distribution of soil aggregates. Indeed, in natural soils small particles (below 20 \( \mu \)m) never occur in free state, but are embedded in larger soil aggregates (up to a few centimetres) by cohesion forces. Chatenet et al. [1996] used the dry sieving technique to derive four typical populations of arid soils. It was suggested that size distribution of any soil could be presented as a combination of these populations according to its mineralogical type. Thus, a continuous multi-modal distribution of soil aggregates can be presented by a combination of lognormal functions:

\[ \frac{dM}{dD_s} = \frac{1}{D_s^{3/2}\pi} \sum_j \frac{\varepsilon_j}{\sigma_j} \exp \left( \frac{-(\ln D_s - \ln D_{s,j})^2}{2\ln^2 \sigma_j} \right), \]  

(3.9)

where \( D_{s,j} \) is mass median diameter (MMD) of the \( j \)th mode; \( \sigma_j \) is its geometric standard deviation; and \( \varepsilon_j \) is mass fraction of particles of the \( j \)th mode.

To derive spatially resolved dataset of soil size distribution we utilized global soil characteristics data from the International Satellite Land-Surface Climatology Project (ISLSCP), Initiative II (http://islscp2.sesda.com). Particularly, we extracted soil texture classification according to content of three major components (sand, silt, and clay). The original data on sand, silt and clay content in soil with \( 1^\circ \times 1^\circ \) spatial resolution were interpolated into the EMEP 50×50 km grid using the kriging technique. Figure 3.3 shows derived spatial distribution of these three components in European and Northern African topsoils.
After the interpolation different soil types were classified according to the classic sand/silt/clay triangle of texture composition [Hillel, 1982]. Resulting spatial distribution of different soil types in Europe is presented in Fig. 3.4. As seen, according to this data, loam soils prevail over Central and Eastern Europe, whereas Southern Europe is dominated by clay loam. Sandy soils are characteristics of Africa and Central Asia. It should be noted that this approach allows only rough information on soil composition and texture. Soil characteristics of natural surfaces are much more variable in reality. Therefore, soil properties data with higher spatial resolution should be used in future.

Each soil type was associated with certain size distribution of soil aggregates based on the dry sieving technique measurements by Chatenet et al. [1996]. We used definitions of soil size distribution derived for major arid soil types by Marticorena and Bergametti [1997]. According to this work size distribution of soil aggregates can be presented as a combination of three soil populations with definite lognormal distribution function. Parameters of the soil populations for eight soil texture types appearing in Europe are presented in Table 3.1. Resulting size distributions of three different soil types (clay loam, loam, and loamy sand) is illustrated in Fig. 3.4. As seen sandy soils are characterized by relatively large soil grains up to a few millimetres. Whereas clay containing soils are dominated by smaller grains of 1-2 hundreds micrometers.
Table 3.1. Parameters of soil size distribution for different soil texture types

<table>
<thead>
<tr>
<th>Soil texture type</th>
<th>Soil population 1</th>
<th>Soil population 2</th>
<th>Soil population 3</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>MMD</td>
<td>σ</td>
<td>%</td>
</tr>
<tr>
<td>Sand</td>
<td>690</td>
<td>1.6</td>
<td>100</td>
</tr>
<tr>
<td>Loamy sand</td>
<td>690</td>
<td>1.6</td>
<td>90</td>
</tr>
<tr>
<td>Sandy loam</td>
<td>690</td>
<td>1.6</td>
<td>80</td>
</tr>
<tr>
<td>Loam</td>
<td>690</td>
<td>1.6</td>
<td>31.25</td>
</tr>
<tr>
<td>Silt loam</td>
<td>520</td>
<td>1.6</td>
<td>75</td>
</tr>
<tr>
<td>Silt</td>
<td>520</td>
<td>1.6</td>
<td>50</td>
</tr>
<tr>
<td>Sandy clay loam</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Clay loam</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>

It should be noted that presented above parameterisation of soil grains size distribution contains significant uncertainty since it is based on very limited measurements data and applies speculative assumptions on attribution of measured size distributions to certain soil texture types. Therefore direct and extensive measurements of size distribution of soil aggregates using the dry sieving technique are highly required for different soil types in different areas of Europe.

3.3. Sandblasting

The sandblasting model for dust suspension has been developed by Alfaro et al. [1997; 1998]. Based on the wind tunnel experiments they derived that the dust particles released by sandblasting from the saltating aggregates of different natural soils can be sorted into three lognormal modes (do not mix up with soil populations). Characteristics of the dust modes are presented in Table 3.2.

Table 3.2. Characteristics of size distribution modes for dust particles released by sandblasting [Alfaro and Gomes, 2001]

<table>
<thead>
<tr>
<th>Mode</th>
<th>$e_i$, kg m$^2$/s$^2$</th>
<th>$d_i$, μm</th>
<th>$\sigma_i$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$3.61 \times 10^{-7}$</td>
<td>1.5</td>
<td>1.7</td>
</tr>
<tr>
<td>2</td>
<td>$3.52 \times 10^{-7}$</td>
<td>6.7</td>
<td>1.6</td>
</tr>
<tr>
<td>3</td>
<td>$3.46 \times 10^{-7}$</td>
<td>14.2</td>
<td>1.5</td>
</tr>
</tbody>
</table>

According to the sandblasting model the vertical dust flux corresponding to $i^{th}$ can be presented in the following form [Alfaro and Gomes, 2001]:

$$ F_{v,i} = \int \frac{F_n(D_z)}{D_z} \alpha_i(D_z) \frac{dM}{dD_z} dD_z, \quad i = \mathbb{N}, $$

(3.10)

where the efficiency of the sandblasting process is given by:

$$ \alpha_i(D_z) = \frac{\pi}{6} \rho_i B \frac{d_i^3}{e_i}. $$

(3.11)

Here $B = 163$ m/s$^2$ is an empirical constant; $\rho_i$ is the fraction of kinetic energy of a soil aggregate required to release dust particles of mode $i$; $d_i$ is the aerosol mass median diameter of mode $i$; and $e_i$ is binding energy of aerosol particles for mode $i$. 

26
The fraction $p_i$ of the aerosol modes release depends on the kinetic energy of an individual soil aggregate:

$$e_c = \frac{100}{3} \pi \rho_i D^2 U^2. \quad (3.12)$$

A scheme of the dependence is presented in Table 3.3. The binding energies $e_i$ correspond to values presented in Table 3.2. As seen from the table the higher kinetic energy of a soil aggregate $e_c$ the more probability to eject finer particles (Mode 1). Figure 3.5 shows relative fractions of the dust aerosol modes as a function of soil aggregates size for given wind friction velocity. As seen from the figure sandblasting of larger soil aggregates, which have higher kinetic energy, releases smaller dust particles.

**Table 3.3.** Fractions ($p_i$) of the dust aerosol modes release as a function of the kinetic energy $e_c$ of an individual soil aggregate

<table>
<thead>
<tr>
<th>Mode 1 ($p_1$)</th>
<th>$e_i$</th>
<th>Mode 2 ($p_2$)</th>
<th>$e_i$</th>
<th>Mode 3 ($p_3$)</th>
<th>$e_i$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$e_i &lt; e_3$</td>
<td>0</td>
<td>$e_i &lt; e_2$</td>
<td>0</td>
<td>$e_i &lt; e_1$</td>
<td>0</td>
</tr>
<tr>
<td>Mode 2 ($p_2$)</td>
<td>0</td>
<td>0</td>
<td>$(e_i-e_2)/(e_i-e_3)$</td>
<td>1-$p_2$</td>
<td>1-$p_1-p_2$</td>
</tr>
</tbody>
</table>

**Fig. 3.5. Fractions of dust aerosol modes as functions of soil aggregates size**

Figure 3.6 presents examples of size distribution of the vertical dust flux from different soil types. As seen dust flux from sand mostly consists of fine particles with size below 5 $\mu m$ (Mode 1). As it was mentioned above sandy soils are characterized by large soil aggregates, which have high enough kinetic energy to eject fine particles. Dust flux from loamy sand contains significant contribution of coarse particles of 10 $\mu m$ and greater (Mode 3). Besides, emission of coarse particles is prevailing for dust flux from loam and clay loam, which mostly consist of smaller soil grains (see Fig. 3.4).

Dependence of dust suspension flux on wind friction velocity for different soil types is illustrated in Fig. 3.7 along with contribution of three aerosol modes to the total flux. As seen from the figure the coarse mode (Mode 3) dominates the dust flux from all soil types at relatively low friction velocities. For sandy soils contribution of the coarse mode is decreasing with increase of friction velocity, whereas contribution of the fine mode is growing. Contribution of different modes of dust suspension flux from loam and clay loam only slightly changes for wind friction velocities up to 100 cm/s.
Fig. 3.6. Size distribution of vertical dust flux for different soil types: (a) – sand; (b) – loamy sand; (c) – loam; (d) – clay loam

Fig. 3.7. Vertical dust flux as a function of the wind friction velocity for different soil types: (a) – sand; (b) – loamy sand; (c) – loam; (d) – clay loam
The described above dust suspension scheme has been applied for calculations of wind blown dust flux in Europe and adjacent territories in 2000. The dust suspension was estimated for areas not covered with vegetation:

- deserts and bare soils;
- agricultural soils (during the cultivation period);
- urban areas.

The calculated mean annual flux of dust suspension from soil is shown in Fig. 3.8 for different ranges of particle sizes (PM2.5, PM10 and PM20). As seen high suspension fluxes are characteristics of deserts in Northern Africa and Central Asia. Elevated fluxes were also obtained for Southern Europe and agricultural regions of Southeastern and Eastern Europe. Comparison of Figs. 3.8a and 3.8b demonstrates that major part of wind dust suspension flux over Europe consists of coarse particles (with particles size greater than 2.5 μm).

![Fig. 3.8. Spatial distribution of calculated mean annual dust suspension flux of different particle size in 2000: (a) – PM2.5; (b) – PM10; (c) – PM20.](image)

### 3.4. Sea-salt aerosol suspension

Description of sea-salt generation and wind suspension from the sea surface has also been included into the model. For this purpose we applied the empirical Gong-Monahan parameterization of the vertical number flux density [Gong, 2003]:

\[
\frac{dF_p}{dR_p} = 1.373U_{10}^{3.44}R_p^{0.017} (1 + 0.057R_p^{3.45}) 10^{1.6\exp(-B^2)},
\]

where \( A = 4.7(1 + \Theta R_p)^{0.017R_p^{-1.44}} \) and \( B = 1 - \frac{\log R_p}{0.433} \).

Here \( R_p \) is the sea-salt aerosol radius; \( U_{10} \) is wind speed at 10 m height; and \( \Theta = 30 \) is an adjustable parameter that controls the shape of the sub-micron size distribution.

The size distribution of the vertical sea-salt aerosol mass flux based on this parameterization is shown in Fig. 3.9a for different wind speeds. Figure 3.9b illustrates dependence of the integral sea-salt aerosol flux on wind speed at 10 m height for different cut-off aerosol diameters. In the following calculations we used the cut-off value of aerosol diameter equal to 10 μm, since larger particles can hardly be transported far from the ocean coastal areas.
3.5. Re-suspension of heavy metals from soil and seawater

To estimate heavy metal emission with dust suspension from soil it is necessary to know content of these metals in erodible soils. For this purpose we used detailed measurement data on heavy metals concentration in topsoil from the Geochemical Atlas of Europe developed under the auspices of the Forum of European Geological Surveys (FOREGS) [Salminen et al., 2005; www.gtk.fi/publ/foregsatlas/]. The data cover most parts of Europe (excluding Eastern European countries) with more than 2000 measurement sites. The kriging interpolation was applied to obtain spatial distribution of heavy metal concentration in soil. For Eastern Europe as well as for the rest of the model domain (Africa, Asia) we used default concentration values 0.2 mg/kg and 15 mg/kg for cadmium and lead, respectively, based on the literature data [Nriagu, 1980; Reimann and Carriat, 1998]. The resulting spatial distributions of Pb and Cd concentration in topsoil of Europe and adjacent territories are presented in Fig. 3.11.

It should be noted that these data mostly reflect background concentrations in residual and sedentary soils. Particularly, soil sampling was avoided from urban areas and areas with agricultural activities. Besides, the organic layer was removed from undisturbed soil samples [Salminen et al., 2005]. On the other hand, Pb and Cd concentrations measured in humus are several times higher than those in underlying soil. Similar enrichment of the upper soil layer with heavy metals by an order of magnitude was obtained by Alriksson [2001] for forest soils and by Linde et al. [2001] for urban soils. Thus, one can expect significant enrichment of the upper skin layer of soil available for wind erosion in comparison with residual concentrations presented in Fig. 3.11. Moreover, road dust in urban areas...
can contain much higher levels of lead because of long-term accumulation of this metal from use of leaded gasoline [Archer and Barret, 1976]. Agricultural soil can be additionally enriched with cadmium due to its input with fertilisers and sewage sludge. Therefore we used conditional enrichment factors for the upper layer of different soil types presented in Table 3.4. More detailed research is required in future for evaluation of heavy metal concentration profile in soil and enrichment of the upper soil layer with heavy metals, particularly, in the urban environment.

In order to estimate heavy metal re-suspension with sea-salt aerosol we used the emission factors: 4 mg/kg for lead and 40 μg/kg for cadmium, derived from the literature [Nriagu, 1980, Richardson et al., 2001]. These emission factors depend on measured heavy metal bulk concentration in seawater and on the estimated enrichment factor of the heavy metal in sea-salt particles comparing to its bulk concentration in seawater. The sea-salt enrichment is commonly connected with elevated concentrations of heavy metals in the sea surface micro-layer.

Estimated wind re-suspension fluxes of lead and cadmium from European soils and seawater in 2000 are presented in Figs. 3.12 and 3.13 in comparison with anthropogenic emissions. Heavy metal re-suspension from soil is presented for dust particles size below 10 μm (PM10). In general, the obtained re-suspension fluxes from soil are comparable with anthropogenic emissions at least in some European countries. High re-suspension fluxes were obtained for desert areas of Africa and Central Asia because of significant dust production in these regions. Elevated fluxes are also characteristics of some areas in Central, Southern and Southeastern Europe, which are conditioned by combination of relatively high concentration in soil and significant dust suspension from agricultural and urban areas.

**Table 3.4.** Expected heavy metal enrichment factors for the upper soil layer

<table>
<thead>
<tr>
<th>Soil types</th>
<th>Pb</th>
<th>Cd</th>
</tr>
</thead>
<tbody>
<tr>
<td>Barren</td>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>Agricultural</td>
<td>5</td>
<td>10</td>
</tr>
<tr>
<td>Urban</td>
<td>50</td>
<td>10</td>
</tr>
</tbody>
</table>

![Fig. 3.11. Spatial distribution of lead (a) and cadmium (b) concentration in topsoil of Europe and adjacent territories](image)
3.6. Uncertainties and further research

Presented above parameterization of heavy metal re-suspension from soil and seawater contains significant uncertainties regarding both wind suspension of dust particles and heavy metal content in soil and suspended aerosol. The most important of them are listed below:

- Soil properties data including spatial distribution of different soil types in Europe and size distribution of soil aggregates;
- Drag partition scheme; particularly, values of roughness elements density for different landuse types;
- Heavy metal content in erodible soils; particularly, enrichment of the upper soil layer with heavy metals in different environments;
- Emission factors of heavy metals suspension from seawater.

These uncertainties are to be addressed in future research. Particularly, it is planned to perform comparative analysis of different soil properties datasets and to utilize soil data with much finer resolution. Besides, in depth analysis of the literature data is required to evaluate critical parameters of the scheme. And finally, a comprehensive sensitivity analysis of the dust suspension scheme is to be performed with regard to selected parameterization along with extensive evaluation against measurement data.
ANALYSIS OF THE MODEL PERFORMANCE

One of the inferences of the Workshop on model review was the conclusion that MSCE-HM model tends to underestimate measured concentrations of some heavy metals (lead and cadmium) in air and precipitation when using the official emissions data for modelling. Similar underestimation was demonstrated by other models participated in the intercomparison studies. An improvement of the agreement was achieved by taking into consideration the wind re-suspension process, however, some underestimation still remains. On the other hand, use of ESPREME expert estimates allows significant reducing the underestimation [Gusev et al., 2006]. To analyse the problem in this Chapter the model results are compared with results of the CMAQ model – one of well-developed contemporary chemical transport models. Besides, the underestimation is analysed using measurements data with high (daily or weekly) temporal resolution.

4.1. Comparison of MSCE-HM and CMAQ modelling results

To analyse peculiarities of the MSCE-HM model performance its modelling results were compared with calculations performed by the CMAQ model under similar conditions. For this purpose both models were applied to calculate the atmospheric transport and depositions of lead and cadmium for the year 2000 using both the official emissions data and the ESPREME estimates. Besides, both models utilized a tentative parameterisation of heavy metal re-suspension from soil and seawater [Gusev et al., 2006] and the same meteorological dataset generated by the MM5 pre-processor. Results of the comparison are presented below.

**Lead**

Figure 4.1 illustrates annual mean concentration of lead in the surface layer calculated by the MSCE-HM and CMAQ models. As seen the patterns of the surface concentration calculated by both models are quite similar. Both models predict elevated concentrations at the same locations in Europe: in southern Poland, western Germany, northern Italy, eastern Ukraine etc. High lead concentrations in Africa are determined by large wind re-suspension fluxes predicted by the dust suspension scheme.

![Spatial distribution of lead mean annual concentration in 2000 calculated with MSCE-HM (a) and CMAQ (b) models based on EMEP official emissions data](image)

*Fig. 4.1. Spatial distribution of lead mean annual concentration in 2000 calculated with MSCE-HM (a) and CMAQ (b) models based on EMEP official emissions data*
Comparison of total annual flux of lead deposition in Europe and adjoint territories calculated by both models is presented in Fig. 4.2. Again as for the air concentration spatial distributions of lead deposition calculated by the MSCE-HM and CMAQ models are very similar. MSCE-HM predicts somewhat higher total deposition fluxes in Europe mostly because of larger estimates of dry deposition over vegetative surfaces. On the other hand, higher dry depositions over bare land and deserts estimated by CMAQ result in higher total depositions in Africa. Besides, the models predict different lead deposition fluxes over the Atlantic because of discrepancy in boundary concentration profiles used in the models.

**Fig. 4.2.** Spatial distribution of lead total (dry and wet) annual deposition in 2000 calculated with MSCE-HM (a) and CMAQ (b) models based on EMEP official emissions data

Figure 4.3 illustrates comparison of lead concentrations in air and precipitation obtained by both models with observations from the EMEP monitoring network. The EMEP official emissions data were used for the calculations. As seen from the figure, both models demonstrate significant correlation with observations (the correlation coefficient is around 0.8) and about 40% underestimation of measurements data. Both models tend to underestimate lower concentrations in precipitation corresponding to Scandinavian sites, and the underestimation is larger for the CMAQ model.

**Fig. 4.3.** Scatter plots of comparison of lead concentrations in air (a) and precipitation (b) calculated by MSCE-HM and CMAQ models based on the official emissions data for 2000 with observations. Red solid line delineates 1:1 ratio, red and green dashed lines show deviation by a factor of two and three, respectively
Agreement of modelling results with measurements is better when the ESPREME emission estimates are used for calculations instead of the official data (Fig. 4.4). The correlation remains significant whereas the underestimation decreases down to 20-25% for both models. Nevertheless, the underestimation of lower values at Scandinavian sites does not change considerably. Thus, variation of anthropogenic emissions does not significantly affect lead concentration in precipitation at these sites. Therefore possible explanations of this can include underestimation of non-anthropogenic emission in this region (e.g. lead re-suspension from seawater) or inaccurate treatment of wet deposition process by both models.

![Scatter plots of comparison of lead concentrations in air (a) and precipitation (b) calculated by MSCE-HM and CMAQ models based on the ESPREME estimates for 2000 with observations. Red solid line delineates 1:1 ratio, red and green dashed lines show deviation by a factor of two and three, respectively.](image)

**Fig. 4.4.** Scatter plots of comparison of lead concentrations in air (a) and precipitation (b) calculated by MSCE-HM and CMAQ models based on the ESPREME estimates for 2000 with observations. Red solid line delineates 1:1 ratio, red and green dashed lines show deviation by a factor of two and three, respectively.

**Cadmium**

Comparison of modelling results obtained by the MSCE-HM and CMAQ models for cadmium is presented in Figs. 4.5 and 4.6. Figure 4.5 shows calculated mean annual concentrations of cadmium in the surface air. Similar to lead, spatial distributions of cadmium air concentration in Europe calculated by two models fairly agree with each other. Elevated concentrations are predicted in Poland, western Germany, some areas of Eastern and Southern Europe.

![Spatial distribution of cadmium mean annual concentration in 2000 calculated with MSCE-HM (a) and CMAQ (b) models based on EMEP official emissions data.](image)

**Fig. 4.5.** Spatial distribution of cadmium mean annual concentration in 2000 calculated with MSCE-HM (a) and CMAQ (b) models based on EMEP official emissions data.
Total annual depositions of cadmium in Europe calculated by MCSE-HM and CMAQ are compared in Fig. 4.6. Spatial patterns of deposition fluxes in Europe calculated by both models are very similar. MSCE-HM predicts somewhat larger depositions, particularly, in remote regions and over the Atlantic Ocean. It can be explained by more significant atmospheric dispersion, larger dry deposition to vegetation and higher boundary concentrations imposed in the MSCE-HM model. CMAQ estimates higher cadmium dry depositions over African deserts.

Fig. 4.6. Spatial distribution of cadmium total (dry and wet) annual deposition in 2000 calculated with MSCE-HM (a) and CMAQ (b) models based on EMEP official emissions data

Comparison of modelled cadmium concentrations in air and precipitation with measurements is illustrated in Fig. 4.7. Modelling results correlates well with the measurement data: the correlation coefficients are around 0.7 for both models. However, underestimation of observed values by the models is greater than for lead and exceeds 60%. The reason for that is much smaller contribution of cadmium re-suspension estimated with the dust production scheme. Taking into account large uncertainty of the tentative parameterisation of heavy metal re-suspension, one can expect possible underestimation of this process contribution. CMAQ demonstrates more significant underestimation of lower cadmium concentration in precipitation at Scandinavian sites.

Fig. 4.7. Scatter plots of comparison of cadmium concentrations in air (a) and precipitation (b) calculated by MSCE-HM and CMAQ models based on the official emissions data for 2000 with observations. Red solid line delineates 1:1 ratio, red and green dashed lines show deviation by a factor of two and three, respectively.
Significant improvement of the comparison with measurements is achieved by both models when the ESPREME emissions estimates are used instead of the official data. The underestimation decreases down to 12% and 35% for concentration in air and precipitation, respectively. CMAQ shows somewhat better correlation in this case, however, considerable underestimation of concentrations in precipitation at Scandinavian site still remains.

Fig. 4.8. Scatter plots of comparison of cadmium concentrations in air (a) and precipitation (b) calculated by MSCE-HM and CMAQ models based on the ESPREME estimates for 2000 with observations. Red solid line delineates 1:1 ratio, red and green dashed lines show deviation by a factor of two and three, respectively.

Thus, both considered models demonstrate quite similar results both for lead and cadmium. Calculated concentration and deposition fields are in good agreement with each other, however, some discrepancies take place because of differences of the models parameterisations of dry deposition and definitions of boundary concentration profiles. Both models demonstrate satisfactory agreement with observations along with some underestimation of the observed values when the official emissions data was used. Use of the ESPREME emissions estimates leads to decrease of the underestimation.

4.2. Modelling results vs. measurements

Comparison of annual mean modelled and measured concentrations in air and in precipitation for 2004 was presented in EMEP/MSC-E technical report [Gusev et al., 2006]. In particular, the effect of wind re-suspension on the results of comparison of modelled and measured parameters was demonstrated. Introduction of wind re-suspension process in modelling scheme markedly improves the model performance for lead, and leads to smaller improvements for cadmium. It was shown that the use of officially reported emission data, even in combination with wind re-suspension, leads to some underestimation of the observed concentrations in air and in precipitation of lead, and to significant underestimation of cadmium. In this report the purpose was to continue this comparison, but taking into account measurement data with the fine temporal resolution. Since the problems of underestimation of the observed data by the model more relevant for lead and cadmium than for mercury, this chapter is focused on these two metals. The results overviewed in this chapter refer to 2004. The anthropogenic emission data used to obtain the discussed model results are based on officially reported data supplemented by TNO expert estimates [Denier van der Gon, 2005] and described in [Ilyin at al., 2006]. The wind re-suspension scheme involved in modelling is presented and discussed in [Gusev et al., 2006].
Lead

Modelled annual mean concentrations of lead in air and in precipitation reasonably well agree with the available measurement data. Correlation coefficient for concentrations in air is 0.80 and for concentrations in precipitation was 0.69 (Fig. 4.9). Regression coefficients for concentrations in air and in precipitation were 0.66 and 0.74, respectively (Fig. 4.9). These regression coefficients imply that the modelled concentrations are mostly below the observed values by about 25 – 30%. Significant correlation coefficients mean that the model manages to capture spatial distribution of lead levels.

\[
\text{C}_{\text{mod}} = 0.66 \times \text{C}_{\text{obs}} \\
R_{\text{corr}} = 0.80
\]

\[
\text{C}_{\text{mod}} = 0.74 \times \text{C}_{\text{obs}} \\
R_{\text{corr}} = 0.69
\]

Fig. 4.9. Comparison of modelled concentrations of lead in air (a) and in precipitation (b) in 2004 with measurements from the EMEP monitoring network. \( R_{\text{corr}} \) is the correlation coefficient, \( C_{\text{mod}} \) – model, \( C_{\text{obs}} \) – observations. Red solid line delineates 1:1 ratio, red and green dashed lines show deviation by a factor of two and three, respectively.

Although for Europe as a whole the model demonstrates satisfactory agreement with measurement data, comparison results for individual stations are different. Annual mean concentrations of lead in air modelled for German, Danish, British stations agree well with measurement data (Fig. 4.10). However, for stations in Slovakia, and Lithuania, some stations in Austria, and Czech Republic the discrepancy between modelled and observed concentrations exceeds 40%.

Fig. 4.10. Comparison of modelled and measured concentrations of lead in air in 2004 at individual EMEP stations

Annual mean value is a result of averaging over numerous realisations of meteorological situations. Annual mean values do not provide enough information to confidently determine the reasons of discrepancies between the model and the measurements or to ensure that minor discrepancies are
not accidental. Therefore, in order to try to understand the model behaviour, comparison on smaller times scales is needed. Comparison made for monthly mean air concentrations shows that at many EMEP stations modelled concentrations in air in winter period significantly underestimate the observed ones, while in summer period they fit measurements much better. This fact is especially clearly seen for stations in Czech Republic, Austria, Germany, and Denmark. Typical examples of monthly mean modelled and measured concentrations of lead in air and in precipitation are stations CZ3 (Kosetice) in Czech Republic and DE7 (Neuglobsow) in Germany shown in Fig. 4.11 and 4.12, respectively. As seen from these figures, in some cases concentrations in precipitation are also underestimated by the model in winter period.

The discrepancy between the model and observations in winter period in 2004 can be caused by inadequate representation of seasonal variations of anthropogenic emissions. The official emission data do not contain information on seasonal variations of the heavy metal releases to the atmosphere. That is why the model does not consider emission variability within a year. In winter time emission of metals can be higher than in summer time because, e.g. of more intensive residential heating. Thus, the introduction of seasonal variability of heavy metal emissions may improve the representation of modelled levels of heavy metals. Another reason may be connected with uncertainties of meteorological driver when simulating atmospheric stability. If the atmospheric boundary layer, parameterised by the meteorological driver is less stable than that at real conditions, the mixing of a pollutant along vertical would be higher and hence near-surface modelled concentrations would deplete. Indirect indication of this can be seen at stations, where underestimation between modelled and measured concentrations in air is much larger than that for concentrations in precipitation.

![Fig. 4.11. Examples of modelled and measured concentrations of lead at station CZ3 (Kosetice, Czech Republic) in air (a) and precipitation (b) in 2004](image)

![Fig. 4.12. Examples of modelled and measured concentrations of lead at station DE7 (Neuglobsow, Germany) in air (a) and precipitation (b) in 2004](image)
For EMEP monitoring stations raw measurement data are available, and some of the stations are characterized by relatively high temporal resolution - one week or one day. In order to investigate possible causes of discrepancies between modelled and measured quantities, modelled data were compared against measurement with high temporal resolution.

Raw air concentrations of lead at station CZ3 in 2004 represent daily mean values sampled once per 2-4 days. Modelled data were extracted from the modelling results with the same frequency for the same dates when the observations took place (Fig. 4.13). As seen, positions of modelled peak concentrations in most cases coincide with those of the observed concentrations. It is also clearly seen that the use of re-suspension markedly improves agreement between modelled and measured concentrations. However, a number of peaks in monitoring data remain underestimated.

Another example of favourable agreement between measured and modelled concentrations is given in Fig. 4.14 for station DK8 (Anholt, Denmark). Majority of peaks were captured by the model, both from viewpoint of location and magnitude. Similar to other stations, the use of re-suspension significantly improves the model performance.

---

**Fig. 4.13.** Comparison of daily mean measured and modelled concentrations of lead in air at station CZ3 (Kosetice, Czech Republic) in 2004. Measured data are daily means sampled once per 2 – 4 days.

**Fig. 4.14.** Comparison of daily mean measured and modelled concentrations of lead in air at station DK8 (Anholt, Denmark) in 2004. Measurements in air are performed on daily basis.
Reproduction of weekly concentrations at station SK6 (Starina, Slovakia) was less successful. Modelled concentrations underestimate measurement values (Fig. 4.15). The discrepancies can be explained by either uncertainties of the emission data, or peculiarities of meteorological data generated by the meteorological driver for mountainous region where the station is situated. It is planned to perform further research in order to approach to understanding of causes of the discrepancies, including analysis of back trajectories for this station.

![Figure 4.15: Comparison of weekly mean measured and modelled concentrations of lead in air at station SK6 (Starina, Slovakia) in 2004.](image)

For most stations in Germany, Denmark, Poland, the Netherlands, Sweden, the United Kingdom (Fig. 4.16) the model matched the observed concentrations in precipitation with acceptable accuracy: bias ranged from -20 to 40%. Significant discrepancy was found for stations in Slovakia, Finland and some Norwegian stations. Similar to air concentrations, comparison for stations was performed on monthly and weekly basis.

![Figure 4.16: Comparison of modelled and measured concentrations of lead in precipitation in 2004 at individual EMEP stations](image)

Comparison of lead concentrations in precipitation on short-term time scale demonstrates different extent of agreement between modelled and measured values at different stations. For example, at station DE9 (Zingst, Germany) temporal variations of modelled concentrations mostly follow the observed ones (Fig. 4.17). It is worth mentioning that the introduction of wind resuspension markedly improve the comparison results. In particular, peaks in March and December were well reproduced. Another example of favourable agreement between modelled and measured values is station NO1.
(Birkenes, Norway): locations of peaks of the modelled and the observed values mostly coincide (Fig. 4.18). For a number of peaks modelled values exceed the observed ones, if re-suspension is switched on, and stay below in opposite cases. One of possible reasons for this can be overestimation of role of re-suspension from seawater for this region.

![Comparison of weekly mean measured and modelled concentrations of lead in precipitation at station DE9 (Zingst, Germany) in 2004](image1)

**Fig. 4.17.** Comparison of weekly mean measured and modelled concentrations of lead in precipitation at station DE9 (Zingst, Germany) in 2004

![Comparison of weekly mean measured and modelled concentrations of lead in precipitation at station NO1 (Birkenes, Norway) in 2004](image2)

**Fig. 4.18.** Comparison of weekly mean measured and modelled concentrations of lead in precipitation at station NO1 (Birkenes, Norway) in 2004

At a number of stations from Scandinavia the model considerably (1.5 - 4 times) underestimates the observations. For example, modelled annual mean concentration at Norwegian station NO56 (Hurdal) is twice lower than the observed one (Fig. 4.19). Comparison of weekly mean values demonstrates that magnitude of a number of peak concentrations was not reproduced by the model. Similar to behaviour of air concentrations at some stations, at NO56 the largest underestimation of the observed values took place in winter season.
**Fig. 4.19.** Comparison of weekly mean measured and modelled concentrations of lead in precipitation at station NO56 (Hurdal, Norway) in 2004

**Cadmium**

Comparison of annual mean modelled and measured concentrations of cadmium in air and in precipitation shows that the model significantly (by 2 - 3 times) underestimates the observed values. This underestimation can be explained by various possible reasons, including underestimation of emission magnitude (anthropogenic and/or re-suspension). In particular, the use of alternative emission data with higher emission values, such as ESPREME, resulted to better agreement between the model and the measurements [Gusev et al, 2006]. Correlation coefficients were relatively high (0.76 for air concentrations, 0.71 for concentrations in precipitation). This means that spatial distribution of the observed cadmium levels was in general reproduced by the model.

As expected, the comparison results for cadmium are less successful than for lead. One of the main reasons for this is much smaller concentrations of cadmium in the environment that makes measurements of cadmium concentration more challenging task. In particular, as follows from the reports of EMEP/CCC, there are much more cadmium samples, which are below the detection limit than those of lead [Berg and Hjellbrekke, 1998; Aas and Hjellbrekke, 2003; Aas and Breivik, 2004]. Laboratories participating in the international analytical intercomparison studies are normally exhibit better results for lead than for cadmium [Berg and Aas, 2000; Uggerud and Skjelmoen, 2001, 2002, 2003].
Almost at all stations modelled concentrations in air underestimate the observed values. Especially high (2 - 4 times) underestimation was found for most stations in Slovakia, Czech Republic, Latvia, Denmark, and Germany. Relatively good agreement on the annual mean levels can be indicated for the United Kingdom, Spain, and the Netherlands (Fig. 4.21).

Similar to lead, reliability of the modelling results on monthly mean level differs for different stations. Comparison of monthly mean modelled and measured air concentrations revealed the effect similar to that observed for lead: at a number of stations modelled and measured concentrations better agree in summer period of year, while in winter period the discrepancy is higher. Comparison results at two stations CZ1 (Svratouch, Czech Republic) and SK5 (Liesek, Slovakia) were selected as examples in order to demonstrate this effect (Fig. 4.22 and 4.23, respectively). Another peculiarity of the results for cadmium is little effect of wind re-suspension on modelled concentrations, according to the applied parameterisation of this process. Besides, modelled emission due to wind re-suspension may be underestimated.

**Fig. 4.21.** Comparison of modelled and measured concentrations of cadmium in air in 2004 at individual EMEP stations

**Fig. 4.22.** Examples of modelled and measured concentrations of cadmium at station CZ1 (Svratouch, Czech Republic) in air (a) and precipitation (b) in 2004

**Fig. 4.23.** Examples of modelled and measured concentrations of cadmium at station SK5 (Liesek, Slovakia) in air (a) and precipitation (b) in 2004
Comparison made for daily mean concentrations of cadmium at station CZ1 demonstrates that the positions of most observed peaks have been captured by the model (Fig. 4.24). Magnitude of the peaks, however, is often underestimated by the model. It is clearly seen, that introduction of wind resuspension in modelling scheme does not have large effect on modelled concentrations at this station. Similar to lead, underestimation of cadmium concentrations can be connected with underestimated emission values at locations from which atmospheric transport to station took place, or with uncertainties in describing atmospheric stability conditions. These assumptions should be analysed in further research.

**Fig. 4.24.** Comparison of daily mean measured and modelled concentrations of cadmium in air at station CZ1 (Svratouch, Czech Republic) in 2004. Measurements in air are sampled once per 2 – 4 days.

Another example of satisfactory ability of the model to simulate daily mean cadmium air concentrations is station NL9 (Kollumerwaard, the Netherlands) (Fig. 4.25). Majority of peak values were caught by the model. Magnitude of peak concentrations was reliably reproduced by the model in the middle of the year. In the beginning and the end of the year peaks were mostly underestimated, and peak in October was overpredicted by the model. Further investigations are needed to explain behaviour of the modelling results. Unlike most of stations, at NL9 the effect of the use of wind resuspension in model calculations is significant and this effect allows to essentially improve the model performance.

**Fig. 4.25.** Comparison of daily mean measured and modelled concentrations of cadmium in air at station NL9 (Kollumerwaard, the Netherlands) in 2004. Measurements in air are sampled once per every second day.
Station SK5 (Liesek, Slovakia) is an example, where the discrepancy between weekly mean modelled and measured air concentrations is significant, especially in the beginning and in the end of the year (Fig. 4.26). This statement is true both for magnitude and temporal variability of the modelled quantities. This station is located in mountainous region. In order to approach to understanding of reasons of these discrepancies it is necessary to analyse meteorological and emission data for the region where the station is located.

Comparison of modelled and measured concentrations of cadmium in precipitation is shown in Fig. 4.27. As seen, for stations in Germany, the United Kingdom, the Netherlands, Iceland and Poland the modelled values relatively well matched the observed values. At stations in Czech Republic, Slovakia, Norway, Finland, Sweden, Denmark, Estonia and Latvia the underestimation is considerable (2-5 times) (Fig. 4.27).

Examples of stations where weekly mean modelled concentrations in precipitation match the observed ones, are GB91 (Banchory, the United Kingdom) and NO1 (Birkenes, Norway) (Fig. 4.28, 4.29). In both cases the model reproduced temporal variability of the observed concentrations and managed to capture most of peaks. Although for Europe as a whole the contribution of wind re-suspension to cadmium levels is not high, at these stations in some weeks the contribution of this process is high and allows to considerably reduce bias between modelled and measured values. The examples are end of March and the beginning of November at station GB91, or end of April and the beginning of December for NO1.
As seen from Fig. 4.27, for a number of stations in Scandinavia the modelled value essentially underestimate the observed ones. Examples of comparison for these stations, made on weekly basis, are shown for stations NO56 (Hurdal, Norway) and LV10 (Rucava, Latvia) in Figs. 4.30 and 4.31, respectively. Although location of some peaks was caught by the model, the magnitude of both peaks and baseline values were underestimated at both stations, with some few exceptions. More detailed investigation is needed to understand the behaviour of modelled concentrations and to explain the causes of these significant discrepancies.
This section of the report presented some examples demonstrating high or weak model performance at some stations where measurements with high (days - weeks) temporal resolution are available. There is a number of reasons which can explain the identified discrepancies between measurements and the model. First, they may be connected with omission of seasonal variability of officially reported emission data. Another group of reasons can be connected with meteorological issues. In particular, the analysis of atmospheric transport and atmospheric stability is needed. Finally, uncertainties of parameterization of wind re-suspension may contribute to discrepancies between the modelled and measured concentrations.
SUMMARY AND FURTHER RESEARCH

This report reflects progress of MSC-E activities on implementation of TFMM recommendations regarding further improvement of the heavy metal MSCE-HM model. In particular, the report presents comparison of different emission inventories, illustrates development and application of the quality control system for input meteorological data, describes further development of the heavy metal wind re-suspension scheme, and presents analysis of the model performance via comparison of the model results with highly resolved observations and with results of the CMAQ model.

Emission inventories

The performed analysis allows concluding that the EMEP official data and TNO and ESPREME estimates of lead and cadmium emissions well correlate with each other. However, significant differences are observed for many countries, particularly, between the EMEP and ESPREME datasets. In general the ESPREME inventory presents more higher emission estimates than two other datasets. The deviation is more pronounced for cadmium emissions. Significant discrepancies take place also in contribution of source categories to national emissions of lead and cadmium estimated by different emission inventories. Participation of national experts is highly required for in depth analysis of discrepancies of the available emission inventories and improvement of emissions data.

Meteorological data

The system of quality control of regional-scale meteorological input data has been developed and applied. The most important input meteorological parameters have been compared with those from the reference data set (ECMWF ERA-40 archive). The verification of meteorological data was focused on the analysis of spatial patterns, temporal variability and frequency distribution of meteorological parameters. Besides, first results of generating meteorological input data on the base of ECMWF analyses with 1°x1° resolution were overviewed. Further activities regarding the supply of MSC-E with regional-scale meteorological data include the following tasks:

- Complete transition from NCEP/DOE Re-analyses to ECMWF analyses data;
- Carry out detailed comparison of pre-processed meteorological data based on NCEP/DOE Re-analysis and ECMWF analysis;
- Perform quality control of input meteorological information on the routine basis;
- Where possible, make use of observational data for the purposes of evaluation of meteorological data.

Re-suspension of heavy metals

The dust production and heavy metal re-suspension scheme have been further developed and improved. Particularly, effect of soil characteristics (soil texture, size distribution of soil grains etc.) on dust production and suspension have been taken into account. Besides, some special effects influencing dust mobilization (such as wind drag partition, inhibition by soil moisture, the Owen effect) have been included or revised in the model. However, the parameterization contains significant
uncertainties regarding both wind suspension of dust particles and heavy metal content in soil and suspended aerosol. The following research is planed to be performed in future:

- Comparative analysis of different soil characteristics datasets and utilizing soil properties data with much finer spatial resolution;
- In depth analysis of the literature data in order to evaluate critical parameters of the dust suspension scheme;
- Comprehensive sensitivity analysis of the dust suspension scheme with regard to the selected parameterization along with extensive evaluation against measurement data.

Model evaluation

Comparison of modelling results obtained with MSCE-HM and CMAQ models demonstrates that the models predict quite similar ambient levels of lead and cadmium. Calculated concentration and deposition fields are in good agreement between the models, however, some discrepancies take place because of differences of the models parameterisations of dry deposition and definitions of boundary concentration profiles. Both models demonstrate satisfactory agreement with observations along with some underestimation of the observed values when the official emissions data were utilized. Use of the ESPREME emissions estimates leads to decrease of the underestimation.

Analysis of the model performance has been performed via comparison of modelling results with measurement data. The major attention is paid to investigation of the seasonal and short-term variation of modelled concentrations in air and precipitation. In general, the model performance is better for lead then for cadmium. It was obtained that the model well reproduce measured values during summertime, but tends to underestimate significantly the observations in winter. Besides, the model manages to reproduce major daily peaks, but often underestimates their amplitudes. Introduction of wind re-suspension significantly improves agreement between modelled and measured values. In order to explain magnitude and the temporal variability of the modelling results at monitoring stations and, where possible, to explain the bias between the modelled and measured values the following research activities are planed:

- Investigation of the effect of seasonal variability of emission data on the modelling results;
- Analysis of the atmospheric boundary layer stability and its influence on the modelled concentrations;
- Application of the back trajectories analysis to study major concentration peaks missed by the model.
REFERENCES

Alriksson A. [2001] Regional variability of Cd, Hg, Pb and C concentrations in different horizons of Swedish forest soils. Water, Air, and Soil Pollution: Focus, vol. 1, No. 3-4, pp.325-341
Annex A

COMPARISON OF THE EMEP OFFICIAL EMISSIONS DATA WITH NON-OFFICIAL TNO AND ESPREME EMISSION ESTIMATES FOR 2000

Lead, t/y

Austria

Belarus

Belgium

Bulgaria

Croatia

Cyprus

Czech Republic

Denmark

Estonia

Finland

France

Germany

Waste disposal
Gasoline combustion
Industrial processes
Industrial and Residential combustion
Public power
Spain

Sweden

Switzerland

United Kingdom
Cadmium, t/y

**Austria**

**Belarus**

**Belgium**

**Bulgaria**

**Croatia**

**Czech Republic**

**Denmark**

**Estonia**

**Finland**

**France**

**Germany**

**Hungary**

- Waste disposal
- Gasoline combustion
- Industrial processes
- Industrial and Residential combustion
- Public power