3. INVERSE MODELLING

Modelling is a powerful method traditionally used for the investigation of environmental pollution. Chemical transport models (CTMs) are used for evaluation of environmental pollution levels (concentrations in the environmental media, deposition fluxes, etc.) provided that emissions of the considered substance are known. Additionally, meteorology, geophysical information and physical-chemical properties of the considered pollutant are used as input data for calculations. Such models will be referred below as direct ones.

Additional information on the environmental pollution can be obtained by applications of adjoint (dual) models (inverse modelling approach). The adjoint model calculates sensitivities of the selected target parameter (TP) to emissions in each grid cell. For example, such usually used characteristics as air concentrations averaged over the selected domain and time period or the amount of the pollutant deposited to the selected domain during the selected time period can be used as target parameters. Generally, target parameter can be chosen as any characteristics of the pollution that can be expressed as a linear combination of concentrations of the considered pollutant in the environmental media. For the sake of simplicity below only linear combinations of air concentrations of the considered pollutant are considered:

\[ TP = \int \int \int c(x, y, z, t) \cdot \chi(x, y, z, t) \, dx \, dy \, dz \, dt \]  (3.1)

Here \( c(x, y, z, t) \) is the pollutant concentration at location \((x, y, z)\) at the moment \(t\), and \( \chi(x, y, z, t) \) is the target parameter density that determines the chosen parameter. For example, target parameter density determining average of air concentrations over the volume \(G\) for the period from \(t_1\) to \(t_2\) equals \(1/\text{Vol}(G)/(t_2-t_1)\) within the volume \(G\) and time period \((t_1, t_2)\) and 0 otherwise.

Application of an adjoint model allows obtaining the formula for calculating values of the selected target parameter directly from emission values omitting calculations of concentrations by means of the direct model:

\[ TP = \int \int \int e(x, y, z, t) \cdot \psi(x, y, z, t) \, dx \, dy \, dz \, dt \]  (3.2)

where \( e(x, y, z, t) \) is emission flux at location \((x, y, z)\) at the moment \(t\), and \( \psi(x, y, z, t) \) is the so-called influence function. The values of influence function at given location and time are in essence sensitivities of the chosen target parameter with respect to the values of emission density.

The values of influence function are calculated by adjoint model using target parameter density as an input. Additional input data for calculations are meteorology, geophysical information and physical-chemical properties of the considered pollutant. It should be stressed that the influence function depends only on the chosen target parameter, not on the values of emission density. Once the influence function is calculated by the adjoint model, formula (2) provides the method of fast calculation of target parameter values through the data on emissions directly avoiding the application of direct model. This is particularly convenient in constructing optimum emission scenarios when the values of one and the same target parameter (or several parameters) should be calculated repeatedly for various emission spatial and temporal distributions.

Applications of adjoint modelling to environmental problems were first outlined by [Marchuk 1986] and [Uliasz 1987]. Further this approach was applied for various pollutants by a lot of authors. In particular, in [Villani et al., 2010] estimation of emissions of CH4 using the TM5 direct and adjoint models [Krol et
al., 2008] was performed. Further, in [Carouge et al., 2010 a, b] inverse modelling approach was used to reconstruct atmospheric CO₂ emissions at 40 km resolution on the basis of observations from a European network. The evaluation of emissions of black carbon by means of adjoint modelling was performed by [Hakami et al., 2005].

Using the inverse modeling approach various problems can be investigated, such as:

- Description of the area for which emissions of the considered pollutant noticeably influence air concentrations averages over predefined time period at particular locations (zones of influence).
- Evaluation of sensitivities of the target parameter to emissions in particular cells or areas.
- Investigation of the pathways of the transport of pollution to selected locations.
- Evaluation of source-receptor relationships for the considered substances.
- Analysis of the reasons of the disagreement between measurements and calculation results obtained by direct model.

Below the application of the adjoint modelling for the analysis of the agreement between calculations and measurements B[a]P and HM is presented.

### 3.1. Heavy metals (HMs)

Inverse modelling approach was applied for analysis of discrepancies between modelled and observed concentrations of lead at the Dutch stations in the framework of the EMEP country-specific case studies. Modelling of Pb concentrations in 2007 was carried out over limited area with spatial resolution 5x5 km². Boundary concentrations were calculated over the EMEP domain with standard resolution 50x50 km².

The comparison of modelled and measured annual mean concentrations demonstrated that the model significantly overestimates the observed annual mean concentrations (Fig. 3.1). Analysis of sources contributing to concentrations showed that the main contributor to the modelled air concentrations is made by wind re-suspension from the urban territories (Fig. 3.2). It is reasonable to assume that the model parameterization of re-suspension from urban lands results to overestimation of observed levels and hence needs refinement.

![Fig. 3.1. Modelled and measured annual mean concentrations of lead at the Dutch stations in 2007.](image1)

![Fig. 3.2. Contributions of anthropogenic and re-suspension sources to air concentrations at the stations](image2)
Comparison of modelled and measured monthly mean levels shows that the overestimation is most pronounced in spring time (March, April) and in December (Fig. 3.3). In spring and winter the absolute contribution of wind re-suspension from urban areas is the highest.

Re-suspension flux of metals is calculated as a product of dust suspension flux and concentration of metals in dust particles. Dust suspension flux strongly depends on meteorological variables such as parameters of surface boundary layer and soil moisture. These factors are not considered in this document. In order to improve the agreement between modelled and measured levels, the analysis was focused on the lead concentrations in dust particles. It is assumed that concentration in dust particles is calculated as follows:

\[
C_{\text{dust}} = C_{\text{soil}} \cdot (1 + EF)
\]

(3.3)

Where \(C_{\text{dust}}\) is concentration of heavy metal in dust particle, \(C_{\text{soil}}\) - concentration in background soil, and \(EF\) is so-called enrichment factor. Concentrations in soil are based on background measurements of heavy metals in top soil (http://weppi.gtk.fi/publ/foregsatlas/). However, re-suspension occurs from the upper soil layer, where concentration of metals is typically enriched. Besides, long-term anthropogenic pollution led to increase of concentrations in soils relative to pre-industrial levels. Therefore, in order to take into account enrichment of soil concentrations in the uppermost soil layer, the enrichment factor was introduced. This factor was estimated for individual land-cover types (urban,
bare lands, arable lands) and spatially distributed taking into account long-term averaged deposition filed of heavy metals for 20 year period [Shatalov et al., 2012].

The values of the enrichment factors for individual land cover types were selected so that the model reproduces reasonably well the annual mean observed levels averaged over the entire EMEP domain. However, in certain regions and/or periods of time this approach can result to significant discrepancies between model and observations. For example, in case of the Dutch stations these are episodes in March, April and December (Fig. 3.4). It should be stressed that the uncertainty of the enrichment factor is high, therefore some correction of it is justified. It is clear that the correction should be focused on urban lands, because contribution from this land-cover type is the most significant. Contribution of other land-cover types to air concentrations in the considered episodes is relatively small and hence its correction will have minor effect on the agreement between modelled and observed values.

![Fig. 3.4. Daily mean observed and modelled Pb air concentrations at station Bilthoven in 2007. Contributions of different sources to modelled concentrations are presented.](image)

For the correction purpose it is necessary to determine regions where the enrichment factor should be reduced. For this purpose the inverse modelling approach was applied. The theoretical overview of the approach is described in this chapter above.

Three spring-time episodes (March, 22\textsuperscript{nd} – April, 03\textsuperscript{rd}; April, 7\textsuperscript{th}-19\textsuperscript{th}; April, 21\textsuperscript{st} – May, 03\textsuperscript{rd}) and one episode in winter (December, 15\textsuperscript{th} – 29\textsuperscript{th}), where the discrepancies are most significant, were examined. Daily mean concentrations at stations were considered as target parameters. For each day of the period and for each station so-called influence functions were calculated. Spatial distribution of influence functions differs for different stations and periods of time. Figure 3.5(a) exemplifies the influence function for daily mean surface concentration of lead at station Bilthoven in April, 15. As seen from the figure, in this day the most significant potential contribution to air concentrations occurs from the sources located to the south-east from the station (Germany, Austria, Hungary etc.). This spatial distribution is comparable with distribution of back trajectories. Figure 3.5b demonstrates probability of presence of air parcel, travelling along the trajectory, in a gridcell. As seen, air parcels are transported to the station in this day from the same regions. However, it should be noted that back trajectories characterize only atmospheric transport, while influence functions take into account both transport and removal processes.
Influence functions multiplied by daily emissions (Fig. 3.6) produce contribution of these emissions to air concentrations of each gridcell. Maps of contribution for 15th of April are exemplified in Fig. 3.7. As seen, in this day the contribution of re-suspension from urban areas is higher than that from anthropogenic sources. The highest contribution occurs from gridcells located near the station. Besides, relatively high contribution of re-suspension occurs from Slovakia and Hungary, because of high re-suspension taken place in these countries in the considered period of time (Fig. 3.7a).
Concentrations evaluated with the usage of influence functions were compared with modelled concentrations (Fig. 3.8). As seen, absolute values and time series of evaluated and modelled concentrations agree reasonably well. Some discrepancies between the two quantities are explained by simplifications introduced in formulation of adjoint model.

**Fig. 3.8.** Concentrations in air at the Dutch stations, evaluated by means of inverse modelling and simulated by the model.
Maps of contribution of different sources were calculated for all stations and all considered periods. On the base of these maps area where enrichment factor was reduced, was identified. Gridcells for days, where overestimation of observed levels by a factor of 1.5 or more is noted for at least four stations, were selected (Fig. 3.9). In these gridcells the enrichment factor was reduced twice. This approach aimed to correct the enrichment factor was intentionally simplified, compared to optimization procedures applied for investigation of B[a]P levels (see section 3.2). However, this simplification is appropriate because the enrichment factor itself is already highly uncertain, and remains uncertain after correction.

Reduction of the decreased enrichment factor in the selected area led to improvement of the agreement between modelled and measured concentrations at the Dutch and Belgian stations (Fig. 3.10). The agreement of annual mean levels is evident for most of the considered stations. The exception is Kollumerwaard, where overestimation was replaced by underestimation of annual mean levels.

Monthly variability of observed and modelled air concentrations (including contributions of different sources) at the Dutch stations is presented at Fig. 3.11. As follows from the figure, after correction of the enrichment factor the modelled levels better agree with the observed levels. The exception is station Kollumerwaard where the model tends to underestimate observed levels in summer and autumn periods. Besides, at station Vlaardingen the modelled peak in April was not removed completely. Most likely it is explained by simplification of the approach aimed at reducing the enrichment factor.

Statistical indexes confirming improvement of modelled levels are demonstrated in Fig. 3.12. Since overestimation of the observed levels was significantly reduced, the mean relative bias and normalized root mean square error (NRMSE) become closer to zero. Correlation coefficient between modelled and observed daily-mean time series exhibits little change at the Dutch stations. However, it increased substantially at Belgian station Koksijde. Most likely, the main reason of the improvement of correlation is explained by refinement of spatial resolution of emissions in the neighbouring Netherlands.
Application of inverse modelling for analysis of heavy metal pollution levels in the Netherlands can be considered as successful. This approach allowed establishing regions which sources have the largest influence on pollution levels at stations. Correction of wind re-suspension in these regions favoured the increasing agreement between modelled and measured levels. However, more detailed investigation of processes governing suspension of dust particles from natural and artificial surfaces, as well as collection of data on concentrations in soils and in soil dust particles is needed for further improvement of quality of model assessment of heavy metal pollution levels both on country scale and in the EMEP region as a whole.
3.2. Benzo[a]pyrene (B[a]P)

For the analysis of the agreement between measurements and model predictions for B[a]P a simplified version of adjoint model for the considered pollutant was elaborated. Namely, taking into account that the processes of gaseous exchange with the underlying surface and wet deposition of gaseous phase are of minor importance for modelling of B[a]P long-range transport, these processes were excluded from the model. The description of B[a]P degradation was performed with the help of effective degradation rate calculated taking into account gas-particle partitioning of the pollutant, reaction of gaseous form of B[a]P with atmospheric OH-radical and photodegradation of B[a]P particulate form. With the exception of this process B[a]P was assumed to be completely in the particle-bound form.

The constructed adjoint model was applied to the analysis of discrepancies between measurements and calculations in 2010. Ten EMEP measurement sites located in the north-western part of Europe were chosen for the analysis. They are: BE13, DE01, DE09, FI96, NL09, NL91, NO02, PL05, SE12 and SE14 (see Fig. 3.13). The comparison of calculated and measured monthly averages of B[a]P air concentrations at the above sites is shown in Fig. 3.14.

Fig. 3.13. Measurement sites chosen for the analysis

Fig. 3.14. The comparison of B[a]P air concentrations at selected EMEP sites (monthly averages, ng/m³) calculated by MSCE-POP model with measurements.
It is seen that essential discrepancies between measurements and model results take place at sites PL05, SE12 and also at DE01, DE09, NO02 and SE14 in some winter months. For other sites the agreement between measurements and model results is more or less reasonable. However, when constructing emission scenario for the refinement of the agreement, one should take into account all sites so that to be sure that the constructed scenario do not lead to worsening of the agreement at sites where this agreement was satisfactory.

To apply the inverse modeling approach to the analysis of the obtained discrepancies between measured and model values of B[a]P air concentrations influence functions were calculated for monthly averages at all the above sites using the simplified adjoint model. As an example, influence function for monthly average of B[a]P air concentrations in February 2010 is shown in Fig. 3.15.

Taking into account simplifications made in the construction of dual model, it is reasonable to compare air concentrations evaluated by means of influence functions with those directly calculated by MSCE-POP model. Such comparison for each of the considered sites was performed on the level of monthly averages of concentrations. The result is presented in Fig. 3.16.

The comparison manifests good agreement between air concentrations obtained by direct modelling with those evaluated by means of influence functions for each site calculated with dual model. Hence,
calculated influence functions can be used for evaluating air B[a]P concentrations for various emission distributions. In addition, values of influence functions allow evaluating the areas within which emissions can be noticeably reflected by air concentrations at particular sites (zones of influence). This is exemplified by the influence function for February averages of B[a]P air concentrations at DE09 (Fig. 3.15).

The values of influence function allow evaluating zone of influence of monthly averages of air concentrations measured at the considered sites, that is, the area for which emissions contribute noticeable to the value of the considered average. For evaluating zones of influence the threshold of 0.05 sec/m can be chosen (marked by light green in Fig. 3.15). Under such threshold value even very high emission level 1.5 tonnes/cell/year (this is maximum value of B[a]P emissions in a cell in a contaminated region of Central Europe) will contribute to annual average air concentrations at the considered site less than 0.001 ng/m³. It can be seen that zone of influence of air concentrations averaged over February at DE09 with the chosen threshold covers most of the Central Europe, some areas in Ukraine, Estonia, Latvia and southern part of Sweden. The contributions of emissions outside this zone make negligible contribution to concentrations at DE09 in February. Of course, since long-range transport potential for B[a]P is lower in summer months (due to higher degradation rates) zones of influence for spring and, especially, summer months will be essentially less (see Fig. 3.17).

To evaluate mutual zone of influence of all monthly averages for all 10 sites used for the analysis, maximum values of influence functions calculated for all monthly averages at these sites can be used. If this maximum exceeds the chosen threshold level in some cell, then emissions in this cell contributes noticeably at least for one month for at least one site. The map of the above maximums is presented in Fig. 3.18. It should be mentioned that any conclusions on emission uncertainties can be applied only to locations included to the zone of influence of the considered sites.

Calculated influence functions together with emission density allow evaluating contributions of all grid cells to monthly averages of air concentrations at the considered sites. This can be done simply by multiplying values of corresponding influence functions by emission flux (cf. formula (3.2)). The obtained maps of contributions can be used for the analysis of the discrepancies between measurement data and model predictions. This will be demonstrated below by the comparison of calculated and measured air concentrations at DE09.

As seen from the plot in Fig. 3.14, maximum underestimation of air concentrations at DE09 takes place in January. To analyze the location of emissions that are possible reason of such underestimation, it is reasonable to compare contributions of various grid cells to monthly averages of air concentrations for all months of 2010 and select cells that contribute essentially to average concentrations.
concentration in January but have much less contributions in other months (where underestimation does not take place). The maps of contributions of emissions in all grid cells to average concentrations at DE09 for all 12 months of 2010 are shown in Fig. 3.19.

From the maps it is seen that, according to model calculations, monthly averages of air concentrations at DE09 in warm months (from May to September) are governed by local emissions. At the same time averages in the rest months can be (in different extent) caused by long-range transport. This should be taken into account in the analysis of underestimation of air concentrations in January.

![Fig. 3.19. Contributions of emissions in all grid cells to average concentrations at DE09 for 12 months of 2010.](image)

It is seen that emissions from the area marked by red oval essentially contribute to monthly average of air concentrations over January. At the same time emissions of this area contribute to averages over other months of 2010 is essentially smaller. It leads to the supposition that underestimation of emissions in the marked area *may* be a reason of underestimation of air concentrations at DE09 in January. At the same time this supposition (being in essence qualitative) should be additionally verified by the analysis of measurement-modelling agreement at sites that are affected by emissions from the selected area (e.g. at PL05).
The map of contributions of emissions in all grid cells to average air concentration at PL05 over January is shown in Fig. 3.20. It is seen that enlarging emissions in the selected area (again marked by red oval in Fig. 3.20) in the framework of a conventional scenario will lead to the enlargement of air concentrations at PL05 in January, which is in agreement with the fact that model strongly underestimates air concentrations in January at PL05 (see Fig. 3.14).

Of course, it is hard to include all 10 sites in the analysis using the above described method since it would require comparison of 120 maps “by eyes”. To perform full analysis for all the considered sites analytical methods should be applied. Here an optimization procedure refining the agreement between measurements and calculations at all considered sites and all months by changing emissions in each grid cell in the framework of a conventional emission scenario is applied. Such scenario can be characterized by the set of correction factors given in each grid cell, that is, coefficients by which initial emissions in each grid cell should be multiplied to obtain emission scenario values. The applied procedure constructs a conventional scenario minimizing root mean square deviation (RMSE) between measurement and modelling data. To avoid unrealistic emission scenarios, additional restrictions to correction factors were introduced (in our case these factors are chosen to be between 0.7 and 3).

Such scenario answers the following two questions. First, it demonstrates to which extent the discrepancies between model predictions and monitoring data can be explained by uncertainties of emission data. Second, it allows selecting the areas within which emission uncertainties most likely occur.

Application of the constructed scenario allowed refining the agreement between measurements and model results at sites DE01, DE09, PL05 and SE14 (Fig. 3.21).

However, for two sites (NO02 and SE12) optimization procedure fails to refine the agreement between model results and calculations (Fig. 3.22). The same situation takes place for November and December at DE01 and DE 09. This manifests that the reasons of discrepancies at these sites and months are different from those originated by emission uncertainties. These can be model uncertainties, peculiarities in location of measurement sites, influence of local sources, etc. To reveal the reasons of measurement/modelling discrepancies at these sites additional work is needed.
At the rest sites (BE13, FI96, NL09 and NL91) usage of emission scenario leads to minor changes not worsening the agreement between measurements and model calculations.

In addition to clarification to what extent measurement/modelling discrepancies can be explained by emission uncertainties, the constructed emission scenario can select areas inside the EMEP grid which can essentially affect the measurement/modelling agreement. To show these areas the map of correction factors can be used (Fig. 3.23). The areas with possible emission uncertainties include those obtained above by qualitative analysis carried out for DE09 site. However, due to inclusion of other sites to the analysis additional areas are selected (located in Belarus and Romania). It should be stressed that that area of possible emission uncertainty in Romania is located at the boundary of zone of influence for the considered 10 sites (see Fig. 3.18) and, hence, the possible uncertainties of emissions inside this area are less justified.

Further, inverse modelling approach allows evaluating the contributions of various source groups to concentrations at considered sites. We recall that five “source groups” with different seasonal variations are used for evaluation of B[a]P concentrations. They are Industry (IND), Residential heating (RES), Road transport (ROT), Waste incineration (WST) and Unknown sources (UNK). The last group includes sources from countries for which the distribution of total emissions between the listed source groups is unknown (these are Central Asian countries). For these sources temporal distribution of emissions is assumed to be homogeneous throughout the entire year. It should be mentioned that the contributions of the last source group to the concentrations at the considered sites is negligible.

The relative contributions of the above source groups to the contamination at the considered sites evaluated with the help of calculated influence functions are shown in Fig. 3.24.

It is seen that the relative contributions of “Residential heating” are prevailing at all sites. These contributions are smaller in warm months and larger in cold ones at most sites. Consequently, relative contribution of “Industry” increases in warm period. The exceptions are sites DE01 and DE09, where the contribution of “Residential heating” group is prevailing over the entire year. This can be conditioned by the shape of zones of influence for these sites.
For examination of possible uncertainties in emissions from particular source category, second emission scenario was elaborated by additional optimization procedure using separate correction factors for each considered category. Though the refinement of the measurement/modelling agreement obtained by application of this scenario is quite the same as with the first scenario, spatial distribution of correction factors are different for different source groups. Spatial distributions of correction factors for Residential heating, Industry and Road transport (as main contributors to B[a]P air contamination) are shown in Fig. 3.25. For Waste incineration changes are less essential.

It is seen that maximum changes predicted by optimization scenario occur for Residential Heating source group, and then comes Industry and Road transport groups.

It is also worth mentioning that the increase of emission totals in countries resulting from the first scenario (changing emission totals in each cell) and second one (using separate correction factors for various source groups) is different (see Table 3.1, where countries with essential increase of emission totals are shown).
Table 3.1. Increase of emission totals in selected countries resulting from the application of the two above emission scenarios.

<table>
<thead>
<tr>
<th>Country</th>
<th>Increase of emission totals, times</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>First scenario</td>
</tr>
<tr>
<td>Poland</td>
<td>2.25</td>
</tr>
<tr>
<td>Lithuania</td>
<td>1.55</td>
</tr>
<tr>
<td>Belarus</td>
<td>2.18</td>
</tr>
<tr>
<td>Romania</td>
<td>1.96</td>
</tr>
</tbody>
</table>

It is seen that one and the same refinement of measurement/modeling agreement can be achieved with (in general) less enlargement of countries’ emission totals if emissions from different source groups are considered separately. So, when investigating emission uncertainties separate consideration of emissions from various source categories is reasonable.

Of course, the obtained results are preliminary and cannot be viewed as final conclusions on emission uncertainties. However, the method described above allows determination possible “hot spots” in emission inventory (from the viewpoint of emission totals, spatial distribution and distribution of emissions between source categories) and areas which can be of interest from the viewpoint of refining emission inventories.

3.3. Concluding remarks

The inverse modelling method is a powerful tool for the investigation of environmental pollution by HMs and POPs. It can be applied to the determination of the areas for which emissions of the considered pollutant noticeably influence air concentrations of the considered pollutant averages over predefined time period at particular locations (zones of influence), analysis of the reasons of discrepancies between monitoring data and model results and other purposes.

The modelling tools for application of inverse modelling approach are adjoint models. At present such models are elaborated for HMs and B[a]P (simplified version). It was found that the results obtained by simplified version of the adjoint model for B[a]P well agree with the results of MSCE-POP model.

Inverse modelling approach was applied to the analysis of discrepancies between modeled and measured levels at the Dutch and Belgian monitoring stations, caused by overestimation of wind re-suspension. The main outcomes of this work can be formulated as follows:

- The inverse modeling allows establishing regions which sources have the largest influence on pollution levels at stations.
- Correction of wind re-suspension in these regions favoured the increasing agreement between modelled and measured levels.

With the help of adjoint B[a]P model the analysis of discrepancies between calculations and measurement results in 2010 for this pollutant was performed. It was found that:

- Uncertainties in emission data can partly explain the disagreement between measurements and model predictions. There exist some areas in Central and Eastern Europe where underestimation of emissions of B[a]P may occur.
It is reasonable to perform the analysis of agreement between measurements and model prediction taking into account sectoral composition of emissions. In particular, sector-specific analysis shows that the uncertainties in emissions from residential heating can cause essential disagreement between measurement data and model calculations.

The found “hot spots” in emission inventories should be a topic of further discussion with emission experts possibly in the framework of TFMM and TFEIP.