EMEP Status Report 3/2011
June 2011

Persistent Organic Pollutants in the Environment

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EXECUTIVE SUMMARY

In accordance with the EMEP Work-plan for 2011, Meteorological Synthesizing Centre East (MSC-E) and Chemical Coordinating Centre (CCC) carried out investigations of the contamination by persistent organic pollutants (PAHs, PCDD/Fs and HCB) within the EMEP region. Main emphasis was made on the refinement of the assessment of POP pollution within the EMEP domain using an integrated approach, further developing of the global scale modelling of POPs, and exploring the links between the climate change and POP pollution. The outcome of these studies is summarized below.

Development of Integrated Approach for the Assessment of POP Pollution

MSC-E continued elaborating the integrated measurement/modelling/emission approach to the evaluation of POP contamination levels. Statistical indicators for the evaluation of agreement between measurements and model calculations used in literature were reviewed. Some new indicators based on the theory of testing statistical hypotheses were considered and the analysis of relations between various statistical indicators was performed. On the basis of this analysis the set of basic and additional indicators was proposed. Methods of emission scenario evaluation based on back trajectories and matrix approach were considered. These methods were tested in course of preliminary analysis of the agreement between calculations and measurements of PAHs, PCDD/Fs and HCB.

Assessment of POP Pollution Levels, Transboundary Transport, and Trends

Evaluation of pollution levels and transboundary transport of POPs in the EMEP region for 2009 was carried on the basis of EMEP measurements, emission data, and modelling of POP long-range transport. Emission datasets for model assessment of POP long-range transport within the EMEP region were prepared by Centre of Emission Inventories and Projections (CEIP) on the basis of officially submitted emission data complemented by expert estimates. For the evaluation of intercontinental transport gridded emissions of PCDD/Fs and HCB for the Northern Hemisphere were prepared. Several conventional scenarios of emissions for considered POPs were constructed for the analysis of emission data uncertainty. Additionally, the sensitivity of the pollution levels to the emissions of particular countries was evaluated and preliminary recommendations for further refinement of assessment of pollution levels within the EMEP domain were formulated.

In 2009 there were twenty three EMEP monitoring sites measuring POPs, among which thirteen sites performed measurements of POP concentrations in both compartments (air and precipitation). Most of the new additions to EMEP were the sites measuring PAHs which were required by the EU air quality directive. New sites in Moldova and Kazakhstan started monitoring of POPs in June 2009. Nevertheless, the spatial coverage of the EMEP monitoring network for POPs still requires further improvement.

Polycyclic Aromatic Hydrocarbons (PAHs)

Modelling of PAH pollution levels within the EMEP region was carried out for indicator PAH congeners, namely, benzo[a]pyrene (B[a]P), benzo[b]fluoranthene (B[b]F), benzo[k]fluoranthene (B[k]F), and indeno[1,2,3-cd]pyrene (IP). Based on available measurements and modelling results, highest levels of air concentrations were found for B[b]F and IP. Slightly lower pollution levels were characteristic of B[a]P and the lowest ones for B[k]F. Elevated levels of contamination by PAHs were obtained for Central and Eastern Europe, Portugal and the western part of Spain.

According to modelling results annual mean B[a]P air concentrations changed differently from 2008 to 2009 increasing or decreasing in some areas of the EMEP region up to 0.7 ng/m^3. Particularly, increase of B[a]P air concentrations was noted for Portugal, Spain, Lithuania, Latvia and Estonia, while levels of air concentrations in the Ukraine, Bulgaria, Italy, and France slightly decreased. These differences were mostly conditioned by the changes in emissions of particular countries and meteorological conditions. Model simulations of B[a]P pollution within the EMEP region showed that transboundary transport was a significant source of pollution for a lot of the EMEP countries contributing from 30% to 70% to total annual deposition. For 25 countries in 2009 its contribution exceeded 50%.

Comparison of modelling results and measurements of B[a]P in air for 2009 showed that for most of the sites differences between the modelled and observed concentrations ranged within 10-30%. Higher discrepancies were found for the sites DE1, NO42, and PL5. Particularly, differences between the measured and computed air concentrations for DE1 and NO42 were accounted for about 70% while underestimation of observed air concentrations at PL5 exceeded a factor of three which may be conditioned by the uncertainties of emission spatial distribution in the region surrounding the site. More detailed analysis of pollution levels in this region applying fine resolution modelling and detailed monitoring of PAH concentrations is required.

According to officially submitted data and expert estimates, emissions of four indicator PAHs within the EMEP domain decreased from 30% to 40% depending on the compound in the period of 1990-2009. Model evaluation of trends in B[a]P pollution levels showed that levels of its annual mean air concentrations declined in this period by about 30%.

Polychlorinated Dibenzo(p)dioxins and Dibenzofurans (PCDD/Fs)

Modelling of PCDD/F pollution levels for 2009 was performed using overall toxicity of 17 toxic PCDD/F congeners. Additionally, model simulations for individual PCDD/F congeners were carried out (2,3,4,7,8-PeCDF, 1,2,3,7,8,9-HxCDD, OCDD and OCDF) for 2006 and 2007 which allowed evaluating possible uncertainties in congener composition of emission data for PCDD/Fs. Analysis of these results was performed by MSC-E in co-operation with experts from Umeå and Stockholm Universities of Sweden.

Modelled levels of concentrations for individual PCDD/F congeners were compared with measurements made at Aspvreten (SE12), Pallas (FI96), and Vindeln (SE35) monitoring sites in 2006-2007. The comparison revealed the underestimation of measured air concentrations by about a factor of 5 for 2,3,4,7,8-PeCDF and larger differences (10 times and higher) for 1,2,3,7,8,9-HxCDD, OCDD, and OCDF congeners. Different level of underestimation for different congeners indicated that there were essential uncertainties in determination of congener composition of PCDD/F emissions. Therefore the data on congener composition of dioxins and furans emissions in the EMEP countries are highly appreciated.

Analysis of agreement between the modelling results and measurements for particular periods of time demonstrated possible discrepancies in spatial distribution of PCDD/F emissions. Particularly, the highest underestimation of air concentrations observed at the site SE12 was associated with atmospheric flows from south-south-west and south-south-east directions for all considered congeners.

At the same time, for other directions the underestimation was much lower, accounting, in example, for a factor of 2.7 for 2,3,4,7,8-PeCDF congener.

Comparison of modelling results for PCDD/F mixture with measurements of the sites Råö (SE14) and Aspvreten (SE12) showed the underestimation of observed PCDD/F air concentrations by approximately a factor of 5, which corresponded to the results obtained in the investigation for selected congeners. Examination of discrepancies between the modelling results and measurements indicated the need of the refinement of emission spatial distribution and organization of additional monitoring campaigns for PCDD/Fs.

Bearing in mind essential differences between the modelling results and measurements for PCDD/Fs, their transboundary transport was characterised using the export of pollution by the countries. Particularly, the export fraction of pollution does not depend on the emission inventory and can be used for evaluation of PCDD/F long-range transport. Thus, it was obtained that transboundary transport of PCDD/Fs significantly contributed to the pollution levels in the EMEP countries. The fraction of pollution exported beyond their boundaries varied typically from 30% to 60%. For 14 countries more than 50% of PCDD/F deposition due their national emission sources took place outside their boundaries.

Hexachlorobenzene (HCB)

In comparison with PAHs and PCDD/Fs evaluation of HCB pollution levels is complicated by more essential uncertainties in information on current sources of HCB release into the environment and historical emissions. Analysis of modelling results on HCB using available measurements of air concentrations revealed that model predictions underestimated observed pollution levels. This underestimation can be related to the incompleteness of available officially submitted emission data and expert estimates as well as with the underestimation of the role of secondary emission sources.

Re-volatilization of HCB from environmental compartments can essentially contribute to the contemporary pollution levels. Underestimated influence of historical emissions, particularly, underestimation of HCB accumulation in soil, was evaluated using the comparison of modelled HCB concentrations in soil with measurements. It was shown that model simulations based on the official emission data and expert estimates led to essentially lower levels of HCB soil concentrations compared to available measurements. To evaluate HCB re-emissions elaboration of scenarios of historical HCB emissions is required.

Model simulations with different conventional emission scenarios confirmed that changes of re-emission contributions and contemporary emissions of the EMEP countries could essentially improve the agreement between calculated and measured HCB air concentrations. Therefore thorough analysis of contemporary and historical emissions is needed to refine the assessment of HCB pollution levels.

Development of Global Modelling Framework GLEMOS for POPs

In order to describe the global-scale transport and accumulation of POPs the Global EMEP Multi-media Modelling System (GLEMOS) is being developed by MSC-E. This year additional processes describing POP fate in seawater were included, namely, POP transport with sea currents, diffusion, degradation, and sedimentation. Preliminary model simulations of POP global scale transport were performed with spatial resolution 1°×1°. Further improvement of POP global scale modelling system GLEMOS will include incorporation of vegetation compartment and its interaction with the atmosphere and soil.

Inter-linkages between Climate Change and POP Pollution

The influence of climate change on POP pollution has recently received increasing attention and is recognized as an important issue by many international organizations (CLRTAP, UNEP, AMAP, etc.). MSC-E has started to work in this direction evaluating sensitivity of POP transport and pollution levels to seasonal variations of selected meteorological parameters and land cover characteristics. Besides, the preparatory work for carrying out modelling experiments with climate change scenarios data was initiated.

Analysis of sensitivity of POP pollution levels to variation of meteorological and environmental factors showed that such factors as temperature, precipitation amount, wind speed and direction, outflow of air masses through the country boundaries, and vegetation cover, can be in most cases sufficient for explaining 90% – 95% of seasonal variability of chemicals air concentrations for a country. The effect of changes of meteorological and environmental factors can be essentially different for different POPs due to wide range of variations of their physical-chemical properties. Besides, the sensitivity of POP pollution levels to variations of meteorological and environmental parameters varies within the EMEP region which can lead to varied response to the climatic changes across Europe.

Presented approach for the evaluation of sensitivity of POP pollution levels to changes in meteorological and environmental factors can be applied to the analysis of model simulations of POP fate based on scenarios of future climate changes. At further stages of this work it is planned to perform a series of modelling experiments to explore both the effect of future changes of emissions, and the influence of projected climate changes on POP fate and behaviour.

Cooperation

This year MSC-E actively cooperated with the CLRTAP subsidiary bodies, EMEP task forces (TF MM, TF HTAP), international organizations (HELCOM, European Commission, UNEP), and national experts. MSC-E informed TF HTAP on the ongoing activities in the field of POP pollution assessment on a global scale and presented an overview of relevant research activities. In the framework of co-operation with HELCOM deposition of dioxins and furans to the Baltic Sea and their long-term trends were evaluated. The Centre participated in the AMAP scientific conference "The Arctic as a Messenger for Global Processes - Climate Change and Pollution" as well as in recent meeting of the EU ArcRisk project and took part in the discussion of the topics related to the linkages between the climate change and POP pollution. MSC-E supported development of local-scale modelling of POP pollution in Italy.

CONTENTS

INTRODUCTION

Persistent organic pollutants (POPs) is a group of substances that have toxic properties, resist degradation in the environment, bioaccumulate through food chains and are transported over long distances from their primary emission locations (by hundreds or thousands of kilometres) via the atmosphere and marine environment. They are known to cause harmful effects on human health and ecosystems, even at locations far from their initial release and for a long time after their emissions are stopped. Projected climate changes may directly and indirectly influence POP long-range transport and fate, and may alter exposure pathways and increase vulnerability for the biotic environment and related health impacts [*Dutchak and Zuber*, 2011].

According to the POP Protocol, the Cooperative Programme for Monitoring and Evaluation of Long-range Transmission of Air Pollutants in Europe (EMEP) provides the Executive Body for the Convention with information on deposition and transboundary transport of POPs within the geographical scope of EMEP. Emission data based on information reported by the EMEP countries is prepared by the Centre of Emission Inventories and Projections (CEIP). Measurements of POP concentrations in air and precipitation are carried out at the EMEP monitoring network under the methodological guidance of the Chemical Coordinating Centre (CCC). Based on this information, the Meteorological Synthesizing Centre – East (MSC-E) performs the assessment of deposition and air concentrations of POPs over the EMEP region linking together monitoring, emission and modelling information. Along with that the transboundary fluxes between the EMEP countries are evaluated.

This Status Report describes the progress in activities of MSC-E and CCC in the evaluation of contamination of the EMEP region by persistent organic pollutants (POPs). Major attention was given to the refinement of the assessment of POP pollution within the EMEP domain using an integrated approach, further developing of the global scale modelling of POPs, and exploring the links between the climate change and POP pollution. These activities were performed in accordance with the EMEP Work-plan for 2011 [ECE/EB.AIR/2010/5].

Evaluation of pollution within the EMEP domain for 2009 was performed for the following POPs: polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and HCB. Input information on emissions for model assessment was based on the most recent officially submitted emission totals and information on spatial distribution of emissions along with available non-Party emission estimates. For the evaluation of contributions of non-EMEP sources to the pollution of the EMEP domain and of re-emissions due to historical accumulation, available emission data for PCDD/Fs and HCB within the Northern Hemisphere were compiled. Measurements of POP concentrations in air and/or in precipitation were available from 23 stations. Evaluation of transboundary fluxes in the EMEP region (country-to-country matrices) was carried out for B[a]P, PCDD/Fs and HCB. The analysis of B[a]P trends for the period from 1990 to 2009 was performed.

The work on elaboration of the integrated measurement/modelling/emission approach to the evaluation of POP contamination levels was continued. The set of statistical indicators for the evaluation of agreement between measurements and model calculations was proposed. Methods of emission scenario evaluation based on back trajectories and matrix approach were considered and tested in the framework of preliminary analysis of the agreement between calculations and measurements of PAHs, PCDD/Fs and HCB.

MSC-E continued the development of the Global EMEP Multi-media Modelling System (GLEMOS) working out the modules describing POP fate in the environment including the processes in the main environmental compartments (soil, seawater) and exchange between these compartments and the atmosphere.

To explore the influence of climate change on POP fate in the environment the sensitivity of POP transport and accumulation in the environment to variations of meteorological and environmental parameters was analyzed for two pollutants: B[a]P and PCB-153. Further work in this direction will include modelling experiments based on the climate change scenarios to explore both the effect of future changes of emissions, and the influence of projected climate changes on POP fate and behaviour.

In the field of evaluation of POP pollution levels within the EMEP region, the EMEP Centres closely co-operated with the subsidiary bodies to the Convention, EMEP task forces, international organizations and programmes as well as with national experts. Special attention was paid to the collaboration with EECCA countries.

Detailed information on the work fulfilled during this year are presented in the Technical Reports of the EMEP Centres [*Shatalov et al.*, 2011; *Travnikov and Jonson*, 2011; *Aas and Breivik*, 2011] as well as on the Internet www.msceast.org and www.emep.int.

Below the content of the report is briefly outlined.

Chapter 1 is devoted to further development of integrated monitoring/modelling/emission approach to the assessment of POP environmental contamination. Statistical indicators applied for the evaluation of agreement between measurements and model calculations are reviewed and the set of basic and supplementary indicators is proposed. Specific tools for the analysis of discrepancies between model predictions and measurements are elaborated using back trajectories and matrix approach. Developed integrated approach is partially applied for the assessment of environmental contamination by PAHs, PCDD/Fs and HCB.

Chapter 2 describes the progress in monitoring activities on POPs carried out by CCC. The spatial coverage of the EMEP monitoring network for POPs is characterised. Particular attention is given to the evaluation of uncertainties of POP measurements and ongoing studies related to the evaluation of these uncertainties are outlined.

Chapter 3 presents results of the assessment of environmental contamination of the EMEP region by PAHs, PCDD/Fs and HCB in 2009. The chapter includes description of pollution levels and estimates of transboundary transport. Agreement between the model predictions and measurements is characterized and analysis of discrepancies is presented. Conventional scenarios of emissions for the considered POPs are constructed to analyze emission data uncertainty. Evaluation of sensitivity of the pollution levels to the emissions of particular countries is presented and preliminary recommendations for further refinement of pollution assessment for the EMEP region are formulated.

Chapter 4 provides information on further development of oceanic module of the global multi-media modelling system GLEMOS for POPs, which includes the processes of transport and diffusion within seawater, degradation, sedimentation, phase partitioning, and exchange with the atmosphere. The results of testing of the developed module are briefly outlined and pilot evaluation of the environmental contamination by PCB-153 on the global scale is presented.

Chapter 5 is devoted to MSC-E activities in the investigation of relationships between the climate change and behaviour of POPs in the environment. In particular, results of the analysis of POP pollution sensitivity on the example of B[a]P and PCB-153 to variation of meteorological and environmental factors are shown. Further work on modelling experiments with climate change scenarios data is outlined.

Chapter 6 highlights the co-operation of MSC-E with CLRTAP subsidiary bodies, EMEP task forces, international organizations, and national experts.

Future activities of the EMEP Centres in the field of POPs are outlined in ***Chapter 7***.

The main results of the EMEP Centres work in the field of the evaluation of pollution levels and transboundary transport of POPs are summarized in Conclusions. Detailed matrices of transboundary fluxes for 2009 calculated using MSCE-POP model for B[a]P, PCDD/Fs and HCB can be found in Annex A of the report.

Acknowledgements. The authors are extremely grateful to all participants of Swedish EPA project BalticPOPs for valuable discussions and comments with regard to the problems of assessment of environmental contamination by PCDD/Fs and for measurement data put on our disposal. In fact, the part of this report concerning PCDD/Fs is written in co-authorship with Karin Wiberg and Ian Cousins.

1. DEVELOPMENT OF INTEGRATED APPROACH FOR ASSESSMENT OF POP POLLUTION

In accordance with the recommendations of HTAP Assessment Report 2010, MSC-E continued the elaboration of integrated monitoring/modelling/emission approach to the evaluation of environmental contamination by toxic substances. This approach allows performing complex analysis of the information on the pollution provided by monitoring and modelling activities and indicating the areas of their further improvement, in particular, necessity to improve emission data, refine modelling approaches, apply fine resolution modelling, and conduct specific monitoring campaigns. The application of this approach allows reducing uncertainties of monitoring data, emission inventories and model predictions by considering interlinks between all these three types of information.

Integrated approach to the assessment of air contamination is carried out in several steps.

Step 1. Initial assessment. At this step assessment of the environmental pollution provided by the available monitoring data is combined with estimates obtained by modelling of transport and accumulation of the considered pollutant in the environment. The necessity of application of model estimates is conditioned by the fact that the coverage of the EMEP domain by monitoring sites measuring POPs is not sufficient for the evaluation of the contamination in the entire domain. In addition, modelling approach allows obtaining information which cannot be directly evaluated by monitoring data such as transboundary transport, life-times in the environment and its compartments, etc.

Step 2. Evaluation of the agreement between measurement data and model predictions. During this step model calculations are compared with available measurements. The agreement between calculation results and monitoring data can be evaluated with the help of statistical *indicators of agreement* such as correlation coefficients, measurement-to-calculation factors, mean square deviation, etc. The ideal situation is when these indicators are within agreed confidentiality intervals showing reasonable level of agreement between measurements and model predictions. However, if essential discrepancies between measurements and model calculations are found at this step, further analysis is required.

Step 3. Analysis of discrepancies. Here the disagreement between measurements and calculation results are analyzed and the reasons of the discrepancies are determined. Such reasons can be related to uncertainties in monitoring data (concerning representativity of monitoring sites, sampling procedures and laboratory analysis), simplifications in model descriptions of environmental processes, uncertainties in the set of physical-chemical properties for the considered pollutant used in modelling, uncertainties of the input data on atmospheric reactants, meteorology data and emissions. Simultaneously, necessary improvements leading to the refinement of the agreement between measurements and model results can also be determined.

Step 4. Refined assessment. At this step the evaluation of environmental contamination by the considered pollutant in the environment is carried out with the help of reliable set of measurements and refined emission data and model parameterization. Steps 2 and 3 can be repeated until the agreement between calculations and measurements meets the agreed set of indicators. So, the process of integrated assessment can be an iterative one.

Initial assessment of pollution (step 1) for the above listed contaminants is presented in Chapter 3. The present Chapter is focused at the procedure of analysis of monitoring/modelling discrepancies (steps 2 and 3). More detailed consideration of this procedure can be found in [*Shatalov et al.*, 2011]. Refined assessment (step 4) can be performed after necessary improvements in model design, monitoring and emission data with participation of national experts.

Evaluation of the agreement between measurement data and model predictions (step 2)

The section is devoted to the analysis of discrepancies between measurements and model calculations. This analysis is usually performed on the basis of statistical indicators of agreement. The overview of such indicators is carried out in this section.

When evaluating the agreement between measurements and model calculations a *target parameter* should be chosen. This parameter can be air concentrations averaged over various time periods (e. g., annual averages of air concentrations at a measurement site location), total deposition to the measurement site during some time period, etc. Further, measurements of the chosen target parameter to be included into the comparison should be selected. Below the number of selected measurements (made at various measurement sites and sampling periods) is denoted by N, y_1, ..., y_N are measured values of the target parameter, and z_1, ..., z_N are model predictions.

In the analysis of the agreement between measured values of the target parameter and their model predictions the calculation deficiency, that is, the differences between measured and calculated values of the parameter $y_j - z_j$, $j = 1, \ldots, N$ will play the crucial role. Since the number of random factors affecting calculation deficiencies $y_j - z_j$ is large, calculation deficiency can be viewed as a normally distributed variable as a first approximation. The following indicators can be used for the characterization of agreement of calculations with measurement data.

First, evaluation of unbiasedness of model predictions will be considered. Unbiasedness means that average values of measurements and calculations are equal:

$$\overline{y} = \overline{z}. \tag{1}$$

In other words, unbiasedness means that the average of calculation deficiency equals zero. Here three indicators of unbiasedness are considered. First is *mean bias* MB[1], that is, average value of calculation deficiency:

$$\mathrm{MB} = \overline{y} - \overline{z}. \tag{2}$$

The value of MB depends both on the closeness of averages of calculated and measured values of the considered parameter and on the absolute value of this parameter. So, some normalization of this indicator is useful. One of the methods of normalization (having possibly some limitations) is to divide the MB value (2) by the average of measurements. The obtained indicator is named *normalized mean bias* (NMB). However, such normalization is not applicable in the case when the average value of the target parameter is close or equal to zero (for example, such situation can take place when evaluating net gaseous flux of a POP through air/soil or air/water interface). Another difficulty in the application of this indicator is that it is hard to find strict justification for the choice of threshold level for this indicator.

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[1] Some of indicators considered below are earlier discussed in [*Derwent et al.,* 2009, *Thunis et al.,* 2011].

For some pollutants the values of NMB ranging between – 0.2 and 0.2 are viewed to be acceptable (see [*Derwent et al.,* 2009]).

Another way of normalization of MB coming from the theory of statistical hypothesis testing is the consideration of the *Student ratio* SR:

$$SR = \frac{\bar{y} - \bar{z}}{S/\sqrt{N}}, \tag{3}$$

where *S* is the square deviation of the calculation deficiency. Since it is assumed that calculation deficiency is distributed normally, the Student Ratio has the Student distribution with N – 1 degrees of freedom. This allows determining the threshold level for indicator (3) as 10% (or 5%) quantile of the Student distribution.

Unbiasedness is rather rough characteristic of the agreement between measurement and model prediction since it compares only average values. To give more detailed characteristics of this agreement the *correlation coefficient Corr* can be used. If this coefficient is close to 1, then the two considered sets of values are likely to be linked with linear relation. However, in this case the absolute values in the considered sets are not obligatory close to one another. Hence, this indicator can be considered as an additional one in evaluating the agreement between measured values of the target parameter and model predictions.

More information on the agreement between model predictions and measured values of the target parameter can be obtained from the consideration of regression relation between calculated and measured values of the parameter. Here the linear regression between these values is considered:

$$y = R \cdot z + B + \alpha, \tag{4}$$

where *R* and *B* are regression coefficients, and α is the approximation error, which is supposed to have zero average. The coefficient *B* is a characteristic of a systematic error. The absence of such an error is to some extent justified by small values of NMB or SR. The coefficient *R* is more important than *B* since this coefficient links variations between measured values and model predictions. Evidently, the best case is when this coefficient equals 1. Statistical testing of such hypothesis is usually performed with the help of the *Regression Coefficient Ratio* (RCR):

$$RCR = \frac{s_y\sqrt{N - 2}}{s_x\sqrt{1 - Corr}}(R - 1). \tag{5}$$

Here, as above, *Corr* means correlation coefficient between *z* and *y* values, and s_z and s_y are standard deviations for z and y, respectively. The RCR has the Student distribution with N – 2 degrees of freedom. This allows determining a threshold level for this quantity as its fractile at some confidence level (usually 10% or 5%).

To characterize the deviation between model calculations and measurements several indicators can be used. The first rough evaluation of the deviation of calculation results from measurements is *the factor*, that is, the ratio of highest from the considered values (calculations or measurements) to the lowest one. For POPs usually deviations within a factor of 2 – 3 are viewed as reasonable taking into account large uncertainties in physical-chemical properties, measurements and emissions.

Some other indicators of the agreement are considered in the literature (see [*Derwent et al.,* 2009, *Thunis et al.,* 2011]). They are: *Mean Gross Error* (MGE) defined as the average of the module of the difference between measured values and model predictions of the target parameter, *Root Mean Square Error* (RMSE) equal to the square root from the average of squares of calculation deficiency values and their normalizations NMGE and NRMSE obtained by the division of MGE and RMSE by the average of measured values. However, normalization by averages of observed values (as in the case of NMB, NMGE and NRMSE) or by square deviation of the difference (as in the case of SR) has its disadvantages. The matter is that under such normalization method possible large relative discrepancies for small values of the target parameter make small contribution to the normalized indicator. This means, for example, that small value of NMB indicates that the model is unbiased for large values of the target parameter but can be biased for small ones.

It should be mentioned also that RMSE is in essence a "mixture" of the standard deviation S of the calculation deficiency $y - z$ and its mean value MB (details can be found in [*Shatalov et al.*, 2011]). Thus, it can be replaced with S as an indicator of absolute deviation together with some indicator of unbiasedness (MB, NMB or SR). The advantage of the usage of S as an indicator is that its value allows evaluating standard deviation σ of calculation deficiency for the target parameter (see [*Shatalov et al.*, 2011]).

One more indicator which can be of use for evaluation of model performance is the sample coefficient of determination R^2. This indicator is defined as

$$R^2 = 1 - \frac{S^2}{S_y^2}, \tag{6}$$

where S^2 is the residual dispersion (the dispersion of calculation deficiency), and S_y^2 is the dispersion of the set of measured values of the target parameter. It can be treated as a fraction of the variability of target parameter reproduced by the model.

It should be taken into account that models can perform differently for different values of the investigated parameter. For example, atmospheric chemistry transport models may be designed for calculations of background concentrations and work worse calculating concentrations in contaminated regions. From this point of view, it seems to be reasonable to provide evaluation of model performance (with the help of the above indicators) separately for high, moderate and low values of the target parameter (e. g., concentrations of a contaminant in the atmosphere).

Taking into account the above discussion, the following conclusions can be made:

The following indicators are proposed as base ones for evaluation of model performance:

- Student Ratio SR given by formula (3) can be used as a characteristic of unbiasedness of the model. The threshold levels for this indicator can be chosen as 5% or 10% quantiles of the Student distribution with corresponding number of degrees of freedom.
- Regression Coefficient Ratio (RCR) can be applied as a measure of closeness of variations of measured values and model predictions of the chosen target parameter. The threshold levels for this indicator can be chosen similar to those for SR.
- Correlation coefficient *Corr* can be used as a measure of the dependence between measured (y) and calculated (z) values of the target parameter.
- Sample coefficient of determination R^2 might be applied as a general measure of the deviation between calculated and model predicted values of the considered parameter.

Additional parameters for evaluation may be:

- Normalized mean bias (NMB).
- Normalized mean gross error (NMGE).
- Normalized Root Mean Square Error (NRMSE).
- Regression coefficients between measured and calculated values of the parameter.

As stated above, it is proposed to perform evaluation of model performance separately for several ranges of magnitudes of the investigated parameter. For example, the question how well the model reproduces air concentrations of a pollutant can be performed separately for highly contaminated, moderately contaminated and background regions.

Below the relations between various indicators of model performance are illustrated by calculations of B[a]P transport in 2009 made by MSCE-POP model. The calculations were performed with the use of emission data compiled on the basis of official data reported to the UN ECE by European countries complemented by expert estimates by TNO [*Denier van der Gon et al.*, 2005]. Annual average of B[a]P air concentrations was used as a target parameter in the evaluation. Measurements and calculations of this parameter at 10 EMEP monitoring sites (BE13, CZ3, DE1, DE9, ES8, LV16, NO1/NO2, NO42, PL5 and SI8) were included in the analysis.

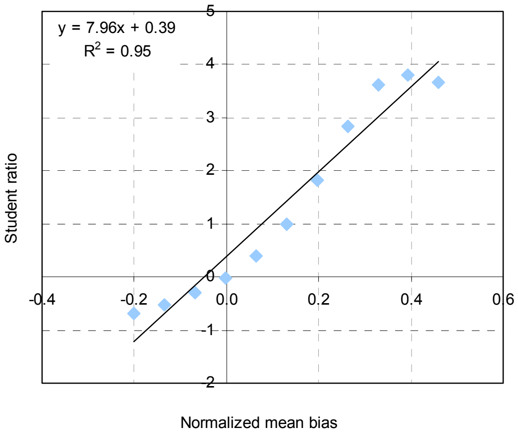


Fig. 1.1. *Comparison of normalized mean bias with the Student ratio*

To illustrate the interdependency of the considered indicators, model calculations with several emission scenarios are considered. These calculations show that there is close relations between some indicators. For example, the comparison of normalized mean bias and Student ratio is shown in the plot in Fig.1.1 together with the regression line. The reason for such strong dependence is that mean square error does not differ much between different scenarios. This once more confirms that only one of these two indicators should be viewed as a main one, and the other is additional.

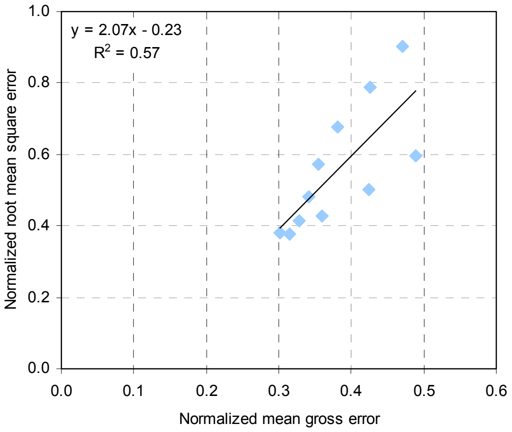


Fig. 1.2. *Comparison of normalized mean error with normalized root mean square error*

Slightly smaller correlation exists between normalized mean gross error and normalized root mean square error (Fig. 1.2). The correlation between these two indicators is explained by the fact that they both characterize one and the same property (closeness of model calculations to the measured values of air concentrations). Possibly, the analysis can be restricted by consideration only one of these indicators.

The application of the described indicators for the analysis of discrepancies between measurements and calculations are demonstrated below for PAHs and PCDD/Fs.

Analysis of discrepancies (step 3)

During model validation step for model calculations of POP transport and accumulation in the environment some discrepancies between model prediction and available measurements for a target parameter are usually found. There are three possible reasons of these discrepancies. First, it is the uncertainties in model description of environmental processes and model parameterization. The evaluation of this type of uncertainty was performed in the framework of EMEP model review and it was found that it typically leads to 50 − 70% uncertainty in model output (that is, calculated values of concentrations and deposition fluxes). Second, disagreement between measurements and model calculations can be conditioned also by possible uncertainties in measurement data concerning representativity of measurement sites as well as uncertainties of measurements at the stages of sampling and laboratory analysis. However, the latter uncertainties are normally not so large accounting for 30 – 50% (see [*Shatalov et al.*, 2005]). Third, the uncertainties of emission data can reach several times or even an order of magnitude in some cases (see evaluation of uncertainties in emission inventories in [*Nielsen et al.*, 2011; *Passant et al.*, 2011; *CITEPA*, 2011; *SEPA*, 2011; *SYKE*, 2011]).

It should be stressed that the refinement of the quality of monitoring data and model formulations is permanently in the focus of the work on the assessment of environmental contamination. Such activities as laboratory intercomparisons, model intercomparison studies, refinement of the data on physical-chemical properties of the considered pollutants on the basis of recent literature data are constantly used for this purpose. The disagreements between measurement and model results often show the direction of investigations in the fields of monitoring and modelling. However, the above estimates of uncertainties show that in the case when the discrepancy between monitoring data and model predictions are out of a factor of 3 – 4, and the uncertainties in model calculations and monitoring data are already investigated being in the above described range, uncertainties in emission inventories seem to be the most possible reasons of the disagreement.

Here the analysis whether the agreement between measurements and calculations can be improved by the refinement of emission data as most valuable source of uncertainty will be considered. The examination of the possibility of refining the agreement between calculations and measurements can be carried out on the basis of calculations with conventional emission scenarios, that is, with changed emission data. These scenarios should be chosen in such a way that the above described indicators have admissible values. The main difficulty in constructing and evaluating emission scenarios using modelling tools is that for the evaluation of each new emission scenario separate model run is required which takes a lot of computational resources. Below we focus on the methods of approximate evaluating of emission scenarios requiring less computational time.

Application of adjoint modelling. First of all, the method of adjoint modelling can be applied for evaluating large number of emission scenarios. This method is now widely used for the analysis of emission inventories based on available measurement data ([*Ustinov,* 2001; *Stohl at al.,* 2009; 2010; *Carouge et al.,* 2010a, b; *Villani et al.,* 2010; *Xiao et al.,* 2010]). Adjoint model calculates the so-called *influence function* whose values are in essence the sensitivities of the target parameter to emission densities at various locations. If the influence function for a target parameter is calculated, the value of this parameter can be easily calculated for any emission data. This allows calculating the values of target parameter under different emission scenarios without re-running the model. However, direct application of adjoint modelling requires in turn large computational time since this model should be run for a lot of target parameters. Such an approach can be used for scientific purposes but is not quite good for operational modelling. However, there exist a number of approximate solutions to the adjoint problem that can be used at the beginning steps of investigations. Below two methods of approximate evaluation of the solution of adjoint problems are described.

1. Back trajectory approach. This approach is based on the consideration of a simplified direct problem taking into account only advection and removal of a pollutant from the atmosphere. The solution of this problem can be explicitly expressed via trajectories of air masses arriving to the given point during the considered period. These expressions can be used for evaluation of the influence function under consideration.

As it is stated above, such approach does not take into account atmospheric diffusion. However, diffusion of a pollutant in the vertical direction can be as essential as its advective transport. So, it seems reasonable to consider two-dimensional transport problem in each horizontal layer with subsequent averaging of the results over height using the vertical profile concentrations characteristic of the considered pollutant. In the applications of this method, averaged vertical profiles of the considered substances obtained from the direct modelling were used. Certainly, this can lead to calculation uncertainties since vertical profile at particular locations can differ from the average one. Similar, usage of average removal rates from the atmosphere can lead to additional uncertainties in calculations. To partially overcome this difficulty, the evaluation procedure allows usage different removal rates for different time periods. The possibility of usage of the approach with the described simplification was checked for B[a]P in EMEP/MSC-E report [*Ilyin et al.*, 2010]. The application of this approach to the analysis of the consistency of emission data with available measurements is illustrated below in the investigation of the agreement between measurements and modelling results for calculations of transport and environmental accumulation of the mixture of 17 toxic PCDD/F congeners.

2. Matrix approach. This approach is based on the ability of direct transport models (e. g., MSCE-POP) to calculate contributions of prescribed groups of emission sources to air concentrations and deposition fluxes. Such emission groups can be, for example, sources of particular countries or sources of the chosen subregions. When this model feature is applied, model output includes spatial distributions of air concentrations and/or deposition fluxes (annual or monthly averages) originated from each source group considered in modelling. This is exemplified by Fig. 1.3 where B[a]P annual averages of air concentrations in 2009 originated from sources of two European countries (France and Finland) are presented.

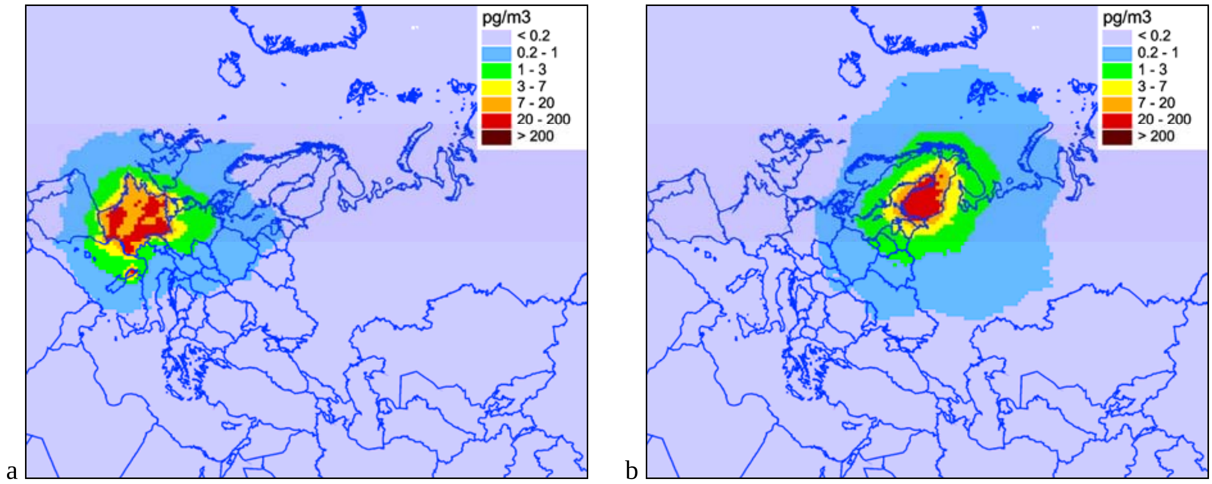


Fig. 1.3. *Spatial distribution of annual means of B[a]P air concentrations originated from sources of France (a) and Finland (b)*

On the basis of these data a lot of information can be obtained. The information most important for evaluation of different emission scenarios is *source-to-site matrix*. This matrix contains the contributions of the considered source groups to air concentrations/deposition fluxes at measurement site locations as calculated by the model. It should be taken into account that these contributions can be different for different years due to the variability of meteorological conditions (wind speed and direction, temperature regime, precipitation amounts, etc.).

As an illustration, contribution of European countries' emissions to air concentrations at BE13 (five main contributors) in January 2009 is shown in the diagram in Fig. 1.4.

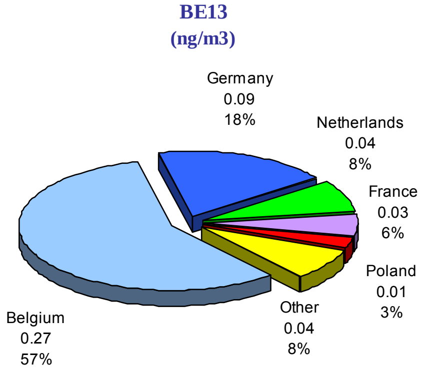


Fig. 1.4. *Contributions of emission sources of European countries to B[a]P air concentrations in January 2009 calculated by MSCE-POP model, ng/m*^{3}

If such information is available for all measurement sites and time periods included into the comparison, air concentrations/deposition fluxes at these sites corresponding to emission scenario can be obtained by scaling the contributions of all source groups in accordance to their new emissions and summing the results of scaling. Calculating air concentrations/deposition fluxes for various scenarios it is possible to choose the scenario for which the agreement between measurements and calculations will be the best possible one from the viewpoint of the indicators described in the previous subsection. This approach is applied below for examination of agreement between measurements and model results for PAHs, PCDD/Fs and HCB.

In the interpretation of the results obtained by the consideration of emission scenarios it is important to take into account that this approach allows changing emission totals for the considered source groups without modification of spatial distribution within these groups. Hence, if the "optimum" emission scenario assumes enlargement of emission total in a country, say, 5 times, this does not mean that emissions of this country are underestimated 5 times everywhere. It is possible that emissions are underestimated in some particular regions of the country only, and the conclusion derived from the results of such scenario is the necessity of more thorough investigation of spatial distribution of emissions for the country with participation of national emission experts. For detailed investigation of spatial emission distribution, source groups within the country should be as small as possible.

Further, scenario approach "as is" cannot provide right values of emissions in the unique way. For example, it is possible that reasonable agreement between measurements and calculations from the viewpoint of indicators can be achieved by large change of emissions in one country. However, the change of emissions needed for the refinement of agreement can exceed the known value of emission uncertainty. Further, usage emissions of sources with small contributions to air concentrations/deposition fluxes at all considered measurement sites for the refinement of the agreement can lead to the loss of stability in calculations. So, this approach allows investigation of the influence of sources to the given measurement sites only provided that the contributions of these sources to air concentrations/deposition fluxes to at least some of the considered sites is essential. This means that to obtain the information on more emission sources the spatial coverage of the EMEP domain by measurement sites should be as complete as possible.

Thus, the results of scenario calculations cannot point out definitely the uncertainty of emission data. Such calculations can be used to select “hot spots” in emission inventories subject to further investigation in collaboration with national experts in emissions and measurements.

The analysis of the discrepancies between measurements and model predictions allows determining main problematic areas in model parameterization and emissions used in modelling and the necessity of additional monitoring campaigns. This approach was tested for B[a]P and PCDD/Fs in Chapter 3. It is highly appreciated to receive comments on the proposed integrated approach and suggestions for its further development.

2. MONITORING OF POP POLLUTION LEVELS

2.1 EMEP measurements of POPs

POPs were included in the EMEP's monitoring program in 1999. However, earlier data has been available and collected, and the EMEP database thus also includes older measurements (see *http://ebas.nilu.no)*). A number of countries have been reporting POPs within the EMEP area in connection with different national and international programmes such as HELCOM, AMAP and OSPARCOM. Data from the open scientific literature are also used for model validation and complements the EMEP data. Detailed information about the sites and the measurement methods are found in EMEP/CCC's data report on heavy metals and POPs [*Aas and Breivik*, 2011].

In 2009 there were thirteen sites measuring POPs in both compartments (air and deposition), and altogether there were twenty-three measurement sites, which are three more than in 2008. Furthermore there are three sites in Spain delivering campaign data of PAHs. Most of the new additions to EMEP are sites measuring PAHs which is required according to the EU air quality directive [*EU*, 2004]. B[a]P, which is a by-product of incomplete combustion processes, is the most frequently measured POP component in EMEP. These results are therefore highlighted herein, whereas additional results for other compounds can be found in the annual data report [*Aas and Breivik*, 2011]. The spatial pattern of the average annual concentration level of B[a]P is shown in Fig. 2.1, where air concentrations seem to decrease when moving towards more remote areas in Europe. Notable differences in air concentrations can be seen between some adjacent sites suggesting that some EMEP sites may be influenced by local emissions of PAHs. In general, elevated concentrations of POPs are often seen in central parts of Europe [*Aas and Breivik*, 2011] reflecting proximity to major sources areas in Europe [*Halse et al.*, 2011; *Denier van der Gon et al.*, 2007].

Even though the spatial coverage has improved, there is still a need for more monitoring sites, especially in south – southeast of Europe to fulfil the goal of the EMEP monitoring strategy [*UNECE*, 2009]. There are, however, some positive developments in this region. At the EMEP sites in Moldova and Kazakhstan there will be one year of air and aerosol measurements of key POPs (PAHs, PCBs, organochlorine pesticides) from June 2009, a campaign financed by the Norwegian Ministry of Foreign Affairs. The results will be published in the next year status report.

2.2 Uncertainty in POP measurements

It is difficult to quantify the uncertainty in the POP measurements since it depends on several factors (methodology, sample handling and preparation etc) and the component in question. *Hayward et al.* [2010] compared high and low volume active samplers with two different types of passive samplers and they concluded that the annually averaged air concentrations determined by the different systems are within a factor of 2.5 for most pesticides. This is similar to what was observed when comparing results obtained on the basis of passive and active air sampling at various EMEP sites [*Halse et al.*, 2011]. The comparability between two identical samplers is usually better, but using different chemical laboratories may decrease the comparability significantly. In the POP laboratory intercomparison performed in 2000-2002 [*Manø and Schaug*, 2003] large uncertainties was seen for some labs – a bias of factor 2 or 3 for some components for some laboratories, though several laboratories were also within 20% of the expected value for most of the species that was included in the intercomparison (PAHs and organochlorine compounds). Due to these large uncertainties in the analytical performance across the EMEP network, it has therefore been a goal to follow up on the past intercomparison to check whether there have been any improvements in the comparability between the European laboratories. Since POPs are global pollutants, in addition to the fact that laboratory intercomparison is a costly and difficult exercise, EMEP CCC has joined forces with the Northern Contaminants Program (NCP) in Canada, which is coordinated by the Laboratory Services Branch (LaSB) of the Ontario Ministry of the Environment (MOE), to perform a new laboratory intercomparison. It will build on experience from the QA/QC program developed in the IPY project Intercontinental Atmospheric Transport of Anthropogenic Pollutants to the Arctic (INCATPA), lead by Environment Canada. In addition to the EMEP QA/QC objectives, this coordination will also allow for the assessment of data comparability between air monitoring programs, especially important also for future effectiveness evaluations under the Stockholm Convention on POP. All the laboratories participating in EMEP, HELCOM, OSPAR or AMAP have been invited to participate. This interlaboratory study aims to assess variability in analyzing standards/sample for four classes of trace organic chemicals, namely, polycyclic aromatic hydrocarbons (PAHs); polybrominated diphenyl ethers (PBDEs); polychlorinated biphenyls (PCBs); and organochlorine pesticides (OCPs). The results are presented and discussed in a separate report [*Schlabach et al.*, 2011]. A preliminary plot of the average results is given in Fig. 2.2. Here the analytical results were compared to ±20% of the target concentrations and the results show that the percentages of chemicals which fell within this range varied greatly among labs, concentrations of standards (high-std vs. low-std), as well as chemical groups. Not all of these labs are EMEP labs, but it shows the large variability of results among laboratories doing POP analysis and confirms the overall results given in *Manø and Schaug* [2003].

Table 2.1. *Measurement sites and program in 2009*

Country	Code	Name	POPs in air and aerosol	POPs in precipitation
Austria	AT0002R	Illmitz	PAHs	
Belgium	BE0013R	Houtem	PAHs	
	BE0014R	Koksijde		Pecticides, HCHs
Cyprus	CY0002R	Ayia Marina	PAHs	
Czech Republic	CZ0003R	Kosetice	PAHs, PCBs, pesticides, HCHs	PAHs, PCBs, pesticides, HCHs
Germany	DE0001R	Westerland	PAHs	PAHs, PCBs, pesticides, HCB, HCHs
	DE0003R	Schauinsland	PAHs	PAHs, PCBs, pesticides, HCB, HCHs
	DE0008R	Schmücke	PAHs	PAHs, PCBs, pesticides, HCB, HCHs
	DE0009R	Zingst	PAHs	PAHs, PCBs, pesticides, HCB, HCHs
Denmark	DK0010G	Nord, Greenland		pesticides, HCB, HCHs
Estonia	EE0009R	Lahemaa	PAH (Benzo[a]pyrene)	
Spain	ES0001R	San Pablo de los Montes	PAHs (campaign)	
	ES0007R	Viznar	PAHs (campaign)	
	ES0008	Niembro	PAHs	
	ES0014R	Els Torms	PAHs (campaign)	
Finland	FI0036R	Pallas/Matorova	PAHs, PCBs, pesticides, HCHs	PAHs, PCBs, HCHs
Great Britain	GB0014	High Muffles	PAHs, PCBs	
Iceland	IS0091R	Storhofdi	PCBs, pesticides, HCB, HCHs	PCBs, pesticides, HCB, HCHs
Latvia	LV0016R	Zoseni	PAH (Benzo[a]pyrene)	
Netherlands	NL0091R	De Zilk		γ-HCH
Norway	NO0042G	Spitsbergen	PAHs, PCBs, pesticides, HCHs, HCB	
	NO0001R	Birkenes	PCBs, HCB, HCHs	PCBs, HCB, HCHs
Poland	PL0005R	Diabla Gora	PAHs	PAHs
Sweden	SE0012R	Aspvreten	PAHs, PCBs, pesticides	PAHs, PCBs, HCHs
	SE0014R	Råö	PAHs, PCBs, pesticides	PAHs, PCBs, HCHs
Slovenia	Sl0008R	Iskbra	PAHs	PAHs

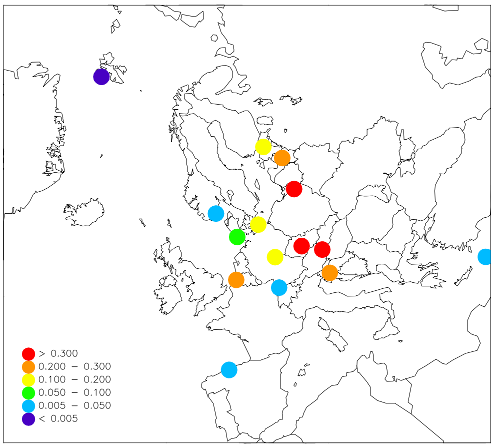


Fig. 2.1. *Spatial distribution of the annual average concentrations of B[a]P in 2009, ng/m^3.*
Note that Cyprus falls outside the map, but the data point is included and shifted somewhat further west

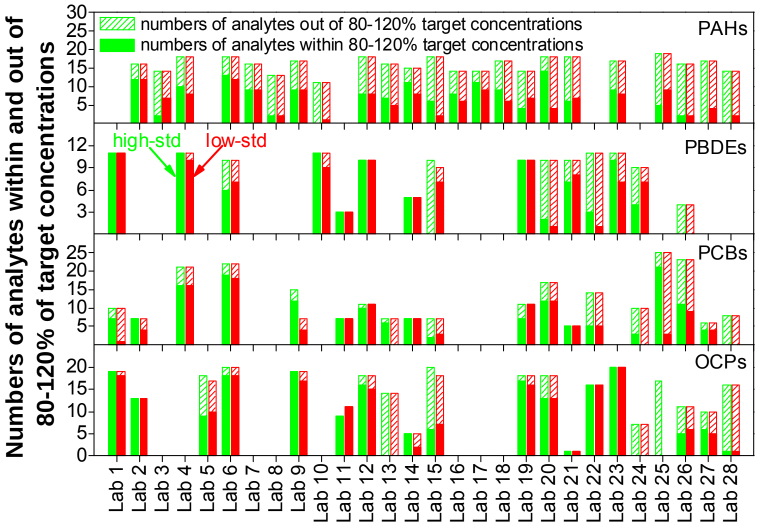


Fig. 2.2. *Preliminary results of the accuracy in analytical performance among the labs participating in the NCP, AMAP and EMEP laboratory intercalibration. In courtesy to Hayley Hung and Yushan Su at Environment Canada who have made the figure*

3. ASSESSMENT OF POP POLLUTION LEVELS, TRANSBOUNDARY TRANSPORT AND TRENDS

This year evaluation of environmental contamination of three pollutants (PAHs, PCDD/Fs and HCB) was performed. The attempt of application of integrated approach for all three pollutants was undertaken. In particular, the analysis of disagreement between measurements and model predictions on the basis of emission scenarios was done. The conclusions of this analysis can be applied in future for the refinement of model parameterization, monitoring data and emission inventories in co-operation with national experts.

3.1. Polycyclic Aromatic Hydrocarbons (PAHs)

According to the EMEP Work-Plan for 2011, modelling of the European contamination by four PAH species, namely, benzo[a]pyrene (B[a]P), benzo[b]fluoranthene (B[b]F), benzo[k]fluoranthene (B[k]F), and indeno[1,2,3-cd]pyrene (IP), in 2009 was performed. The four mentioned species are listed in the POP Protocol as indicator species for evaluation of POP contamination. Since the considered PAHs are mostly particle-bound, transport from emission sources located outside the EMEP region and re-emission from the underlying surface were not taken into account at this stage, and therefore modelling of PAHs was carried out without setting initial and boundary conditions. On the basis of model simulations, performed for the period 1990 – 2009 by the regional version of MSCE-POP model, analysis of trends in B[a]P contamination was carried out and B[a]P transboundary transport for 2009 was estimated.

Emissions of PAHs

Officially submitted data. Official data on the emission totals of polycyclic aromatic hydrocarbons (PAHs) were submitted by 39 countries for 1990-2009 (for at least one year). In comparison with the previous year additional two countries – Albania and Armenia – reported their emission data.

Emission totals for each of the four indicator PAHs for the considered period (for at least one year) were submitted by 27 countries, namely, Albania, Armenia, Belarus, Bulgaria, Croatia, Cyprus, the Czech Republic, Denmark, Estonia, France, Germany, Hungary, Iceland, Ireland, Latvia, Liechtenstein, Lithuania, Montenegro, the Netherlands, Poland, Republic of Moldova, Romania, Slovakia, Slovenia, Sweden, Switzerland and the UK.

Compared to the emission values of B[a]P used in modelling for 2008, emissions in Germany, Poland, Romania declined by 11, 8 and 7 tonnes, respectively. Significant increase of the emission value of the mixture of four indicator PAHs (PAH-4) was noted for Portugal (14 times) in comparison with previously reported data.

The information on PAH emission spatial distributions was provided by 26 countries (Austria, Belarus, Belgium, Bulgaria, Croatia, Cyprus, the Czech Republic, Denmark, Estonia, Finland, France, Germany, Hungary, Iceland, Ireland, Italy, Latvia, Lithuania, the Netherlands, Norway, Poland, Slovakia, Spain, Sweden, Switzerland, the UK). In comparison with the previous year Slovakia and Switzerland resubmitted information on spatial distribution for 1990, 1995, 2000 and 2005. Finland and Spain submitted gridded data for 2009.

Official information on B[a]P emissions by sectors for 2009 is available for 27 countries. According to these data the sector *1A4bi Residential - Stationary plants* represents the most significant source category of B[a]P emissions (Table 3.1).

Table 3.1. *Key source categories for B[a]P emissions in 2009*

NFR Code	NFR Category	Contributions to emission total, %	Cumulative Total, %
1 A 4 b i	Residential: Stationary plants	79.2%	79.2%
2 C 3	Aluminium production	4.5%	83.7%
1 B 1 b	Fugitive emission from solid fuels: Solid fuel transformation	3.6%	87.3%
1 A 4 a i	Commercial / institutional: Stationary	2.2%	89.5%
1 A 3 b iii	Road transport: Heavy duty vehicles	1.7%	91.2%
1 A 1 a	Public electricity and heat production	1.7%	92.9%

Officially reported information on uncertainties of PAH emissions for 2009 is available for Denmark, Finland, France, Sweden and the UK. According to the data submitted by the countries, the uncertainty of the Danish B[a]P emissions is 928% [*Nielsen et al.*, 2011], whereas for the UK uncertainty of B[a]P emissions is in the range of -60% to 200% [*Passant et al.*, 2011]. The uncertainty of French and Swedish PAH-4 emissions is 75% [*CITEPA*, 2011] and 583% [*SEPA*, 2011], respectively. For the Finnish PAH-4 emissions the uncertainty is in the range from -78% to 170% [*SYKE*, 2011].

Emission data used for modelling. The data on emission totals from the EMEP countries for 2009 used for modelling were based on the official data received from the EMEP Centre on Emission Inventories and Projections (CEIP) [http://www.emep-emissions.at/ceip/]. For European countries, which did not report their emissions, unofficial data of emission inventories [*Denier van der Gon et al.*, 2005; *MEPA*, 2007] were used. The gridded emissions for 2009 were prepared by CEIP for EMEP countries with spatial resolution 50×50 km^2.

The official information on emissions for the Asian part of the EMEP domain was not available. The emission data for this region were prepared by MSC-E. The B[a]P emissions in the Asian part of Russia were estimated using official emission data for the European part of the country and the ratio between the population of the European and the Asian parts of the country. The B[a]P emission values for Tajikistan, Turkmenistan and Uzbekistan were taken from the global atmospheric emission inventory of PAHs prepared by *Y.Zhang and S.Tao* [2009]. For Kazakhstan and Kyrgyzstan unofficial emission data from TNO emission inventory [*Denier van der Gon et al.*, 2005] were applied. The spatial distribution of B[a]P emissions in the Central Asian countries and the Asian part of Russia was determined on the basis of data on population density [*Li*, 1996] obtained from the web site of Canadian Global Emissions Interpretation Centre [*http://www.ortech.ca/cgeic*].

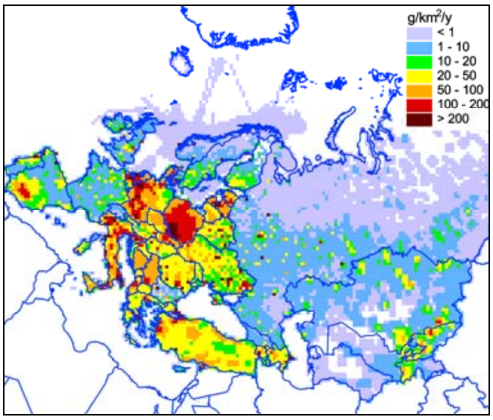


Fig. 3.1. *Spatial distribution of B[a]P emissions in 2009 over the EMEP domain with resolution 50×50 km^2, g/km^2/y*

The spatial distribution of B[a]P emissions for 2009 is illustrated in Fig. 3.1. Elevated levels of B[a]P emissions (20 – 200 g/km^2/y) can be noted for the central, southern and eastern parts of Europe. Countries of Northern and Western Europe, Russia, and the Central Asian countries are characterised by relatively low emission fluxes (1 - 20 g/km^2/y).

Total B[a]P emission within the EMEP grid in 2009 used in model simulations is estimated as 502 tonnes. This value includes 470 tonnes from emission sources located in European countries and 32 tonnes – from the Central Asian region. Maximum contribution to the total B[a]P emission within the EMEP domain in 2009 was made by the Ukraine (20%) followed by Poland (8%), Portugal (8%), Romania (8%) and Turkey (8%).

Emission trends. According to the official and unofficial information, emissions of four indicator PAHs within the EMEP domain decreased by about 30% - 40% depending on the compound in the period from 1990 to 2009. Temporal variations of total emissions of four indicator PAHs within the EMEP region are displayed in Fig. 3.2.

Among the countries submitted official data on PAH emissions for 2009, maximum emission reduction within the considered period took place in the UK (95%), Norway (85%), Germany (81%), Republic of Moldova (77%) and the Netherlands (75%). At the same time in Denmark, Estonia, Italy, Latvia, Cyprus and Portugal PAH emissions were increased in comparison with the level of emission in 1990.

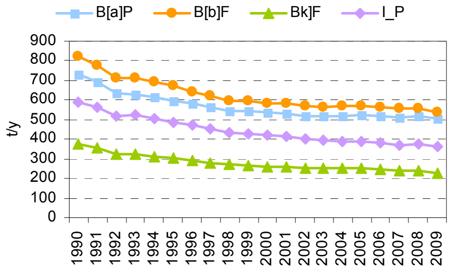


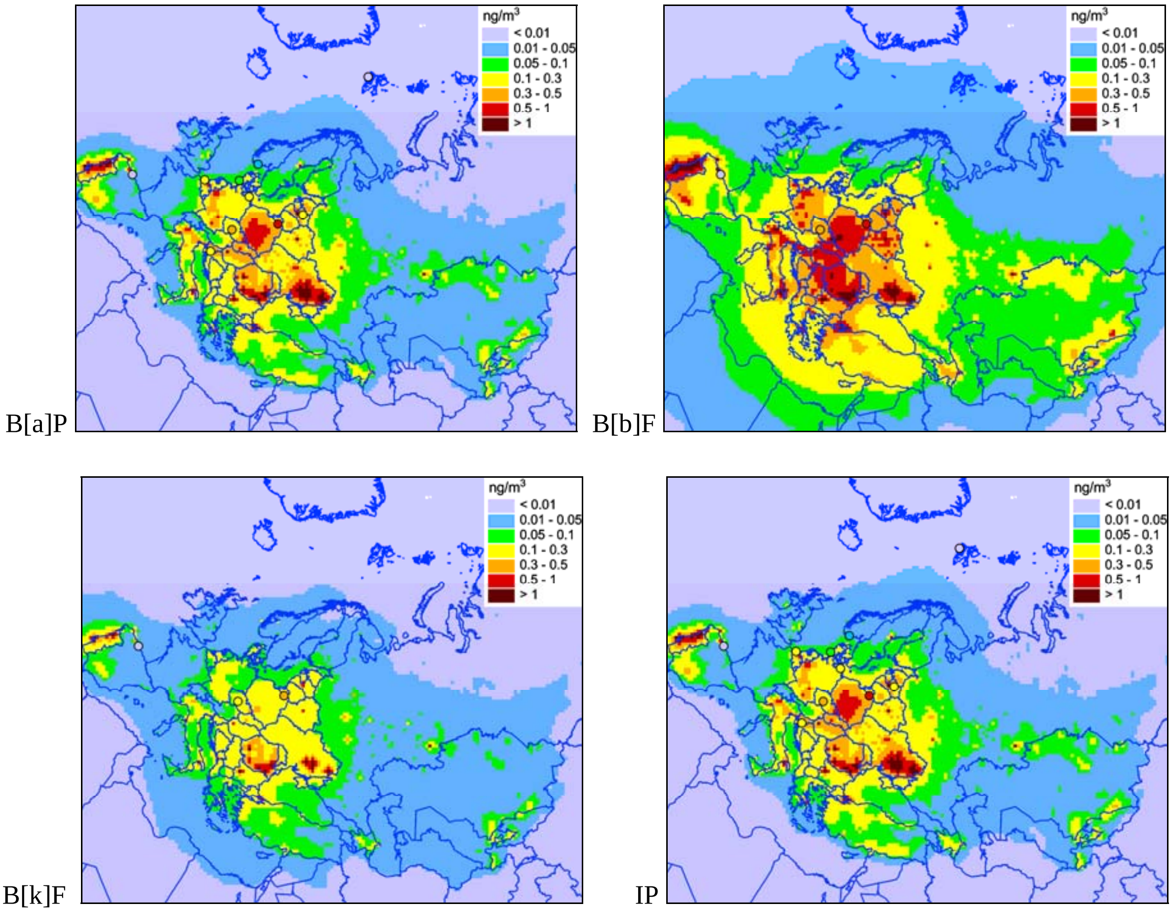
Fig. 3.2. *Temporal variations of PAH emissions within the EMEP grid in 1990-2009, t/y*

Evaluation of contamination in the EMEP region.

Contamination levels. Spatial distributions of annual means of air concentrations of four considered species as predicted by the model are displayed in Fig. 3.3.

It is seen that the highest concentration levels among the considered four PAH species are found for B[b]F and IP. Slightly lower contamination levels are characteristic of B[a]P. The lowest levels of contamination are obtained for B[k]F. This is confirmed by measurements available at EMEP measurement sites.

For all the considered pollutants, the areas with high contamination levels are Central and Eastern Europe, Portugal and the western part of Spain. Clean regions are located on the Scandinavian Peninsula, in the UK and partly in France. However, spatial distributions of contamination are also different for the considered PAHs. This is illustrated by Table 3.2 where average air concentrations for five countries with maximum contamination for each of the four PAHs are given.



B[a]P B[b]F

B[k]F IP

Fig. 3.3. *Spatial distribution of air concentrations of four indicator PAH species (B[a]P, B[b]F, B[k]F and IP) in 2009 as predicted by MSCE-POP model and given by measurement data, ng/m*^{3}

Table 3.2. *Average air concentrations of the four considered PAHs for five most polluted European countries, ng/m*^{3}

Benzo[a]pyrene		Benzo[b]fluoranthene		Benzo[k]fluoranthene		Indeno[1,2,3-cd]pyrene	
Country	Concentration	Country	Concentration	Country	Concentration	Country	Concentration
PT	0.71	PT	1.31	RO	0.44	PT	0.76
UA	0.60	RO	0.81	PT	0.36	PL	0.60
RO	0.52	SK	0.63	MD	0.25	UA	0.57
PL	0.46	PL	0.62	UA	0.25	RO	0.48
SK	0.40	HU	0.61	BG	0.22	SK	0.45

It is seen that the list of five countries with highest contamination levels is different for different PAHs. For example, the Ukraine is present among these five countries for B[a]P, B[k]F and IP and is not in the list of five countries with maximum air concentrations for B[b]F. The order of countries in the list is also different. For example, Romania is the most contaminated country for B[k]F and stands on the fourth place for IP.

Compared with the calculations of the previous year, total European emissions are almost unchanged. However, there are essential differences in spatial distribution of emissions over the EMEP domain. The map of differences in air concentrations calculated for 2008 and 2009 is presented in Fig. 3.4.

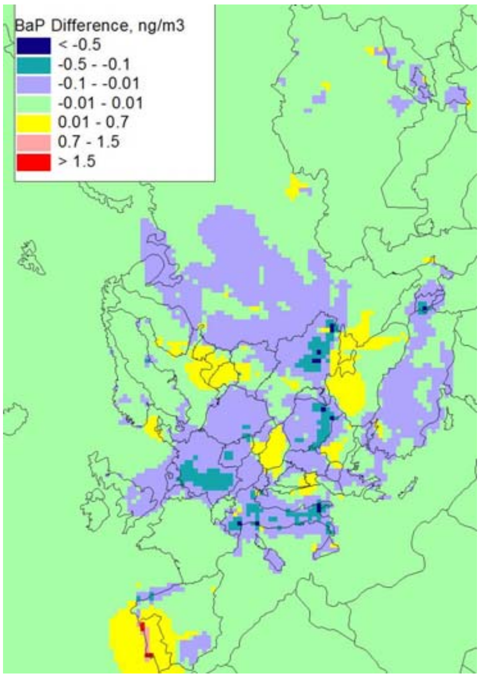


Fig. 3.4. *Spatial distribution of differences in B[a]P air concentrations as predicted by MSCE-POP model for 2008 and 2009, ng/m*^{3}

Essential increase of air concentrations (about 0.5 ng/m^3) is calculated for Portugal and part of Spain, Latvia, Lithuania and Estonia, Slovakia and Hungary. Inversely, the concentrations are dropped essentially (by 0.1 − 0.5 ng/m^3) in the Ukraine, Romania, Germany and Italy. The reason of changes in air concentrations is both in the change of meteorological conditions and in emissions of the countries. For example, emissions of Portugal are enlarged by the country about 14 times, what was the reason of enlargement of calculated air concentrations in Portugal and (partly) Spain.

Most measurements confirm the predictions made by the model. For most sites the difference of measured and calculated values of B[a]P air concentrations range from 10 to 30%. At DE1 and NO42 the difference is about 70%. However, at two sites (ES8 and PL5) measurements and calculations differ from each other. At ES8 model results occur to be higher than measurements from 6 to 14 times depending on the chemical in question. It should be mentioned that about 85% of measurements at this site is below the detection limit. The same situation takes place at sites ES1, ES7 and ES14 where almost all measurements of air concentrations are below the detection limit. The additional measurement data in Spain, Portugal, France and Italy could be of use for more thorough analysis of contamination in the south-west of Europe.

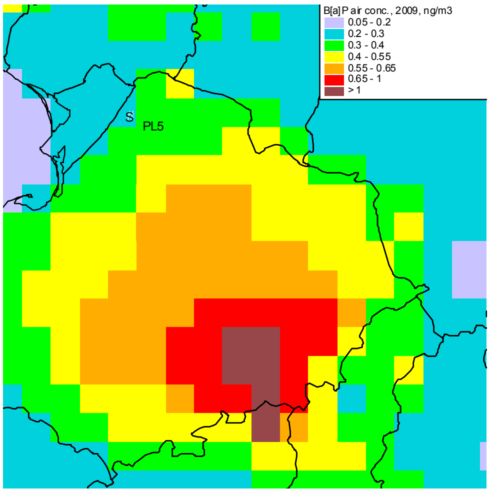


Fig. 3.5. *Spatial distribution of B[a]P air concentrations near site PL5, ng/m*^{3}

On the opposite, model predictions of air concentrations at PL5 are 2 – 4 times lower than measurements. Since site PL5 is located on the border of highly contamination region (see Fig. 3.5), it could be assumed that the disagreement between measurements and model predictions at this site could be conditioned by uncertainties in emission spatial distribution in this region. For the investigation of this hypothesis emission sources of countries surrounding the site can be split in several source groups and calculations with higher spatial resolution can be used for evaluation of contamination levels in the considered region. Such consideration can be performed with participation of national experts on emissions and measurements. Similar analysis is performed for heavy metals for a number of countries.

Source-receptor relationships. Modelling of long-range transport and deposition of PAHs within the EMEP domain allowed evaluating source-receptor relationships, that is, contributions of national emission sources, transboundary transport, and re-emission to air concentrations and deposition fluxes in various countries or/and at different locations. Source-receptor relationships of PAHs will be illustrated by those of B[a]P. The information on source-receptor relationships for other PAHs can be found in the Annex A to this report.

<u>*Transboundary transport.*</u> One of the applications of evaluated source-receptor relationships is calculation country-to-country matrices for deposition fluxes. These fluxes include contributions of national sources, transboundary transport from other European countries and re-emissions. Estimates of total annual deposition of B[a]P over the European and the Central Asian countries in 2009 are presented in Fig. 3.6 along with contributions of the above emission source groups.

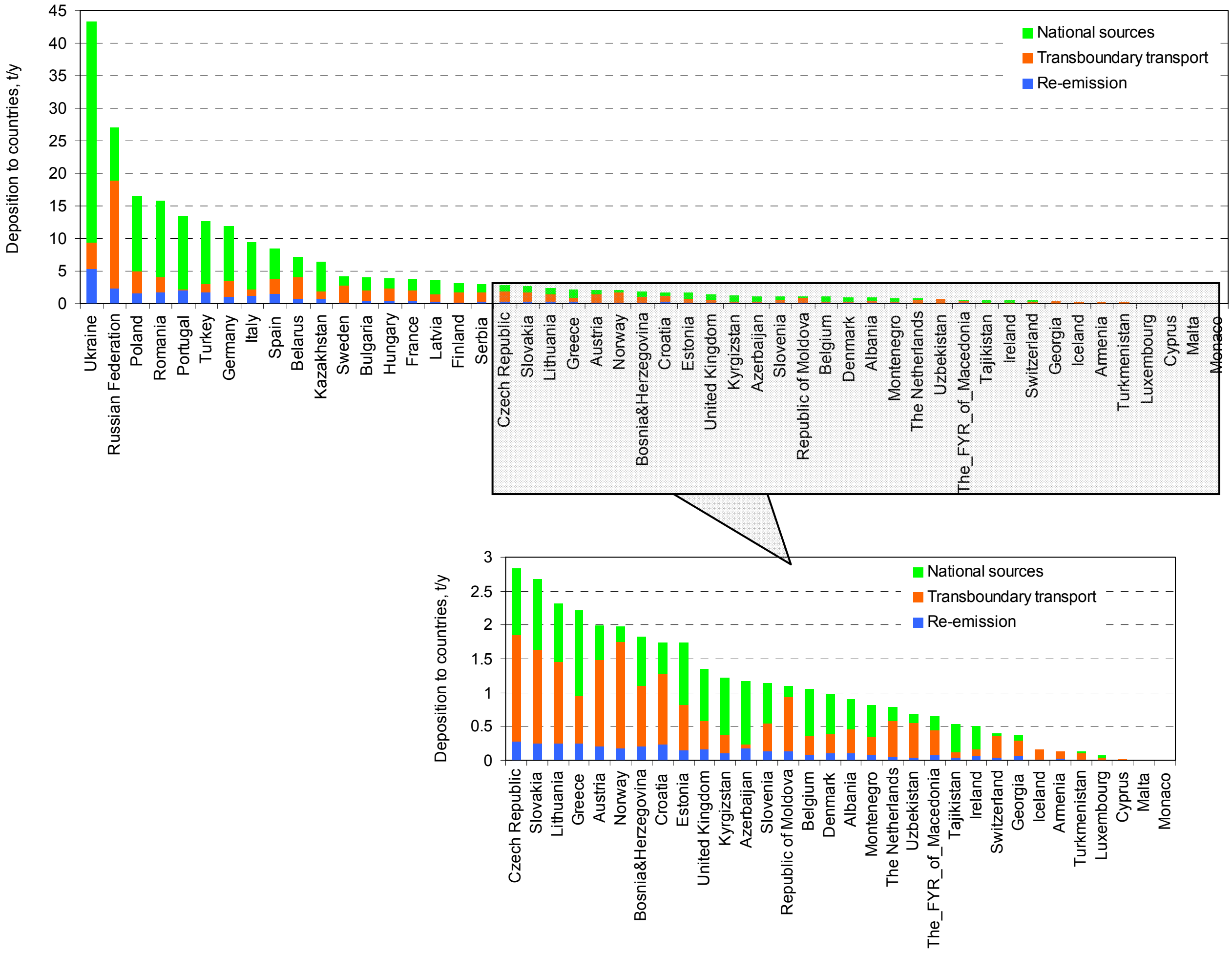


Fig. 3.6. *Total annual deposition of B[a]P over the European and the Central Asian countries in 2009 and contributions of national emission sources, transboundary transport within the EMEP region and re-emission, t/y*

It can be seen that transboundary transport of B[a]P is a significant source of pollution for a lot of European countries with typical contribution from 20% to 70%. For 12 countries the contribution of B[a]P transboundary transport (import fractions) exceeds 70%, and for 30 countries – 40%. Transboundary transport and its contribution to total deposition depends on a number of factors like the size of a country territory, peculiarities of meteorological conditions, and magnitude of domestic emission of a given country. In particular, for countries with significant national emissions, in comparison with the emissions of surrounding countries, the contribution of transboundary transport is typically low, like for instance, for the Ukraine (11%), Germany and Poland (22%). At the same time for countries with relatively small territory or low emission the contribution of transboundary transport can be essential (Switzerland – 94%, Norway – 88%).

It should be taken into account that the contributions from external sources are subject to spatial variability, so that in some subregions of a country they can be essentially higher than the average over the country. This phenomenon is illustrated by spatial distributions of transboundary fractions of air concentrations in Spain and France (Fig. 3.7).

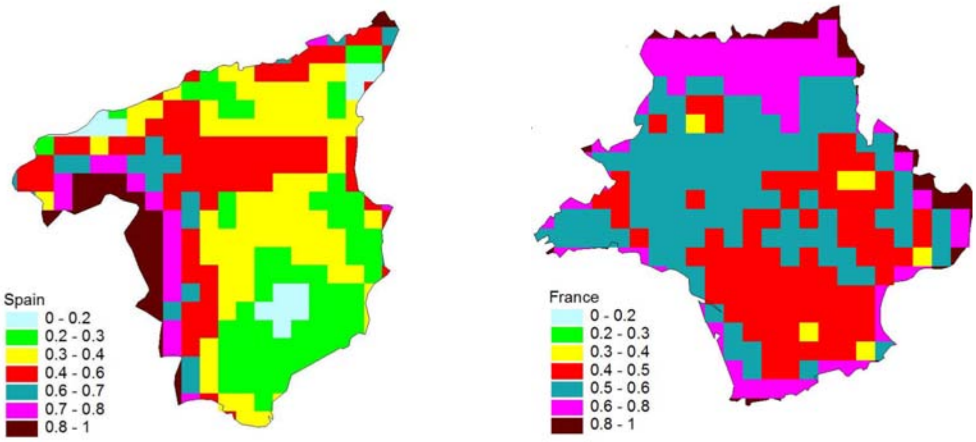


Fig. 3.7. *Spatial distributions of import fractions for air concentrations in Spain and France*

It can be seen that though average fraction of the imported pollution in Spain and France is 32% and 50%, respectively, this fraction can reach 80% and more in some particular regions in these countries. This shows that the investigation of import fractions in the European countries with finer spatial resolution is reasonable.

The contribution of a particular country to the transboundary transport of pollution can be also characterised by the fractions of total deposition originated from its national emission sources deposited outside (export) and inside its territory. This information is shown in Fig. 3.8. It is seen that essential fraction of pollutant emitted from the country sources can be transported outside their boundaries. In particular, this fraction varies from almost 100% for Monaco to about 8% for Iceland. It is worth mentioning that the fraction of transboundary fractions of deposition in a country depends on the meteorological conditions of the considered year. As an example, transboundary fraction of Switzerland changed form about 95% in 2008 to 60% in 2009.

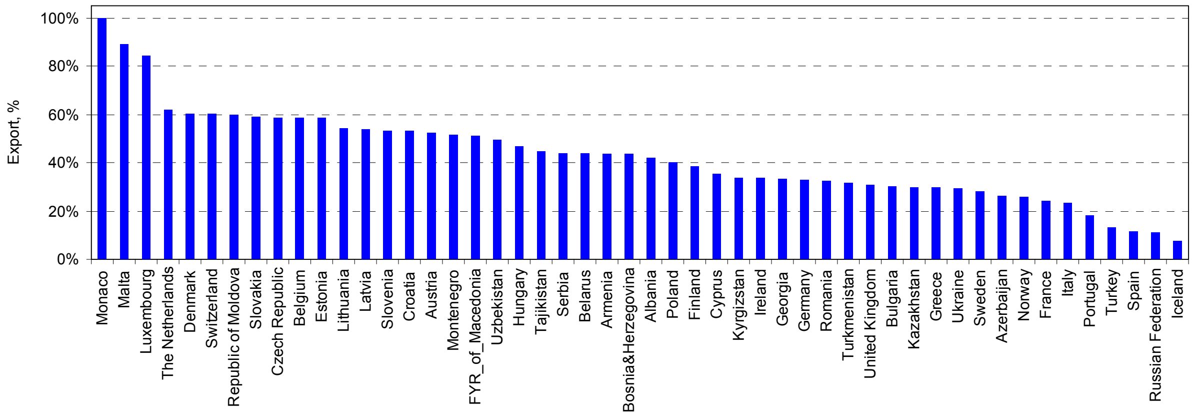


Fig. 3.8. *Fractions of B[a]P deposition originated from countries emission sources and occurred outside their territories in 2009 (export), %*

On the basis of country-to-country matrices, more detailed information on import and export is prepared for each EMEP country. The information on import consists of fractions of deposition to the country originated by emission sources of all other European countries (import charts). An example of import and export charts for the Netherlands is presented in Fig. 3.9.

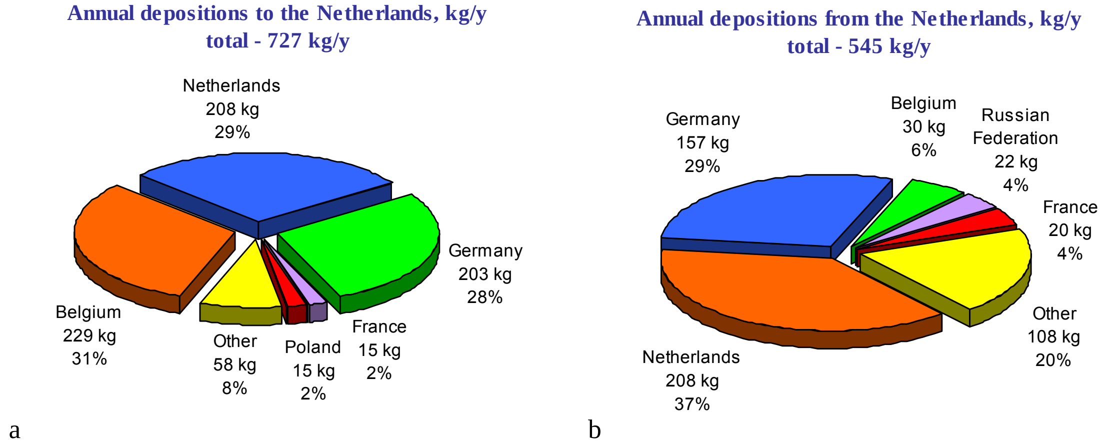


Fig. 3.9. *Import (a) and export (b) of B[a]P deposition for the Netherlands (2009), kg/y*

Source contributions at measurement sites. Another important application of calculated source-receptor relationships is the possibility to evaluate various emission scenarios in the course of the analysis of the agreement between measurements, calculation results and emission data in the framework of the integrated monitoring/modelling/emission assessment of environmental pollution. The evaluation of emission scenarios is performed on the basis of contributions of the considered emission sources to air concentrations at the locations of measurement sites (country-to-site matrix) which can be calculated for all measurement sites with the information on the considered pollutant available. This information allows evaluating the sensitivity of calculated air concentrations at the location of measurement sites with respect to emissions of all considered sources. The contributions of various sources to calculated B[a]P air concentration values at site locations is exemplified by two EMEP sites BE13 and PL5 (Fig. 3.10).

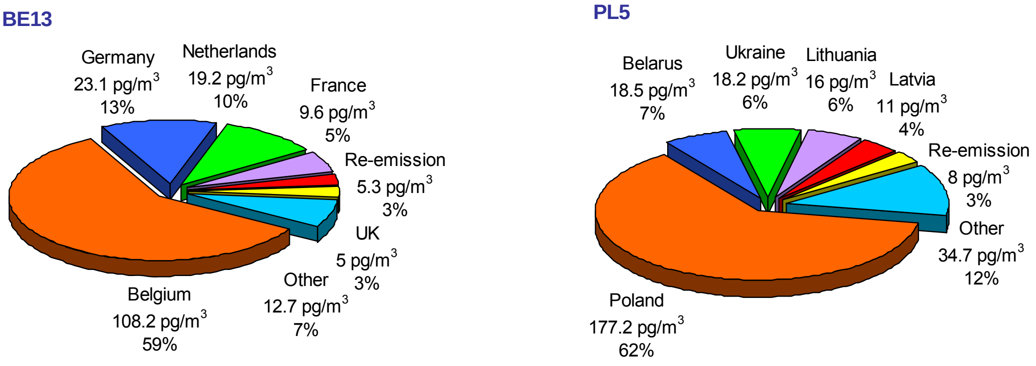


Fig. 3.10. *Contributions of various emission sources to the calculated values of air concentrations at BE13 and PL5, pg/m^3*

With this information, it is possible to evaluate changes in air concentrations due to the change of emissions of this or that source and to construct emission scenarios refining the agreement between calculations and measurements at the given site or at a number of sites. This approach will be used in the next section.

Application of integrated approach: analysis of measurement/calculation discrepancies

This section is devoted to the analysis of the discrepancies between measurements and model predictions. Since for B[a]P maximum information on measurements is available in comparison with the other considered PAHs, the analysis will be performed for B[a]P only.

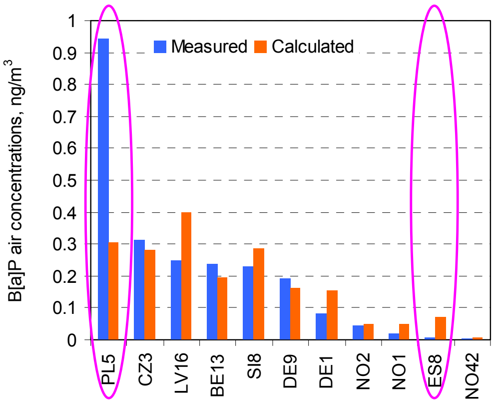


Fig. 3.11. *B[a]P contamination levels (air concentrations) obtained by measurements at EMEP monitoring sites in 2009 in comparison with model predictions, ng/m^3*

<u>*Spatial resolution.*</u> Annual averages of B[a]P air concentrations obtained by measurements at 9 EMEP monitoring sites are presented by the plot in Fig. 3.11.

The values of air concentrations measured at almost all monitoring sites show the same contamination levels (0.1 – 0.3 ng/m^3 in the Central and Western Europe and lower than 0.05 ng/m^3 in the remote regions) as the model does. As mentioned above, the exceptions are sites PL5 and ES8 (marked by red ovals in Fig. 3.11). It should be noticed that at ES8 about 85% of measurements are below the detection limit.

To reveal the reasons of underestimation of air concentrations at PL5 it should be taken into account that the uncertainties due to model parameterization is estimated as 40 – 50%. Hence, large discrepancy between measurements and model predictions at PL5 (almost three times) can be (at least partially) explained by uncertainties in the emission inventory. To examine the possibility of refinement of the agreement between measurements and calculations by changing emission data calculations with emission scenarios were carried out. For the construction of these scenarios the approach based on the country-to-site matrices was used.

It is found that enlarging emissions in Poland 3.2 times allows reducing the disagreement between calculation and modelling at PL5 up to about 30% with minor changes of the agreement at other EMEP sites. The results of the scenario calculations together with the results of initial calculations and measurement data are shown in Fig. 3.12 (the results at ES8 are excluded).

Under such scenario for almost all measurement sites measurement data agree with model prediction within a factor of two. The exceptions are DE1 and NO42 where measurement-to-calculation factor equals 2.3 and 2.7, respectively. The application of this scenario allows enlarging correlation coefficient between measurements and model predictions from 0.57 to 0.94. Regression coefficient becomes 1.17 compared with 1.32 for initial calculations. The full set of statistical parameters for initial calculations in comparison with those for scenario calculations is presented in Table 3.3. The values of the Student ratio and Regression coefficient ratio for the scenario calculations are below the threshold level.

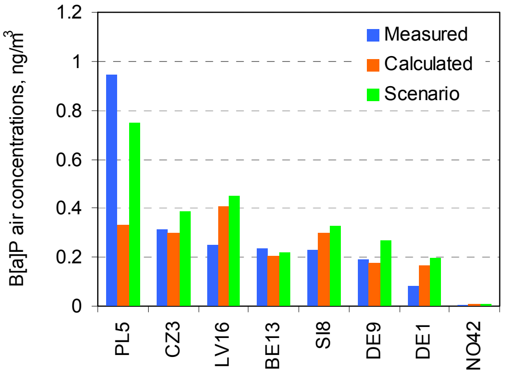


Fig. 3.12. *B[a]P contamination levels (air concentrations) obtained by measurements at EMEP monitoring sites in 2009 in comparison with model predictions and scenario calculations, ng/m^3*

Table 3.3. *The change of statistical parameters of the agreement between calculations and measurements due to the consideration of emission scenario*

	Initial calculations	Scenario calculations
Student ratio	0.56	-1.50
Regression coefficient ratio	2.74	2.05
Correlation coefficient	0.57	0.94
Coefficient of determination R^2	0.40	0.88
Normalized mean bias	0.16	-0.19
Normalized mean gross error	0.48	0.38
Normalized root mean square error	0.23	0.11
Regression slope	1.32	1.17
Regression intercept	-0.03	-0.11

The results of the above analysis do not mean in essence that the emissions in Poland are underestimated about three times in total. As it has been already noted, the reason of the underestimation of air concentrations at PL5 by the model can be conditioned also by uncertainties in spatial distribution of the emissions in the region surrounding the considered site. To refine the evaluation of contamination in this region modelling with finer spatial resolution can be of use. However, for such kind of modelling the data on spatial distribution of emissions in Poland (and possibly in neighbouring countries) are needed.

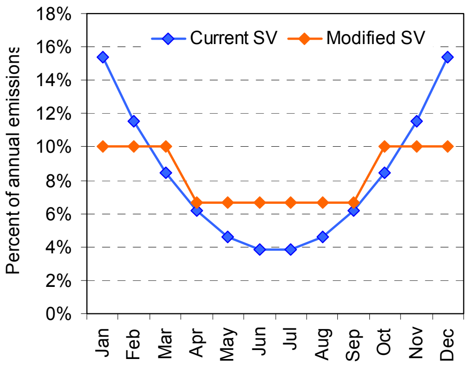


Fig. 3.13. *Currently used and modified seasonal variation of B[a]P emissions*

Temporal resolution. Except for annual means of B[a]P concentrations, seasonal variations of this pollutant are of importance since the level of air concentrations can essentially change within the year. Seasonal variations (SV) of the pollution are conditioned by two factors. First is temperature dependence of PAH degradation and deposition fluxes, and the second is seasonal variations of emissions. The first factor is taken into account in model calculations. However, under the existing assumption on emission seasonal variations (see [*Baart et al.*, 1995]) the model essentially underestimate seasonal variations of pollution obtained at measurement sites. To refine the agreement between measurements and model results at the level of monthly averages special emission scenario with stronger seasonal variations of emissions was considered. The plot with currently used and modified emission seasonal variations is displayed in Fig. 3.13.

The comparison of calculation results using previous and modified emission seasonal variations with measurements at two EMEP monitoring sites (BE13 and SI8) is shown in Fig. 3.14.

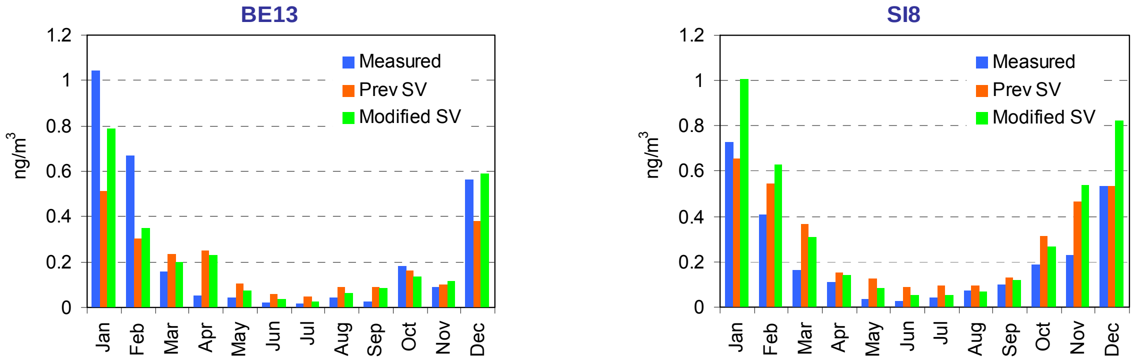


Fig. 3.14. *Comparison of calculation results using previous and modified seasonal variation (SV) of B[a]P emissions with measurements at two EMEP monitoring sites, ng/m^3.*

Correlation coefficient between measurements and model results has been enlarged due to seasonal variation change from 0.92 to 0.94 for BE13 and from 0.92 to 0.98 for SI8.

Trend analysis

This section is focused on the analysis of trends of B[a]P contamination from 1990 to 2009. Calculations were performed by regional version of MSCE-POP model using official emission data complemented by expert estimates when necessary.

The trend of emission density averaged over the EMEP domain is shown in Fig. 3.15a.

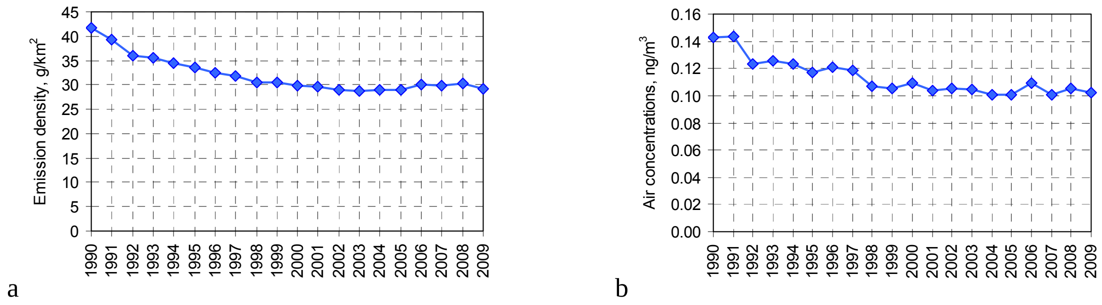


Fig. 3.15. *Trends of B[a]P emission density, g/km^2/y (a) and air concentrations, ng/m^3 (b) averaged over the entire EMEP domain*

The reduction of emission density from 1990 to 2009 amounts to about 30%. The reduction of air concentrations in the EMEP domain is almost the same (see Fig. 3.15b). This indicates that for levels of air concentrations for the considered PAHs are determined rather by current emissions than by re-emission of earlier accumulated contaminant from the underlying surface.

It should be mentioned that the reduction of emissions and air concentrations are different in different countries. To illustrate possible dynamics of contamination in particular EMEP countries three countries were chosen: Finland (FI), France (FR) and Denmark (DK). These three countries are characterized by different dynamics of contamination.

In Finland emissions are almost stable within the considered period with slight variations (Fig.3.16a).

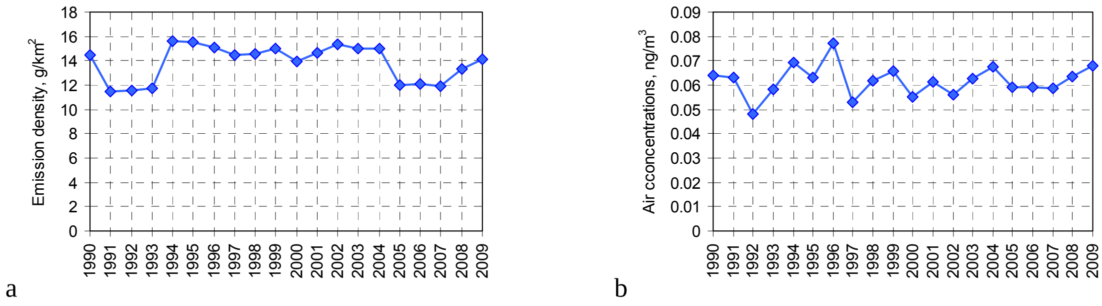
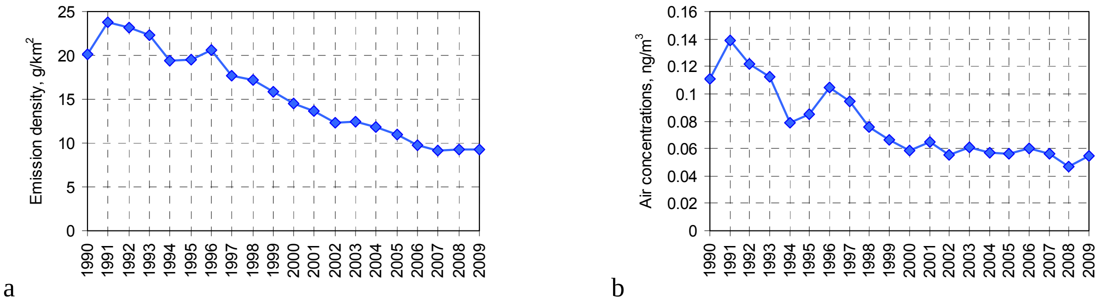


Fig. 3.16. *Trends of B[a]P emission density, g/km^2/y (a) and air concentrations, ng/m^3 (b) in Finland*

The same situation takes place for air concentrations (Fig. 3.16b). Temporal variability of air concentrations differs from that of emissions in Finland. It can be conditioned by the natural annual variability of meteorological conditions.

On the opposite, emissions in France drop 2.2 times, that is much more than in Europe as a whole (Fig. 3.17a). This leads to the reduction of air concentrations in the country (Fig. 3.17b). The reduction of air concentrations in France is 2 times that is slightly less than the reduction of emissions. The difference in reduction rates of air concentrations and emissions in the country is conditioned by the influence of transboundary transport though it is clear that for France the influence of domestic sources is prevailing.

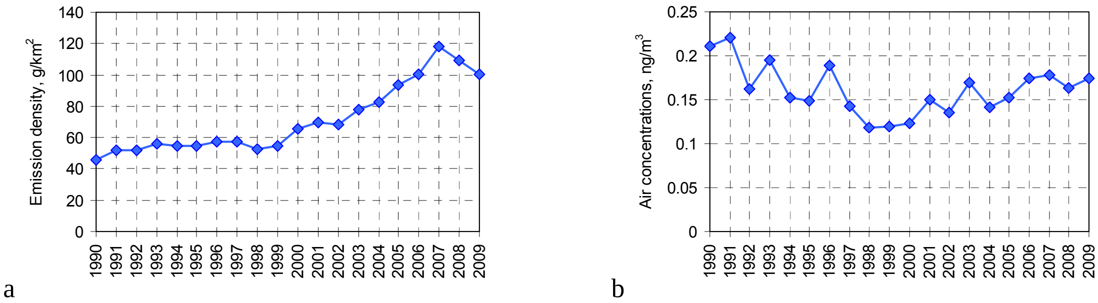


a

b

Fig. 3.17. *Trends of B[a]P emission density, g/km^2/y (a) and air concentrations, ng/m^3 (b) in France*

Finally, the trend of emission density in Denmark is characterized by enlarging emissions during the considered period (Fig. 3.18a) except for the two last years (2008 and 2009).



a

b

Fig. 3.18. *Trends of B[a]P emission density, g/km^2/y (a) and air concentrations, ng/m^3 (b) in Denmark*

The increase of emissions in Denmark amounts to about 2 times. However, air concentrations are slightly reduced during this period (by about 10%), see Fig. 3.18b. The reduction of air concentrations is conditioned by the decline of emissions in the neighbouring countries and by the fact that about 60% of national emissions are exported outside the country (see Fig. 3.19).

This indicates that for the considered country transboundary transport within the EMEP region plays essential role for B[a]P contamination.

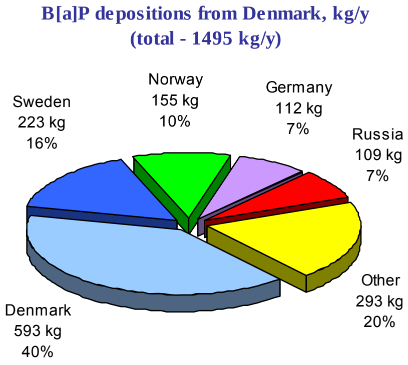


Fig. 3.19. *Export of B[a]P deposition from Denmark, kg/y*

3.2. Polychlorinated Dibenzo(p)dioxins and Dibenzofurans (PCDD/Fs)[2]

This year calculations of the overall toxicity of 17 toxic PCDD/F congeners with properties of the "indicator" congener 2,3,4,7,8-PeCDF were performed for 2009. Emission data for modelling were generated on the basis of official emission data complemented by non-Party emission estimates. Preliminary calculations from 1970 to 2009 were made to generate initial and boundary conditions for the EMEP domain on the basis of the hemispheric transport model. Measurement data for 2009 were available at EMEP sites Råö (SE14) and Aspvreten (SE12) (four months at each site). Additionally, model calculations of environmental levels of four PCDD/F congeners for 2006 and 2007 were performed in co-operation between Umeå University of Sweden and Meteorological Synthesizing Centre East of EMEP (MSC-E). The comparison of the calculation results with measurements at Aspvreten, Pallas and Vindeln allowed evaluating possible uncertainties in congener composition of emission data for PCDD/Fs.

Emissions

Officially submitted emissions. Data on total emissions of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) (sum of toxicities of 17 toxic PCDD/F congeners) were officially reported by 39 European countries and Canada for the period from 1990 to 2009 (for at least one year). In comparison with the previous reporting year additional two countries – Albania and Montenegro – reported their emission data.

Compared to the emission values of PCDD/Fs used in modelling for 2008, emissions in Italy, the UK, Romania, Spain, and Slovakia declined by 85, 43, 32, 28 and 23 g I-TEQ. Significant increase of the emission value was noted for Bulgaria (4 times) in comparison with previously reported data.

The information about spatial distribution of dioxin emissions at least for one year of the period 1990-2009 was provided by 26 countries (Austria, Belarus, Belgium, Bulgaria, Croatia, Cyprus, the Czech Republic, Denmark, Estonia, Finland, France, Germany, Hungary, Iceland, Ireland, Italy, Latvia, Lithuania, Netherlands, Norway, Poland, Slovakia, Spain, Sweden, Switzerland and the UK). Slovakia and Switzerland resubmitted information on spatial distribution for 1990, 1995, 2000 and 2005. Finland and Spain submitted gridded data for 2009.

Official information on PCDD/F emissions by sectors in 2009 was available for 34 countries. The maximum contribution to the total PCDD/F emissions was made by sector *1A4bi Residential - Stationary plants* (Table 3.4).

Officially reported information on uncertainties of PCDD/F emissions for 2009 was available for Denmark, Finland, France, Sweden and the UK. The uncertainty of Danish, French and Swedish dioxin emissions for 2009 was 585% [*Nielsen et al.*, 2011], 67% [*CITEPA*, 2011] and 116% [*SEPA*, 2011], respectively. For the Finnish dioxin emissions the uncertainty was in the range from -42% to 54% [*SYKE*, 2011], whereas the uncertainty of the UK dioxin emissions was estimated to vary from -50% to 200% [*Passant et al.*, 2011].

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[2] This section is written in co-authorship with K.Wiberg and I.Cousins.

Table 3.4. *Key source categories for PCDD/F emissions in 2009*

NFR Code	NFR Category	Contributions to emission total, %	Cumulative Total, %
1 A 4 b i	Residential: Stationary plants	30.7%	30.7%
2 C 1	Iron and steel production	10.7%	41.4%
1 A 3 c	Railways	6.9%	48.3%
6 D	Other waste	6.0%	54.3%
1 A 2 a	Stationary combustion in manufacturing industries and construction: Iron and steel	6.0%	60.3%
6 C e	Public electricity and heat production	5.6%	65.9%
1 A 2 f i	Stationary combustion in manufacturing industries and construction: Other	5.5%	71.4%
1 A 1 a	Public electricity and heat production	4.6%	76.0%
6 C b	Industrial waste incineration	4.3%	80.3%
1 A 2 b	Stationary Combustion in manufacturing industries and construction: Non-ferrous metals	2.7%	83.0%

Emission data used for modelling. The preparation of annual PCDD/F emission values for 2009 used for modelling was carried out on the basis of official information submitted by CEIP. In absence of officially reported information unofficial data of emission inventories [*Denier van der Gon et al*., 2005; *Pulles et al.,* 2006] were applied. The gridded emissions for 2009 were prepared by CEIP for EMEP countries with spatial resolution 50×50 km^2.

The official information on emissions for the Asian part of the EMEP domain and the USA was not available. The emission data for these regions were prepared by MSC-E. The PCDD/F emission for the Asian part of Russia was estimated using officially reported rates for the European part of the country and data on population density similar to PAHs. The PCDD/F emissions of Tajikistan, Turkmenistan and Uzbekistan for 2009 were taken from the non-Party inventory of PCDD/F emissions in the Central Asian countries made in the framework of the global International POPs Elimination Project (IPEP) [*Hodjamberdiev,* 2006]. The latest available information on PCDD/F emission in the USA was taken from the dioxin and furan inventories prepared by [*UNEP*, 1999] for 1995. The spatial distribution of PCDD/F emissions in the Central Asian countries and the Asian part of Russia was constructed on the basis of data on population density [*Li*, 1996].

The spatial distribution of PCDD/F emissions in the EMEP domain for 2009 is shown in Fig. 3.20. Significant levels of PCDD/F emissions (0.5 - 5 ng I-TEQ/m^2/y) can be seen in countries of Central, Southern, and Eastern Europe. Other parts of Europe, in particular, Northern and Western Europe, are characterised by lower emission fluxes varying from 0.01 to 0.5 ng I-TEQ/m^2/y.

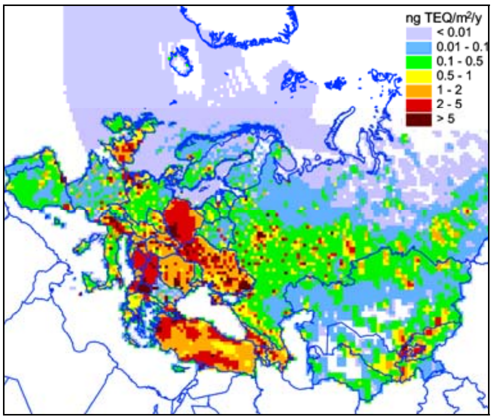

Fig. 3.20. *Spatial distribution of PCDD/F emissions in 2009 over the EMEP domain with resolution 50×50 km^2, ng I-TEQ/m^2/y*

The total emissions of PCDD/Fs within the Northern Hemisphere in 2009 amounted 9.4 kg I-TEQ, including 6.6 kg I-TEQ from emission sources located within the EMEP domain and 2.8 kg I-TEQ - from North America. Considering the PCDD/F annual emissions of individual countries for 2009 it can be noted that maximum contributions to the total dioxin emissions within the EMEP countries have been made by Turkey (11%), the Ukraine (11%) and the Russian Federation (9%).

Emission trends. According to the official and unofficial emission data, total emissions of PCDD/Fs within the EMEP domain decreased by 58% in the period from 1990 to 2009. PCDD/F emissions within the Northern Hemisphere (EMEP region, the USA and Canada) decreased by 50% during the same period (Fig. 3.21).

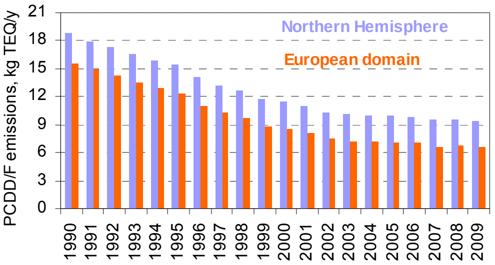

Fig. 3.21. *Temporal trends of PCDD/F emissions in the EMEP domain and the Northern Hemisphere in 1990-2009, kg TEQ/y*

Among the countries submitted official data on PCDD/F emissions for 2009, maximum emission reduction within the considered period took place in the Netherlands (96%), France (95%), Germany (92%), Belgium (91%), Switzerland (91%), Romania (91%) and the Czech Republic (89%). At the same time in Republic of Moldova, Belarus, Latvia and Liechtenstein dioxin emissions were increased in comparison with the level of emission in 1990.

Congener-specific emissions. For the evaluation of the environmental contamination by particular PCDD/F congeners emission data for 2006 and 2007 were generated for four PCDD/F congeners (2,3,4,7,8-PeCDF, 1,2,3,7,8,9-HxCDD, OCDD and OCDF). These data were calculated using total emissions of PCDD/F mixture (in I-TEQ) for modelling evaluated on the basis of official emission data submitted by EMEP countries to the UN ECE Secretariat and emission expert estimates made by TNO. In addition, emission data for the north-western part of the Russian Federation were updated using official information on B[a]P emissions in this region and regression relations between B[a]P and PCDD/F emissions. Shipping emissions (including spatial distribution) were evaluated on the basis of available emissions of NO_x [*Bartnicki et al.,* 2009] and using the data from EMEP/EEA emission inventory guidebook, 2009. Emissions of selected congeners were evaluated on the basis of total toxicity of the mixture of 17 toxic PCDD/F congeners and congener profile of PCDD/F emissions in each European country according to the estimates of POPCYCLING-Baltic project [*Pacyna et al.,* 2003].

Evaluation of contamination by particular congeners.

Prior to carrying out the assessment of contamination by PCDD/Fs for 2009, examination of possible uncertainties of emission data for PCDD/F mixture including its congener composition was performed by model calculations of the transport and environmental levels of four PCDD/F congeners (2,3,4,7,8-PeCDF, 1,2,3,7,8,9-HxCDD, OCDD and OCDF) for 2006 and 2007 in co-operation with Umeå University of Sweden and Meteorological Synthesizing Centre East of EMEP (MSC-E). To take into account the contributions of long-term accumulation in the environmental media and of PCDD/F sources located outside the EMEP region, initial and boundary conditions for calculations of environmental levels in the EMEP region were generated by calculations with the help of hemispheric MSCE-POP model for the period from 1970 to 2007.

The results of calculations were compared with measurements made at Aspvreten (SE12), Pallas (FI96), and Vindeln (SE35) monitoring sites provided to MSC-E by Umeå University. Measurements at Aspvreten and Pallas are one-day measurements for selected days in the end of 2006 and first half of

2007 [*Sellström et al.*, 2009]. These data were accompanied with information on wind pattern, temperature and precipitation amount at measurement sites for the corresponding time periods. The presence of such information was rather helpful for the interpretation of the comparison of measured data with calculation results. At the site Vindeln measurements of monthly deposition flux for the period from December 2006 to November 2007 were performed.

Below the comparison of modelling results for "indicator congener [3] " 2,3,4,7,8-PeCDF with measurements at SE12 and FI96 is considered.

Air concentrations. The comparison results for air concentrations of this congener are shown in Fig. 3.22. Measurements are grouped by the direction of atmospheric transport which occurred in the corresponding period (below the following abbreviations are used: E – East, N – North, S – South, W – West so that E – East, NNE – North-North-East, etc.).

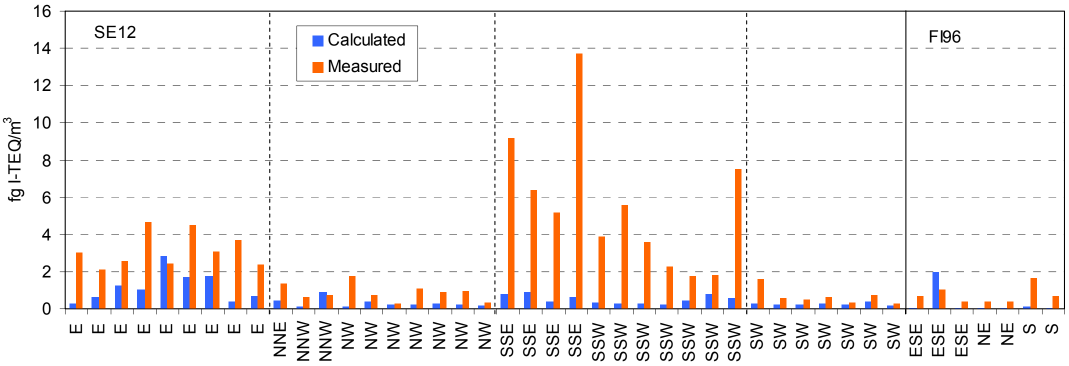

Fig. 3.22. *Comparison of modelling results with measurements (air concentrations) for 2,3,4,7,8-PeCDF at SE12 and FI96 in 2006 – 2007, fg I-TEQ/m^3. The values are grouped by the compass sectors (E – East, N – North, S – South, W – West) from which air transport occurs in the corresponding periods*

The comparison of modelling results and measurements of 2,3,4,7,8-PeCDF shows essential underestimation of observed air concentrations by the model by about a factor of five on the average. About 40% of calculated concentrations agree with measurements within a factor of three. The relations between calculations and measurements strongly depend on the compass sectors from which the contamination has arrived. The agreement for all sectors except for SSE and SSW seems to be reasonable. If measurements corresponding to these two sectors are excluded from the comparison, measurement-to-calculation ratio is lowered down to 2.7 on the average.

Deposition flux. The results of the comparison of monthly deposition fluxes of 2,3,4,7,8-PeCDF at the site Vindeln (SE35) are shown in Fig. 3.23a.

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[3] The contribution of this congener to the total toxicity of PCDD/F mixture amounted to 30% – 40% of total toxicity.

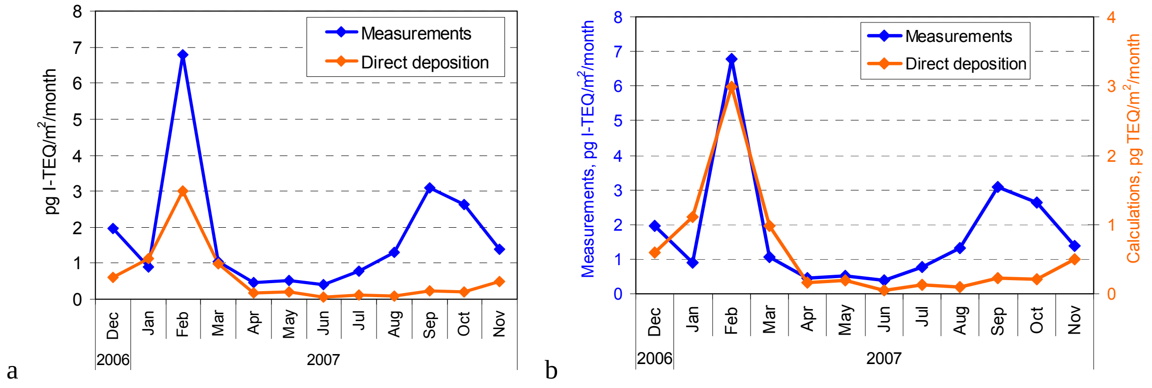


Fig. 3.23. *Comparison of calculation results with measurements (deposition flux) at Vindeln (SE35) in 2006 – 2007: a – one scale, b – different scales, pg I-TEQ/m^2*

For the convenience to compare seasonal variations of calculated flux with measurements the comparison with different scales is shown in Fig. 3.23b. The comparison shows that the model is capable to capture the observed seasonal variations of deposition fluxes. However, general underestimation is 2.9 times on the average. This corresponds to the results obtained from the analysis of the agreement between air concentrations at the sites Aspvreten and Pallas with SSE and SSW sectors excluded. It can be seen that in the end of 2006 and beginning of 2007 the model well represents variations of PCDD/F deposition flux. The discrepancies in the second half of 2007 can be conditioned by model description of gas/particle partitioning (including the data on concentrations of the atmospheric aerosol).

For the rest three congeners, the comparison of measured and calculated values of air concentrations at sites Aspvreten (SE12) and Pallas (FI96) shows that model estimates at this site are lower than measurements approximately 10 times for 1,2,3,7,8,9-HxCDD, 20 – 30 times for OCDD and 15 – 20 times for OCDF. Again, strong underestimation takes place for SSE and SSW compass sectors.

For the interpretation of the obtained results, it should be kept in mind that rather coarse spatial resolution used in model simulations (50×50 km) could be an additional reason of the underestimation of observed concentrations. Besides, for reproducing short-time contamination episodes the emission data with corresponding temporal resolution are needed. Different underestimation for different congeners can be conditioned by uncertainties in congener profile used in modelling in addition to general underestimation of emissions.

Application of integrated approach: analysis of measurement/calculation discrepancies

For the analysis of the reasons of model underestimation of PCDD/F air concentrations matrix approach was applied. The contributions of several groups of emission sources to the contamination in the grid cells with the measurement sites were evaluated by the model. These emission groups are: several European countries that can affect the contamination in the Baltic region where the considered measurement sites are located (BY, DK, FI, FR, DE, LV, LT, NO, PL and SE), other European countries considered as one source group, two specific regions (Black Triangle (BT) and north-western part of the Russian Federation (RW)), shipping emissions, emissions of non-EMEP sources and re-emissions due to long-term accumulation in the environment. These groups of sources are expected to contribute noticeably to the contamination of the three considered measurement sites.

The reasons of model underestimation of the contamination levels (air concentrations and deposition fluxes) are both uncertainties of emission totals and uncertainties of evaluation of congener composition of emissions. To reveal the sensitivity of calculated values of air concentrations to emissions from the above listed source groups and to emission congener composition several emission scenarios are prepared and considered. To reflect the above two reasons of emission uncertainty, each emission scenario is determined by two types of coefficients:

Source-specific coefficients defined for each source, by which the emissions from this source are multiplied. Source-specific coefficients do not depend on the considered congener but are different for different sources. These coefficients enumerate possible uncertainties in emission totals for different source groups.

Congener-specific coefficients defined for each congener, by which emissions of the given congener from every source are multiplied. The congener-specific coefficients are chosen the same for all emission sources but differs from one congener to another. This coefficient enumerates possible uncertainty related to emission congener composition.

So, emission total for the given congener and each source group is obtained from the emission total from the initial emission inventory by multiplication by two coefficients: congener-specific one and source-specific one.

Below, two emission scenarios are considered. Congener-specific coefficients and source-specific coefficients for the considered sources for the two considered scenarios are shown in Table 3.5.

Table 3.5. *Congener-specific and source-specific scenario coefficients for minimum and maximum emission scenarios*

Scenario	Congener	Congener-specific coefficient	Source-specific coefficients *)				
			PL	DE	FR	BT	RW
Minimum	2,3,4,7,8-PeCDF	1.5	2	1	1	1	2
	1,2,3,7,8,9-HxCDF	3					
	OCDD	6					
	OCDF	5					
Maximum	2,3,4,7,8-PeCDF	1.5	10	10	10	10	2
	1,2,3,7,8,9-HxCDF	3					
	OCDD	5					
	OCDF	5					

*) For all other sources source-specific coefficients equal 1

Air concentrations. The comparison of measurements with calculations made under the minimum scenario for 2,3,4,7,8-PeCDF is shown in Fig. 3.24.

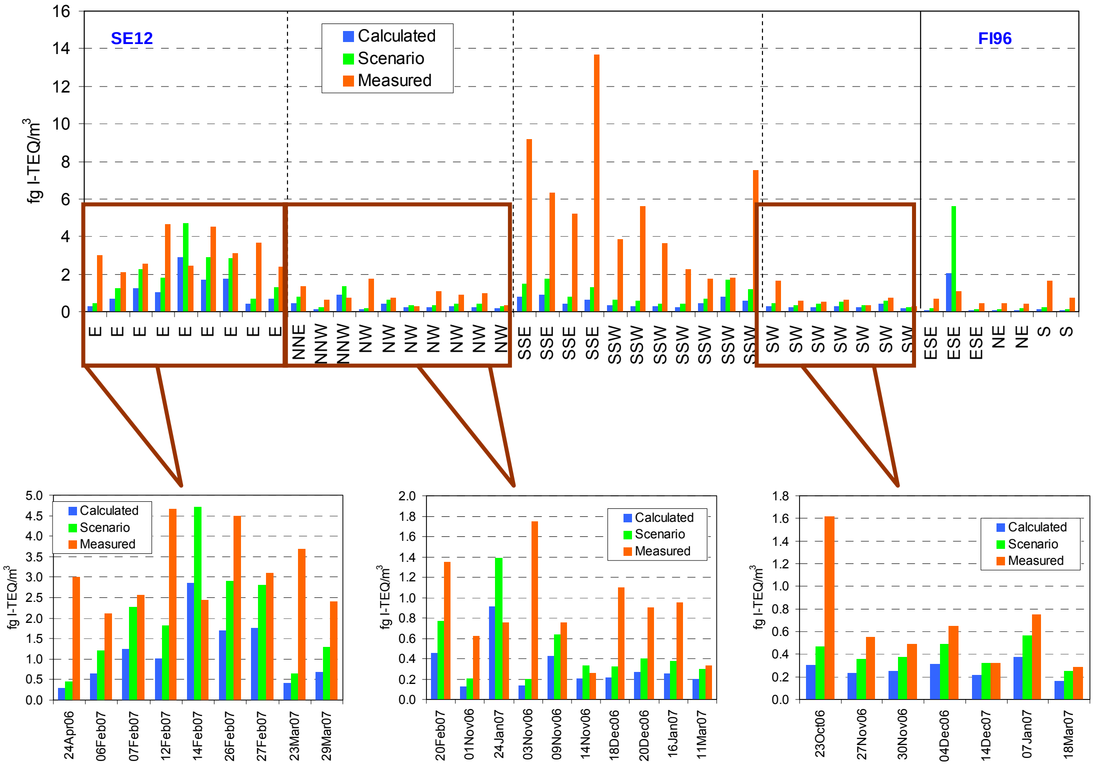


Fig. 3.24. *Comparison of observed air concentrations of 2,3,4,7,8-PeCDF in 2006 – 2007 with those calculated under minimum scenario, fg I-TEQ/m*^{3}

It is seen that even such moderate emission scenario can improve the agreement between measurements and calculation results for almost all measurement except for those corresponding to the dates when air masses come from South-South-East (SSE) and South-South-West (SSW) compass sectors. For this scenario about 52% of measurement data agree with calculations within a factor of three and among them about 41% of measurements agree with calculations within a factor of two.

However, this scenario cannot improve the agreement for measurements when air masses come from SSE and SSW compass sectors. If these measurements are excluded from the comparison, the share of measured values that agree with calculations within a factor of three is grown up to about 77%.

Similar situation with calculations made under the minimum scenario takes place for 1,2,3,7,8,9-HxCDD and OCDF as well. For 1,2,3,7,8,9-HxCDD about 64% of measurements are with within a factor of three with respect to calculations (including 46% within a factor of two) and for OCDF about 61% of measurements are within a factor of three with respect to calculations (including 41% within a factor of two). Maximum disagreement for these pollutants takes place again for compass sectors SSE and SSW.

However for OCDD the situation is somewhat different (see Fig. 3.25).

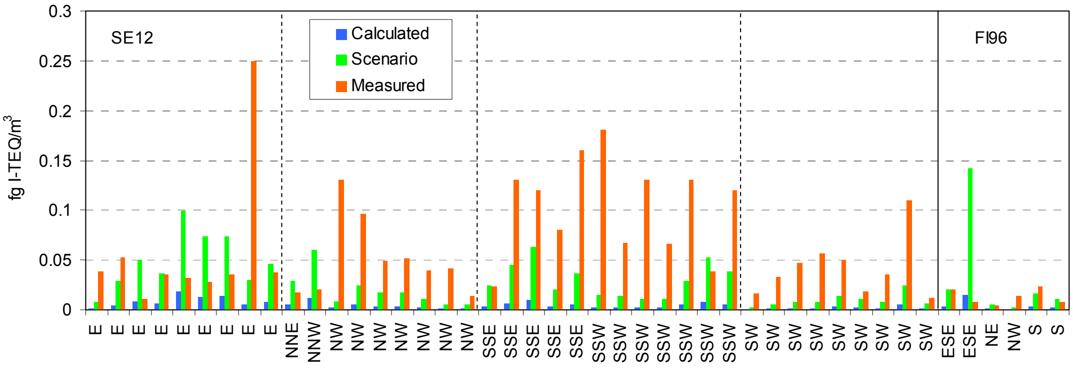


Fig. 3.25. *Comparison of observed air concentrations of OCDD with those calculated under minimum scenario, fg I-TEQ/m^3*

For this pollutant only 42% of measurements agree with calculations within a factor of three. Here minimum scenario leads to underestimation of measured values for South-West (SW) and North-West (NW) compass sectors and to overestimation of measured concentrations for East (E) compass sector. This leads to the assumption that sources of OCDD are different from sources of other three considered congeners. One of possible reasons of the difference is additional source of OCDD emissions from atmospheric transformations of pentachlorophenol. More detailed discussion on this subject can be found in [*Shatalov et al.*, 2011].

The comparison of measurements with calculations made under the maximum scenario for 2,3,4,7,8-PeCDF is shown in Fig. 3.26.

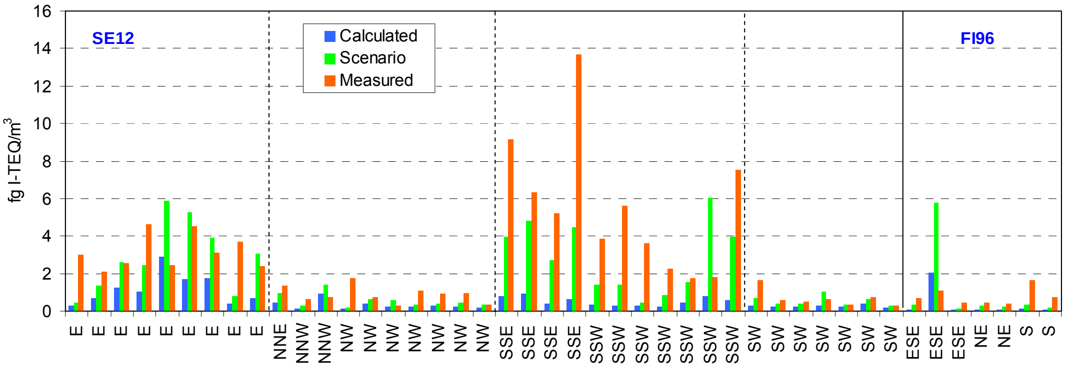


Fig. 3.26. *Comparison of observed air concentrations of 2,3,4,7,8-PeCDF with those calculated under maximum scenario, fg I-TEQ/m^3*

It is seen that the application of maximum scenario leads to essential improvement of the agreement between measurements and calculated values of air concentrations. The agreement within a factor of two for this scenario becomes 50% for 2,3,4,7,8-PeCDF, 67% for 1,2,3,7,8,9-HxCDD and 68% for OCDF. However, even in this case the disagreement between measurements and calculations can reach as much as 7.5 times. Besides, for some cases when high discrepancies between model predictions and measurements take place measured values of total toxicity are high enough (exceed

10 fg I-TEQ/m^3). For the comparison we note that the values of total PCDD/F toxicity at the Canadian background site Eagle Harbor ranges from 0.06 to 4.9 fg I-TEQ/m^3. This gives rise to the assumption that in addition to underestimation of the European emissions, possible influence can also be expected from the emission sources located close enough to the measurement site (local sources).

Deposition flux. The comparison of measurements of deposition flux of 2,3,4,7,8-PeCDF at Vindeln (SE35) with calculations using minimum and maximum emission scenarios is displayed in Fig. 3.27.

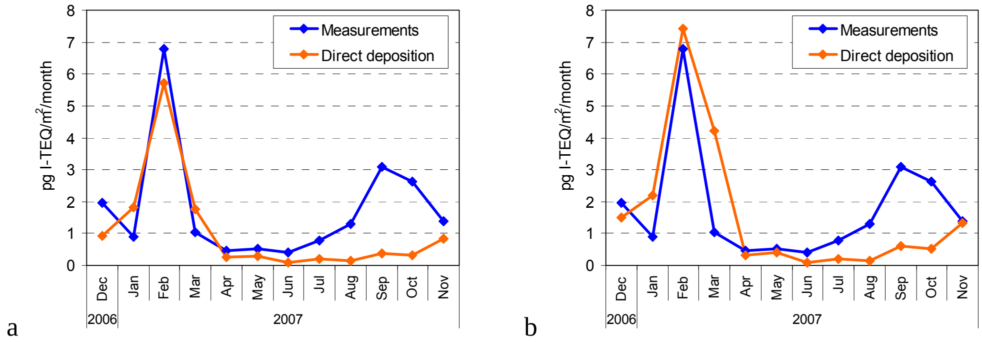


Fig. 3.27. *Comparison of observed deposition flux of 2,3,4,7,8-PeCDF with those calculated under minimum (a) and maximum (b) scenarios, pg I-TEQ/m^2/month*

The results obtained manifest that the calculations based on the considered scenarios agree with measurements at SE35 (again, with the exception for the end of the year).

The comparison of measured and calculated deposition fluxes for the rest three congeners can be found in [*Shatalov et al.*, 2011].

The above analysis allows concluding that:

- Emissions of PCDD/Fs may be totally underestimated up to 5 times. Underestimation of emissions is different for different congeners.
- For better evaluation of the agreement between measurements and modelling results measurement sites with more homogeneous distribution over the EMEP domain are desirable.
- After refinement of emission data direct calculations with refine emissions can give reasonable estimates of PCDD/F environmental contamination.

Evaluation of contamination of the EMEP region by PCDD/Fs (overall toxicity)

Here the results of calculations of overall PCDD/F toxicity in the EMEP domain in 2009 are described. Spatial distribution of annual means of air concentrations of PCDD/F mixture as predicted by the model calculations is displayed in Fig. 3.28.

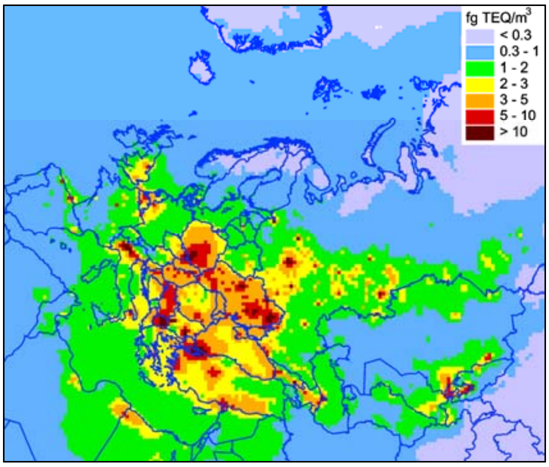


Fig. 3.28. *Spatial distribution of air concentrations of PCDD/F mixture in 2009 as predicted by MSCE-POP model, fg I-TEQ/m*^{3}

As follows from calculations, highest concentrations levels of PCDD/Fs (3 – 10 fg I-TEQ/m^3 and higher) take place in Central, Southern and Eastern Europe. More clean regions are located in Western Europe (the western part of France, Germany and the UK) except for the eastern part of the UK, Belgium and the Netherlands. Here typical contamination levels are in the range from 1 to 3 fg I-TEQ/m^3. The areas on the Scandinavian Peninsula, Portugal and Spain are considered to be clean regions with air concentrations less than 1 fg I-TEQ/m^3.

Modelled air concentrations of PCDD/F mixture were compared with measurements made at Råö (SE14) and Aspvreten (SE12) in 2009 (four months at each site). These data were kindly put at our disposal by Eva Brorström Lundén. The results of the comparison are displayed in the plot in Fig. 3.29 (a):

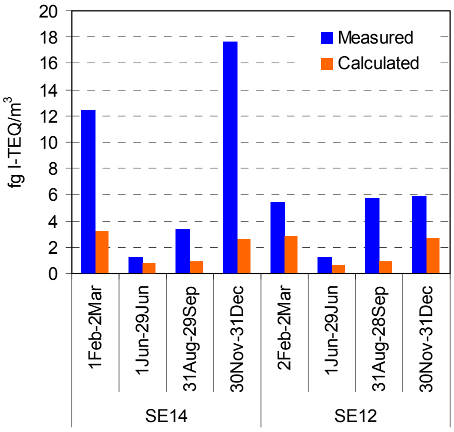


Fig. 3.29. *Comparison of calculation results with measurements (air concentrations) at two Swedish sites, fg I-TEQ/m*^{3}

According to the comparison, measurement-to-calculation factor ranges from 1.5 to 9 depending on the considered month. On the average, the model underestimates air concentrations of PCDD/Fs 4.7 times. As mentioned above, uncertainties of calculated concentrations due to the model description and monitoring data seem to be in the range of factor 2 – 3. Hence, the possibility of refining the agreement by changing emission data (emission scenarios) should be examined. This is made in the next subsection.

Analysis of measurement/calculation discrepancies. Annual averages of PCDD/F air concentrations obtained by measurements at SE12 and SE14 for four months in 2009 are presented above by the plot in Fig. 3.29 together with the results of model calculations. As it was mentioned above, the model underestimates air concentrations 4.7 times on the average.

First, calculations with total emissions in all European countries enlarged 4.7 times (Total enlargement scenario) were carried out. Such scenario does not solve the problem of emission underestimation but is used as a first rough approximation. The results of scenario calculations in comparison with measurements are presented in Fig. 3.30.

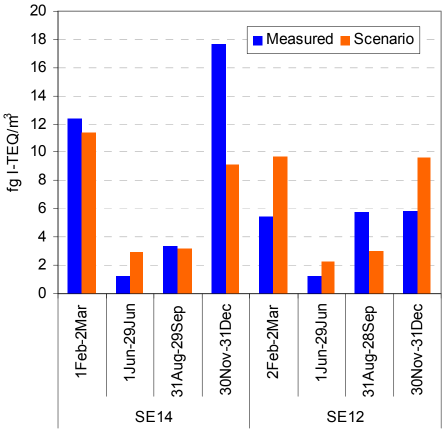


Fig. 3.30. *Comparison of calculation results obtained by Total enlargement scenario with measurements (air concentrations) at two Swedish sites, fg I-TEQ/m^3*

The application of such scenario allows refining the comparison from the viewpoint of unbiasedness. Namely, the value of the Student ratio for the comparison of measurements with initial calculations equals 2.8 which is higher that the threshold level, which in this case equals to 2.36. For Total enlargement scenario this value becomes –0.11 which shows that the model reproduces air concentrations without significant bias. The value of normalized mean bias NMB changes from 0.78 for initial calculations to 0.1 for Total enlargement scenario, and regression slope changes from 5.24 to 1.24. So, calculations under this scenario meet the criteria of unbiasedness. However, this scenario does not refine the value of correlation coefficient (its value is 0.65 as for initial calculations) and coefficient of determination R^2 for this scenario is rather low (0.49). Hence, it is reasonable to try to construct emission scenario with changes of emissions different for different countries.

Similar to the B[a]P case, the evaluation of emission scenarios is performed on the basis of country-to-site matrix for monthly averages of air concentrations which can be calculated by the model for all measurement sites and time periods for which the information on the considered pollutant is available. This information evaluates the sensitivity of calculated air concentrations at the location of measurement sites with respect to emissions of all considered sources. The contributions of various sources to calculated PCDD/F air concentration values at site locations is exemplified by two EMEP sites SE12 and SE14 in February 2009 (Fig. 3.31).

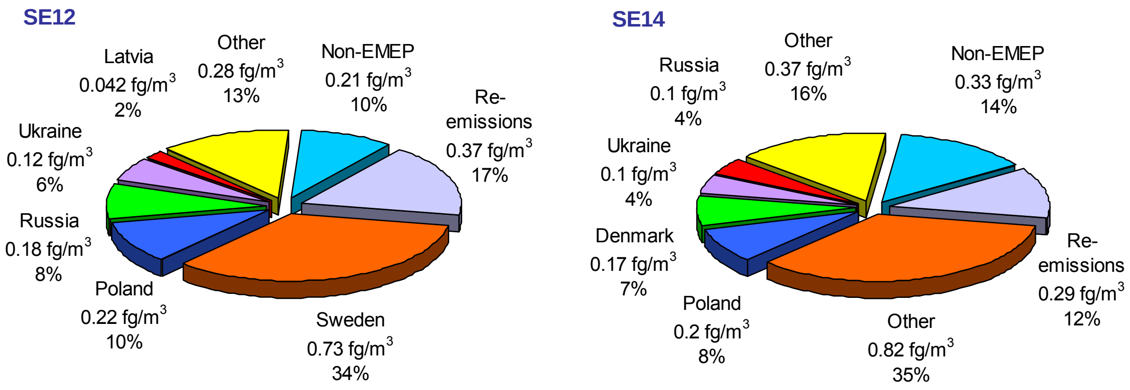


Fig. 3.31. *Contributions of various emission sources to the calculated values of air concentrations at SE12 and SE14 in February, 2009, fg I-TEQ/m*^{3}

Using this information it is possible to evaluate changes in air concentrations due to the change of emissions of this or that source and to construct emission scenarios further refining the agreement between modelling results and measurements at the given site or at a number of sites for the considered months. Besides, it is clear that for the refinement of the agreement at sites SE12 and SE14 it is reasonable to change emission totals only for countries with essential contributions to air concentrations at these sites. For further refinement of emission field in the EMEP region it is desirable to obtain measurements at sites more homogeneously distributed over the region.

The scenario that leads to noticeable refinement of the agreement between measurements at selected sites and modelling results was constructed on the basis of the indicators described in Chapter 1. This scenario includes total increase of PCDD/F emissions by 50% and additionally enlarges emissions of five European countries: Germany – 2 times, Denmark – 8 times, the UK – 8 times, Poland – 4 times and Sweden – 5 times. The comparison of model predictions under such scenario with measurements at SE12 and SE14 is displayed in Fig. 3.32.

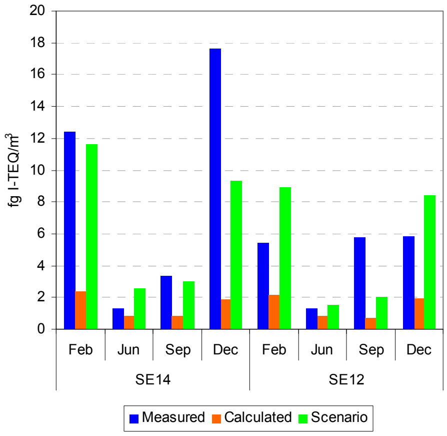


Fig. 3.32. *Comparison of air concentrations calculated under emission scenario with the results of base calculations and with measurements at SE12 and SE14, fg I-TEQ/m*^{3}

Such scenario allows further refining the agreement between calculations and measurements. For this scenario the value of Student ratio becomes 0.51, correlation coefficient is equal to 0.74 and regression coefficient became 1.04. Further, for almost all considered months calculations agree with measurements within a factor of two except for September at SE12 where measurement-to-calculation factor exceeds 2 (it is equal to 2.9). The latter explains relatively low value of coefficient of determination (0.59). It was found that changing emission totals in European countries it is not possible to refine the agreement in September without essential worsening the agreement within other months. Hence, the discrepancies in the mentioned months are probably conditioned by the uncertainties in spatial distributions of emissions inside countries. To analyze this situation, trajectory approximation of the influence function for monthly averages of air concentrations for four considered months at SE12 were constructed (Fig. 3.33).

SE12

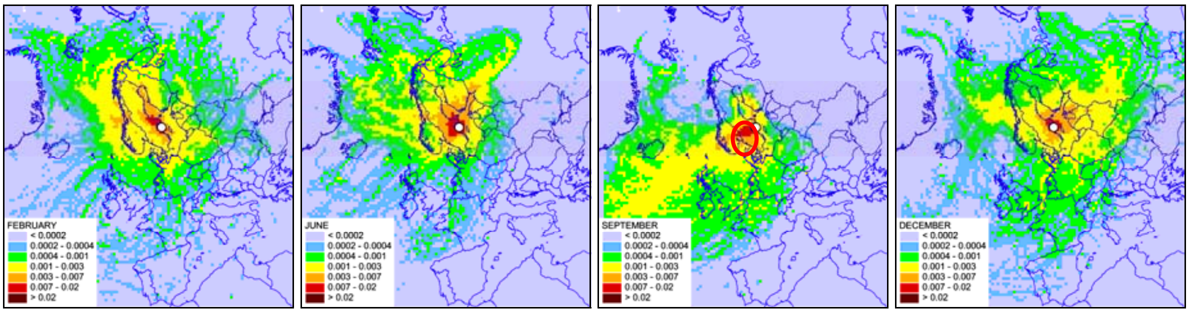


Fig. 3.33. *Influence functions for monthly averages of PCDD/F air concentrations for February, June, September and December at SE12, min/m*

It can be seen that in September at SE12 the contamination is partly determined by sources located in the part of Sweden marked by red ovals whereas this area does not affect contamination at the considered site in other months. It is reasonable to perform more thorough examination of this area from the viewpoint of spatial distribution of emissions.

The results of the above analysis do not mean in essence that the emissions in five above listed countries are underestimated in total. As was already noted, the reason of underestimation of air concentrations at these sites by the model can be conditioned also by uncertainties in spatial distribution of emissions. To refine the evaluation of contamination in this region modelling with finer spatial resolution can be of use. However, for such kind of modelling the data on spatial distribution of emissions in the considered countries (and possibly in their neighbours) should be refined with participation of emission experts.

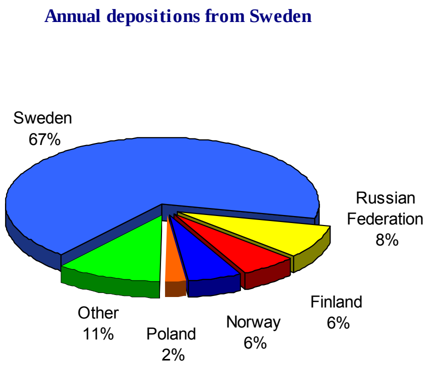


Fig. 3.34. *Export of PCDD/F deposition for Sweden (2009)*

Evaluation of the transboundary transport. Modelling of long-range transport and deposition of PCDD/Fs within the EMEP domain allows evaluation of source-receptor relationships including contributions of national emission sources, transboundary transport within EMEP, non-EMEP sources and re-emission to air concentrations and deposition fluxes in various countries or/and at different locations. It should be taken into account that the information on the import of PCDD/Fs to the countries is essentially emission-dependent whereas the information on export is independent of emission data at least from the viewpoint of relative values. So, keeping in mind essential uncertainties in emissions of PCDD/Fs it is reasonable to present here the information on the export only. The example of export chart for Sweden is presented in Fig. 3.34.

The fractions of PCDD/F mixture deposited outside a country in total deposition flux originated from national sources are shown in Fig. 3.35.

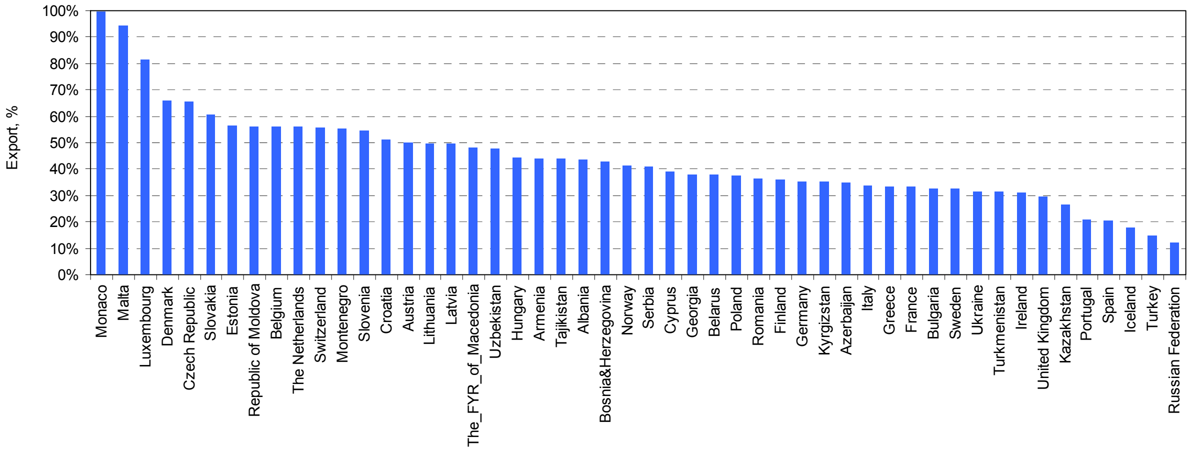


Fig. 3.35. *Fractions of PCDD/F deposition originated from countries emission sources and occurred outside their territories in 2009 (export), %*

Calculations show that the export from European countries is typically in the range from 30% to 60%. It should be stressed that the export fractions depend on the meteorological conditions of the considered year.

3.3. Hexachlorobenzene (HCB)

This section of the report describes the progress in the evaluation of HCB pollution levels within the EMEP domain. In comparison with PAHs and PCDD/Fs considered in the previous sections evaluation of HCB pollution levels is complicated by more essential uncertainties in information on the current sources of HCB release into environment and historical emissions. The work on the evaluation of HCB pollution within the European region and the Northern Hemisphere was started several years ago and is described in the MSC-E reports [*Gusev et al.*, 2009; *Shatalov et al.*, 2010].

In particular, evaluation of HCB pollution within the EMEP domain [*Gusev et al.*, 2009] revealed essential underestimation of observed HCB air concentrations by the model. This study was based on official HCB emission data complemented by the expert estimates of TNO. To consider the influence of distant emission sources a scenario of HCB emission within the Northern Hemisphere was constructed. This scenario reflected the level of emissions in the mid-1990s and thus did not allow taking into account previous agricultural application of HCB in substantially larger amounts which led to the underestimation of the influence of historical emissions. Comparison of modelling results with available measurements showed underestimation of observed concentrations by a factor of 2-4 and higher in some regions indicating that the level of HCB emission in Europe was likely more significant than that officially reported by the EMEP countries. Additionally it was shown that application of higher HCB emission (TNO emission inventory) in modelling essentially improved the agreement between the measurements and model estimates. Thus, it was concluded that the underestimation of observed HCB concentrations could be related to the incompleteness of available officially submitted emission data and expert estimates as well as with the underestimation of the role of secondary emission sources.

Importance of secondary HCB emissions was considered in further study presented in [*Shatalov et al.*, 2010]. Following available information the application of HCB in various activities was started from 1945 and reached its maximum in 1980-s. The major source of its release into the environment in that period was the use in agriculture as a fungicide which likely led to the accumulation of HCB in soils and subsequent re-emission. To examine these three simple emission scenarios of historical HCB emissions (low, average and high) for the period 1945-2008 were constructed and model simulations of HCB transport and accumulation in the environment at hemispheric scale were carried out. Model evaluation of HCB fate indicated that the major part of HCB mass at the end of calculation period was stored in soil (more than 90%). Other media contained only small share of the total environmental burden. These estimates of HCB distribution in the environment were close to other modelling studies [*Zhang et al.,* 2003; *MacLeod and Mackay,* 1999; *Barber et al.* 2005] which showed that major part of HCB in the environment was likely accumulated in soil. Starting from 1980-s the agricultural use of HCB was banned in many countries world-wide resulting in considerable decrease of primary emission of HCB and the increase of relative importance of HCB re-volatilization. Thus, re-emission of HCB from the environmental compartments (e.g. soils, seawater) can exceed nowadays primary anthropogenic HCB emission.

At current stage of the work the evaluation and analysis of HCB pollution within the EMEP region was continued using the developing integrated approach (see Chapter 1). Particularly, modelling of HCB pollution levels was performed using available official emission data and expert estimates. The level of agreement between the modelled air concentrations and measurements was examined. To explain the discrepancies found between the modelling results and measurements several conventional scenarios of HCB emissions were constructed. Additionally the sensitivity of the pollution levels to emissions of particular countries was analyzed and preliminary recommendations for further refinement of the assessment of HCB pollution within the EMEP region were formulated.

Emissions of HCB

Officially submitted emissions. Official data on total national HCB emissions were submitted by 27 European countries as well as by Canada and the USA for the period from 1990 to 2009 (for at least one year). The number of countries reporting their data on HCB emissions is gradually increasing. In particular, this year Albania officially reported information on national HCB emissions. Among these countries 18 ones (Austria, Belgium, Croatia, Cyprus, Denmark, Estonia, France, Germany, Italy, Latvia, Montenegro, Portugal, Romania, Slovakia, Slovenia, Sweden, the UK and the USA) recalculated their official emissions for at least one year within the period from 1990 to 2008. For instance, HCB emissions of Italy for 2008 were increased from 0.03 to 31 kg/y, and HCB emission of the UK was decreased from 91 to 58 kg/y for the same year.

Among the countries officially submitted data for both years 1990 and 2009 the most significant emission decrease was reported by the UK (from 3170 to 33 kg) and France (from 1200 to 15 kg). At the same time HCB emissions of Estonia and Belarus were increased 3 and 2 times, respectively.

The information on spatial distribution of HCB emissions was submitted by 17 countries (Austria, Belarus, Belgium, Bulgaria, Cyprus, the Czech Republic, Denmark, Estonia, Finland, France, Germany, Hungary, Ireland, Latvia, Poland, Slovakia and Spain).

Official information on HCB emissions by sectors for 2009 is available for 27 countries. According to these data the most significant source category of HCB emissions is *2C1 Iron and steel production* sector (Table 3.6) followed by *1A4bi Residential - Stationary plants* sector.

Table 3.6. *Key source categories of HCB emission in 2009 and their contributions to total emission, %*

NFR Code	NFR Category	Contribution to Total Emission, %	Cumulative Total, %
2 C 1	Iron and steel production	59.4%	59.4%
1 A 4 b i	Residential: Stationary plants	9.4%	68.8%
4 G	Other Agricultural processes	4.7%	73.5%
1 A 1 a	Public electricity and heat production	4.4%	77.9%
6 C b	Industrial waste incineration	3.9%	81.7%
2 B 5 a	Other chemical industry	3.8%	85.6%
2 E	Production of POPs	1.8%	87.4%
1 A 3 b i	Road transport: Passenger cars	1.8%	89.1%
2 C 5 e	Other metal production	1.7%	90.8%

Analysis of sectoral data of individual countries shows that the contribution of key source categories to total HCB emissions can essentially vary among the European countries. Detailed information on relative contribution of key source categories mentioned above to national emission totals is exemplified by several countries in table 3.7. It can be seen that data of a number of countries do not consider the key source categories given above. Particularly, the information on contribution of *Iron and Steel production* sector to the emissions of a number of countries is not presented (e.g. Germany and the UK). Besides, the most part of HCB emissions of the countries is determined by a few sectors and information on some HCB sources is absent in national inventories. More significant contribution to HCB emissions originated from the production of chlorinated solvents and pesticides, wastes and sewage sludge incineration, metals smelting, sintering process, steel manufacturing, production of magnesium and cement as well as combustion of fossil fuel can be expected. Thus, it is possible to assume that officially reported HCB emissions can be underestimated.

Table 3.7. *Contributions of key source categories to total HCB emissions of European individual countries*

NFR Code	BG	HR	DK	EE	DE	IE	ES	GB
2 C 1	98.8%						95.3%	
1 A 4 b i	0.7%		24.7%	63.7%	62.8%			
4 G						98.9%		71.4%
1 A 1 a			66%	26.7%	25.6%		1.4%	26.9%
6 C b	0.2%	100%						
2 E							3.2%	
Total	99.7%	100%	90.7%	90.4%	88.4%	98.9%	99.9%	98.3

BG – Bulgaria; HR – Croatia; DK – Denmark; EE – Estonia; DE – Germany; IE – Ireland; ES – Spain; GB – United Kingdom.

Officially reported information on uncertainties of HCB emissions for 2009 is available for Denmark, Finland, France and the UK. The uncertainty of Danish and French HCB emissions is 718% [*Nielsen et al.*, 2011] and 60% [*CITEPA*, 2011]. For the Finnish HCB emissions the uncertainty is in the range from -73% to 134% [*SYKE*, 2011], whereas the uncertainty of the UK HCB emissions is in the range from -40% to 90% [*Passant et al.*, 2011].

Emission data used for modelling. Modelling of HCB pollution within the EMEP domain for 2009 was performed using official emission data received from the EMEP Centre on Emission Inventories and Projections (CEIP) [http://www.emep-emissions.at/ceip/]. Officially submitted data were complemented by unofficial data of TNO emission inventory [*Denier van der Gon et al*., 2005] for countries which did not provide official emission data. The gridded HCB emissions within the EMEP domain for 2009 with spatial resolution 50×50 km^2 were made by CEIP. Additional two datasets of HCB emissions for the EMEP domain describing the level of emissions in 2000 and 2009 were prepared on the basis of TNO emission inventory.

To compile the distribution of HCB emission outside the EMEP region national data for Canada and the USA were used. The HCB emission for Japan, China, Pakistan, the Republic of Korea, and India was taken from various sources [*Toda*, 2005; *Bailey*, 2001; *Shatalov et al.*, 2005]. However, as it was mentioned above, the information on HCB emissions compiled for Northern Hemisphere is subject of essential uncertainties.

Evaluation of contamination within the EMEP region

Modelling of HCB long-range transport and accumulation in the environment for 2009 was performed by the regional version of MSCE-POP model using official emission data complemented by the data of TNO emission inventory. The contribution of emission sources located outside the EMEP region and the influence of HCB accumulation in the environmental media were evaluated using the hemispheric scale model simulations for the period from 1990 to 2009. These modelling results were used to prepare initial and boundary concentrations for modelling of HCB within the EMEP region. Spatial distribution of HCB emission fluxes and annual mean air concentrations within the EMEP region for 2009 are shown in Fig. 3.36.

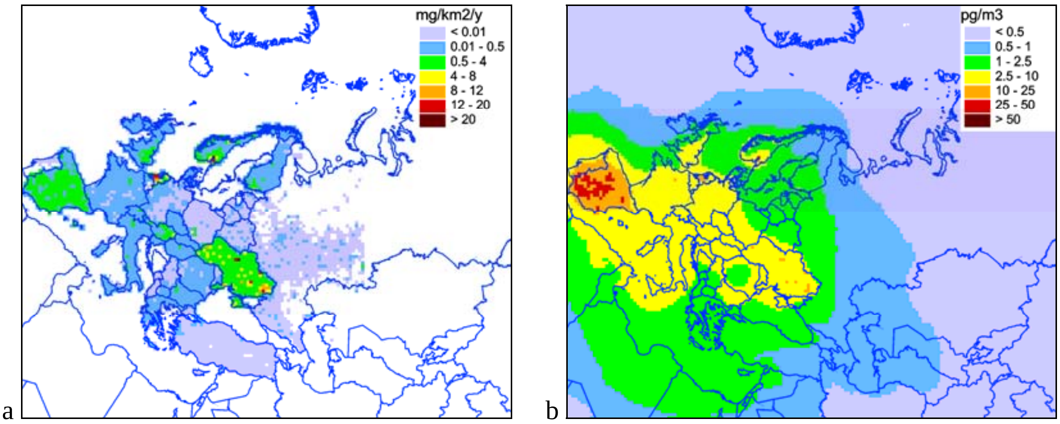


Fig. 3.36. *Spatial distribution of HCB emission fluxes, mg/*km^2*/y (a) and annual mean air concentrations of HCB,* pg/m^3 *(b) in 2009 obtained by MSCE-POP model on the basis of official emission data complemented by the TNO inventory*

It is seen that application of official emission data for 2009 in model simulations leads to essentially low values of HCB annual mean air concentrations. Particularly, air concentrations characteristic of Western, Southern and Central Europe are in the range from 2 to 20 pg/m^3, whereas in Northern Europe the concentrations are below 2 pg/m^3. This can be seen also from the comparison of modelled HCB air concentrations with measurements of EMEP monitoring sites shown in Fig. 3.37. The model essentially underestimates measured levels of air concentrations at all considered sites. A number of factors can contribute to the underestimation of HCB pollution levels. Comparing to the previous study of HCB pollution [*Gusev et al.*, 2009], where similar tendency to underestimate measured HCB concentrations was revealed, the differences between the modelled and observed values obtained for 2009 are even larger. These can be explained first of all by the decreasing trend in the HCB emissions reported by the EMEP countries. Particularly, according to officially submitted data complemented by the expert estimates of TNO total annual HCB emission within the European region for 2009 is accounted for 1.5 tonnes which is essentially lower than the estimates of TNO for 2009, namely, 9 tonnes, and is also lower than the total annual HCB emissions (27 tonnes) used in the model simulations described in [*Gusev et al.*, 2009].

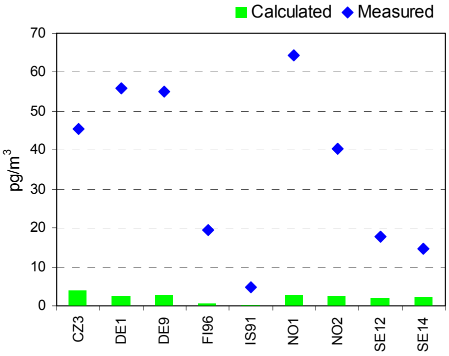


Fig. 3.37. *Comparison of calculated HCB air concentrations, pg /m^3 in 2009 calculated on the basis of official emissions with available measurements at EMEP sites*

Taking into account large differences between modelling results and measurements, model simulations with additional two emission datasets based on the HCB emission inventory of TNO and meteorological data for 2009 were carried out. Particularly, these emission datasets represent the levels of HCB emission within the European region for 2000 and 2009. Spatial distribution of annual emission fluxes and corresponding modelled annual mean air concentrations of HCB are shown in Figs. 3.38 and 3.39.

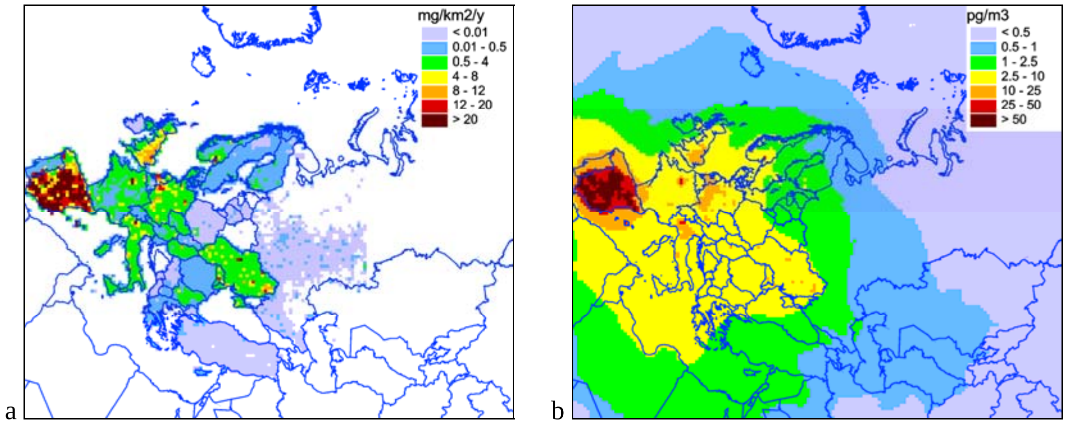


Fig. 3.38. *Spatial distribution of HCB emissions, mg/km^2/y (a) and air concentrations, pg /m^3 (b) in 2009 calculated by MSCE-POP model on the basis of the TNO emission inventory for 2009*

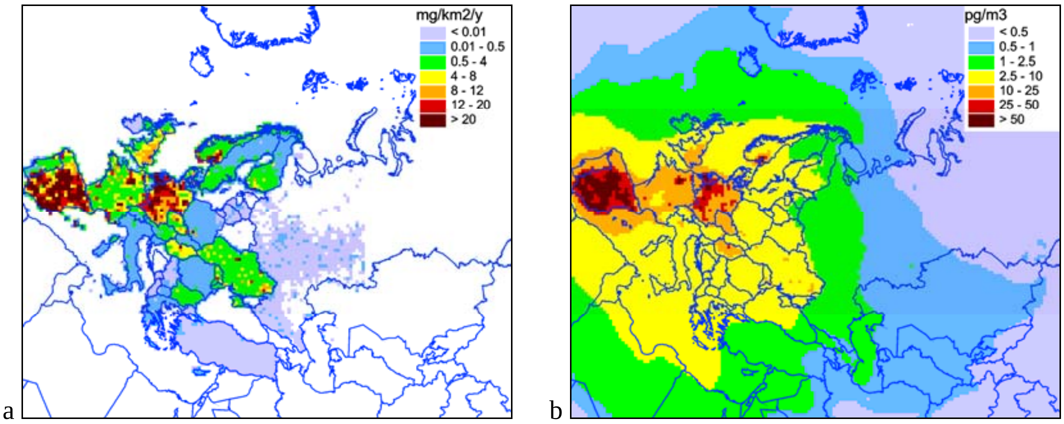


Fig. 3.39. *Spatial distribution of HCB emissions, mg/km^2/y (a) and air concentrations, pg /m^3 (b) in 2009 calculated by MSCE-POP model on the basis of the TNO emission inventory for 2000 (with meteorology of 2009)*

It is seen that contamination levels obtained using TNO emission inventory for 2009 (Fig. 3.38b) are, in general, higher than that simulated with the official emission data. However, significantly high HCB emission fluxes in Spain comparing to other European countries can be noted, which is the implication to the uncertainties of currently available information on HCB emissions. According to this inventory, about 60% of European emissions were contributed by Spain.

Model simulations based on HCB emissions of TNO for 2000 show much higher concentrations in Central Europe in comparison to modelling results obtained using emission data for 2009. Elevated values of air concentrations are found in Spain and Germany (30 – 40 pg/m^3 and higher). In the rest part of Europe HCB air concentrations range from 2 to 10 pg/m^3, and lower in the remote regions.

The comparison of calculated air concentrations obtained with all three emission inventories is presented in Fig. 3.40.

Confronting modelling results obtained with the use of three considered emission datasets, namely, official emissions and expert estimates for 2009 and 2000, it is seen that modelled air concentrations essentially lower than measured ones. Application of HCB emission data for 2000 based on TNO emission inventory leads to relatively higher concentrations improving the agreement with measurements. However, even in this case underestimation about a factor 2 - 7 (except for sites FI96 and NO42 where underestimation is higher) takes place.

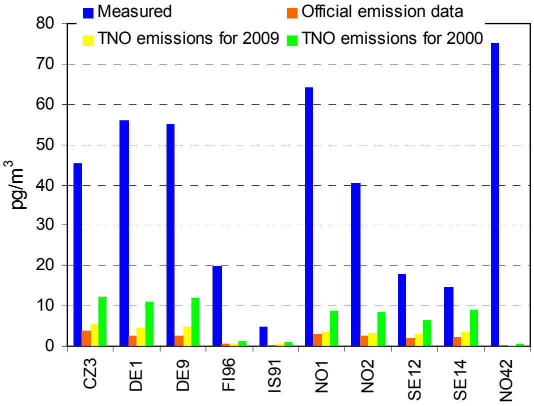


Fig. 3.40. *Comparison of calculated HCB air concentrations, pg /m^3 in 2009 calculated on the basis of three emission inventories with available measurements at EMEP sites*

Application of integrated approach: analysis of measurement/calculation discrepancies

Taking into account essential level of the differences between the modelling results and measurements for HCB there is a need to perform analysis of likely reasons of the underestimation and of the level of uncertainties of measurements, emission data, and modelling approach. Possible reasons of such underestimation of observed HCB air concentrations by the model are discussed below.

Uncertainties of measurement data. To evaluate the possibility of such uncertainty, measurements of HCB air concentrations at a number of locations within the Northern Hemisphere for several years in period 1980 – 2002 were used. These measurement data were collected by *J.Barber, A.Sweetman and K.Jones* in 2005 EuroChlor Science Dossier “Sources, Environmental Fate and Risk Characterization” (see Annexes to this Dossier) and summarized in [*Barber et al.,* 2005]. According to these data, obtained in various monitoring campaign, background levels of HCB air concentrations range typically from 25 to 85 pg/m^3. At the same time, in the contaminated regions, mostly outside the EMEP region, the levels of HCB in air can be essentially higher, reaching about 400 pg/m^3. This indicates the presence of significant emissions of HCB originating from primary or secondary sources. Thus uncertainties of measurement data are likely not the predominant reason of the discrepancies between the model predictions and measurements for HCB.

Uncertainties in model description and parameterization. The comparison of model parameterizations for HCB used in MSCE-POP model with those used in other models showed that the values of the parameters describing HCB behaviour were close to one another. For example, degradation half-lives of HCB in the atmosphere and soil used by MSCE-POP are 1.7 and 4.2 years, respectively, which corresponds to the values reported in [*Barber et al.,* 2005]. Further, uncertainties of model output (air concentrations and deposition fluxes) due to uncertainties in model parameterization is estimated as 50 – 70% (see EMEP Status Report [*Gusev et al.,* 2005]). It should be noted that MSCE-POP model was successfully applied to describe pollution levels of other POPs like, for instance, PAHs, PCBs, HCHs, and showed reasonable agreement with available measurements. Besides, close modelling approach, implemented in the MSCE-HM model, was applied to the evaluation of such long-lived chemical like mercury showing again good agreement with observed concentrations. Therefore, uncertainties of model parameterization can hardly be a reason of 2 – 7-fold underestimation of HCB air concentrations.

Uncertainties of historical HCB emissions. As mentioned above, according to previous investigations of MSC-E [*Shatalov et al.*, 2010] re-emission flux can be one of the most essential sources for contemporary HCB pollution levels. Similar conclusions were made in the study [*Barber et al.,* 2005] where it was mentioned that the amount of HCB emitted from soil to air at the peak of its usage may be accounted for hundreds to thousands tonnes per year, which is essentially higher compared with present levels of anthropogenic emissions in Europe (several tonnes). Thus this can make it a significant source of HCB to the environment. Since re-emission from soil is determined by historical accumulation of HCB, underestimation of re-emission flux can be a consequence of underestimation of historical emissions of the pollutant. For additional substantiation of the possibility of underestimating re-emissions, modelled concentrations of HCB in soil were compared with measured ones. A lot of measurements in soil for various years and locations are compiled in the EuroChlor Science Dossier cited above. It should be stressed that due to strong variability of soil concentrations and organic carbon content in soil as well as variability of soil concentrations in time, direct comparison of calculated and measured soil concentrations is rather difficult. Nevertheless some rough comparison can be made. Particularly, most of modelled soil concentrations (about 95%), obtained for 2009, do not exceed the level of 0.5 pg/g. At the same time, background HCB concentrations in soil measured in period 1998-2000 were about 100 pg/g. Taking into account possible decline of soil concentrations during 2000-2009 (up to approximately 10 times based on the study [*Barber et al.,* 2005]) and essential variability of HCB soil content due to the mentioned above reasons it can be concluded that soil concentrations (and, consequently, re-emission flux) can be underestimated by the model up to 20 times.

Uncertainties in definition of boundary and initial concentrations in media. The underestimation of observed HCB pollution levels in model simulations can be additionally caused by the uncertainties in definition of boundary and initial concentrations for modelling within the EMEP domain. The set of boundary and initial concentrations is generated in course of modelling of HCB long-range transport and fate at hemispheric scale. Thus, bearing in mind essential long-range transport potential of HCB, the knowledge on the emissions, both historic and contemporary, on global/hemispheric scale is of importance for reasonable description of HCB pollution levels at European scale.

Uncertainties of contemporary HCB emissions. The disagreement between model calculations and measurements can also be conditioned by uncertainties in contemporary emissions both in Europe and in the rest parts of the Northern Hemisphere for 2009. As it was mentioned above emissions of some European countries can be underestimated. This can be indirectly confirmed by substantial differences between emission totals of European countries and by the fact that countries recalculate their emission data backward in time (for example, Spain has recently reported 14-fold decrease of its HCB emission). As it was mentioned above compiled HCB emissions for the entire Northern Hemisphere are also subject of essential uncertainties. The uncertainties in the HCB emissions of non-EMEP sources can lead to the underestimation of intercontinental transport of the considered pollutant.

Thus, it can be seen that most essential reasons of underestimation HCB air concentrations in model simulations can be related to underestimated influence of secondary emission sources (via underestimation of historical emissions) and underestimated levels of contemporary HCB emissions in Europe and in the Northern Hemisphere. To examine the influence of these factors a number conventional emission scenarios were constructed and analyzed. Particularly, the increase of HCB re-emission, uniform and non-uniform increase of countries HCB emissions were considered.

Evaluation of emission scenarios was carried out on the basis of matrix approach described in Chapter 1. For this purpose the contributions of sources of all European countries to HCB air concentrations were evaluated using the model simulations with TNO emissions of HCB for 2009. On the basis of these calculations "country-to-site" matrix for HCB was generated. Using this matrix, it is possible to evaluate the changes in air concentrations at the locations of monitoring sites corresponding to emission totals of all European countries. The examples of contributions of European countries to the three EMEP monitoring sites are given in Fig. 3.41.

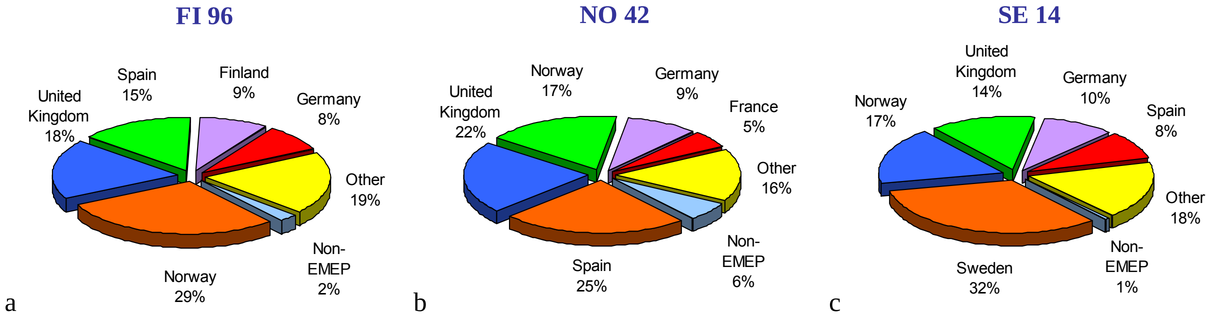


Fig. 3.41. *Contributions of various emission sources to annual mean HCB air concentrations for the location of two EMEP monitoring sites: FI96 (a), NO42 (b), SE14 (c)*

With the help of such information for all sites, it is possible to recalculate air concentrations at these sites in accordance with the changes of emission totals in the emission scenario. It should be stressed that contribution of HCB historically accumulated in the underlying surface from sources of entire Northern Hemisphere and contribution of non-EMEP sources also can be changed in the framework of emission scenarios.

The effects of the changes of HCB re-emission and countries HCB emissions in accordance with particular scenario on modelled HCB air concentrations and their agreement with measurements are summarized in the Table 3.8.

Table 3.8. *Conventional scenarios of HCB emission and statistical characteristics*

Initial calculations/scenario	Correlation of modelled and observed concentrations	Residual square deviation	Sites with agreement out of factor of 2
Initial calculations	0.75	38.4	All sites
Increase of re-emission	0.66	17.8	FI96 (2.4), NO1 (2.3)
Uniform increase of countries emissions	0.73	14.8	FI96 (3.4), SE14 (2.3)
Non-uniform increase of countries emissions	0.86	12.4	FI96 (3.4)

Increase of re-emissions. First scenario is aimed at the evaluation of the possibility to refine the agreement between measurements and model predictions by enlarging re-emission contribution (Re-Emission scenario). To do this, the contribution of re-emissions is enlarged up to the level for which the discrepancies between measurements and modelling can be explained by random factors at the significance level 95%. The contributions of the rest sources remain unchanged under this scenario. The required enlargement of re-emission contribution for such scenario is 14 times. The comparison of measured and calculated values of HCB air concentrations under this scenario is shown in Fig. 3.42. It should be noted that measurements of NO42 were not included in the analysis due to essentially high values of measured HCB concentrations (higher than observed concentrations in Europe). The reason of this phenomenon should be further considered in cooperation with national experts.

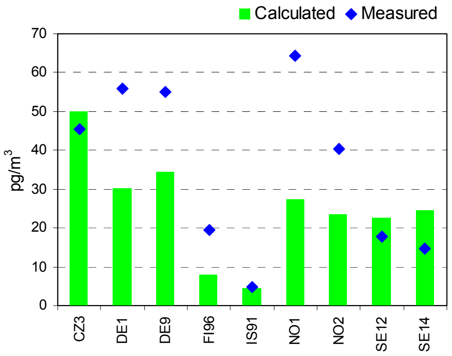


Fig. 3.42 *Comparison of modelled HCB air concentrations, pg /m^3 for 2009 (on the basis of scenario 'Increase of re-emission') with available measurements at EMEP sites (NO42 excluded)*

For the interpretation of the obtained characteristics it should be taken into account that initial correlation coefficients equals 0.75 and initial square deviation is 38.4. So, the application of this scenario has led to 2-fold reduction of square deviation from measurements to calculations with slight decrease of correlation coefficient. Further, at all sites but two the agreement between measurements and calculations is within a factor of 2.

The results of this scenario indicate that re-emission flux can be an important reason of underestimation of air concentrations by the model. Hence, the improvement of historical HCB emission inventory is needed for the refinement of the quality of model assessment (this conclusion confirms the conclusion made on the basis of the calculations made in the previous year). However, the decrease of correlation coefficient that takes place for this scenario shows that further refinement of the agreement cannot be achieved with the help of this factor only.

Uniform increase of countries emissions. The second scenario includes enlargement of emissions of all European countries 3 times with simultaneous enlargement of re-emission contribution (Uniform enlargement scenario). The enlargement coefficient for the countries emissions is chosen in accordance with the results obtained in [*Gusev et al.,* 2009] (see above). Similar to the previous scenario, the contribution of re-emission is enlarged up to the level for which the discrepancies between measurements and modelling can be explained by random factors at the significance level 95%. The enlargement coefficient in this case occurs to be 7. The comparison of measurements and calculations obtained under this scenario is shown in the diagram in Fig. 3.43.

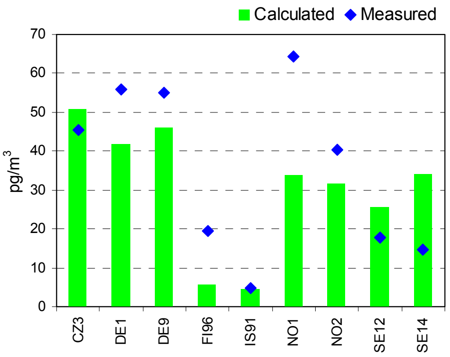


Fig. 3.43. *Comparison of modelled HCB air concentrations, pg /m*^{3} *for 2009 (on the basis of scenario 'Uniform increase of countries emissions' with available measurements at EMEP sites (NO42 excluded)*

This scenario leads to better refinement from the viewpoint of the square deviation. Again, calculated values at the considered sites but two agree with measurements within a factor of 2. Besides, the agreement at the German and Norwegian sites is better than under the first scenario (increase of re-emissions). It should be mentioned that the change of European emissions under this scenario is performed in all countries with one and the same coefficient, so that the spatial distribution of emissions remains unchanged.

Non-uniform increase of countries emissions. The third emission scenario supposes non-uniform enlargement of emission totals in European countries. For the construction of this scenario two statistical characteristics of the agreement between modelled and measured concentrations were taken into account, namely, correlation coefficient K_{corr} and the coefficient of multiple determination R^2. Of course, no optimization with regard to these two parameters was performed, so the scenario coefficients were chosen in such a way that the values of K_{corr} and R^2 are as good as possible. Here scenario coefficients are numbers at which total emissions of countries are multiplied to obtain scenario emissions.

Using this method, a scenario was constructed for which emissions of four European countries (Finland, Germany, Norway and the UK) are raised 4 times and re-emission contribution is raised 7 times (Spatial distribution scenario). This choice is compatible with the results obtained in [*Gusev et al.,* 2009] (see above). The comparison of measurements and calculations obtained under this scenario is shown in the diagram in Fig. 3.44.

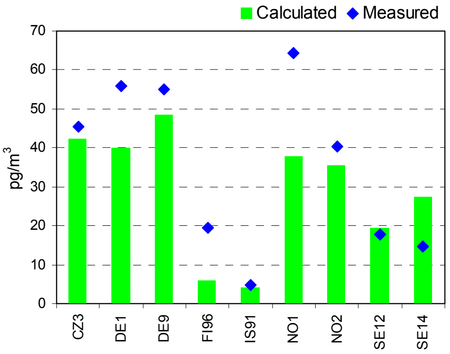


Fig. 3.44. *Comparison of modelled HCB air concentrations, pg /m*^{3} *for 2009 (on the basis of scenario 'Non-uniform increase of countries emissions' with available measurements at EMEP sites (NO42 excluded)*

It is seen that the results obtained under this scenario lead to better agreement with measurements in comparison with two previous scenarios (Table 3.8). This means that in order to refine description of HCB pollution levels in Europe and to improve the agreement between modelling results with measurements both the refinement of historical emissions and contemporary anthropogenic emissions are important.

The last scenario was elaborated on the basis of annual averages of air concentrations at the considered sites. However, it can be interesting to examine the agreement between modelled monthly averages of air concentrations under this scenario and observed concentrations. It should be noted that monthly mean HCB air concentrations at some of considered sites contain outliers, that is, the values of air concentrations lying outside 95% confidence interval with respect to other monthly averages at the same site. Such phenomenon is noticed at four sites: CZ3, IS91, SE12 and SE14. Seasonal variations of monthly averaged values of measured and calculated air concentrations of HCB for CZ3 and DE1 (with the excluded outliers) are shown in Fig. 3.45. It can be seen that, in spite of rough approach in the construction of scenarios modelling results reasonably describe seasonal variations of concentrations observed at a number of sites.

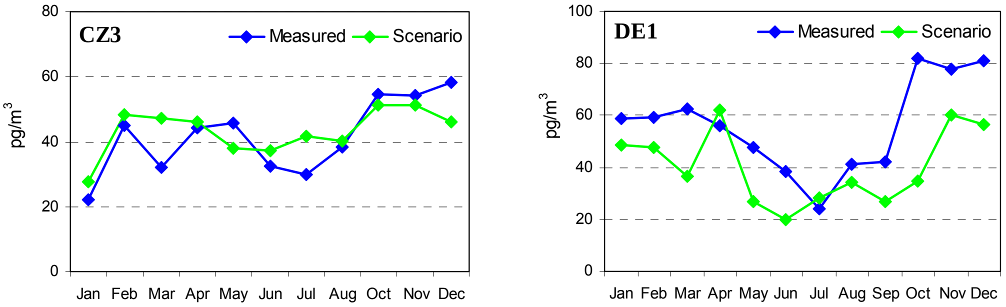


Fig. 3.45. *Comparison of modelled monthly averages of HCB air concentrations, pg /m^3 for 2009 (on the basis of scenario 'Non-uniform increase of countries emissions') with measurements at EMEP sites CZ3 and DE1*

At the same time, essential discrepancies between seasonal variations of measured and calculated air concentrations are found for the site FI96 (Fig. 3.46).

As seen from the plot, at this site seasonal variations of measurements and calculations are opposite. Namely, calculated concentrations are characterized by higher values in warm months whereas measured concentrations drop in summer. To analyze the reasons of this phenomenon influence functions [*Ilyin et al.*, 2010] for several months of 2009 are constructed (Fig. 3.47).

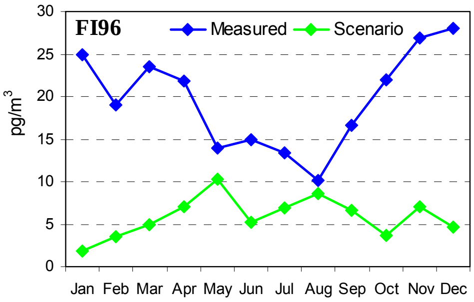


Fig. 3.46. *Comparison of modelled monthly averages of HCB air concentrations, pg /m^3 for 2009 (on the basis of scenario 'Non-uniform increase of countries emissions') with measurements at EMEP site FI96*

Calculations of the influence functions show that for the months with essential underestimation at FI96 (January, February, March, October, November and December) the transport from north-west of Russia or from north of Russia through the Arctic takes place. So, it can be supposed that emissions in these regions are underestimated in the inventory used in modelling.

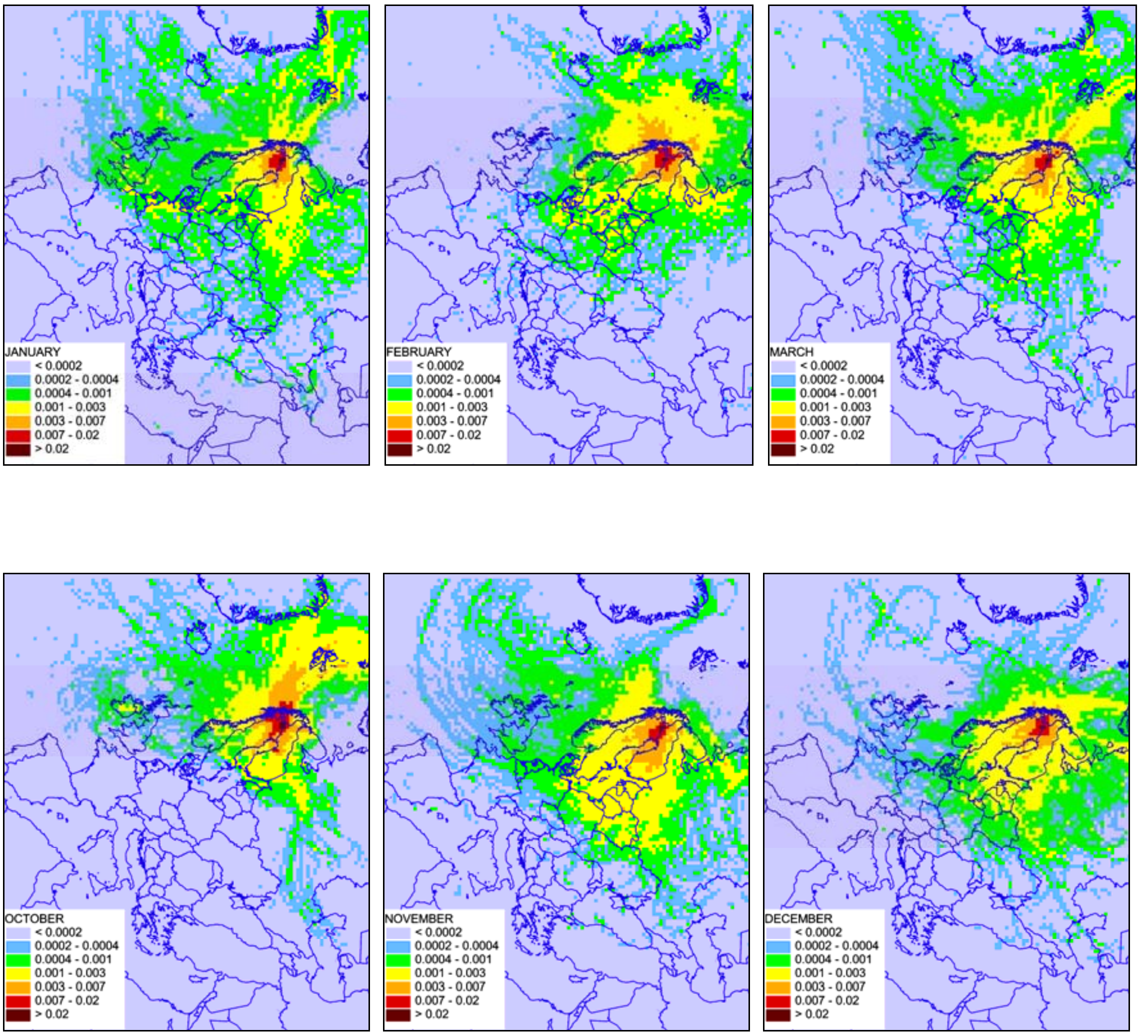


Fig. 3.47. *Influence functions for monthly averages of HCB air concentrations for EMEP site FI96, min/m*

It should be stressed once more that the results of scenario calculations do not indicate that emissions in the countries, for which emission totals were enlarged, are underestimated according to the used scenario coefficients. Each considered scenario operates with emission totals and does not change the spatial distribution of emissions within the countries. The scenario results just indicate the areas where additional examination should be performed. Such examination includes more detailed evaluation of emissions and their splitting into source groups considered in the model, refinement of model parameters, characterization of measurement data, etc. For more precise representation of spatial distribution of contamination modelling with finer spatial resolution is required. Such activity can be performed in the framework of case studies carried out in collaboration with national experts in emissions and monitoring.

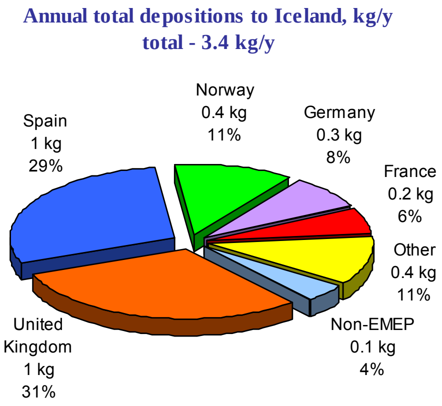


Fig. 3.48. *Import chart for Iceland*

Concerning the influence of intercontinental transport on the contamination of European countries, the calculations showed that it can be noticeable at remote locations (about 6% of anthropogenic contamination at NO42, see Fig. 3.41b) and in the countries located close to the EMEP boundary (e.g. Iceland, see Fig. 3.48).

So, according to the calculations with the used emission inventory, the contribution of intercontinental transport can be evaluated as about 5 – 10% in some areas. However, as it was mentioned above, refinement of emission inventories in North America and South-east Asian countries can lead to larger values of calculated contributions of the intercontinental transport.

Evaluation of transboundary transport. Modelling of long-range transport and deposition of HCB within the EMEP domain with calculating contributions of particular countries to air concentrations and deposition fluxes allows in principle evaluating of source-receptor relationships, that is, contributions of national emission sources, transboundary transport, and re-emission to air concentrations and deposition fluxes in various countries or/and at different locations. However, under strong uncertainty in the emission data the calculations of *import* (the contribution of other countries to deposition flux in the considered countries) is also highly uncertain. Another situation takes place for *export*, that is, the fraction of national emission deposited to the areas of other European countries, which is not dependent on emission totals. As an example, calculated spatial distribution of deposition flux originated from German sources is shown in Fig. 3.49.

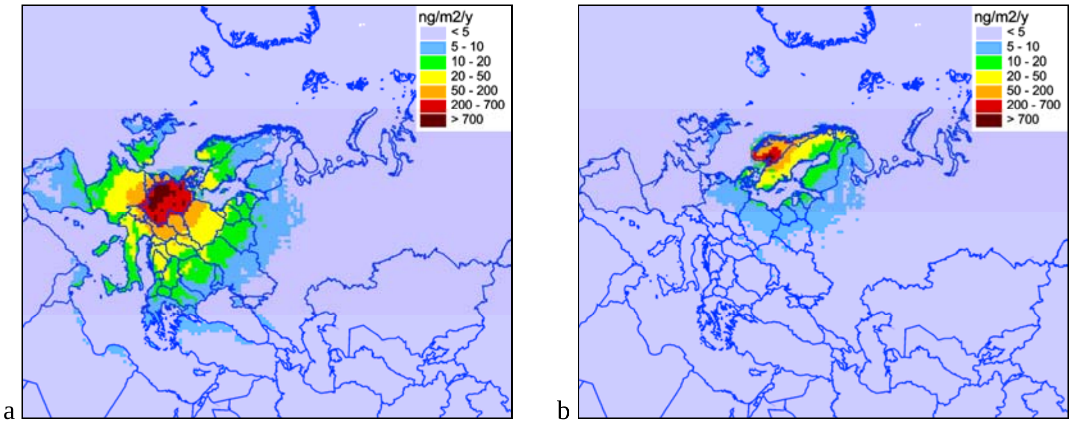


Fig. 3.49. *Spatial distribution of HCB deposition fluxes originated from sources of a particular country as calculated by the model: (a) – Germany, (b) – Norway, ng/m2/year*

Of course, absolute values of deposition flux depend on the total German emissions. However, the scaling of emissions will lead to the corresponding scaling of the values of deposition flux without a change of spatial distribution. Therefore, the fractions of deposition originated from emission sources of the given country that take place over the areas of other countries do not depend on emission total of the considered country. These fractions (export) are presented by the plot in Fig. 3.50.

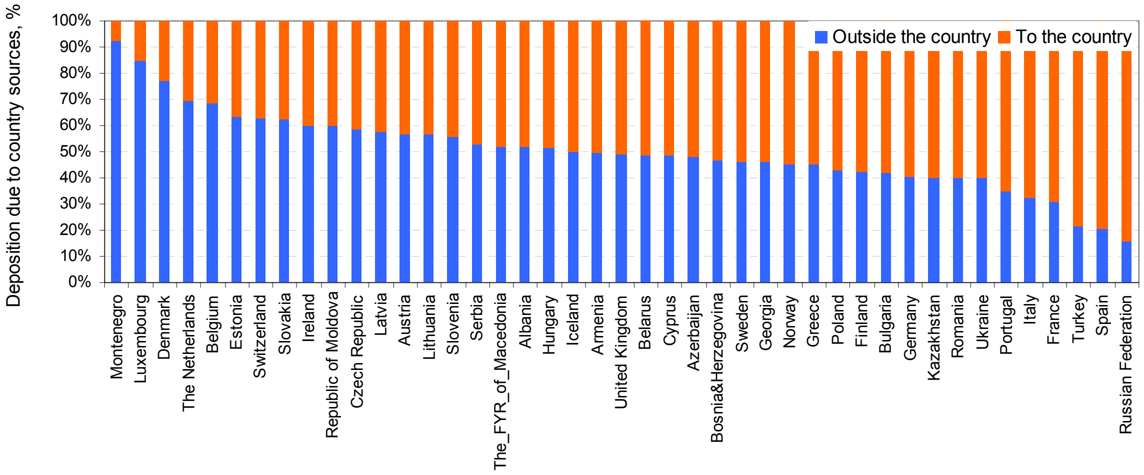
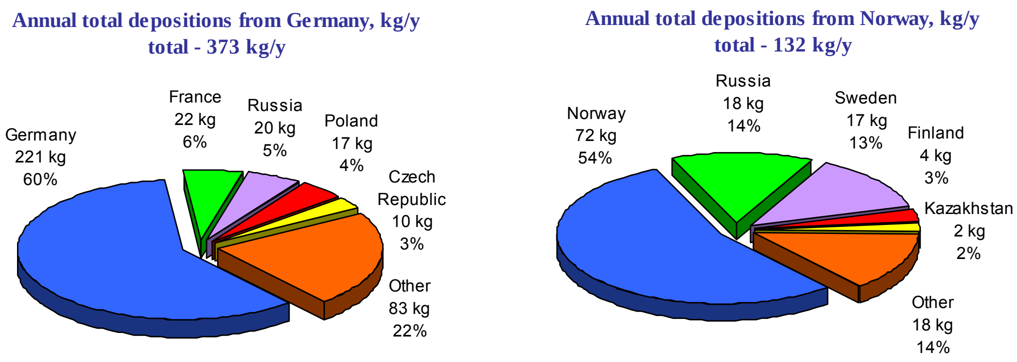


Fig. 3.50. *Fractions of deposition flux originated by emission sources of a country and deposited to the area of other countries (export)*

It can be seen that typically 30 – 60% of total deposition originated from sources of a particular country are deposited outside the area of this country. This fraction is strongly dependent on geographic location and the area of a country. For example, countries with small area located upwind form a lot of other European countries (e. g., Denmark, the Netherlands, Belgium) are characterized by high value of export fraction. From the other hand, lower export fractions are characteristic of countries with large territory from which the transport is directed outside the EMEP domain (e. g., Germany, Romania, the Ukraine, Portugal). Of course, export fractions can depend on meteorological situation of a given year.

More detailed information on export from European countries can be obtained from the export charts showing fractions of total deposition originated from emission sources of the country and deposited to the area of other European countries. Two examples of such export charts are shown in Fig. 3.51 for Germany and Norway. It should be stressed that, unlike the import charts, which change essentially with the change of emission scenario, export charts are independent of the considered emission scenario concerning relative shares of deposition flux deposited to this or that country.



a b

Fig. 3.51. *Fractions of depositions due to the country sources deposited to the areas of various European countries: (a) – Germany, (b) – Norway*

In general, the set of spatial distributions of deposition fluxes originated by all European countries can serve as a kind of influence function for depositions. Namely, if emissions of all countries are given, deposition flux over the entire area of the EMEP domain can be calculated as follows. Spatial distributions of deposition fluxes originated by country sources can be obtained by scaling spatial distributions of each country according to the given emission totals. Then, summing all scaled spatial distributions, total spatial distribution of deposition fluxes can be obtained. Of course, the same approach can be applied for the calculation of air concentrations. The disadvantage of this approach is that spatial distribution of emissions inside each country cannot be changed without the actual modelling with more detailed splitting of emissions into set of sources.

To exemplify the possibility to use matrix calculations for the evaluation of air concentrations in the entire EMEP domain, spatial distribution of air concentrations under the above constructed emission scenario (Non-uniform increase of countries emissions) was evaluated (Fig. 3.52).

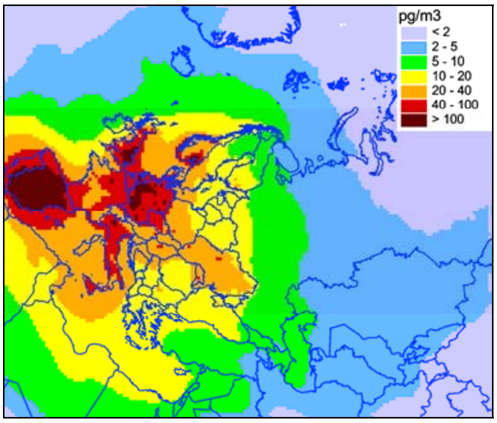


Fig. 3.52. *Spatial distribution of HCB air concentrations, pg /m^3 (b) in 2009 calculated by MSCE-POP model on the basis of the scenario 'Non-uniform increase of countries emissions'*

It should be mentioned once more that spatial distribution shown in Fig. 3.52 is conventional since it is based on the conventional emission scenario. In addition, it should be taken into account that all changes in emissions involved in the constructed scenario are made in the countries that noticeably contribute to air concentrations at EMEP sites with data on HCB available.

This study represents an attempt to evaluate current level of uncertainties involved in the assessment of HCB pollution of European region and to discuss possible directions of its improvement. Further activity in this respect can be performed in framework of specialized case studies for selected European countries with participation of national experts in emissions and monitoring. Particularly, such studies can make it possible to organize additional monitoring campaigns, to improve HCB emissions on the example of particular European countries, to refine model parameterization for HCB and apply fine resolution modelling, which finally can lead to the refinement of pollution assessment for the whole European region.

4. DEVELOPMENT OF GLOBAL MODELLING FRAMEWORK GLEMOS FOR POPS

Investigation of POP global transport is an important field of activity aimed at evaluation of the contribution of global emission sources to the pollution levels with the EMEP domain. To investigate the role and extent of the influence of global sources the EMEP has initiated the development of relevant modelling approach starting from hemispheric scale and moving recently to the global scale modelling. In framework of this activity MSC-E continued development of the Global EMEP Multi-media Modelling System (GLEMOS) for POPs. Particularly, main attention was paid to the description of the behaviour of POPs in the marine environment including transport of the pollutants with sea currents. To prepare global-scale input data on ocean currents Parallel Ocean Program has been chosen as oceanic preprocessor and its output was adapted for usage in the GLEMOS. New oceanic advection-diffusion module has been developed, tested, and implemented in the model. Detailed description of the progress in the development of GLEMOS can be found in [*Travnikov and Jonson*, 2011].

Below the information on Parallel Ocean Program, its adaptation as an oceanic preprocessor, spin-up and testing is presented. The description of the new oceanic advection-diffusion block for GLEMOS is given. Some preliminary results of POP ocean transport modelling are discussed.

Preprocessing of oceanic data

Parallel Ocean Program (http://climate.lanl.gov/Models/POP/) was chosen as the oceanic pre-processor for the GLEMOS model. It is freely available model developed at Los Alamos National Laboratory. This program is derived from the Bryan-Cox-Semtner class of ocean models [*Semtner*, 1986] first developed by Kirk Bryan and Michael Cox at the NOAA. Parallel Ocean Program is the ocean component of the Community Climate System Model - CESM (http://www.cesm.ucar.edu/) - a fully-coupled, global climate model that provides state-of-the-art computer simulations of the Earth's past, present, and future climate states. This subsection is devoted to the description of the use of this model at the MSC-E.

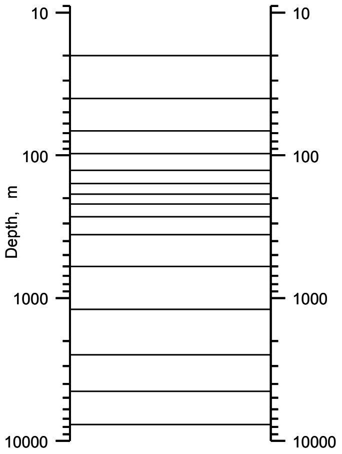


Fig. 4.1. *15-layer vertical structure of POP ocean model*

Grids and bathymetry

The Parallel Ocean Program model was compiled and launched at the MSC-E at global scale with 3^0x3^0 and 1^0x1^0 spatial resolutions. The data with 3^0x3^0 resolution is supposed to be used as input information for long-term historical modelling of persistent organic pollutants fate aimed media saturation and the preparation of input data for 1^0x1^0 modelling.

The vertical structure of the model grid chosen both for 3^0x3^0 and 1^0x1^0 resolutions is shown on Fig. 4.1. 15 irregular layers are used, their boundaries coincide with the appropriate boundaries of 29-layer ECMWF ORA-S3 ocean re-analysis layers for better data assimilation.

Gridded bathymetry data sets have been compiled on the base of the following two information sources:

1. ISLSCP2 ¼ degree dataset (http://daac.ornl.gov/ISLSCP_II/islscpii.shtml): land-ocean mask.
2. ETOPO2v2 global gridded 2-minute database (http://www.ngdc.noaa.gov/mgg/global/etopo2.html): global relief.

Spin-up

To initialize global-scale oceanic calculations monthly climatological data on potential temperature and salinity of the water were used (NOAA NODC World Ocean Atlas 2005: http://www.nodc.noaa.gov/OC5/WOA05/pr_woa05.html). The velocity field was set zero at the start. The model physics spin up a velocity field in balance with the density field.

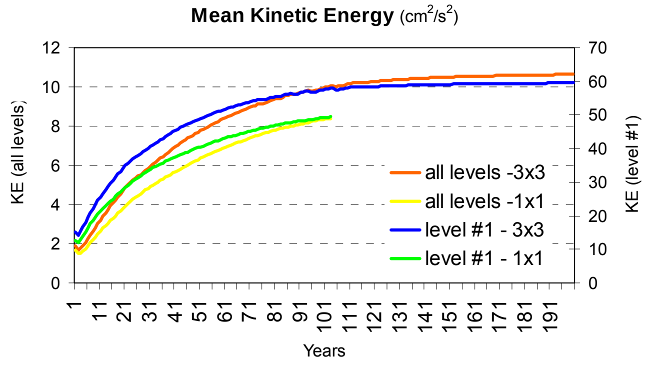


Fig. 4.2. *Mean kinetic energy for 1^0x1^0 and 3^0x3^0 spin-up runs for the upper model layer (right vertical axis) and for all the layers (left axis)*

Spin-up is performed for the time period sufficient for an ocean model to reach a state of statistical equilibrium under the applied forcing. It is usually difficult for global general circulation models to reach this state. The deep ocean requires hundreds of years to adjust. The upper ocean only requires about 50-100 years. We have performed 200-year spin-up for 3^0x3^0 grid and 100-year one – for 1^0x1^0 grid.

Mean kinetic energy (KE) of water is traditionally used as an indicator of statistical equilibrium. Fig. 4.2 shows time trends of total mean KE and KE of the upper water layer during spin-up periods for 1^0x1^0 and 3^0x3^0 resolutions. It can be seen that upper ocean reached quasi-steady state earlier than Deep Ocean. KE depends on grid resolution, but the curves look similar.

The result of the spin-up on climatological data is set of statistically balanced gridded variables which could be used for subsequent short-term simulations. It should be noted that the field of currents velocity (Fig. 4.3) was formed by the gravity and the Coriolis force. Surface wind stress was not taken into account. For this reason, velocity field is smooth and slowly changing. To take into consideration the friction between the wind and the water's surface along with the influence of other surface meteorological parameters (atmospheric pressure and temperature) it is needed to assimilate high temporal resolution analysis data. ECMWF 6-hour meteorological re-analysis (**http://www.ecmwf.int/**) was used for this purpose at the second stage of spin-up (Fig. 4.4). In addition to this, 3-D daily data on ocean potential temperature and salinity from ECMWF ORA S3 ocean re-analyses were assimilated at this stage. The inclusion of new data required several years to establish new equilibrium.

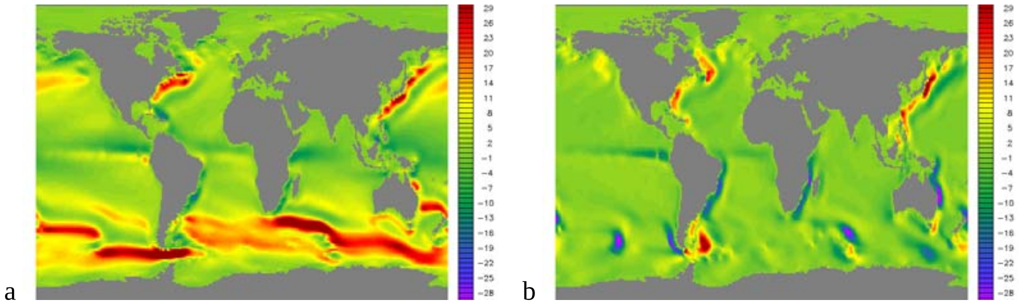


Fig. 4. 3. *Spatial distributions of zonal (a) and meridional (b) current velocities (cm/s) in the upper ocean layer after 100-year spin-up with 1^0x1^0 spatial resolution*

Results for 2009

The last phase of the complete computation cycle – the preprocessing of ocean parameters distribution for the GLEMOS model (Fig. 4.4) - was carried out using the forcing to ECMWF data too. 1^0x1^0 and 3^0x3^0 data sets for 2009 have been prepared. To evaluate the performance of Parallel Ocean Program model and reliability of these data ocean currents fields were analyzed. The velocity of currents was not assimilated from analysis. For this reason, it can be used as an objective and independent measure of data quality.

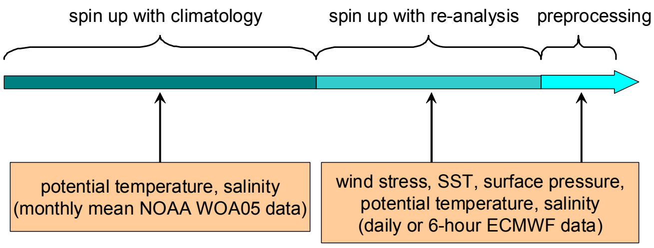


Fig. 4.4. *Calculation cycle of POP model*

The spatial distributions of currents velocity components in the upper ocean layer (Fig. 4.5) are much more complicated then those without wind stress influence (Fig. 4.3). The major currents (Equatorial, Gulf Stream, Kuroshio, Antarctic Circumpolar etc.) were reproduced.

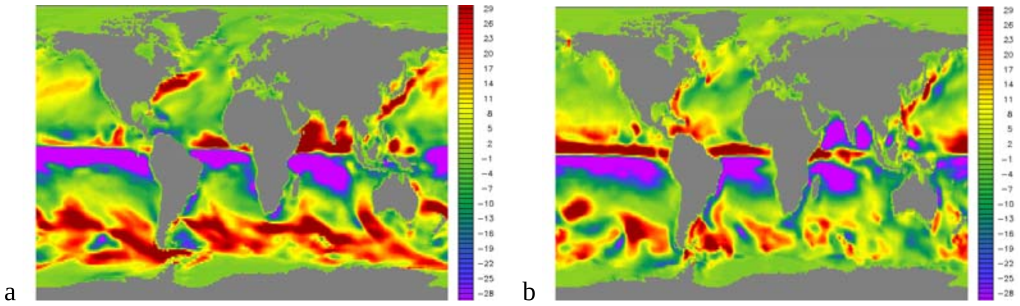


Fig. 4.5. *Spatial distributions of zonal (a) and meridional (b) current velocities (cm/s) in the upper ocean layer on 31 Dec 2009*

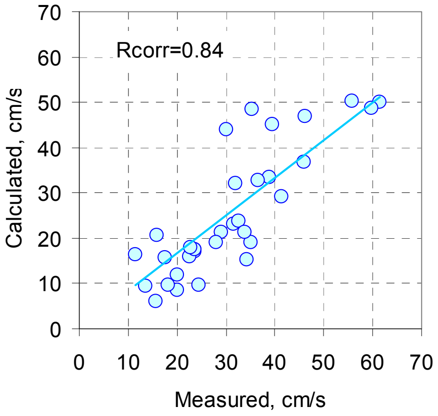


***Fig. 4.6.** Scatter plot for annual mean calculated and measured ocean current velocities*

Calculated ocean currents have been compared with fixed depth measurement data of Tropical Atmosphere Ocean (TAO) project (http://www.pmel.noaa.gov/tao/). 33 of 39 TAO measurement sites were involved in the comparison. 5 sites were excluded due to insufficient number of measurements, 1 site – due to very strong variations of currents in the vertical direction which could not be reproduced by 15-layer model. Annual mean modelled (1^0x1^0) and measured values at 10m depth are in good agreement (Fig. 4.6). Daily computed data for most of the stations correlates with measurements (Fig. 4. 7 - example for equatorial station in the Indian Ocean)

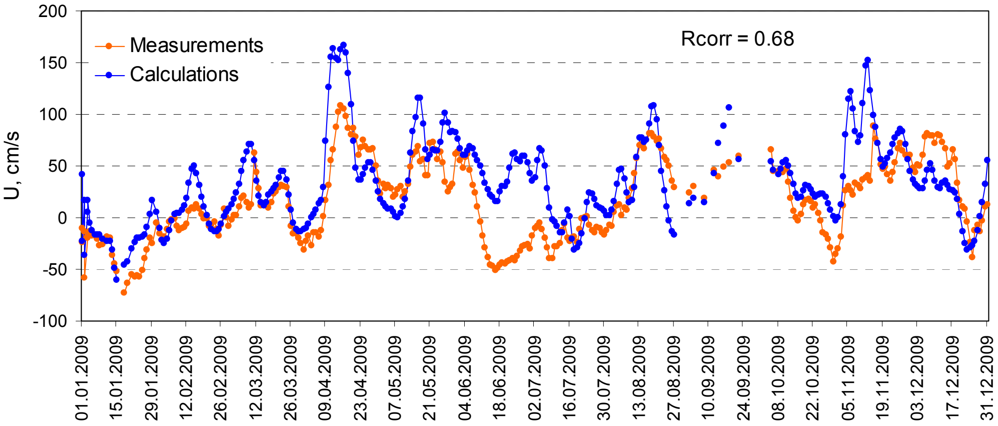


***Fig. 4.7.** Calculated and measured daily averaged zonal ocean current velocities in the Indian Ocean (0^0 N, 80.5^0 E) at depth 10 m*

Development of ocean transport module for GLEMOS

Parallel Ocean Program provides a means of modelling the advection and diffusion of passive tracers in the ocean. The modelling of oceanic transport of passive tracers is performing using the similar methods that are used for the main ocean parameters such as temperature, salinity and currents velocity. It is reasonable to employ the same numerical methods and discretization for the description of oceanic tracer transport in the GLEMOS model to ensure the compatibility of the models and to provide better adaptation of input data.

The numerical scheme of tracer advection and diffusion used in the Parallel Ocean Program model was implemented into the oceanic block of the GLEMOS model (which also contains partitioning, degradation and sedimentation modules) and tested. Below some results of the testing of the new

GLEMOS oceanic transport module are presented. More information can be found in [*Travnikov and Jonson*, 2011].

Rotational flow field test of the advection scheme

Several simple one- and two-dimensional advection-diffusion tests of transport module have been performed. One of them is rotational flow experiment for the evaluation of the diffusion of numerical scheme first proposed by *Smolarkiewicz* [1982]. It has been carried out in Cartesian and latitude-longitude coordinates.

a. Cartesian coordinates

A cone with base radius $15\Delta x$ and maximum height $\psi_{\max 0} = \sqrt{15\Delta x}$ was originally located in background field $\psi_0 = 1$ in two-dimensional domain of 200×200 grid points with $\Delta x = \Delta y = 1$. The cone was rotated with constant angular velocity $\omega = 0.1$ clockwise around the point $(100\Delta x, 150\Delta y)$. (Fig. 4.8a). The integration was carried out with the time step $\Delta t = 0.05$. The test has shown that numerical scheme diffusion is not very high (Fig. 4.8, 4.9). After one full rotation the cone became 14% lower. Its form was somewhat disturbed. Low-amplitude wavelike disturbance of background field took place on the leeward side of the cone (Fig. 4.9).

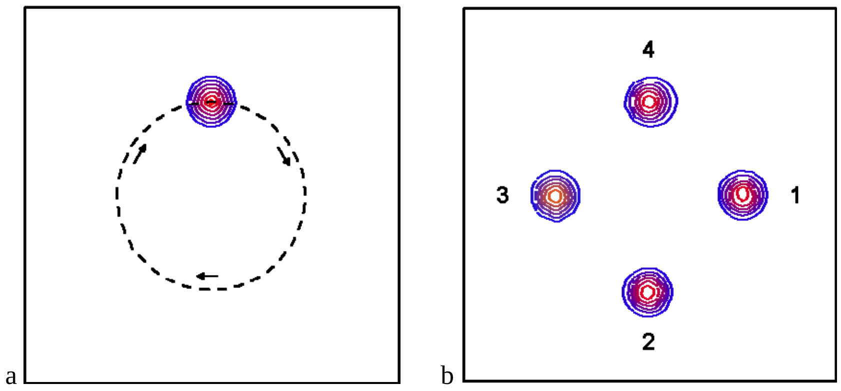


Fig. 4.8. *Initial conditions (a) and results (b) of rotational field flow test. Concentric circles denote isolines of ψ value with step $\Delta\psi=0.5$ after ¼ of full revolution (1), ½ of full revolution (2), ¾ of full revolution (3), full revolution (4)*

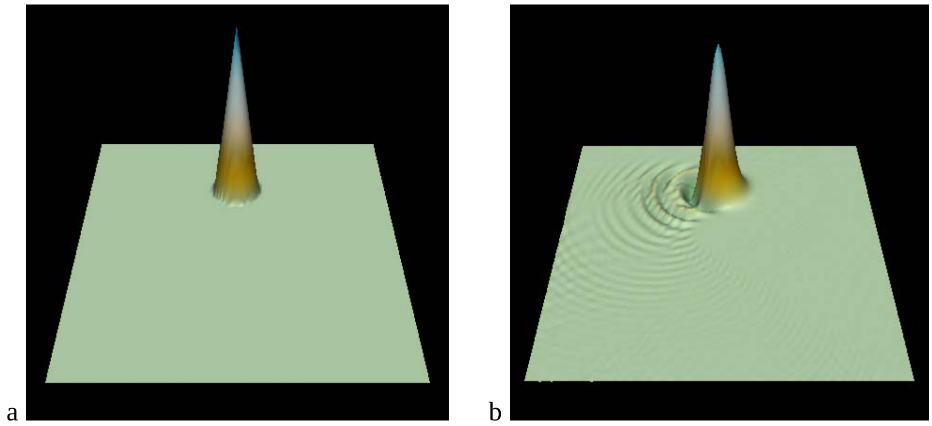


Fig. 4.9. *Spatial distributions of ψ value in rotational field flow test: a – initial, b – after one full rotation*

b. Latitude-longitude coordinates

The same test was carried out on 1^0x1^0 latitude-longitude grid. The center of rotation was places on the equator. A cone with radius $\pi R_e/180$ (where R_e is the radius of the Earth) and height $\psi_{\max_0} = \sqrt{15}$ began to rotate in background field $\psi_0 = 1$ from the initial point located at latitude φ =45^0 and the same longitude as the rotational center ($\Delta\lambda$ =0). The components of rotation velocity for this case was defined as

$$V_\varphi = -\omega R_e \sin(\Delta\lambda)$$
$$V_\lambda = \omega R_e |\cos(\Delta\lambda)| \sin\varphi$$

where $\omega = 2\pi$ min^{-1} – the angular velocity.

The results of integration with time step $\Delta t = 0.05$ (Fig. 4.10) are similar to those in Cartesian coordinates. The cone height reduction was the same 14%.

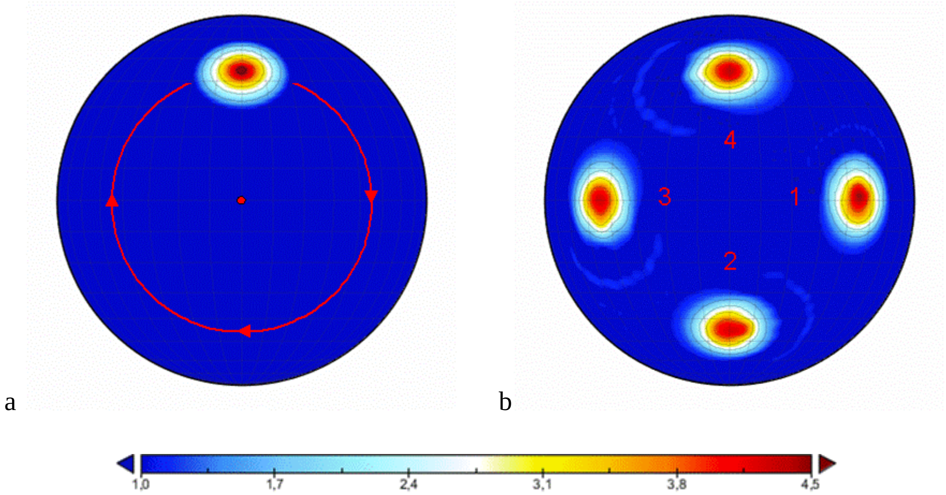


Fig. 4.10. *Initial conditions (a) and results (b) of rotational field flow test. Concentric circles denote isolines of ψ value after ¼ of full revolution (1), ½ of full revolution (2), ¾ of full revolution (3), full revolution (4)*

Tracer test

Annual calculation of tracer ocean transport in the real field of currents (calculated by the POP model for 2009) has been performed. It was supposed that four point sources located near the coast of the USA, Italy, Japan, and Nigeria released tracer to the upper ocean layer with constant rate. Tracer was assumed to be diphasic: dissolved and particulate phases were considered. The following oceanic processes were examined: advection, vertical and horizontal diffusion, partitioning, degradation, and sedimentation. The properties of tracer related to these processes were supposed to be the same as that of PCB-153. The exchange with other media was neglected.

Spatial distributions of tracer water concentration for two time moments in the middle and at the end of the year are presented in Fig. 4.11. It can be seen, that tracer was transported from the sources relatively slowly. For example, being released in Gulf Stream, it did not reach the European coast by the end of the year (Fig. 4.11b). Most of tracer mass remained in the upper ocean during all the period of simulation (Fig. 4.12).

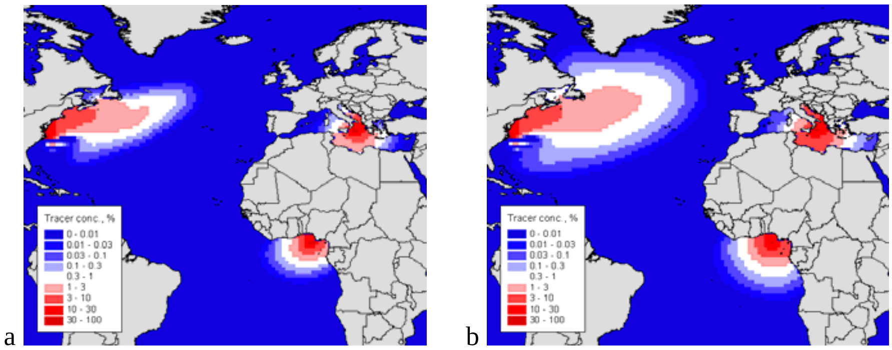


Fig. 4.11. *Tracer test results. Spatial distributions of tracer ocean concentrations in the upper model layer on Jul 31 (a) and Dec 31 (b). Units: percent of the maximum value on Dec 31. Japanese source plum is not presented*

Almost the entire released tracer mass was preserved in the ocean. Only 1 percent degraded. The share of sedimented mass was negligible.

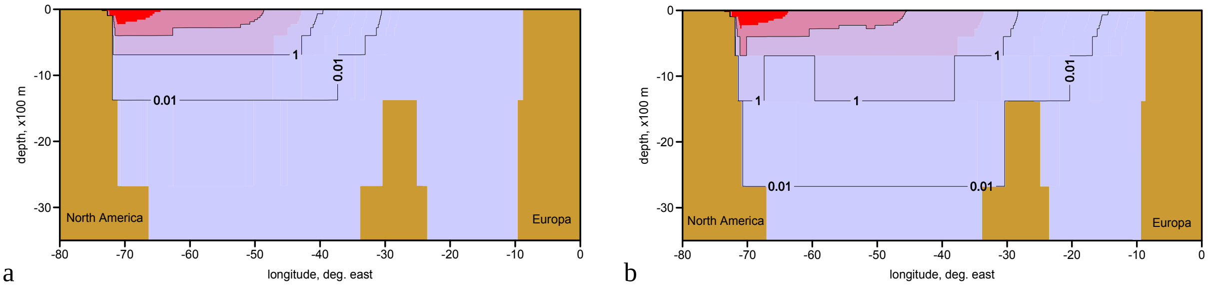


Fig. 4.12. *Tracer test results. Spatial distributions of tracer ocean concentrations in vertical plane located at 40^0 N on Jul 31 (a) and Dec 31 (b). Units: percent of the maximum value on Dec 31. Solid lines denote levels 0.01%, 1%, 10%. Tracer released near the coast of the USA (75^0 W, 35.5^0 N)*

Modelling of PCB-153 global transport

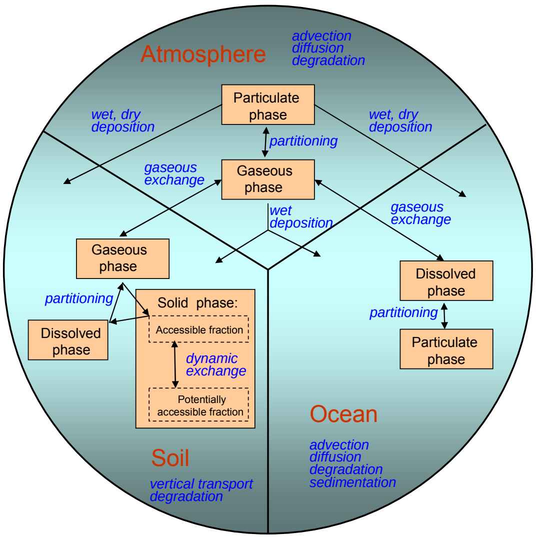


Fig. 4.13. *The scheme of the main processes related to POPs in the current version of the GLEMOS model (the dissolved phase includes also POPs sorbed on the dissolved organic matter)*

The scheme of the main processes related to POPs in current GLEMOS version after the implementation of the new ocean module is illustrated in Fig. 4.13. Several phases of POPs (shown as rectangles) are considered in each medium. These phases are involved in different physical-chemical processes (highlighted blue), namely, transport, degradation, phase partitioning, and inter-media exchange (arrows).

To evaluate the capability of GLEMOS to reproduce POPs environmental contamination levels at the current stage of model development annual (2009) global-scale simulation of PCB-153 transport on 1^0x1^0 grid was performed.

Calculated spatial distributions of PCB-153 annual mean concentrations in air, ocean and soil are presented in Fig. 4.14-4.16 correspondingly.

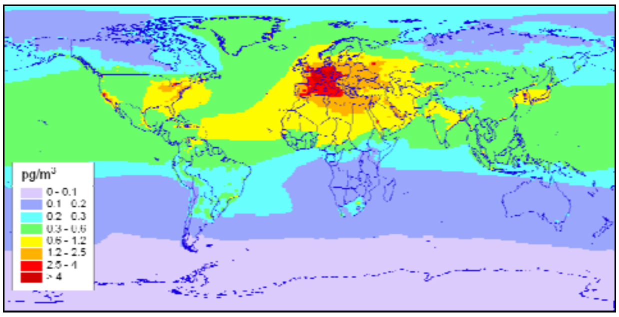


Fig. 4.14. *Spatial distributions of PCB-153 annual mean concentration in surface air for 2009*

The atmosphere. Elevated level of contamination is a characteristic of the Europe and North America (Fig.4.14). In the most polluted central part of Europe air concentrations exceed 4 pg/m^3. Over the most part of the Southern Hemisphere PCB-153 air concentrations are relatively low (0.1-0.3 pg/m^3).

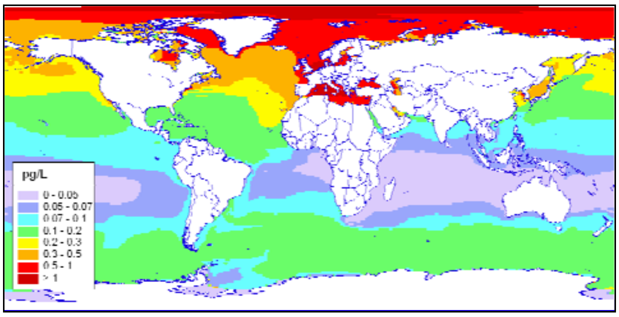


Fig. 4.15. *Spatial distributions of PCB-153 annual mean concentration in seawater for 2009, pg/L*

The ocean. There is evident latitudinal dependence of pollution level (Fig. 4.15). The highest values of PCB-153 content in seawater (more then 0.5 pg/L) were obtained for high-latitude regions. The reason for this is strong temperature dependence of the gaseous flux from air to water: lower air temperatures correspond to higher air-water flux. Seawater PCB-153 concentrations in the Antarctic region are higher than those in equatorial regions.

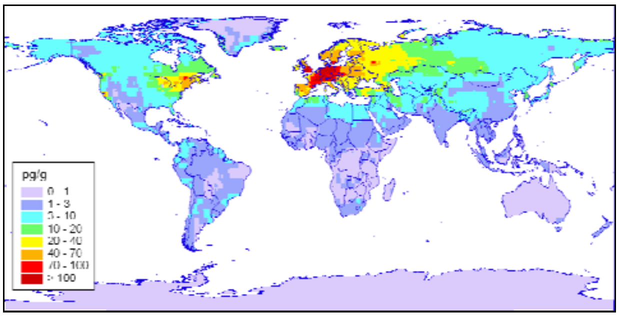


Fig. 4.16. *Spatial distributions of PCB-153 annual mean concentration in the top 5 cm of soil for 2009*

Soil. Elevated levels of PCB-153 in soil occurred in the regions with maximum anthropogenic emissions: they are Europe and Northern America (Fig. 4.16). Maximum concentrations (over 100 pg/g) take place in the Central Europe. The range 20-70 pg/g can be assumed as background for Europe, 3-10 pg/g – for North America.

Thus, three environmental media have been included in the GLEMOS model: the atmosphere, the ocean and soil. The global-scale modelling of POP fate in the ocean with sea currents, diffusion, degradation, phase partitioning, and sedimentation processes are available now. Further work on the elaboration of the multimedia approach within the GLEMOS modelling system will be directed, in particular, to the description of POP fate in vegetation and the interaction of this compartment with the atmosphere and soil.

5. INTER-LINKAGES BETWEEN CLIMATE CHANGE AND POP POLLUTION

The influence of climate variability and change on POP pollution has recently received increasing attention and is recognized as an important issue by many international organizations (CLRTAP, UNEP, AMAP, etc.). Particularly, the TF HTAP Assessment 2010 [*Dutchak and Zuber*, 2011], summarizing the information on intercontinental transport of POPs, concludes that climate change has the potential to affect all pathways of POPs in the atmosphere and in other environmental compartments (hydrosphere, cryosphere, soils, and biosphere). Changing climate may alter exposure pathways and increase vulnerability for the biotic environment and related health impacts.

The Stockholm Convention in cooperation with the AMAP has conducted a study on climate change and POPs inter-linkages. The outcome of the study, summarized in the report [*UNEP/AMAP*, 2011], was presented at the AMAP Conference "The Arctic as a Messenger for Global Processes – Climate Change and Pollution" in May 2011. The study concludes that climate change is expected to increase planet's vulnerability to POP pollution. The main factors, directly related to climate change and influencing POP fate and long-range transport, include increasing temperature, altering of atmospheric and oceanic transport pathways and their intensity, melting of sea and land ice, and increasing frequency and strength of extreme events.

Taking into account the importance of this issue Parties to the LRTAP Convention recognized the necessity to establish work on the links between climate change and air pollution by mercury and POPs. Thus, recent Executive Body session recommended to include this activity as one of the priority tasks into the long-term strategy for the Convention [*ECE/EB.AIR/106/add.1*].

Evaluation of effects of climate variability on POP pollution represents a challenging task due to the complex nature of their cycling in the environment which depends on interaction of many factors. For example, most of processes governing POP fate, namely, volatilization, phase partitioning and degradation, are sensitive to the changes of temperature and precipitation. Potential effects of climatic changes on POP transport and fate were reviewed in a number of studies [*MacDonald et al.*, 2005; *Dalla Valle et al.*, 2007; *Lamon et al.*, 2009a]. It is noted that possible effects of climate change on POP pollution can be connected with the changes of primary and secondary emission rates, atmospheric circulation and distribution of atmospheric constituents (atmospheric particles, reactive species), efficiency of removal processes (degradation, dry and wet deposition), characteristics of underlying surface (changes of land use, organic carbon content), ocean currents, and snow/ice cover.

Bearing this in mind, it is important to use models in order to evaluate responses of POP pollution to the climate variability. Particularly, application of models allows accounting complex interaction of various processes governing POP fate, examining sensitivity of POP transport and levels to variations of climate parameters, and performing modelling experiments based on the climate change scenarios.

A few modelling studies have been performed recently to analyze the fate of POPs in response to the climate variability and change [*MacLeod et al.*, 2005; *Dalla Valle et al.*, 2007; *Lamon et al.*, 2009b]. For example, *MacLeod et al* [2005] examined relationships in variations of modelled PCB concentrations and North Atlantic Oscillation index as an indicator of climate variability. It was found that variations of NAO index correlated with modelled concentrations at some locations, besides similar correlations could be seen with measurements in North America and Europe. *Lamon et al.* [2009b] found increased intercontinental transport of PCBs and higher emissions of POPs to air from primary sources due to projected enhanced wind speed and higher temperature under the scenario representing larger climatic changes (SRES (Special Report on Emission Scenarios) A2 scenario) in comparison to present conditions. Spatial attention to this issue is paid within the ongoing EU project ArcRisk where a number of modelling studies are carried out to explore the influence of climate change on POP transport to and fate in the Arctic.

MSC-E has started to work in this direction evaluating sensitivity of POP long-range transport potential to seasonal variations of selected meteorological parameters and land cover characteristics. This activity is continued with the examining of sensitivity of POP air concentrations and net deposition fluxes to variability of wider range of environmental parameters. Besides, the preparatory work for carrying out modelling experiments with climate change scenarios data is initiated. The progress in this work is presented below.

Analysis of sensitivity of POP pollution to variation of climate related parameters

Analysis of sensitivity of POP pollution levels to variation of meteorological and environmental factors is of importance for understanding POP transport and fate as well as their links with climate variability. POPs physical-chemical properties vary in wide range so behaviour of different POPs will be characterized by different responses to climate changes. Besides, projected changes of meteorological parameters, e.g. temperature and precipitation, will likely have different character in different parts of Europe that will also affect the levels of pollution.

Previous stage of this work, described in [*Shatalov et al.*, 2010], was devoted to the evaluation of POP transport distance and its dependence on the selected meteorological and environmental factors using successive regression analysis. In particular, the influence of temperature, precipitation amount, and land cover characteristics were considered. Thus, it was shown that such factors as temperature and vegetation cover had essential effect on the transport distance of POPs.

At current stage similar analysis is applied to the relationships between wider range of meteorological and environmental factors and variability of POP air concentrations and net deposition fluxes as target parameters. Two POPs were selected for this analysis, namely, B[a]P and PCB-153, representing the POPs with different physical-chemical properties. For example, B[a]P is mainly presented in the atmosphere in particulate phase whereas for PCB-153 the gaseous phase is more essential. The study considers seasonal variations of POP pollution levels in a form of spatial averages of air concentrations and net deposition fluxes over individual countries as target parameters. Examples of the results are given below for air concentrations of the selected POPs. Detailed information on both target parameters can be found in the Technical Report [*Shatalov et al.*, 2011].

The following meteorological and environmental factors are used in the analysis, namely:

- Temperature;
- Precipitation amount;
- Wind speed;
- Wind direction;

- Outflow of air masses through the boundary of the considered country;
- Leaf area index of forests;
- Leaf area index of low vegetation.

In comparison with the previous study [*Shatalov et al.*, 2010] additional factors were included in the analysis. In particular, to characterize the effect of the variance of transport pathways the wind direction, wind speed, and outflow of air masses from a country were added. Additionally, such factors as temperature, precipitation amount and wind speed were considered both for a country and EMEP domain levels. Detailed description of this approach can be found in the Technical Report [*Shatalov et al.*, 2011].

The analysis applied in this study allows ranking of meteorological and environmental factors by their influence on chosen target parameters. Exploring of this influence and ranking of the factors are performed for individual countries of the EMEP domain to describe their spatial variations. The ranking is carried out according to the results of the regression analysis.

Averaged ranks over all European countries for B[a]P and PCB-153 air concentrations as target parameter are shown in Fig. 5.1.

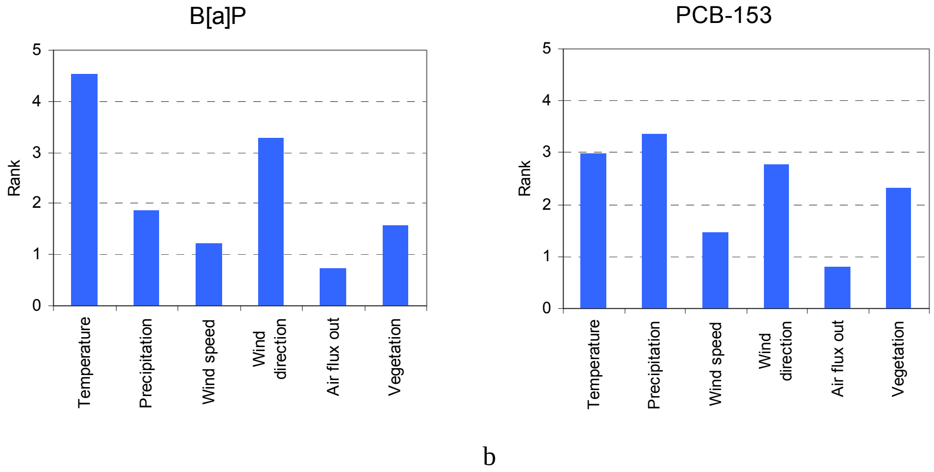


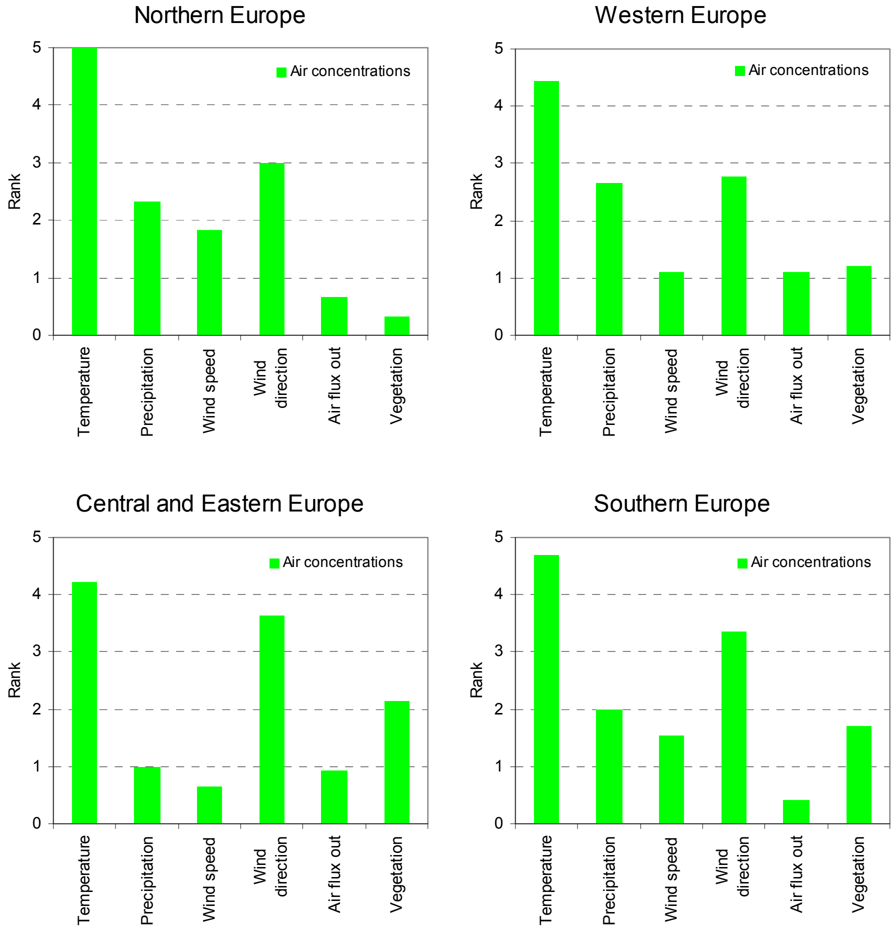
Fig. 5.1. *Ranking of meteorological/environmental factors for air concentrations of B[a]P (a) and PCB-153 (b)*

Essential variability of physical-chemical properties of POPs may lead to different changes in their fate in response to climate changes. Particularly, it can be seen that for B[a]P the highest priority belongs to temperature which is followed by wind direction, precipitation amount and vegetation cover. For PCB-153 the most essential factor is precipitation followed by temperature, wind direction and vegetation cover. The influence of temperature is substantially higher for B[a]P air concentrations comparing to PCB-153 which might be connected with higher rate of degradation in the atmosphere for B[a]P. For PCB-153, characterized by longer atmospheric half-life and more intensive inter-media cycling, the importance of temperature variations is close to precipitation and wind direction. The vegetation cover has fourth priority among the other factors both for B[a]P and PCB-153.

Ranking of factors for different parts of Europe. Ranking of meteorological and environmental factors can essentially vary in different parts of Europe. Therefore, it is interesting to explore how geographic location affects the importance of considered factors. To do that, four groups of European countries, divided by their geographic locations, were examined.

- Northern Europe: Estonia, Finland, Ireland, Iceland, Norway and Sweden;
- Western Europe: Belgium, Germany, Denmark, Spain, France, the UK, Luxembourg, the Netherlands and Portugal;
- Central and Eastern Europe: Austria, Bulgaria, Belarus, Switzerland, the Czech Republic, Hungary, Lithuania, Latvia, the Republic of Moldova, Poland, Romania, the Russian Federation and the Ukraine;
- Southern Europe: Albania, Armenia, Azerbaijan, Bosnia and Herzegovina, Cyprus, Serbia, Montenegro, Greece, Italy, Kyrgyzstan, Kazakhstan, the FYR of Macedonia, Slovenia, Slovakia, Tajikistan, Turkey and Uzbekistan.

The results of the ranking of considered factors for the above country groups for B[a]P air concentrations are presented in Fig. 5.2.


Northern Europe

Western Europe

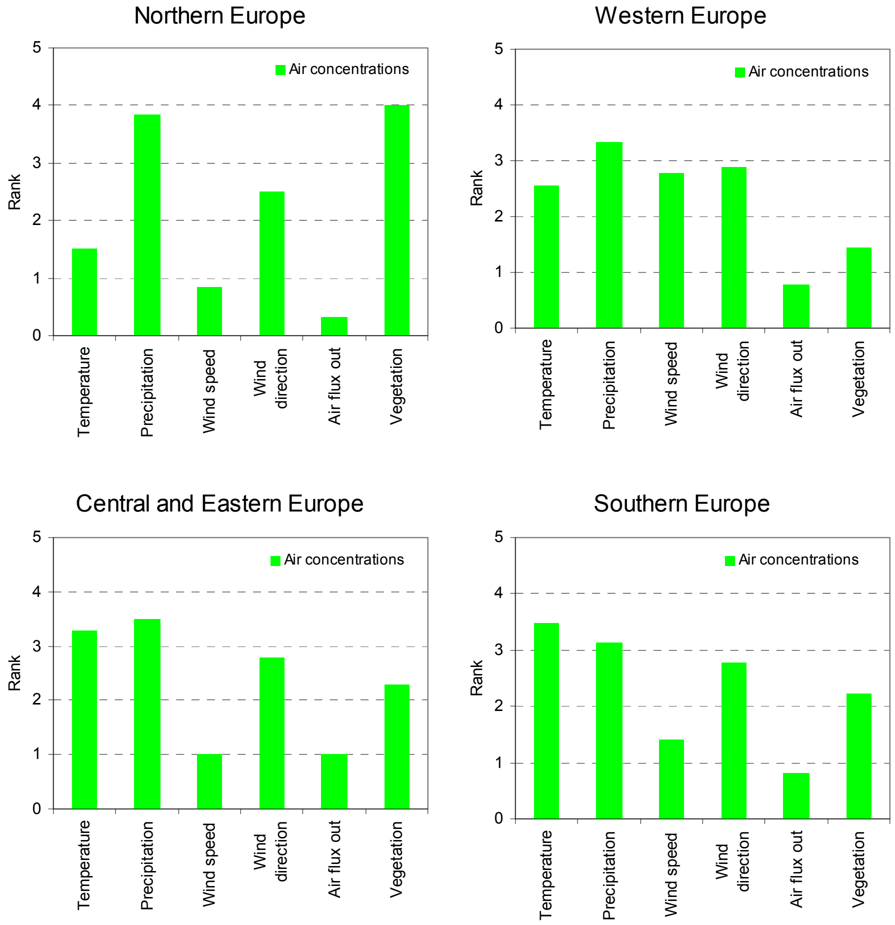
Central and Eastern Europe

Southern Europe

Fig. 5.2. *Ranking of meteorological and environmental factors for B[a]P air concentrations over the European countries with different geographical locations*

It is seen that in all considered groups of the European countries temperature and wind direction are the two main factors. The factor of the third priority is precipitation amount (for Northern, Western and Southern Europe) and vegetation (for Central and Eastern Europe). It can be noted that vegetation cover has relatively higher priority for the countries of two groups: Central and Eastern Europe and Southern Europe comparing to other countries.

The results of factors ranking for PCB-153 air concentrations for the above country groups are displayed in Fig. 5.3.



Northern Europe

Western Europe

Central and Eastern Europe

Southern Europe

Fig. 5.3. *Ranking of meteorological and environmental factors for PCB-153 air concentrations over the European countries with different geographical locations*

In case of PCB-153 the variance of ranking in different groups of countries is higher comparing to that of B[a]P. Thus, the temperature is a factor of the first priority for the countries of Southern Europe. For Northern Europe the factor of the first priority is vegetation followed by the precipitation amount. In case of Western, Central, and Eastern Europe the precipitation amount is the factor of the first priority while the second and third priorities belong to temperature, wind direction, and wind speed.

Ranking of factors for individual European countries. The analysis of ranking on the level of individual European countries reveals that relative importance of the factors can also essentially vary from country to country within the considered above groups. Particularly, the differences in ranking are connected not only with the geographic location of countries, but also with the influence of differences in vegetation cover, closeness to water bodies, the value of the country area, etc. To illustrate this, the aggregation of countries into the groups by the similar ranking pattern was performed (see Technical Report [*Shatalov et al.*, 2011] for details). Results of this aggregation for B[a]P and PCB-153 air concentrations are shown in Figs. 5.4 and 5.5.

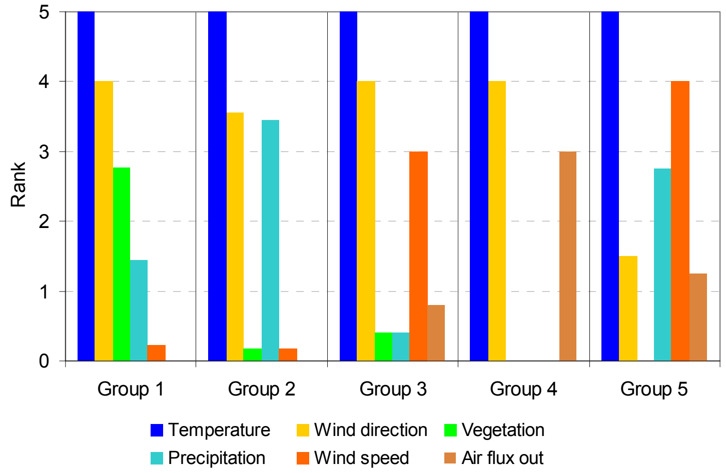


Fig. 5.4. *Typical patterns of ranking of meteorological and environmental factors for B[a]P air concentrations over Europe*

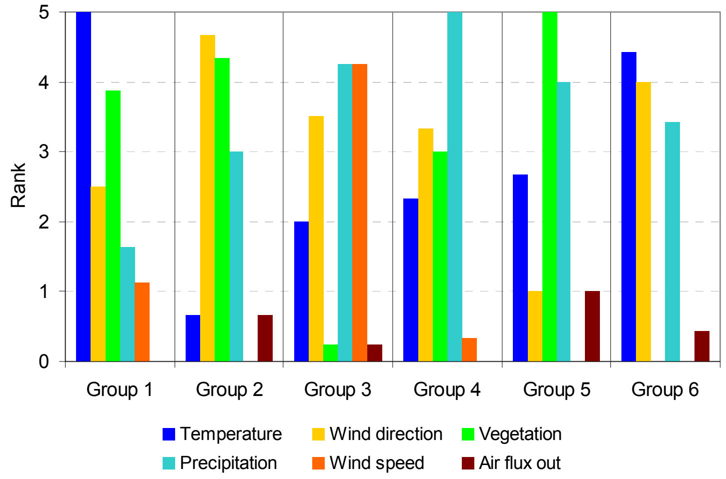


Fig. 5.5. *Typical patterns of ranking of meteorological and environmental factors for PCB-153 air concentrations over Europe*

In general, the distribution of the factors within these groups is similar to that obtained above (Figs. 5.2 and 5.3). At the same time, the groups with typical ranking pattern can contain the countries located in different parts of Europe. For instance, in case of B[a]P, the second group includes such countries as Finland, Germany, Portugal, and Poland, and in case of PCB-153 the first group includes Turkey, Poland, and Romania.

As seen from Fig. 5.4 for B[a]P the most important factor is temperature followed by the wind direction in most of cases. Precipitation amount is relatively important for three patterns from five and vegetation cover is a factor of the third priority in one of the patterns. Additionally, two of the patterns show relatively high importance of the wind speed.

Contrary to B[a]P, temperature is the first important factor only in two typical patterns from six in case of PCB-153 (Fig. 5.5). Precipitation amount is among three most important factors in five typical patterns. Wind direction and vegetation are among the three major factors in most of typical patterns.

These results show that the sensitivity of POP pollution levels to variations of meteorological and environmental parameters is different in different parts of Europe which can be also reflected in varied response to the climatic changes across Europe. In particular, the analysis of future variability and extremes in temperature and precipitation, performed using high resolution modelling [*Dankers and Hiederer*, 2008], shows that future changes might not be uniform in Europe. Elevated winter temperatures are characteristic of Eastern Europe and in the Alps, while higher summer temperatures mostly affect Southern Europe. Among the summer months the August can have the largest increase in extreme summer temperatures and the occurrence of heat waves. The changes in precipitation can be very different between the southern and northern parts of Europe. For Southern Europe the annual precipitation is projected to decrease, along with higher risk of longer dry periods, while for Northern Europe the precipitation is generally increasing, particularly in winter. Thus, for B[a]P with high sensitivity of its air concentrations to temperature variations projected changes in climate characteristics for Southern Europe (extreme summer temperatures, heat waves, and dry periods) might lead to the increase of pollution. Similarly, for PCBs these changes can lead to more intense volatilization and long-range transport.

Further analysis of links between climate change and POP behaviour. Due to the complex character of POP cycling in the environment the analysis of future changes in POP pollution levels requires experimental simulations on the basis of multimedia POP transport models and climate change scenarios.

Working in this direction the MSC-E has started preparation of necessary modelling tools and collection of input information. It is planned to perform experimental model simulations of POP fate using climate change scenarios data and the global scale multimedia modelling system GLEMOS being developed at the MSC-E. For preparation of meteorological input to drive GLEMOS the global advanced research WRF model is applied [*Travnikov and Jonson*, 2011].

Input information on meteorological parameters representing future climate conditions will be obtained on the basis of the output of global climate models (GCMs) provided by WMO-WCRP CMIP3 multi-model database. Several datasets generated by GCMs on the basis of the scenarios of future greenhouse gases emissions (e.g. SRES B2, A1B, A2) can be selected, which represent different extent of future climate changes. Some of these scenarios were used in a number of other modelling studies (e.g. [*Lamon et al.*, 2009b]). To analyze the effect of future climatic changes base-case model simulations applying the 20C3M scenario, characterizing the state of the climate during the recent several decades, will be carried out. It should be noted that, in order to reduce uncertainties, it would be reasonable to use the model output of several global climate models which showed good agreement with measurements while simulating climate variability during several recent decades.

It is planned to perform a series of modelling experiments with the GLEMOS model to explore both the effect of future changes of emissions using available projections, and the influence of projected climate changes on POP long-range transport, source-receptor relationships, strength of secondary emission sources, and distribution of POPs in media. It is reasonable to examine the response of pollution levels to the climate-induced changes for several POPs, for instance, PAHs, PCDD/Fs, and PCBs, which differ by their physical-chemical properties and behaviour in the environment and for which projections of future emission levels are available.

Concluding remarks

- Presented analysis shows that considered list of meteorological and environmental factors, including temperature, precipitation amount, wind speed and direction, outflow of air masses through the country boundaries, and vegetation cover, is in most cases sufficient for explaining 90% – 95% of seasonal variability of chemicals air concentrations for a country.
- Due to the differences in physical-chemical properties the effect of variations of meteorological and environmental factors is different for the considered POPs. In particular, variability of temperature and wind direction has the highest priority for the B[a]P and similar pollutants. For PCB-153, with longer atmospheric half-life and more intensive inter-media cycling, the precipitation, temperature and wind direction are the factors of major priority.
- Geographical location of a country alone does not determine the ranking of meteorological and environmental factors. It can be additionally affected by vegetation cover, closeness to water bodies, the value of the country area, etc.
- Presented approach can be applied also to the analysis of model simulations of POP fate based on scenarios of future climate changes. It is planned to perform a series of modelling experiments to explore both the effect of future changes of emissions, and the influence of projected climate changes on POP fate and behaviour.

6. COOPERATION

6.1. Helsinki Commission (HELCOM)

In accordance with the Memorandum of Understanding between the Baltic Marine Environment Protection Commission (HELCOM) and the United Nations Economic Commission for Europe on cooperation in the field of monitoring of air pollutants EMEP Centres prepared an annual joint report on the assessment of airborne pollution load to the Baltic Sea [*Bartnicki et al*., 2010]. MSC-E contributed to the report with the evaluation of atmospheric input of dioxins and furans to the Baltic Sea. Officially reported emission data on PCDD/Fs to EMEP for 1990-2008 were used in model simulations. Modelling results on PCDD/F deposition to the Baltic Sea along with the contributions of surrounding countries were presented. The report was welcomed and endorsed by the Contracting Parties at the HELCOM MONAS 13 meeting in October 2010 and was recommended to be published on the EMEP and HELCOM websites.

Along with the joint report, the indicator fact sheets with updated information on the temporal variations of PCDD/F emissions to air and their deposition over the Baltic Sea in the period from 1990 to 2008 were prepared. These indicator fact sheets are available in the Internet at the HELCOM web site [*www.helcom.fi*].

Annual emissions of dioxins and furans in HELCOM countries have decreased from 1990 to 2008 by 22% (Fig. 6.1). The most significant drop of PCDD/F emissions can be seen in Finland (58%) and Denmark (54%). Some decrease of emission can also be noted for Lithuania (46%), Sweden (37%), Germany (33%), Poland (25%), Russia (15%), and Estonia (11%). For some of the HELCOM countries the level of PCDD/F emissions in 2008 is higher than emission of 1990. In particular, Latvia reported higher values of emissions for 2008 in comparison with the emissions for 1990 which is most likely connected with more essential uncertainties of emission estimates for earlier years. In 2008 the total annual PCDD/F emissions of HELCOM countries amounted to 1.4 kg I-TEQ. Among the HELCOM countries the largest contributions to the total annual PCDD/F emission of HELCOM countries

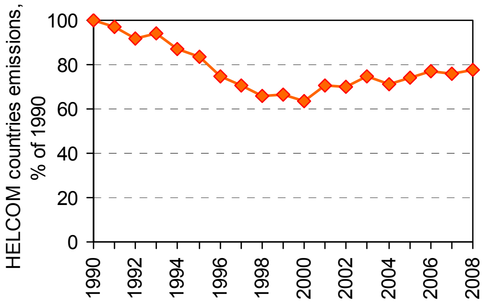


Fig. 6.1. *Trend of dioxins and furans emissions from HELCOM countries in 1990-2008 according to official emissions data*

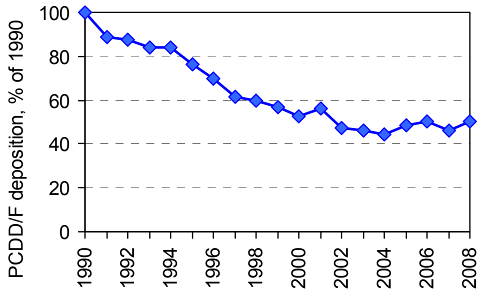


Fig. 6.2. *Temporal variations of PCDD/F deposition to the Baltic Sea in 1990-2008*

belong to Russia (58%) followed by Poland (28%) and Germany (5%).

The level of annual atmospheric deposition of PCDD/Fs to the Baltic Sea has decreased from 1990 to 2008 by 50% (Fig. 6.2). The most significant change in PCDD/F atmospheric deposition can be noted for the Kattegat (57%) and the Baltic Proper (54%). For other sub-basins the decrease of deposition varies from 28% to 50% (Fig. 6.3). The highest level of PCDD/F atmospheric deposition fluxes (0.43 ng I-TEQ/m^2/y) over the Baltic Sea in 2008 can be seen in its southern-western part (the Belt Sea) while the lowest one (0.07 ng I-TEQ/m^2/y) over the Gulf of Bothnia. In other sub-basins the level of deposition fluxes varies from about 0.10 to 0.23 ng I-TEQ/m^2/y. Among the HELCOM countries the most essential contributions to deposition over the Baltic Sea belong to Poland and Denmark.

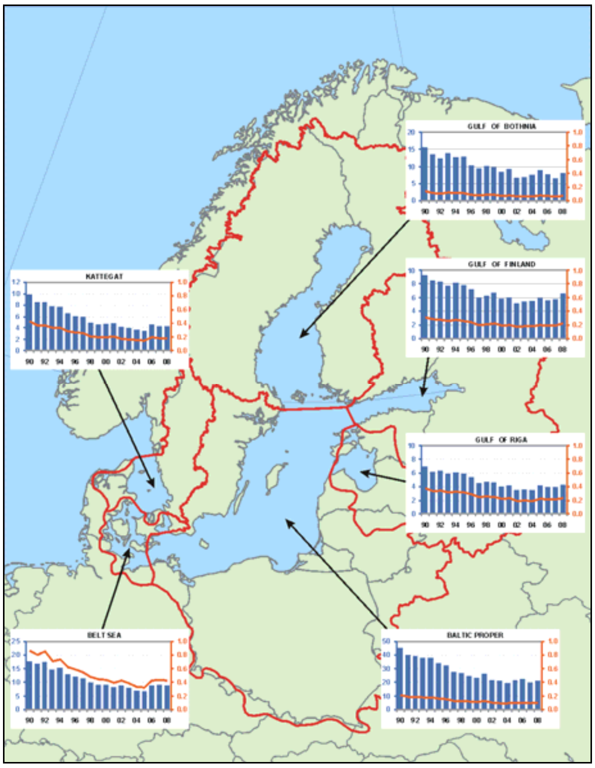


Fig. 6.3. *Computed annual deposition of PCDD/Fs to the six sub-basins of the Baltic Sea for the period 1990-2008 in t/y as bars (left axis) and deposition fluxes in ng I-TEQ/m^2/y as lines (right axis)*

6.2. Arctic Monitoring and Assessment Program (AMAP)

MSC-E continues cooperating with the Arctic Monitoring and Assessment Program (AMAP). This year the Centre participated in the work of the AMAP scientific conference "The Arctic as a Messenger for Global Processes - Climate Change and Pollution" held on 4-6 May 2011 as well as in recent meeting of EU ArcRisk project and took part in the discussion of the topics related to the linkages between the climate change and POP pollution. This conference represented the most comprehensive compilation of scientific knowledge on the effects of climate change on the Arctic.

Main scope of the conference was the climate change phenomena, its global features, and recent studies of changes in Arctic, namely accelerating warming and melting of land and sea ice. Besides, the recent AMAP/UNEP report on climate change and POPs was presented. The report highlighted key scientific findings with regard to the links between the climate change and POPs. Particularly, it is concluded that expected climate changes has the potential to increase the vulnerability of the planet to the pollution by POPs. Foreseen climatic changes may affect POP fate through the increasing of temperature, altering of atmospheric and oceanic transport pathways, melting of sea and land ice, and increasing of frequency and strength of extreme events.

Along with that updated information on the pollution of the Arctic by legacy and new POPs compiled in the recent AMAP Assessment of POP pollution was demonstrated. Special attention was paid to the temporal and spatial trends of new POPs important for the Arctic, namely, per- and polyfluorinated compounds and their degradation products (PFSAs and PFCAs), current use pesticides (PCP, trifluralin, endosulfan), and brominated flame retardants (PBDEs, HBCD).

6.3. Task Force on Hemispheric Air Pollution (TF HTAP)

MSC-E continued to co-operate with the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) and took part in the discussion of further activities on POPs within TF HTAP and development of working plan for next years during the Task Force meeting in Arona, Italy in May 2011.

Particularly, the Centre presented an overview of available data and ongoing EMEP activities aimed at development of new emission inventories, extension of monitoring networks and application of chemical transport models for the assessment of environment pollution by POPs on a global scale. Besides, the importance of elaboration and application of the multi-media modelling approach to the assessment of POP cycling and accumulation in the environment was stressed. The assessment could be the most effectively performed within the framework of integrated analysis involving combined use of emissions, monitoring and modelling aspects.

The Task Force decided to continue its work on the assessment of intercontinental pollution by selected POPs and agreed that its work would benefit from close cooperation with on-going activities within relevant projects and international programmes (EU ArcRisk project, UNEP, AMAP, Stockholm Convention etc.)

6.4. Contribution to Development of Local-scale Modelling in Italy

Cooperation with national experts on environmental pollution is one of the important directions of the activity of MSC-E. This year in framework of collaboration with the ENEA institute of Italy a set of model simulations of transport and fate of PCDD/Fs, PCBs, PAHs, HCB, and γ-HCH was carried out. Three-dimensional air concentrations with spatial resolution of 50x50 km and temporal resolution of 6 hours for the agreed area were prepared. Information produced by MSC-E was used by the national modelling system MINNI as the initial and boundary conditions for modelling of POP pollution in Italy with fine resolution. Examples of the spatial distribution of air concentrations for selected POPs over Italy are shown in Fig. 6.4.

Modelling results obtained by the MSCE-POP model were compared with available measurements of EMEP monitoring network and data of various POP monitoring campaigns carried out in Italy. Particularly, modelling results on PCBs were compared with the observations made at the Ispra site in 2005-2006 [*Castro-Jimenez et al.*, 2008; *Castro-Jimenez et al.*, 2009]. Examples of the comparison results for PCB-118 and PCB-153 are given in Fig.6.5.

In general, for most of the POPs, considered in this study, levels of concentrations provided by the MSCE-POP model reasonably agreed with measurements. At the same time, for HCB and γ-HCH larger deviations were found between modelled and observed concentrations which indicated necessity of further work on the refinement of information on their emissions as within Italy and in surrounding countries.

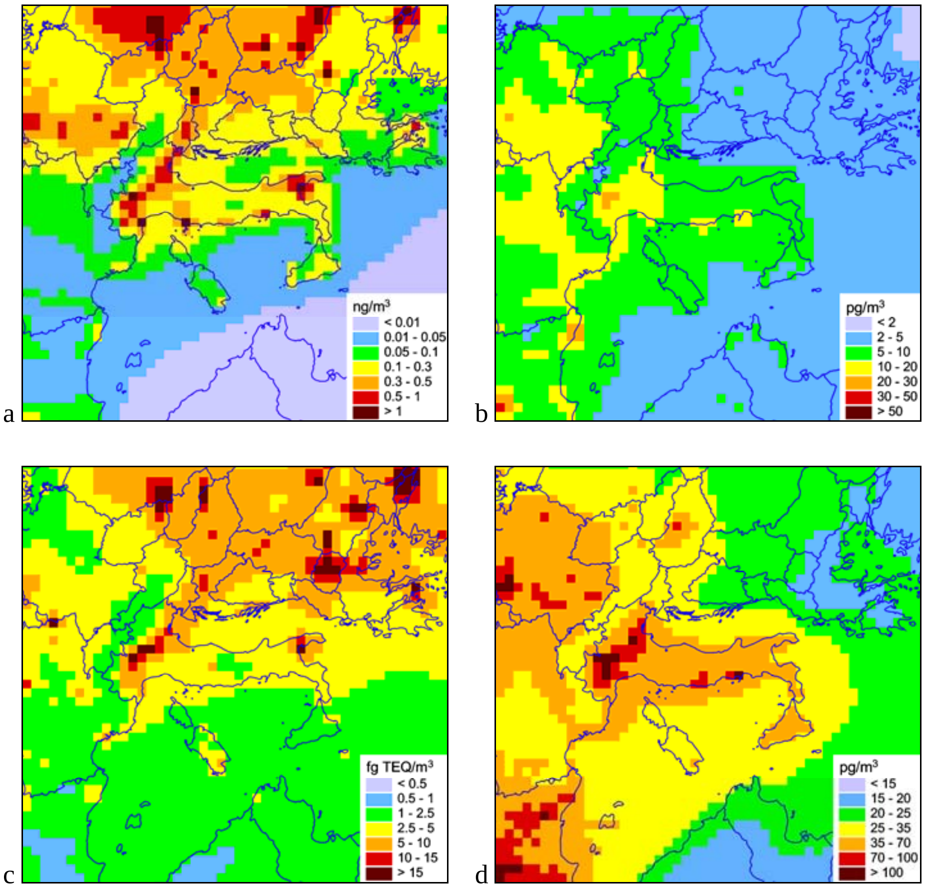


Fig. 6.4. *Spatial distribution of B[a]P (a), PCB-153 (b), PCDD/Fs (c), and HCB (d) air concentrations over Italy for 2005*

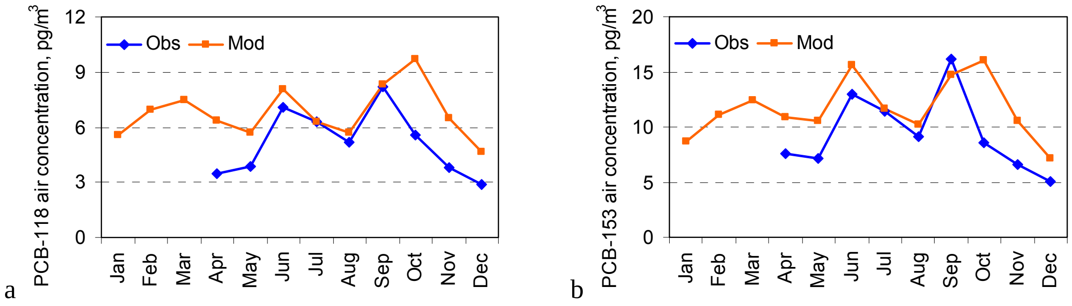


Fig. 6.5. *Comparison of modelled seasonal variations of PCB-118 (a) and PCB-153 (b) with measurements performed at the Ispra site in Italy for 2005, pg/m^3*

6.5. Evaluation of contamination of Baltic Sea region by PCDD/Fs

This year the work on evaluation of contamination in the Baltic region by PCDD/Fs was undertaken in the co-operation with experts from Umeå and Stockholm Universities (Sweden). In the framework of this activity, model assessment of transport and accumulation of four particular PCDD/F congeners was carried out. The comparison of these calculations with congener-specific measurements of PCDD/F air concentrations allowed estimating the uncertainties in PCDD/F emission congener composition. The results of these investigations are described above in Chapter 3.

7. FUTURE ACTIVTIES

In order to further improve evaluation of POP pollution of the EMEP region the following activities of MSC-E and CCC are proposed for 2012/2013:

Ongoing activities

- Review, store and make available these monitoring data for the modelling Centres and Parties;
- Publish the validated annual data and contribute to preparation, review and assessments of observation data presented in the series of EMEP reports;
- Provide training/guidance to Parties to establish monitoring activities in compliance with the EMEP monitoring strategy;
- Arrange laboratory intercomparisons for main components, heavy metals and elemental carbon/organic carbon (EC/OC); and use the results from the EMEP and the Arctic Monitoring and Assessment Programme (AMAP) 2010 laboratory intercomparison of POPs together with field intercomparison between passive and active sampling to assess the uncertainties in the POP measurements;
- Address global scale integration of quality assessment/quality control (QA/QC) activities of regional monitoring programmes, including standards for metadata provision and intercomparisons (in collaboration with the Task Force on Hemispheric Transport of Air Pollution);
- Maintain close interaction with relevant organizations and bodies in relation to integration of observations, including monitoring efforts under other Convention bodies (e.g., the ICPs and national monitoring obligations to European Commission Directives, as well as activities undertaken by EEA, WMO, the OSPAR Commission, the Baltic Marine Environment Protection Commission (HELCOM), the United Nations Environment Programme (UNEP), AMAP, Nitrogen in Europe(NinE), GMES/GEOSS and others;
- Evaluate of air concentrations, deposition fluxes and transboundary transport of POPs (PAHs, PCDD/Fs, and HCB) for 2010-2011;
- Calculate POP dispersion on a global scale with the help of global EMEP model (GLEMOS) for the evaluation of initial and boundary conditions and contributions of intercontinental transport to pollution levels in the EMEP domain and in remote regions (the Arctic) with spatial resolution 1°x 1°;
- Perform model assessment of transboundary pollution within the EMEP region by POPs for 2010-2011 including contamination of marginal seas with spatial resolution 50km x 50km;
- Prepare input data required for global/regional/local modelling (emission, meteorological, and geophysical data);

- Contribute to the effect community work with information on ecosystem-dependent deposition fluxes of POPs to different land use types to support evaluation of the pollutants adverse effect on human health and the environment;
- Support countries with information required for air quality management in and implementation of the CLRTAP Protocols on POPs with special emphasis to EECCA countries;
- Cooperate with the CLRTAP subsidiary bodies (WGSR, WGE), EMEP task forces (TFHTAP, TFMM), and relevant international organizations.

New (research and development) activities

Monitoring of POPs:

- Improve the EMEP database to include more statistical opportunities for aggregated data, further develop the plotting routines and develop improved export routines for data download for modellers;
- Evaluate the one year campaign data POPs from Kazakhstan and Moldova to assess the relative importance of the different pollutants and the main source in this region;
- Explore the use of passive POP measurements used in among others the MONET (RECETOX) project o validate the EMEP model and other transport models to evaluate source contribution;
- Evaluate new measurements data of POPs from Eastern Europe, the Caucasus and Central Asia to assess the relative importance of the different pollutants and the main source regions;

Global/regional/local modelling:

- Further develop the multimedia GLEMOS modelling framework including improvement of the multi-media approach and refinement of the pollutant specific processes;
- Incorporate data on aerosols and atmospheric reactants based on external datasets or simplified chemical modules for improving evaluation of POP pollution levels;
- Further develop and test the integrated monitoring/modelling/emission approach for POPs including the adjoint modelling;

EMEP Case Study:

- Initiate model assessment of POP pollution on a country scale;

Climate change impact on POP long-range transport and fate:

- Evaluate sensitivity of POP contamination to variation of meteorological parameters, atmospheric constituents (aerosols, reactants), and environmental factors influenced by climate change;
- Perform modelling of climate change effects on POP transport and fate for selected periods using the climate change scenarios data.

CONCLUSIONS

This Status Report presents the progress in the evaluation of pollution levels and transboundary transport of POPs within the EMEP region and at the hemispheric/global scales achieved in 2011. The main conclusions of the work carried out by CCC and MSC-E are summarized below.

Development of Integrated Approach for Assessment of POP Pollution

- In accordance with the recommendations of HTAP Assessment Report 2010, MSC-E continued the elaboration of integrated monitoring/modelling/emission approach to the evaluation of environmental contamination by toxic substances. The approach is aimed at the improvement of quality of pollution assessment and reducing uncertainties of monitoring data, emission inventories, and model predictions. It includes several steps, namely, initial assessment of pollution, evaluation of agreement between measurement data and model predictions, analysis of discrepancies, and refined assessment.

- Statistical indicators used for analysis of the agreement between measurement data and model predictions were reviewed and tested in application to the evaluation of modelled POP pollution levels. Additional indicators based on the theory of statistical hypothesis testing were introduced. Dependencies between considered indicators were examined and the set of indicators for the analysis of the agreement of modelling results with measurements for POPs was proposed.

- In cases when the level of discrepancies between the model predictions and measurements exceeds a factor of 2 – 3, developed integrated approach presumes the application of complex analysis of emission data, monitoring data, and modelling approach applied in the assessment of pollution. This analysis is intended to determine the reasons of elevated discrepancies and to recommend further steps to reduce the uncertainties and thus improve pollution assessment.

- To analyze the uncertainties of emission data consideration of conventional emission scenarios can be performed. Some approaches for the evaluation of emission scenarios based on the backward trajectory and matrix approaches were developed and tested. The effect of the use of constructed emission scenarios is evaluated using the set of statistical indicators. The application of these tools was demonstrated for the evaluation of environmental contamination by PAHs, PCDD/Fs and HCB.

Assessment of POP Pollution Levels, Transboundary Transport, and Trends

- Evaluation of PAHs, PCDD/Fs and HCB pollution levels and transboundary transport in the EMEP region for 2009 was carried out on the basis of EMEP measurements, emission data, and modelling of POP long-range transport and fate.

- The number of EMEP sites measuring POPs within the EMEP region was increased in 2009 by three additional sites comparing to previous year. Thus, in 2009 the EMEP monitoring network contained 23 sites for POPs among which thirteen sites performed POP measurements in both compartments (air and deposition). Additional sites in Moldova and Kazakhstan started air and aerosol measurements of key POPs (PAHs, PCBs, organochlorine pesticides) from June 2009. At the same time, the spatial coverage of the EMEP monitoring network for POPs still requires further improvement, especially in south – southeast of Europe to fulfil the goal of the EMEP monitoring strategy.

- Emission datasets for the assessment of POP pollution within the EMEP region were prepared on the basis of official data submitted by the EMEP Centre on Emission Inventories and Projections (CEIP) and non-Party expert estimates. For the evaluation of intercontinental transport gridded emissions of PCDD/Fs, PCB-153, and HCB for Northern Hemisphere were constructed.

- Assessment of POP pollution levels was performed using the developed integrated approach. The level of agreement between the modelling results and measurements was examined and the discrepancies found were analyzed. Several conventional scenarios of emissions were constructed for the analysis of emission data uncertainty. Additionally the sensitivity of the pollution levels to the emissions of particular countries was evaluated and preliminary recommendations for further refinement of assessment of pollution levels within the EMEP region were formulated.

- Further improvement of contamination assessment can be achieved by the refinement of emission and monitoring data for all considered pollutants in the course of specific case studies with application of fine resolution modelling and detailed monitoring of POP concentrations in cooperation with national experts in the assessment of pollution.

Polycyclic Aromatic Hydrocarbons (PAHs)

- Evaluation of PAH pollution within the EMEP region for 2009 was performed for the four indicator PAHs (B[a]P), B[b]F, B[k]F, and IP). According to available measurements and model calculations, highest concentrations levels among the considered four PAH species were found for B[b]F and IP. Slightly lower levels of contamination were characteristic of B[a]P. The lowest levels of pollution were obtained for B[k]F.

- For all the considered PAHs, the areas with high contamination levels were characteristic of Central and Eastern Europe, Portugal and western part of Spain. Moderate and low PAH pollution levels were indicated for the Scandinavian Peninsula, the UK and France. Spatial distributions of contamination are different for the considered PAHs.

- Spatial distribution of annual mean B[a]P air concentrations changed differently from 2008 to 2009 increasing or decreasing in some areas of the EMEP region up to 0.7 ng/m^3. Particularly, increase of B[a]P air concentrations was noted for Portugal, Spain, Lithuania, Latvia and Estonia, while levels of air concentrations in the Ukraine, Bulgaria, Italy, and France slightly decreased. These differences were mostly conditioned by the changes in emissions for particular countries and changes in meteorological conditions for the considered two years.

- For most of the sites measuring B[a]P in air the difference between the modelled and observed air concentrations ranged from 10 to 30%. Higher discrepancies were found for the sites DE1, NO42, ES8, and PL5. Particularly, differences between the measured and computed air concentrations for DE1 and NO42 were accounted for about 70%. Observed concentrations provided by ES8 were in most of the cases below the detection limit.

- In case of Polish site PL5 the underestimation of observed B[a]P air concentrations by the model exceeded a factor of 3 which may be conditioned by the uncertainties of emission spatial distribution in the region surrounding the site. More thorough analysis of pollution levels in this region applying fine resolution modelling and detailed monitoring of PAH concentrations is required.

- On the basis of model simulations transboundary transport of B[a]P in the form of import and export of total annual depositions was estimated. Following these results transboundary transport of B[a]P was a significant source of pollution for most of European countries contributing about 30 - 70% to their annual total deposition. For 25 countries in 2009 its contribution exceeded 50%. The export of pollution for European countries ranged typically from 30% to 60%.

- The analysis of trends of B[a]P contamination in period from 1990 to 2009 was carried out. Total B[a]P emissions of EMEP countries decreased in this period by about 30%. Similar rate of decline was obtained for annual mean B[a]P air concentrations. This indicates that for particle-bound PAHs levels of air concentrations are determined rather by current emissions than by re-emission of earlier accumulated contaminant from the underlying surface.

Polychlorinated Dibenzo(p)dioxins and Dibenzofurans (PCDD/Fs)

- Evaluation of PCDD/F pollution levels for 2009 was performed using overall toxicity of 17 toxic PCDD/F congeners. In the model simulations physical-chemical properties of the “indicator” congener 2,3,4,7,8-PeCDF were used. Additionally, model calculations of pollution levels of four PCDD/F congeners (2,3,4,7,8-PeCDF, 1,2,3,7,8,9-HxCDD, OCDD and OCDF) for 2006 and 2007 were performed in co-operation between Umeå University of Sweden and MSC-E. These calculations allowed evaluating possible uncertainties in congener composition of emission data for PCDD/Fs.

- Modelling results for individual PCDD/F congeners were compared with measurements made at Aspvreten (SE12), Pallas (FI96), and Vindeln (SE35) monitoring sites in 2006-2007 put at MSC-E disposal by Umeå University. Results of the comparison revealed underestimation of measured air concentrations by the model about a factor of 5 for 2,3,4,7,8-PeCDF. Larger differences (10 times and higher) were obtained for 1,2,3,7,8,9-HxCDD, OCDD, and OCDF congeners. Different level of underestimation for different congeners manifested that there were essential uncertainties in determination of congener composition of PCDD/F emissions. Therefore the data on congener composition of dioxins and furans emissions in European countries are highly appreciated.

- The analysis of the agreement between the modelling results and measurements showed possible discrepancies in spatial distribution of PCDD/F emissions. In particular, for all considered congeners maximum underestimation at the site SE12 was associated with atmospheric flows from south-south-west and south-south-east directions. In case of other directions the underestimation of observed concentrations by the model was much lower, accounting, in example, for a factor of 2.7 for 2,3,4,7,8-PeCDF congener.

- The analysis of the agreement between air concentrations of PCDD/F mixture calculated for 2009 with available measurements at the sites Råö (SE14) and Aspvreten (SE12) showed the underestimation of observed pollution levels by the model about 5 times, which corresponded to the results obtained in the investigation for selected congeners. Examination of discrepancies between the modelling results and measurements indicated necessity of the refinement of emission spatial distribution and organization of additional monitoring campaigns for PCDD/Fs.

- Taking into account essential discrepancies between the modelling results and measurements of PCDD/Fs, evaluation of PCDD/F transboundary transport was described using the export of pollution by the countries. The export fractions do not depend on the emission inventories. It was obtained that the fraction of PCDD/F deposition, originated from countries emission sources and occurred outside their boundaries, varied typically from 30% to 60% and exceeded 50% for 14 countries.

Hexachlorobenzene (HCB)

- Evaluation of HCB pollution within the EMEP region for 2009 was performed using available official emission data and expert estimates. Analysis of modelling results using available measurements of HCB air concentrations revealed that modelling results significantly underestimated observed pollution levels. The underestimation can be related to the incompleteness of available officially submitted emission data and expert estimates as well as with the underestimation of the role of secondary emission sources.

- Re-emission of HCB from environmental compartments can essentially contribute to the contemporary pollution levels. To evaluate HCB re-emissions elaboration of scenarios of historical HCB emissions is required. Underestimation of the influence of historical emissions (and, as a consequence, of re-emission) was additionally substantiated by the comparison of modelled HCB concentrations in soil with measurements. Particularly, it was shown that the model simulations based on the official emission data and expert estimates led to essentially lower levels of HCB soil concentrations compared to available measurements.

- Essential improvement of the agreement between calculated and measured HCB air concentrations can be achieved by changing contribution of re-emission and emission totals of EMEP countries using conventional emission scenarios. Thus, further analysis of contemporary and historical emissions is needed to refine the assessment of HCB pollution levels.

Development of Global Modelling Framework for POPs

Further improvement of the description of global scale POP cycling was carried out in framework of development of global POP modelling system GLEMOS. Particularly, additional processes describing POP fate in seawater were included, namely, POP transport with sea currents, diffusion, degradation, and sedimentation. The implemented advection and diffusion scheme was tested. Experimental model simulations of PCB-153 global scale transport were performed with spatial resolution 1°×1°. Further improvement of POP global scale modelling system GLEMOS will include incorporation of vegetation compartment and its interaction with the atmosphere and soil.

Inter-linkages between Climate Change and POP Pollution

- In order to improve understanding POP transport and fate as well as their links with climate variability the analysis of sensitivity of POP pollution levels to variation of meteorological and environmental factors was carried out. It was shown that such factors as temperature, precipitation amount, wind speed and direction, outflow of air masses through the country boundaries, and vegetation cover, can in most cases sufficient for explaining 90% – 95% of seasonal variability of chemicals air concentrations for a country.
- Due to the differences in physical-chemical properties the effect of variations of meteorological and environmental factors differs depending on particular group of POPs. In particular, variability of temperature and wind direction has the most high priority for the B[a]P and similar pollutants. For PCB-153, with longer atmospheric half-life and more intensive inter-media cycling, the precipitation, temperature and wind direction are the factors of major importance.
- The sensitivity of POP pollution levels to variations of meteorological and environmental parameters is different in different parts of Europe which can be also reflected in varied response to the climatic changes across Europe. Geographical location of a country alone does not determine the ranking of meteorological and environmental factors. It can be additionally affected by vegetation cover, closeness to water bodies, the value of the country area, etc.
- Presented approach can be applied to the analysis of model simulations of POP fate based on scenarios of future climate changes. It is planned to perform a series of modelling experiments to explore both the effect of future changes of emissions, and the influence of projected climate changes on POP fate and behaviour.

Cooperation

The work on the assessment of POP pollution including evaluation of long-range transport and fate, improvement of global scale modelling and development of integrated approach was performed in close collaboration with CLRTAP subsidiary bodies (TFMM, TF HTAP), international organizations (AMAP, EU, HELCOM, OSPAR, UNEP, WMO, etc) and national experts.

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ANNEX A

COUNTRY-TO-COUNTRY DEPOSITION MATRICES FOR 2009

***Table A.1**. Codes of countries*

Country/Region/Sea	Code	Country/Region/Sea	Code
Albania	AL	Monaco	MC
Armenia	AM	Montenegro	ME
Austria	AT	Netherlands	NL
Azerbaijan	AZ	Norway	NO
Belarus	BY	Poland	PL
Belgium	BE	Portugal	PT
Bosnia and Herzegovina	BA	Republic of Moldova	MD
Bulgaria	BG	Romania	RO
Croatia	HR	Russian Federation (European part)	RU
Cyprus	CY	Russian Federation (Asian part)	RUA
Czech Republic	CZ	Serbia	RS
Denmark	DK	Slovakia	SK
Estonia	EE	Slovenia	SI
Finland	FI	Spain	ES
France	FR	Sweden	SE
Georgia	GE	Switzerland	CH
Germany	DE	The Former Yugoslav Republic of Macedonia	MK
Greece	GR	Tajikistan	TJ
Hungary	HU	Turkey	TR
Iceland	IS	Turkmenistan	TM
Ireland	IE	Ukraine	UA
Italy	IT	United Kingdom	GB
Kazakhstan	KZ	Uzbekistan	UZ
Kyrgyzstan	KY	Baltic Sea	BAS
Latvia	LV	Black Sea	BLS
Lithuania	LT	Caspian Sea	CAS
Luxembourg	LU	North Sea	NOS
Malta	MT	Mediterranean Sea	MDT

***Table A.2**. Matrix of B[a]P country-to-country deposition in 2009, kg/y*

Receptors ↓ Emitters ⟶

	AL	AM	AT	AZ	BA	BE	BG	BY	CH	CY	CZ	DE	DK	
AL	**445.6**	0.00	0.57	0.05	4.19	0.17	18.48	0.41	0.02	0.00	1.17	2.34	0.24	**AL**
AM	0.01	**0.01**	0.01	70.58	0.01	0.01	0.06	0.10	0.00	0.01	0.04	0.10	0.02	**AM**
AT	1.87	0.00	**500.7**	0.06	10.75	15.63	4.80	5.62	1.83	0.00	108.61	429.39	8.17	**AT**
AZ	0.02	0.00	0.05	**934.4**	0.03	0.04	0.18	0.49	0.00	0.01	0.18	0.43	0.07	**AZ**
BA	22.34	0.00	9.58	0.07	**729.1**	1.89	17.93	2.19	0.09	0.00	16.00	26.78	2.00	**BA**
BE	0.04	0.00	0.81	0.01	0.15	**712.2**	0.08	1.11	0.19	0.00	2.04	118.49	2.98	**BE**
BG	11.88	0.00	2.52	0.60	6.86	1.10	**2040.67**	4.35	0.05	0.07	7.60	13.63	1.54	**BG**
BY	2.42	0.00	6.82	1.55	4.92	6.48	14.71	**3138.21**	0.18	0.02	29.61	71.70	16.95	**BY**
CH	0.18	0.00	8.91	0.01	0.96	8.87	0.33	0.76	**22.78**	0.00	2.71	87.80	1.38	**CH**
CY	0.01	0.00	0.01	0.00	0.01	0.00	0.05	0.02	0.00	**10.42**	0.01	0.03	0.01	**CY**
CZ	1.27	0.00	70.20	0.06	5.48	16.82	5.61	9.14	0.81	0.00	**974.45**	402.03	12.78	**CZ**
DE	1.10	0.00	99.56	0.15	3.82	304.37	4.55	23.06	14.03	0.00	191.82	**8405.76**	111.81	**DE**
DK	0.12	0.00	1.61	0.02	0.38	13.41	0.57	4.16	0.11	0.00	6.64	109.41	**593.29**	**DK**
EE	0.38	0.00	1.04	0.21	1.03	3.34	1.05	48.84	0.05	0.00	3.85	22.31	8.97	**EE**
ES	0.33	0.00	2.54	0.06	1.77	4.48	0.86	3.75	0.28	0.00	4.96	33.89	2.56	**ES**
FI	0.59	0.00	3.81	1.03	2.38	9.23	2.20	106.29	0.16	0.01	13.01	61.63	25.64	**FI**
FR	1.19	0.00	12.36	0.11	5.47	148.77	2.17	10.95	8.15	0.00	20.30	419.94	13.70	**FR**
GB	0.32	0.00	1.83	0.05	0.77	32.51	1.22	5.63	0.22	0.00	5.04	65.74	13.65	**GB**
GE	0.08	0.00	0.12	73.06	0.10	0.11	0.64	1.22	0.00	0.04	0.41	1.03	0.19	**GE**
GR	36.76	0.00	1.04	0.47	3.92	0.38	83.70	2.03	0.03	0.06	2.77	5.07	0.73	**GR**
HR	9.34	0.00	23.10	0.07	180.16	2.23	12.12	3.01	0.12	0.00	20.31	36.19	1.79	**HR**
HU	7.42	0.00	52.98	0.11	39.14	5.32	32.05	8.30	0.24	0.01	59.21	79.02	4.68	**HU**
IE	0.06	0.00	0.35	0.01	0.18	3.56	0.24	1.09	0.04	0.00	0.94	12.38	2.83	**IE**
IS	0.05	0.00	0.59	0.04	0.15	4.67	0.34	3.34	0.04	0.00	2.53	22.77	9.47	**IS**
IT	17.50	0.00	49.31	0.12	49.15	7.27	18.19	5.10	3.15	0.02	20.74	110.17	4.28	**IT**
KY	0.00	0.00	0.00	0.49	0.00	0.01	0.01	0.04	0.00	0.00	0.01	0.06	0.01	**KY**
KZ	0.32	0.00	1.60	19.32	0.71	2.36	2.52	21.05	0.06	0.02	5.45	19.23	4.37	**KZ**
LT	0.75	0.00	2.79	0.26	1.89	3.54	2.53	202.41	0.10	0.00	11.30	40.27	13.33	**LT**
LU	0.00	0.00	0.11	0.00	0.02	6.02	0.01	0.10	0.03	0.00	0.28	17.11	0.21	**LU**
LV	0.81	0.00	2.27	0.33	1.88	4.48	2.46	139.43	0.10	0.00	8.74	42.79	16.49	**LV**
MC	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	**MC**
MD	0.59	0.00	0.47	0.19	0.82	0.35	5.00	5.32	0.01	0.01	1.79	3.96	0.65	**MD**
ME	57.76	0.00	0.69	0.02	27.44	0.21	8.52	0.29	0.01	0.00	1.39	2.74	0.27	**ME**
MK	46.79	0.00	0.47	0.07	2.22	0.16	77.78	0.47	0.01	0.01	1.28	2.15	0.27	**MK**
MT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	**MT**
NL	0.06	0.00	1.04	0.01	0.18	229.16	0.17	1.24	0.11	0.00	3.09	202.61	4.97	**NL**
NO	0.65	0.00	5.94	0.31	2.03	35.49	3.06	46.44	0.39	0.01	26.35	215.82	154.68	**NO**
PL	5.10	0.00	44.86	0.52	15.70	31.07	20.62	282.74	1.06	0.01	433.66	516.94	82.11	**PL**
PT	0.02	0.00	0.22	0.01	0.09	0.68	0.07	0.60	0.02	0.00	0.53	4.24	0.41	**PT**
RO	18.28	0.00	12.25	0.85	35.71	4.83	190.50	19.82	0.23	0.09	30.25	59.93	5.90	**RO**
RS	51.81	0.00	6.97	0.18	83.64	2.10	164.01	2.86	0.08	0.01	16.71	27.71	2.71	**RS**
RU+ RUA	7.86	0.00	28.32	117.72	18.45	55.16	51.60	1003.60	1.23	0.27	100.63	422.79	109.43	**RU+ RUA**
SE	0.89	0.00	9.26	0.86	2.74	31.16	3.96	112.01	0.51	0.01	40.47	233.44	233.04	**SE**
SI	1.75	0.00	41.12	0.02	14.27	1.03	3.09	1.52	0.08	0.00	8.57	22.62	0.89	**SI**
SK	2.75	0.00	26.02	0.08	10.62	4.39	12.99	10.57	0.18	0.00	122.45	62.20	4.92	**SK**
TJ	0.00	0.00	0.00	0.37	0.00	0.00	0.01	0.01	0.00	0.00	0.01	0.02	0.00	**TJ**
TM	0.01	0.00	0.08	5.56	0.02	0.09	0.05	0.98	0.00	0.00	0.31	0.81	0.16	**TM**
TR	3.46	0.00	2.22	22.51	4.00	1.25	47.34	8.00	0.06	4.82	6.10	14.09	1.67	**TR**
UA	8.28	0.00	14.08	8.30	18.37	8.78	65.49	363.11	0.31	0.15	51.98	104.04	18.93	**UA**
UZ	0.01	0.00	0.09	2.59	0.03	0.09	0.07	1.01	0.00	0.00	0.34	0.79	0.18	**UZ**
	AL	**AM**	**AT**	**AZ**	**BA**	**BE**	**BG**	**BY**	**CH**	**CY**	**CZ**	**DE**	**DK**	

***Table A.2**. Matrix of B[a]P country-to-country deposition in 2009, kg/y (continued)*

Receptors ↓ Emitters ⟶

	EE	ES	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	KY	
AL	0.16	0.67	0.09	0.56	0.12	0.01	46.08	1.60	2.89	0.04	0.00	94.08	0.00	**AL**
AM	0.01	0.01	0.00	0.01	0.01	3.07	0.05	0.01	0.05	0.00	0.00	0.07	0.02	**AM**
AT	1.78	4.17	1.65	10.32	5.45	0.01	2.21	22.10	87.50	1.49	0.00	136.77	0.00	**AT**
AZ	0.05	0.04	0.03	0.04	0.03	5.32	0.14	0.03	0.19	0.01	0.00	0.26	0.24	**AZ**
BA	0.96	1.78	0.66	1.84	0.84	0.01	9.50	100.80	60.92	0.23	0.00	167.60	0.00	**BA**
BE	0.50	6.23	0.27	36.07	13.55	0.00	0.04	0.18	0.46	2.47	0.00	3.71	0.00	**BE**
BG	1.33	0.88	0.66	1.03	0.48	0.12	85.31	3.61	19.57	0.11	0.00	39.13	0.01	**BG**
BY	45.33	2.82	17.08	3.80	3.42	0.26	5.23	4.32	28.82	0.82	0.01	22.98	0.03	**BY**
CH	0.19	6.18	0.16	33.40	3.41	0.00	0.19	1.34	1.67	0.95	0.00	141.03	0.00	**CH**
CY	0.00	0.01	0.00	0.01	0.00	0.00	0.18	0.01	0.03	0.00	0.00	0.14	0.00	**CY**
CZ	2.84	3.87	2.57	8.50	6.56	0.01	1.67	8.45	66.54	1.50	0.01	34.73	0.00	**CZ**
DE	9.75	41.92	7.33	146.96	59.43	0.02	1.84	5.11	24.69	12.71	0.04	105.62	0.01	**DE**
DK	2.23	4.00	1.42	5.32	8.40	0.00	0.17	0.40	2.26	1.88	0.01	3.58	0.00	**DK**
EE	**906.31**	1.58	38.59	1.81	1.65	0.03	0.53	0.87	3.76	0.43	0.00	5.92	0.01	**EE**
ES	0.82	**4694.7**	0.42	51.38	3.53	0.01	0.29	2.12	3.86	1.91	0.02	30.41	0.00	**ES**
FI	226.74	6.40	**1483.2**	5.59	7.67	0.10	1.11	2.62	11.05	2.18	0.04	15.94	0.08	**FI**
FR	2.96	223.73	1.92	**1605.2**	40.41	0.01	1.02	7.04	11.14	11.54	0.04	220.20	0.01	**FR**
GB	2.76	24.49	2.39	26.41	**765.35**	0.01	0.41	0.67	3.00	91.89	0.07	8.05	0.01	**GB**
GE	0.15	0.08	0.08	0.08	0.08	**66.95**	0.54	0.09	0.54	0.02	0.00	0.75	0.02	**GE**
GR	0.66	1.33	0.34	0.98	0.22	0.10	**1270.5**	2.03	6.29	0.07	0.00	66.22	0.01	**GR**
HR	0.71	2.22	0.54	2.44	1.00	0.01	6.16	**453.8**	141.23	0.29	0.00	173.72	0.00	**HR**
HU	2.31	1.80	1.49	3.22	2.25	0.02	11.10	75.96	**1664.1**	0.58	0.00	79.71	0.00	**HU**
IE	0.53	6.07	0.59	3.48	28.83	0.00	0.07	0.16	0.68	**342.73**	0.03	1.90	0.00	**IE**
IS	2.08	3.63	2.53	2.56	7.37	0.00	0.11	0.15	1.10	3.59	**8.52**	1.24	0.02	**IS**
IT	1.34	23.81	1.01	36.21	3.17	0.02	21.95	62.85	51.67	0.94	0.00	**7218.8**	0.00	**IT**
KY	0.01	0.01	0.01	0.01	0.01	0.01	0.03	0.00	0.01	0.00	0.00	0.06	**849.4**	**KY**
KZ	6.53	1.73	5.67	1.65	1.53	0.66	1.10	0.68	5.09	0.44	0.01	4.83	233.12	**KZ**
LT	25.76	1.68	9.80	2.31	1.95	0.04	0.94	1.67	9.10	0.48	0.00	9.29	0.00	**LT**
LU	0.03	0.60	0.02	4.41	0.47	0.00	0.01	0.02	0.05	0.11	0.00	0.48	0.00	**LU**
LV	143.45	2.21	21.66	2.80	2.42	0.05	1.03	1.58	7.55	0.62	0.00	10.27	0.01	**LV**
MC	0.00	0.00	0.00	0.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.00	**MC**
MD	0.77	0.24	0.41	0.27	0.21	0.04	2.05	0.57	2.86	0.04	0.00	4.75	0.00	**MD**
ME	0.15	0.43	0.09	0.35	0.11	0.00	7.75	2.86	3.96	0.04	0.00	74.83	0.00	**ME**
MK	0.19	0.32	0.10	0.28	0.08	0.01	73.81	0.98	3.32	0.02	0.00	28.34	0.00	**MK**
MT	0.00	0.01	0.00	0.01	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.14	0.00	**MT**
NL	0.80	5.57	0.41	14.63	14.22	0.00	0.07	0.18	0.74	2.46	0.01	2.91	0.00	**NL**
NO	42.30	18.54	55.46	17.55	40.67	0.05	1.40	2.15	11.36	11.23	0.13	16.33	0.04	**NO**
PL	33.98	11.46	20.46	17.90	14.12	0.09	7.26	19.59	134.89	3.31	0.02	80.69	0.02	**PL**
PT	0.18	130.55	0.11	1.36	0.49	0.00	0.02	0.11	0.27	0.28	0.01	1.49	0.00	**PT**
RO	4.93	3.17	2.63	3.89	2.42	0.17	43.23	20.05	154.31	0.50	0.00	109.04	0.02	**RO**
RS	1.40	1.45	0.82	1.68	0.93	0.03	42.56	30.85	82.43	0.22	0.00	110.07	0.00	**RS**
RU+ RUA	561.55	34.66	478.81	35.49	33.96	16.18	24.15	17.33	89.50	9.39	0.17	112.88	25.95	**RU+ RUA**
SE	129.49	15.14	229.46	17.19	24.72	0.13	1.94	3.70	20.18	6.80	0.08	26.41	0.05	**SE**
SI	0.29	1.07	0.23	1.35	0.45	0.00	1.52	74.60	42.42	0.14	0.00	115.98	0.00	**SI**
SK	2.58	1.38	1.57	2.38	1.97	0.01	4.47	14.99	223.63	0.44	0.00	35.77	0.00	**SK**
TJ	0.00	0.01	0.00	0.00	0.00	0.01	0.02	0.00	0.01	0.00	0.00	0.04	36.41	**TJ**
TM	0.11	0.05	0.07	0.06	0.06	0.12	0.03	0.03	0.22	0.02	0.00	0.20	2.06	**TM**
TR	1.76	1.67	0.91	1.53	0.71	4.99	97.78	2.52	11.70	0.19	0.00	33.98	0.04	**TR**
UA	23.14	5.09	12.11	6.22	5.21	1.87	28.18	15.64	120.19	1.24	0.01	79.95	0.13	**UA**
UZ	0.13	0.06	0.10	0.06	0.06	0.07	0.06	0.03	0.28	0.02	0.00	0.23	134.96	**UZ**
	EE	**ES**	**FI**	**FR**	**GB**	**GE**	**GR**	**HR**	**HU**	**IE**	**IS**	**IT**	**KY**	

***Table A.2**. Matrix of B[a]P country-to-country deposition in 2009, kg/y (continued)*

Receptors ↓ Emitters ⟶

	KZ	LT	LU	LV	MC	MD	ME	MK	MT	NL	NO	PL	PT	
AL	0.05	0.15	0.02	0.38	0.00	0.23	55.58	32.44	0.02	0.06	0.01	5.69	0.54	**AL**
AM	0.17	0.02	0.00	0.04	0.00	0.02	0.01	0.01	0.00	0.00	0.00	0.31	0.01	**AM**
AT	0.10	2.06	2.30	3.85	0.00	0.42	3.20	0.88	0.00	5.47	0.40	147.19	4.38	**AT**
AZ	3.19	0.08	0.00	0.16	0.00	0.09	0.02	0.02	0.00	0.02	0.01	1.45	0.05	**AZ**
BA	0.11	0.89	0.20	2.23	0.00	0.64	115.24	4.29	0.02	0.70	0.11	51.41	1.38	**BA**
BE	0.03	0.64	17.52	1.54	0.00	0.02	0.06	0.01	0.00	30.28	0.21	11.97	6.87	**BE**
BG	0.72	1.39	0.11	3.28	0.00	6.47	8.28	33.42	0.01	0.44	0.09	46.48	0.92	**BG**
BY	3.11	188.49	0.51	210.10	0.00	10.78	3.31	1.81	0.00	2.92	1.02	666.71	3.67	**BY**
CH	0.02	0.31	1.62	0.60	0.00	0.04	0.31	0.07	0.00	2.01	0.10	10.93	5.94	**CH**
CY	0.00	0.01	0.00	0.01	0.00	0.01	0.01	0.01	0.00	0.00	0.00	0.10	0.01	**CY**
CZ	0.14	3.76	1.83	6.81	0.00	0.50	2.13	0.88	0.00	6.74	0.64	637.83	4.67	**CZ**
DE	0.40	13.18	56.51	29.21	0.00	0.84	1.62	0.66	0.00	156.74	3.85	652.89	47.20	**DE**
DK	0.07	2.43	0.85	5.65	0.00	0.15	0.19	0.07	0.00	7.32	2.51	54.28	5.57	**DK**
EE	0.61	27.09	0.22	285.24	0.00	0.66	0.60	0.20	0.00	1.35	0.68	65.36	2.47	**EE**
ES	0.12	1.48	0.72	2.78	0.00	0.18	0.59	0.14	0.00	1.20	0.21	30.91	2023.9	**ES**
FI	3.87	50.37	0.59	211.84	0.00	1.50	1.00	0.31	0.00	3.75	5.33	185.68	7.91	**FI**
FR	0.24	4.74	41.44	9.31	0.01	0.33	2.02	0.42	0.00	20.02	1.07	98.83	169.13	**FR**
GB	0.23	2.52	1.54	6.56	0.00	0.12	0.48	0.19	0.00	9.06	2.45	35.45	44.73	**GB**
GE	0.53	0.26	0.01	0.48	0.00	0.28	0.07	0.06	0.00	0.04	0.01	3.69	0.09	**GE**
GR	0.43	0.62	0.04	1.58	0.00	1.91	4.66	40.52	0.05	0.15	0.05	17.54	1.12	**GR**
HR	0.10	0.87	0.26	1.69	0.00	0.61	18.63	2.57	0.01	0.84	0.10	60.91	1.63	**HR**
HU	0.19	2.87	0.61	5.95	0.00	1.95	13.67	5.10	0.01	2.00	0.27	228.18	1.65	**HU**
IE	0.08	0.47	0.20	1.23	0.00	0.02	0.11	0.04	0.00	1.33	0.40	6.43	9.22	**IE**
IS	0.35	1.79	0.27	4.74	0.00	0.19	0.09	0.04	0.00	1.88	2.25	25.44	10.41	**IS**
IT	0.17	1.73	1.07	3.45	0.00	0.80	23.41	4.80	0.20	2.39	0.25	74.79	15.42	**IT**
KY	141.57	0.01	0.00	0.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.13	0.02	**KY**
KZ	**4469.9**	5.12	0.19	12.43	0.00	1.52	0.42	0.23	0.00	0.97	0.47	58.56	3.14	**KZ**
LT	0.53	**868.20**	0.32	307.47	0.00	1.48	1.12	0.43	0.00	1.52	0.68	292.56	2.18	**LT**
LU	0.00	0.06	**26.92**	0.13	0.00	0.00	0.01	0.00	0.00	0.45	0.01	1.36	0.61	**LU**
LV	0.79	184.32	0.38	**2182.35**	0.00	1.35	1.20	0.45	0.00	1.84	0.97	170.02	3.36	**LV**
MC	0.00	0.00	0.00	0.00	**0.00**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	**MC**
MD	0.52	1.17	0.03	2.04	0.00	**167.01**	0.59	0.48	0.00	0.16	0.04	23.81	0.28	**MD**
ME	0.03	0.14	0.02	0.37	0.00	0.13	**448.45**	5.31	0.01	0.08	0.02	6.23	0.31	**ME**
MK	0.06	0.17	0.02	0.43	0.00	0.36	5.35	**215.75**	0.01	0.06	0.02	6.39	0.28	**MK**
MT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	**0.06**	0.00	0.00	0.01	0.00	**MT**
NL	0.03	0.68	2.19	1.91	0.00	0.03	0.09	0.03	0.00	**207.98**	0.33	14.59	6.71	**NL**
NO	1.46	31.00	2.37	93.94	0.00	1.31	0.97	0.41	0.00	14.88	**221.51**	313.19	19.90	**NO**
PL	1.21	96.29	2.87	110.55	0.00	5.97	7.55	3.25	0.01	14.74	3.17	**11636.7**	14.13	**PL**
PT	0.03	0.26	0.10	0.53	0.00	0.02	0.03	0.01	0.00	0.19	0.03	3.59	**11198.4**	**PT**
RO	1.83	5.85	0.53	13.12	0.00	71.55	24.22	15.22	0.01	1.85	0.34	218.38	2.94	**RO**
RS	0.23	1.44	0.22	3.51	0.00	1.65	148.61	53.08	0.01	0.80	0.14	68.67	1.15	**RS**
RU+ RUA	1542.99	265.40	4.23	854.23	0.00	25.89	10.79	5.55	0.01	21.84	14.46	1361.36	50.58	**RU+ RUA**
SE	2.50	74.25	1.96	248.45	0.00	2.28	1.26	0.54	0.00	14.01	32.37	568.37	21.39	**SE**
SI	0.04	0.45	0.13	0.73	0.00	0.18	2.97	0.60	0.00	0.40	0.04	28.08	0.80	**SI**
SK	0.14	3.77	0.45	7.04	0.00	0.98	4.39	2.02	0.00	1.77	0.29	524.65	1.50	**SK**
TJ	10.62	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.06	0.01	**TJ**
TM	17.38	0.19	0.01	0.35	0.00	0.08	0.01	0.00	0.00	0.04	0.01	3.26	0.07	**TM**
TR	1.58	2.48	0.12	5.32	0.00	6.47	3.13	2.95	0.03	0.50	0.11	53.30	1.47	**TR**
UA	16.28	41.52	0.78	69.09	0.00	96.67	10.43	6.64	0.01	3.91	1.24	982.26	6.12	**UA**
UZ	135.99	0.20	0.01	0.36	0.00	0.08	0.01	0.01	0.00	0.04	0.01	3.57	0.07	**UZ**
	KZ	**LT**	**LU**	**LV**	**MC**	**MD**	**ME**	**MK**	**MT**	**NL**	**NO**	**PL**	**PT**	

***Table A.2**. Matrix of B[a]P country-to-country deposition in 2009, kg/y (continued)*

Receptors↓ Emitters ⟶

	RO	RS	RU+ RUA	SE	SI	SK	TJ	TM	TR	UA	UZ	Total	
AL	19.09	42.18	0.36	0.11	0.79	1.58	0.00	0.00	6.35	13.03	0.00	**798.2**	**AL**
AM	0.66	0.02	0.47	0.01	0.01	0.05	0.01	0.03	25.71	4.29	0.02	**106.1**	**AM**
AT	26.25	9.85	1.55	2.51	119.22	47.20	0.00	0.00	2.40	34.75	0.00	**1774.8**	**AT**
AZ	2.24	0.08	7.65	0.04	0.04	0.24	0.13	0.50	19.17	22.22	0.27	**999.9**	**AZ**
BA	95.41	94.91	1.15	0.80	13.28	19.49	0.00	0.00	5.62	31.52	0.00	**1612.5**	**BA**
BE	0.70	0.12	0.35	0.72	0.33	0.65	0.00	0.00	0.13	3.00	0.00	**976.7**	**BE**
BG	869.32	78.20	5.34	0.74	2.51	12.91	0.00	0.01	154.55	247.46	0.01	**3715.8**	**BG**
BY	201.34	10.29	88.07	11.40	5.48	39.37	0.01	0.03	35.39	1427.64	0.04	**6340.0**	**BY**
CH	2.52	0.67	0.23	0.34	3.08	1.37	0.00	0.00	0.23	3.66	0.00	**357.3**	**CH**
CY	0.27	0.02	0.02	0.00	0.01	0.02	0.00	0.00	5.09	0.62	0.00	**17.2**	**CY**
CZ	36.60	9.27	2.46	4.12	15.44	150.29	0.00	0.00	2.08	37.66	0.00	**2559.8**	**CZ**
DE	36.16	5.36	7.21	18.34	10.24	31.40	0.00	0.00	3.59	69.86	0.00	**10720.7**	**DE**
DK	6.12	0.62	1.40	9.34	0.60	3.37	0.00	0.00	0.38	12.86	0.00	**873.2**	**DK**
EE	16.70	1.59	30.52	11.06	1.07	4.12	0.00	0.00	3.09	70.33	0.01	**1575.5**	**EE**
ES	9.46	1.45	0.89	0.79	3.79	3.33	0.00	0.00	0.33	14.07	0.00	**6941.3**	**ES**
FI	35.66	3.11	155.01	133.00	3.74	12.59	0.04	0.03	8.32	183.65	0.04	**2996.1**	**FI**
FR	18.09	3.98	2.73	4.05	13.81	10.62	0.00	0.00	1.62	35.02	0.00	**3205.8**	**FR**
GB	8.38	1.23	2.82	4.26	1.01	3.23	0.00	0.00	0.56	11.88	0.00	**1189.2**	**GB**
GE	8.52	0.25	12.62	0.10	0.10	0.58	0.01	0.05	83.63	57.12	0.03	**314.9**	**GE**
GR	98.82	18.37	2.94	0.38	1.33	4.17	0.00	0.01	146.62	134.25	0.01	**1961.2**	**GR**
HR	70.08	56.85	1.08	0.70	139.72	28.83	0.00	0.00	4.27	35.55	0.00	**1495.9**	**HR**
HU	427.57	99.59	2.78	1.86	56.90	315.18	0.00	0.00	12.27	144.91	0.00	**3454.5**	**HU**
IE	1.71	0.29	0.75	0.85	0.23	0.68	0.00	0.00	0.14	2.33	0.00	**433.3**	**IE**
IS	4.98	0.29	3.15	3.95	0.26	1.66	0.01	0.00	0.49	15.00	0.00	**154.1**	**IS**
IT	78.36	28.90	1.91	1.50	136.76	24.17	0.00	0.00	16.52	49.85	0.00	**8173.3**	**IT**
KY	0.11	0.01	0.23	0.01	0.01	0.02	88.80	0.25	0.34	1.07	40.13	**1123.0**	**KY**
KZ	37.91	1.46	237.71	3.71	0.90	6.53	57.41	3.68	20.52	372.07	43.57	**5678.5**	**KZ**
LT	37.15	3.32	20.58	10.63	2.16	11.11	0.00	0.00	4.46	162.61	0.01	**2070.7**	**LT**
LU	0.09	0.01	0.03	0.05	0.04	0.07	0.00	0.00	0.01	0.31	0.00	**60.3**	**LU**
LV	35.16	3.22	31.00	15.38	1.98	8.68	0.00	0.01	5.21	153.10	0.01	**3214.2**	**LV**
MC	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	**0.2**	**MC**
MD	376.21	1.98	4.46	0.34	0.59	2.85	0.00	0.00	17.52	330.87	0.00	**962.3**	**MD**
ME	16.90	49.06	0.22	0.11	0.84	1.80	0.00	0.00	2.37	6.41	0.00	**728.7**	**ME**
MK	28.05	46.83	0.44	0.11	0.47	1.90	0.00	0.00	13.01	16.64	0.00	**575.5**	**MK**
MT	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	**0.3**	**MT**
NL	1.28	0.25	0.47	1.05	0.29	0.88	0.00	0.00	0.16	3.34	0.00	**726.9**	**NL**
NO	40.65	3.16	29.24	144.47	3.50	17.05	0.02	0.01	6.00	138.43	0.01	**1791.8**	**NO**
PL	254.02	27.36	35.95	29.88	30.81	354.96	0.01	0.01	16.18	582.63	0.01	**15006.4**	**PL**
PT	0.70	0.10	0.19	0.15	0.21	0.29	0.00	0.00	0.05	1.54	0.00	**11348.2**	**PT**
RO	**11656.3**	168.03	17.64	2.58	16.34	68.32	0.01	0.02	154.37	875.82	0.02	**14038.2**	**RO**
RS	432.92	**1127.3**	2.07	1.11	7.29	26.60	0.00	0.00	22.32	59.69	0.00	**2590.1**	**RS**
RU+ RUA	593.37	33.02	**8225.7**	155.22	23.00	106.68	7.75	2.52	368.86	7626.50	6.22	**24663.2**	**RU+ RUA**
SE	57.63	4.08	68.49	**1496.9**	6.46	29.69	0.02	0.02	11.48	289.31	0.03	**4075.1**	**SE**
SI	16.98	7.61	0.46	0.34	**586.1**	12.52	0.00	0.00	1.22	13.65	0.00	**1006.3**	**SI**
SK	133.47	21.49	2.44	1.91	20.66	**1029.7**	0.00	0.00	5.50	102.80	0.00	**2411.3**	**SK**
TJ	0.05	0.00	0.11	0.00	0.00	0.01	**428.61**	0.63	0.25	0.53	16.86	**494.7**	**TJ**
TM	1.11	0.04	2.41	0.08	0.05	0.34	13.89	**31.36**	1.59	26.22	17.79	**127.4**	**TM**
TR	270.69	12.85	19.23	0.92	2.20	9.08	0.02	0.07	**9749.1**	521.68	0.05	**10936.6**	**TR**
UA	1249.79	38.57	221.09	10.45	17.16	118.79	0.06	0.15	262.15	**33842.6**	0.18	**37956.7**	**UA**
UZ	1.43	0.05	3.30	0.10	0.04	0.44	179.60	6.39	1.31	24.57	**127.64**	**626.5**	**UZ**
	RO	RS	RU+ RUA	SE	SI	SK	TJ	TM	TR	UA	UZ	Total	

***Table A.3**. Matrix of PCDD/F country-to-country deposition in 2009, g TEQ/y*

Receptors ↓ Emitters ⟶

	AL	AM	AT	AZ	BA	BE	BG	BY	CH	CY	CZ	DE	DK	
AL	**12.36**	0.00	0.04	0.01	0.21	0.01	0.88	0.01	0.01	0.00	0.12	0.03	0.01	**AL**
AM	0.01	**1.17**	0.004	2.27	0.01	0.002	0.03	0.002	0.001	0.00	0.01	0.004	0.002	**AM**
AT	0.07	0.00	**15.51**	0.01	0.26	0.30	0.20	0.03	0.39	0.00	3.43	1.12	0.10	**AT**
AZ	0.01	0.20	0.01	**44.87**	0.01	0.01	0.07	0.01	0.004	0.00	0.04	0.01	0.01	**AZ**
BA	0.48	0.00	0.27	0.01	**23.56**	0.05	0.62	0.01	0.02	0.00	0.77	0.14	0.04	**BA**
BE	0.00	0.00	0.02	0.001	0.01	**18.31**	0.01	0.004	0.04	0.00	0.07	0.75	0.04	**BE**
BG	0.38	0.002	0.13	0.06	0.31	0.04	**105.3**	0.03	0.02	0.002	0.56	0.10	0.03	**BG**
BY	0.06	0.002	0.16	0.10	0.12	0.11	0.58	**21.01**	0.03	0.00	1.52	0.22	0.22	**BY**
CH	0.02	0.00	0.30	0.002	0.07	0.17	0.05	0.01	**6.13**	0.00	0.12	0.32	0.02	**CH**
CY	0.01	0.00	0.00	0.004	0.01	0.002	0.04	0.001	0.001	**0.18**	0.01	0.003	0.001	**CY**
CZ	0.04	0.00	1.83	0.01	0.13	0.28	0.17	0.04	0.12	0.00	**42.41**	1.09	0.15	**CZ**
DE	0.05	0.00	2.48	0.02	0.12	5.28	0.19	0.10	2.40	0.00	4.10	**34.32**	1.35	**DE**
DK	0.01	0.00	0.03	0.004	0.01	0.22	0.02	0.02	0.02	0.00	0.20	0.35	**6.29**	**DK**
EE	0.01	0.00	0.02	0.02	0.02	0.06	0.04	0.18	0.01	0.00	0.14	0.08	0.09	**EE**
ES	0.05	0.00	0.18	0.01	0.17	0.35	0.11	0.03	0.19	0.00	0.37	0.43	0.10	**ES**
FI	0.03	0.002	0.08	0.10	0.07	0.19	0.17	0.37	0.03	0.00	0.50	0.20	0.25	**FI**
FR	0.11	0.00	0.55	0.02	0.33	4.74	0.25	0.07	2.17	0.00	1.11	3.17	0.31	**FR**
GB	0.02	0.00	0.09	0.01	0.06	1.18	0.09	0.05	0.10	0.00	0.37	0.53	0.33	**GB**
GE	0.02	0.14	0.02	3.28	0.03	0.01	0.18	0.02	0.01	0.002	0.08	0.02	0.01	**GE**
GR	1.28	0.002	0.13	0.05	0.36	0.05	10.45	0.03	0.03	0.003	0.41	0.11	0.03	**GR**
HR	0.25	0.00	0.65	0.01	4.96	0.05	0.42	0.02	0.03	0.00	1.19	0.19	0.04	**HR**
HU	0.10	0.00	1.31	0.01	0.79	0.09	0.61	0.05	0.04	0.00	3.26	0.22	0.07	**HU**
IE	0.00	0.00	0.02	0.003	0.01	0.16	0.02	0.01	0.02	0.00	0.07	0.10	0.06	**IE**
IS	0.00	0.00	0.02	0.01	0.01	0.13	0.02	0.01	0.01	0.00	0.09	0.10	0.09	**IS**
IT	0.93	0.00	1.57	0.03	2.09	0.24	1.28	0.04	0.78	0.001	1.49	0.67	0.12	**IT**
KY	0.01	0.01	0.01	0.24	0.01	0.01	0.03	0.004	0.003	0.00	0.02	0.01	0.003	**KY**
KZ	0.09	0.04	0.17	2.63	0.16	0.18	0.66	0.30	0.06	0.003	0.83	0.24	0.15	**KZ**
LT	0.02	0.00	0.07	0.02	0.04	0.06	0.09	0.88	0.02	0.00	0.46	0.13	0.19	**LT**
LU	0.00	0.00	0.003	0.00	0.001	0.18	0.001	0.00	0.01	0.00	0.01	0.12	0.004	**LU**
LV	0.01	0.00	0.05	0.02	0.04	0.07	0.08	0.57	0.02	0.00	0.32	0.14	0.18	**LV**
MC	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	**MC**
MD	0.02	0.001	0.02	0.03	0.03	0.01	0.34	0.04	0.004	0.00	0.15	0.02	0.01	**MD**
ME	1.12	0.00	0.03	0.00	0.79	0.01	0.28	0.00	0.01	0.00	0.10	0.02	0.01	**ME**
MK	1.23	0.00	0.04	0.01	0.12	0.01	2.87	0.01	0.01	0.00	0.14	0.03	0.01	**MK**
MT	0.00	0.00	0.00	0.00	0.001	0.00	0.001	0.00	0.00	0.00	0.001	0.00	0.00	**MT**
NL	0.00	0.00	0.02	0.002	0.01	5.08	0.01	0.01	0.02	0.00	0.10	1.46	0.09	**NL**
NO	0.05	0.001	0.15	0.05	0.09	0.84	0.21	0.17	0.09	0.00	0.95	0.92	1.65	**NO**
PL	0.10	0.001	0.93	0.05	0.30	0.50	0.56	1.18	0.15	0.00	31.45	1.70	1.09	**PL**
PT	0.00	0.00	0.02	0.002	0.01	0.05	0.01	0.004	0.02	0.00	0.04	0.05	0.01	**PT**
RO	0.36	0.003	0.32	0.11	0.83	0.10	7.48	0.12	0.05	0.002	1.63	0.20	0.08	**RO**
RS	1.18	0.00	0.24	0.02	2.47	0.06	3.43	0.02	0.02	0.00	1.02	0.14	0.05	**RS**
RU	0.40	0.12	0.74	8.68	0.71	1.01	3.85	4.84	0.28	0.01	4.36	1.39	1.16	**RU**
RUA	0.10	0.03	0.19	1.22	0.18	0.35	0.81	0.49	0.08	0.002	0.91	0.41	0.31	**RUA**
SE	0.05	0.002	0.23	0.09	0.11	0.68	0.27	0.42	0.10	0.001	1.72	0.84	3.18	**SE**
SI	0.04	0.00	1.05	0.003	0.30	0.02	0.11	0.01	0.02	0.00	0.52	0.08	0.01	**SI**
SK	0.05	0.00	0.61	0.01	0.22	0.07	0.32	0.05	0.03	0.00	11.54	0.17	0.07	**SK**
TJ	0.00	0.004	0.004	0.15	0.01	0.003	0.02	0.002	0.002	0.00	0.01	0.003	0.001	**TJ**
TM	0.01	0.02	0.02	1.79	0.02	0.02	0.09	0.02	0.01	0.001	0.10	0.03	0.01	**TM**
TR	0.50	0.31	0.30	1.33	0.57	0.16	9.49	0.13	0.10	0.07	1.06	0.27	0.10	**TR**
UA	0.23	0.01	0.42	0.67	0.49	0.21	3.36	2.32	0.09	0.003	3.66	0.42	0.30	**UA**
UZ	0.01	0.01	0.02	0.67	0.02	0.02	0.07	0.03	0.01	0.001	0.10	0.03	0.01	**UZ**
	AL	**AM**	**AT**	**AZ**	**BA**	**BE**	**BG**	**BY**	**CH**	**CY**	**CZ**	**DE**	**DK**	

***Table A.3**. Matrix of PCDD/F country-to-country deposition in 2009, g TEQ/y (continued)*

Receptors ↓ Emitters ⟶

	EE	ES	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	KY	
AL	0.001	0.06	0.001	0.04	0.04	0.02	0.74	0.23	0.11	0.003	0.00	1.64	0.00	**AL**
AM	0.00	0.01	0.00	0.01	0.01	1.46	0.02	0.01	0.01	0.001	0.00	0.05	0.002	**AM**
AT	0.003	0.17	0.01	0.39	0.55	0.01	0.06	0.96	1.17	0.02	0.002	2.80	0.00	**AT**
AZ	0.00	0.02	0.001	0.02	0.03	3.51	0.04	0.02	0.02	0.002	0.00	0.10	0.02	**AZ**
BA	0.002	0.11	0.003	0.08	0.12	0.02	0.16	6.17	1.13	0.01	0.00	2.24	0.00	**BA**
BE	0.00	0.13	0.001	1.87	1.04	0.001	0.003	0.01	0.01	0.03	0.001	0.07	0.00	**BE**
BG	0.003	0.10	0.004	0.07	0.12	0.15	1.41	0.39	0.54	0.01	0.001	1.11	0.002	**BG**
BY	0.07	0.10	0.06	0.13	0.39	0.17	0.12	0.23	0.47	0.02	0.003	0.47	0.004	**BY**
CH	0.001	0.21	0.001	0.67	0.34	0.003	0.02	0.19	0.05	0.02	0.001	3.99	0.00	**CH**
CY	0.00	0.01	0.00	0.01	0.01	0.01	0.07	0.01	0.01	0.001	0.00	0.07	0.00	**CY**
CZ	0.004	0.10	0.01	0.28	0.52	0.01	0.04	0.39	1.06	0.02	0.002	0.52	0.00	**CZ**
DE	0.02	0.68	0.03	7.37	4.68	0.02	0.06	0.27	0.36	0.16	0.01	1.58	0.00	**DE**
DK	0.004	0.08	0.01	0.17	0.74	0.004	0.01	0.02	0.03	0.03	0.003	0.07	0.00	**DK**
EE	**1.71**	0.04	0.13	0.07	0.15	0.02	0.01	0.04	0.04	0.01	0.001	0.10	0.001	**EE**
ES	0.003	**73.41**	0.00	1.64	1.07	0.01	0.05	0.40	0.14	0.08	0.01	1.74	0.001	**ES**
FI	0.30	0.17	**5.35**	0.19	0.77	0.10	0.05	0.14	0.15	0.04	0.01	0.35	0.01	**FI**
FR	0.01	7.38	0.01	**46.95**	6.03	0.02	0.11	0.88	0.35	0.29	0.02	6.05	0.001	**FR**
GB	0.01	0.79	0.01	1.52	**92.90**	0.01	0.02	0.11	0.10	1.67	0.03	0.44	0.001	**GB**
GE	0.001	0.04	0.002	0.03	0.05	**40.48**	0.10	0.05	0.04	0.004	0.001	0.20	0.003	**GE**
GR	0.003	0.19	0.004	0.14	0.16	0.12	**24.73**	0.47	0.33	0.01	0.002	2.72	0.002	**GR**
HR	0.002	0.10	0.003	0.08	0.13	0.01	0.11	**32.04**	2.74	0.01	0.001	2.33	0.00	**HR**
HU	0.004	0.08	0.01	0.10	0.21	0.02	0.12	4.14	**37.35**	0.01	0.001	1.01	0.00	**HU**
IE	0.002	0.26	0.003	0.28	2.46	0.003	0.00	0.03	0.02	**7.66**	0.01	0.11	0.00	**IE**
IS	0.003	0.14	0.01	0.21	1.04	0.01	0.01	0.02	0.02	0.09	**1.21**	0.06	0.001	**IS**
IT	0.004	1.01	0.01	1.08	0.62	0.04	0.61	5.17	1.27	0.04	0.005	**102.8**	0.001	**IT**
KY	0.00	0.02	0.001	0.01	0.02	0.12	0.02	0.02	0.01	0.002	0.00	0.08	**33.07**	**KY**
KZ	0.03	0.29	0.06	0.26	0.67	1.27	0.25	0.26	0.32	0.04	0.01	0.91	9.41	**KZ**
LT	0.04	0.04	0.03	0.07	0.22	0.02	0.02	0.07	0.13	0.01	0.001	0.15	0.001	**LT**
LU	0.00	0.01	0.00	0.94	0.05	0.00	0.00	0.002	0.001	0.002	0.00	0.01	0.00	**LU**
LV	0.26	0.05	0.07	0.09	0.23	0.03	0.02	0.07	0.09	0.01	0.002	0.17	0.00	**LV**
MC	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	**MC**
MD	0.002	0.02	0.002	0.02	0.04	0.04	0.05	0.04	0.08	0.002	0.00	0.12	0.001	**MD**
ME	0.00	0.03	0.00	0.02	0.02	0.01	0.10	0.20	0.09	0.00	0.00	0.89	0.00	**ME**
MK	0.001	0.04	0.001	0.03	0.04	0.02	1.09	0.14	0.13	0.002	0.00	0.67	0.00	**MK**
MT	0.00	0.001	0.00	0.001	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	**MT**
NL	0.002	0.12	0.002	0.64	1.41	0.002	0.003	0.01	0.01	0.04	0.002	0.06	0.00	**NL**
NO	0.05	0.52	0.16	0.77	4.03	0.07	0.07	0.18	0.18	0.19	0.03	0.57	0.004	**NO**
PL	0.05	0.27	0.07	0.48	1.28	0.08	0.13	0.79	2.04	0.05	0.01	1.13	0.003	**PL**
PT	0.00	2.00	0.001	0.13	0.13	0.002	0.004	0.03	0.01	0.01	0.002	0.13	0.00	**PT**
RO	0.01	0.19	0.01	0.16	0.30	0.21	0.60	1.10	3.01	0.02	0.003	1.85	0.005	**RO**
RS	0.00	0.10	0.00	0.08	0.14	0.04	0.55	2.13	2.04	0.01	0.00	1.71	0.00	**RS**
RU	1.02	1.10	1.33	1.25	3.44	10.31	0.99	1.26	1.58	0.19	0.04	3.67	0.29	**RU**
RUA	0.07	0.42	0.16	0.42	1.40	0.82	0.25	0.32	0.35	0.09	0.02	1.05	0.83	**RUA**
SE	0.17	0.45	0.72	0.64	2.49	0.12	0.09	0.24	0.29	0.11	0.02	0.68	0.005	**SE**
SI	0.001	0.04	0.001	0.04	0.05	0.00	0.03	3.93	0.62	0.003	0.00	1.88	0.00	**SI**
SK	0.004	0.06	0.01	0.08	0.18	0.01	0.07	0.72	5.46	0.01	0.001	0.52	0.00	**SK**
TJ	0.00	0.01	0.00	0.01	0.01	0.07	0.01	0.01	0.01	0.001	0.00	0.04	1.74	**TJ**
TM	0.001	0.04	0.003	0.03	0.07	0.57	0.05	0.04	0.03	0.005	0.001	0.16	0.27	**TM**
TR	0.01	0.55	0.02	0.40	0.59	3.65	3.43	0.88	0.71	0.04	0.01	3.96	0.01	**TR**
UA	0.05	0.30	0.06	0.30	0.74	1.35	0.60	0.93	2.41	0.04	0.01	1.91	0.03	**UA**
UZ	0.002	0.04	0.003	0.03	0.07	0.30	0.04	0.03	0.03	0.005	0.001	0.13	5.30	**UZ**
	EE	**ES**	**FI**	**FR**	**GB**	**GE**	**GR**	**HR**	**HU**	**IE**	**IS**	**IT**	**KY**	

Table A.3. *Matrix of PCDD/F country-to-country deposition in 2009, g TEQ/y (continued)*

Receptors↓ Emitters ⟶

	KZ	LT	LU	LV	MC	MD	ME	MK	MT	NL	NO	PL	
AL	0.003	0.002	0.00	0.004	0.003	0.02	0.10	8.60	0.02	0.01	0.01	0.19	**AL**
AM	0.02	0.00	0.00	0.001	0.00	0.004	0.00	0.03	0.001	0.00	0.002	0.03	**AM**
AT	0.005	0.01	0.01	0.02	0.01	0.02	0.01	0.26	0.01	0.16	0.04	2.52	**AT**
AZ	0.28	0.002	0.00	0.004	0.001	0.01	0.001	0.05	0.003	0.004	0.01	0.09	**AZ**
BA	0.01	0.01	0.001	0.02	0.01	0.03	0.22	0.99	0.02	0.03	0.02	0.90	**BA**
BE	0.001	0.00	0.13	0.01	0.001	0.001	0.00	0.01	0.00	1.10	0.03	0.18	**BE**
BG	0.03	0.01	0.001	0.02	0.004	0.31	0.03	9.09	0.02	0.03	0.03	1.02	**BG**
BY	0.08	0.89	0.003	1.04	0.004	0.29	0.01	0.42	0.01	0.08	0.10	9.40	**BY**
CH	0.001	0.002	0.01	0.01	0.02	0.003	0.002	0.07	0.004	0.06	0.02	0.22	**CH**
CY	0.001	0.00	0.00	0.001	0.00	0.002	0.001	0.04	0.003	0.00	0.00	0.02	**CY**
CZ	0.005	0.02	0.01	0.03	0.004	0.02	0.004	0.21	0.004	0.17	0.07	10.42	**CZ**
DE	0.01	0.06	0.32	0.17	0.02	0.02	0.004	0.22	0.01	4.98	0.39	10.32	**DE**
DK	0.003	0.01	0.00	0.04	0.001	0.004	0.00	0.03	0.001	0.21	0.10	0.91	**DK**
EE	0.01	0.09	0.001	1.35	0.001	0.01	0.001	0.04	0.001	0.04	0.04	0.70	**EE**
ES	0.01	0.01	0.01	0.03	0.03	0.01	0.01	0.16	0.01	0.17	0.15	0.76	**ES**
FI	0.11	0.14	0.003	0.75	0.004	0.04	0.003	0.14	0.01	0.13	0.30	2.06	**FI**
FR	0.01	0.03	0.44	0.07	0.90	0.02	0.01	0.33	0.02	1.05	0.24	2.14	**FR**
GB	0.01	0.02	0.02	0.07	0.01	0.01	0.002	0.11	0.003	0.60	0.47	1.06	**GB**
GE	0.05	0.004	0.00	0.01	0.001	0.04	0.002	0.11	0.01	0.01	0.02	0.22	**GE**
GR	0.02	0.01	0.001	0.02	0.01	0.15	0.03	14.18	0.09	0.03	0.04	0.77	**GR**
HR	0.005	0.01	0.002	0.01	0.01	0.03	0.04	0.65	0.01	0.03	0.02	1.22	**HR**
HU	0.01	0.01	0.003	0.04	0.01	0.07	0.01	0.69	0.01	0.06	0.03	3.75	**HU**
IE	0.004	0.004	0.003	0.01	0.002	0.002	0.001	0.02	0.00	0.11	0.12	0.19	**IE**
IS	0.02	0.01	0.002	0.02	0.001	0.005	0.00	0.02	0.001	0.09	0.30	0.32	**IS**
IT	0.01	0.01	0.01	0.04	0.40	0.06	0.08	2.22	0.25	0.14	0.11	1.92	**IT**
KY	5.69	0.001	0.00	0.002	0.001	0.00	0.001	0.03	0.003	0.004	0.004	0.05	**KY**
KZ	**181.5**	0.07	0.004	0.16	0.01	0.19	0.01	0.44	0.02	0.12	0.22	2.38	**KZ**
LT	0.01	**4.76**	0.002	1.63	0.001	0.03	0.002	0.08	0.002	0.05	0.05	4.14	**LT**
LU	0.00	0.00	**0.25**	0.001	0.00	0.00	0.00	0.00	0.00	0.02	0.002	0.02	**LU**
LV	0.02	0.91	0.002	**12.71**	0.002	0.03	0.001	0.08	0.002	0.05	0.06	2.03	**LV**
MC	0.00	0.00	0.00	0.00	**0.004**	0.00	0.00	0.00	0.00	0.00	0.00	0.00	**MC**
MD	0.02	0.01	0.00	0.02	0.00	**8.95**	0.001	0.12	0.002	0.01	0.01	0.54	**MD**
ME	0.00	0.00	0.00	0.00	0.00	0.01	**0.87**	1.03	0.01	0.01	0.00	0.14	**ME**
MK	0.004	0.002	0.00	0.004	0.002	0.02	0.02	**70.24**	0.01	0.01	0.01	0.21	**MK**
MT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	**0.06**	0.00	0.00	0.001	**MT**
NL	0.001	0.004	0.01	0.01	0.001	0.001	0.00	0.01	0.00	**9.66**	0.06	0.28	**NL**
NO	0.07	0.11	0.02	0.40	0.01	0.04	0.004	0.21	0.01	0.57	**10.72**	4.17	**NO**
PL	0.04	0.44	0.01	0.56	0.01	0.15	0.01	0.60	0.01	0.40	0.30	**215.2**	**PL**
PT	0.001	0.002	0.002	0.00	0.002	0.001	0.00	0.01	0.001	0.02	0.02	0.09	**PT**
RO	0.07	0.03	0.003	0.06	0.01	2.71	0.04	2.74	0.03	0.06	0.06	3.65	**RO**
RS	0.01	0.01	0.00	0.02	0.01	0.07	0.31	13.44	0.02	0.03	0.02	1.27	**RS**
RU	13.17	1.01	0.02	3.60	0.03	1.08	0.04	2.36	0.07	0.66	1.17	17.79	**RU**
RUA	34.08	0.13	0.01	0.36	0.01	0.18	0.01	0.51	0.02	0.23	0.62	3.08	**RUA**
SE	0.08	0.30	0.01	1.24	0.01	0.07	0.004	0.25	0.01	0.50	1.85	8.45	**SE**
SI	0.00	0.002	0.001	0.005	0.004	0.01	0.005	0.14	0.004	0.01	0.01	0.52	**SI**
SK	0.01	0.01	0.002	0.04	0.003	0.04	0.01	0.35	0.005	0.05	0.03	8.95	**SK**
TJ	0.46	0.00	0.00	0.001	0.00	0.00	0.00	0.02	0.002	0.002	0.00	0.02	**TJ**
TM	2.09	0.005	0.00	0.01	0.001	0.02	0.001	0.06	0.01	0.01	0.02	0.25	**TM**
TR	0.15	0.04	0.005	0.09	0.03	0.65	0.04	2.73	0.14	0.10	0.11	2.51	**TR**
UA	0.59	0.22	0.01	0.42	0.01	4.96	0.02	1.62	0.03	0.15	0.22	17.49	**UA**
UZ	7.86	0.01	0.00	0.01	0.001	0.02	0.00	0.05	0.004	0.01	0.02	0.26	**UZ**
	KZ	**LT**	**LU**	**LV**	**MC**	**MD**	**ME**	**MK**	**MT**	**NL**	**NO**	**PL**	

Table A.3. *Matrix of PCDD/F country-to-country deposition in 2009, g TEQ/y (continued)*

Receptors ↓ Emitters ⟶

	PT	**RO**	**RS**	**RU**	**RUA**	**SE**	**SI**	**SK**	**TJ**	**TM**	**TR**	**UA**	**UZ**	**Total**	
AL	0.002	0.23	2.97	0.08	0.00	0.01	0.01	0.04	0.00	0.001	0.72	0.46	0.001	**30.06**	**AL**
AM	0.00	0.02	0.02	0.11	0.001	0.001	0.001	0.004	0.001	0.02	2.68	0.20	0.01	**8.25**	**AM**
AT	0.005	0.24	0.57	0.14	0.00	0.05	0.67	0.82	0.00	0.001	0.29	0.93	0.001	**34.37**	**AT**
AZ	0.001	0.07	0.04	1.20	0.01	0.00	0.00	0.01	0.02	0.24	2.42	0.68	0.16	**54.34**	**AZ**
BA	0.003	0.73	5.24	0.13	0.00	0.03	0.10	0.28	0.00	0.002	0.54	0.83	0.002	**46.16**	**BA**
BE	0.003	0.01	0.01	0.02	0.00	0.02	0.002	0.01	0.00	0.00	0.02	0.05	0.00	**24.00**	**BE**
BG	0.003	6.20	5.32	0.73	0.00	0.03	0.03	0.25	0.001	0.01	9.27	5.93	0.01	**149.30**	**BG**
BY	0.004	1.01	0.50	6.23	0.04	0.22	0.03	0.45	0.003	0.01	2.15	25.22	0.02	**74.59**	**BY**
CH	0.004	0.05	0.11	0.03	0.00	0.01	0.04	0.03	0.00	0.00	0.08	0.14	0.00	**13.62**	**CH**
CY	0.00	0.02	0.02	0.02	0.00	0.001	0.001	0.004	0.00	0.00	1.49	0.08	0.00	**2.15**	**CY**
CZ	0.003	0.24	0.44	0.17	0.00	0.08	0.07	2.21	0.00	0.001	0.20	0.95	0.001	**64.54**	**CZ**
DE	0.02	0.23	0.31	0.55	0.01	0.39	0.05	0.38	0.00	0.003	0.38	1.43	0.004	**85.92**	**DE**
DK	0.003	0.04	0.04	0.13	0.00	0.32	0.00	0.03	0.00	0.00	0.05	0.21	0.001	**10.48**	**DK**
EE	0.002	0.07	0.06	1.78	0.01	0.20	0.01	0.04	0.00	0.002	0.20	0.87	0.003	**8.53**	**EE**
ES	0.90	0.13	0.26	0.14	0.01	0.05	0.06	0.08	0.00	0.002	0.17	0.41	0.003	**84.12**	**ES**
FI	0.01	0.23	0.18	8.75	0.13	1.66	0.02	0.12	0.004	0.01	0.89	2.95	0.03	**28.29**	**FI**
FR	0.09	0.28	0.54	0.32	0.01	0.14	0.15	0.22	0.00	0.003	0.52	1.03	0.004	**89.48**	**FR**
GB	0.04	0.12	0.15	0.39	0.02	0.14	0.02	0.08	0.001	0.002	0.20	0.55	0.005	**104.53**	**GB**
GE	0.001	0.19	0.09	1.81	0.00	0.01	0.01	0.03	0.002	0.04	6.73	1.94	0.03	**56.15**	**GE**
GR	0.01	1.22	2.04	0.58	0.00	0.03	0.04	0.16	0.001	0.005	12.55	3.95	0.01	**77.78**	**GR**
HR	0.002	0.64	3.57	0.12	0.00	0.03	0.93	0.48	0.00	0.001	0.41	1.03	0.002	**54.61**	**HR**
HU	0.002	2.93	4.60	0.24	0.00	0.05	0.31	7.52	0.00	0.002	0.61	4.67	0.002	**75.20**	**HU**
IE	0.01	0.03	0.03	0.09	0.01	0.02	0.00	0.02	0.00	0.00	0.04	0.11	0.001	**12.15**	**IE**
IS	0.01	0.03	0.02	0.24	0.03	0.05	0.00	0.02	0.001	0.002	0.10	0.21	0.004	**4.82**	**IS**
IT	0.02	0.99	2.85	0.34	0.01	0.08	0.90	0.50	0.00	0.004	2.16	1.90	0.004	**136.94**	**IT**
KY	0.001	0.03	0.02	0.16	0.02	0.00	0.00	0.01	3.87	0.21	0.51	0.20	13.70	**58.24**	**KY**
KZ	0.01	0.93	0.46	29.45	13.48	0.18	0.03	0.26	2.10	1.70	5.20	12.48	15.20	**285.89**	**KZ**
LT	0.002	0.17	0.13	1.51	0.01	0.20	0.01	0.11	0.00	0.002	0.30	2.25	0.003	**18.26**	**LT**
LU	0.00	0.001	0.001	0.002	0.00	0.001	0.00	0.001	0.00	0.00	0.002	0.006	0.00	**1.65**	**LU**
LV	0.002	0.14	0.11	1.69	0.01	0.27	0.01	0.08	0.00	0.003	0.34	1.84	0.004	**22.99**	**LV**
MC	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	**0.01**	**MC**
MD	0.001	2.60	0.12	0.49	0.00	0.01	0.01	0.06	0.00	0.003	1.40	10.11	0.005	**25.59**	**MD**
ME	0.00	0.15	2.44	0.03	0.00	0.00	0.01	0.03	0.00	0.00	0.24	0.19	0.00	**8.92**	**ME**
MK	0.001	0.27	3.42	0.08	0.001	0.01	0.01	0.05	0.00	0.001	0.96	0.52	0.001	**82.47**	**MK**
MT	0.00	0.00	0.001	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.002	0.001	0.00	**0.08**	**MT**
NL	0.003	0.01	0.02	0.04	0.00	0.03	0.00	0.01	0.00	0.00	0.02	0.08	0.00	**19.36**	**NL**
NO	0.02	0.28	0.26	2.09	0.13	1.63	0.02	0.17	0.002	0.01	0.77	2.06	0.01	**35.81**	**NO**
PL	0.01	1.32	1.07	2.73	0.02	0.62	0.12	3.88	0.002	0.01	1.13	15.51	0.01	**288.62**	**PL**
PT	**4.90**	0.01	0.02	0.02	0.002	0.01	0.004	0.01	0.00	0.00	0.02	0.04	0.00	**7.86**	**PT**
RO	0.01	**77.98**	8.69	1.66	0.01	0.07	0.10	1.05	0.002	0.01	8.89	21.94	0.02	**148.65**	**RO**
RS	0.00	3.01	**80.17**	0.23	0.00	0.03	0.06	0.45	0.00	0.00	1.44	1.63	0.00	**117.68**	**RS**
RU	0.04	4.60	2.47	**611.9**	9.10	1.98	0.16	1.30	0.19	1.00	26.46	117.5	1.65	**873.37**	**RU**
RUA	0.02	0.95	0.53	44.63	**155.5**	0.43	0.04	0.27	0.35	0.44	4.53	11.56	1.64	**271.36**	**RUA**
SE	0.02	0.38	0.30	4.01	0.10	**19.71**	0.04	0.30	0.003	0.01	1.23	4.02	0.02	**56.62**	**SE**
SI	0.001	0.16	0.45	0.04	0.00	0.01	**3.66**	0.19	0.00	0.00	0.12	0.37	0.001	**14.48**	**SI**
SK	0.002	0.83	0.89	0.19	0.00	0.04	0.10	**15.95**	0.00	0.001	0.40	3.35	0.002	**51.51**	**SK**
TJ	0.00	0.01	0.01	0.07	0.01	0.00	0.00	0.00	**20.87**	0.24	0.28	0.09	6.50	**30.70**	**TJ**
TM	0.001	0.09	0.06	1.10	0.09	0.01	0.00	0.03	0.98	**15.43**	1.49	1.42	10.14	**36.73**	**TM**
TR	0.02	3.91	2.03	4.41	0.02	0.08	0.10	0.40	0.005	0.07	**680.0**	21.09	0.06	**747.39**	**TR**
UA	0.01	8.88	2.00	21.56	0.10	0.28	0.11	2.03	0.02	0.08	16.15	**619.6**	0.14	**717.64**	**UA**
UZ	0.001	0.09	0.05	1.12	0.18	0.01	0.00	0.03	8.76	2.85	0.99	1.38	**54.48**	**85.19**	**UZ**
	PT	**RO**	**RS**	**RU**	**RUA**	**SE**	**SI**	**SK**	**TJ**	**TM**	**TR**	**UA**	**UZ**	**Total**	

Table A.4*. Matrix of HCB country-to-country deposition in 2009, kg/y*

Receptors ↓ Emitters ⟶

	AL	AM	AT	AZ	BA	BE	BG	BY	CH	CY	CZ	DE	DK	
AL	**0.012**	0.000	0.043	0.000	0.000	0.002	0.138	0.000	0.003	0.000	0.029	0.295	0.010	**AL**
AM	0.000	**0.001**	0.006	0.000	0.000	0.001	0.009	0.000	0.001	0.001	0.005	0.065	0.004	**AM**
AT	0.000	0.000	**10.89**	0.000	0.000	0.027	0.053	0.000	0.096	0.000	1.157	9.066	0.090	**AT**
AZ	0.000	0.000	0.014	**0.001**	0.000	0.001	0.020	0.000	0.002	0.001	0.011	0.161	0.012	**AZ**
BA	0.001	0.000	0.280	0.000	**0.000**	0.008	0.139	0.000	0.010	0.000	0.172	1.309	0.042	**BA**
BE	0.000	0.000	0.023	0.000	0.000	**0.909**	0.003	0.000	0.008	0.000	0.027	3.904	0.040	**BE**
BG	0.000	0.000	0.136	0.000	0.000	0.008	**12.20**	0.000	0.010	0.001	0.126	1.122	0.051	**BG**
BY	0.000	0.000	0.135	0.000	0.000	0.016	0.080	**0.002**	0.012	0.001	0.206	2.574	0.280	**BY**
CH	0.000	0.000	0.228	0.000	0.000	0.013	0.014	0.000	**1.298**	0.000	0.050	3.300	0.022	**CH**
CY	0.000	0.000	0.003	0.000	0.000	0.000	0.006	0.000	0.000	**0.094**	0.002	0.027	0.001	**CY**
CZ	0.000	0.000	1.462	0.000	0.000	0.026	0.040	0.000	0.040	0.000	**9.209**	10.18	0.122	**CZ**
DE	0.000	0.000	2.789	0.000	0.000	0.482	0.073	0.000	0.630	0.000	2.158	**221.5**	1.107	**DE**
DK	0.000	0.000	0.018	0.000	0.000	0.013	0.005	0.000	0.003	0.000	0.033	2.069	**3.420**	**DK**
EE	0.000	0.000	0.012	0.000	0.000	0.004	0.006	0.000	0.001	0.000	0.018	0.469	0.083	**EE**
ES	0.000	0.000	0.201	0.000	0.000	0.040	0.053	0.000	0.056	0.000	0.128	3.346	0.114	**ES**
FI	0.000	0.000	0.064	0.000	0.000	0.019	0.028	0.000	0.008	0.000	0.082	1.902	0.331	**FI**
FR	0.000	0.000	0.520	0.000	0.000	0.432	0.080	0.000	0.617	0.000	0.376	21.8	0.333	**FR**
GB	0.000	0.000	0.045	0.000	0.000	0.064	0.013	0.000	0.010	0.000	0.057	2.359	0.254	**GB**
GE	0.000	0.000	0.019	0.000	0.000	0.002	0.032	0.000	0.002	0.002	0.016	0.215	0.017	**GE**
GR	0.002	0.000	0.110	0.000	0.000	0.006	1.083	0.000	0.010	0.002	0.080	0.844	0.034	**GR**
HR	0.000	0.000	0.520	0.000	0.000	0.007	0.081	0.000	0.011	0.000	0.210	1.361	0.035	**HR**
HU	0.000	0.000	1.238	0.000	0.000	0.013	0.146	0.000	0.016	0.000	0.547	2.466	0.076	**HU**
IE	0.000	0.000	0.007	0.000	0.000	0.007	0.002	0.000	0.001	0.000	0.009	0.329	0.039	**IE**
IS	0.000	0.000	0.007	0.000	0.000	0.004	0.003	0.000	0.001	0.000	0.009	0.266	0.035	**IS**
IT	0.001	0.000	1.399	0.000	0.000	0.035	0.268	0.000	0.263	0.001	0.433	6.314	0.117	**IT**
KY	0.000	0.000	0.019	0.000	0.000	0.002	0.016	0.000	0.003	0.001	0.013	0.202	0.011	**KY**
KZ	0.000	0.000	0.246	0.000	0.000	0.034	0.211	0.000	0.031	0.006	0.232	3.827	0.329	**KZ**
LT	0.000	0.000	0.039	0.000	0.000	0.007	0.015	0.000	0.004	0.000	0.066	1.185	0.181	**LT**
LU	0.000	0.000	0.004	0.000	0.000	0.011	0.000	0.000	0.002	0.000	0.004	0.635	0.004	**LU**
LV	0.000	0.000	0.027	0.000	0.000	0.007	0.012	0.000	0.003	0.000	0.043	0.956	0.162	**LV**
MC	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	**MC**
MD	0.000	0.000	0.023	0.000	0.000	0.002	0.069	0.000	0.002	0.000	0.028	0.312	0.026	**MD**
ME	0.000	0.000	0.036	0.000	0.000	0.001	0.137	0.000	0.002	0.000	0.030	0.216	0.008	**ME**
MK	0.001	0.000	0.043	0.000	0.000	0.002	0.455	0.000	0.003	0.000	0.033	0.313	0.012	**MK**
MT	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	**MT**
NL	0.000	0.000	0.024	0.000	0.000	0.254	0.003	0.000	0.005	0.000	0.033	6.161	0.074	**NL**
NO	0.000	0.000	0.080	0.000	0.000	0.035	0.031	0.000	0.012	0.000	0.113	3.382	0.868	**NO**
PL	0.000	0.000	0.709	0.000	0.000	0.059	0.119	0.000	0.050	0.001	2.820	16.77	0.932	**PL**
PT	0.000	0.000	0.021	0.000	0.000	0.005	0.006	0.000	0.006	0.000	0.016	0.411	0.019	**PT**
RO	0.000	0.000	0.340	0.000	0.000	0.019	1.922	0.000	0.023	0.002	0.346	2.867	0.146	**RO**
RS	0.002	0.000	0.231	0.000	0.000	0.009	0.875	0.000	0.011	0.001	0.190	1.381	0.048	**RS**
RU (all)	0.001	0.000	0.907	0.000	0.000	0.174	0.761	0.001	0.111	0.012	1.080	19.73	2.382	**RU (all)**
SE	0.000	0.000	0.125	0.000	0.000	0.047	0.044	0.000	0.018	0.001	0.188	5.245	2.299	**SE**
SI	0.000	0.000	0.828	0.000	0.000	0.004	0.024	0.000	0.008	0.000	0.117	0.907	0.017	**SI**
SK	0.000	0.000	0.601	0.000	0.000	0.009	0.060	0.000	0.011	0.000	0.947	1.900	0.070	**SK**
TJ	0.000	0.000	0.010	0.000	0.000	0.001	0.009	0.000	0.001	0.001	0.007	0.100	0.005	**TJ**
TR	0.001	0.000	0.277	0.000	0.000	0.022	1.050	0.000	0.030	0.047	0.224	2.837	0.162	**TR**
TU	0.000	0.000	0.042	0.000	0.000	0.005	0.042	0.000	0.005	0.002	0.034	0.542	0.040	**TU**
UA	0.000	0.000	0.379	0.000	0.000	0.034	0.522	0.000	0.032	0.003	0.494	5.184	0.458	**UA**
UZ	0.000	0.000	0.038	0.000	0.000	0.004	0.033	0.000	0.005	0.001	0.031	0.504	0.039	**UZ**
	AL	**AM**	**AT**	**AZ**	**BA**	**BE**	**BG**	**BY**	**CH**	**CY**	**CZ**	**DE**	**DK**	

***Table A.4**. Matrix of HCB country-to-country deposition in 2009, kg/y (continued)*

Receptors ↓ Emitters →

	EE	**ES**	**FI**	**FR**	**GB**	**GE**	**GR**	**HU**	**IE**	**IS**	**IT**	
AL	0.000	2.392	0.004	0.181	0.211	0.000	0.020	0.065	0.000	0.000	1.161	**AL**
AM	0.000	0.604	0.003	0.044	0.068	0.000	0.001	0.007	0.000	0.000	0.101	**AM**
AT	0.000	7.131	0.030	1.439	1.795	0.000	0.003	0.670	0.001	0.000	3.967	**AT**
AZ	0.000	1.294	0.010	0.102	0.178	0.000	0.002	0.016	0.000	0.000	0.209	**AZ**
BA	0.000	4.764	0.016	0.463	0.696	0.000	0.006	0.594	0.000	0.000	2.447	**BA**
BE	0.000	1.891	0.007	4.293	2.504	0.000	0.000	0.008	0.001	0.000	0.132	**BE**
BG	0.000	5.112	0.031	0.469	0.855	0.000	0.043	0.325	0.000	0.000	1.458	**BG**
BY	0.000	4.652	0.250	0.751	2.155	0.000	0.004	0.197	0.001	0.000	0.763	**BY**
CH	0.000	5.288	0.006	2.452	0.788	0.000	0.001	0.036	0.000	0.000	4.886	**CH**
CY	0.000	0.349	0.001	0.022	0.025	0.000	0.001	0.004	0.000	0.000	0.072	**CY**
CZ	0.000	4.725	0.036	1.144	1.851	0.000	0.002	0.494	0.001	0.000	0.946	**CZ**
DE	0.000	21.8	0.158	13.15	15.16	0.000	0.005	0.287	0.004	0.000	3.583	**DE**
DK	0.000	0.974	0.029	0.407	1.656	0.000	0.000	0.013	0.000	0.000	0.089	**DK**
EE	**0.002**	0.735	0.443	0.162	0.606	0.000	0.000	0.015	0.000	0.000	0.095	**EE**
ES	0.000	**2248**	0.027	8.592	3.529	0.000	0.004	0.124	0.002	0.000	3.584	**ES**
FI	0.000	4.160	**14.85**	0.828	3.325	0.000	0.002	0.066	0.001	0.000	0.509	**FI**
FR	0.000	125.4	0.071	**159.6**	17.61	0.000	0.006	0.225	0.006	0.000	9.839	**FR**
GB	0.000	5.866	0.050	3.211	**143.8**	0.000	0.001	0.030	0.019	0.000	0.336	**GB**
GE	0.000	1.491	0.012	0.125	0.239	**0.001**	0.002	0.022	0.000	0.000	0.268	**GE**
GR	0.000	7.200	0.020	0.536	0.671	0.000	**0.453**	0.164	0.000	0.000	2.460	**GR**
HR	0.000	4.252	0.014	0.444	0.626	0.000	0.003	**1.123**	0.000	0.000	2.638	**HR**
HU	0.000	4.936	0.029	0.628	1.139	0.000	0.004	13.70	**0.000**	0.000	1.606	**HU**
IE	0.000	1.355	0.009	0.383	3.748	0.000	0.000	0.005	0.045	**0.000**	0.055	**IE**
IS	0.000	1.000	0.017	0.203	1.028	0.000	0.000	0.005	0.001	0.000	**0.056**	**IS**
IT	0.000	38.22	0.045	5.046	2.859	0.000	0.021	0.713	0.001	0.000	155.3	**IT**
KY	0.000	2.316	0.010	0.149	0.188	0.000	0.002	0.018	0.000	0.000	0.263	**KY**
KZ	0.000	21.80	0.443	2.151	4.896	0.000	0.015	0.257	0.002	0.000	2.717	**KZ**
LT	0.000	1.495	0.138	0.314	1.064	0.000	0.001	0.048	0.000	0.000	0.219	**LT**
LU	0.000	0.237	0.001	0.447	0.154	0.000	0.000	0.001	0.000	0.000	0.020	**LU**
LV	0.000	1.336	0.235	0.285	1.022	0.000	0.001	0.033	0.000	0.000	0.190	**LV**
MC	0.000	0.002	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.003	**MC**
MD	0.000	0.970	0.017	0.116	0.270	0.000	0.002	0.047	0.000	0.000	0.221	**MD**
ME	0.000	0.812	0.003	0.077	0.129	0.000	0.002	0.149	0.000	0.000	0.319	**ME**
MK	0.000	1.845	0.005	0.150	0.221	0.000	0.030	0.081	0.000	0.000	0.668	**MK**
MT	0.000	0.012	0.000	0.001	0.001	0.000	0.000	0.000	0.000	0.000	0.006	**MT**
NL	0.000	1.476	0.011	1.578	3.023	0.000	0.000	0.010	0.001	0.000	0.110	**NL**
NO	0.000	5.354	0.379	1.389	7.842	0.000	0.002	0.063	0.003	0.000	0.575	**NO**
PL	0.000	11.68	0.259	2.445	5.766	0.000	0.005	0.889	0.002	0.000	1.872	**PL**
PT	0.000	145.4	0.005	0.769	0.493	0.000	0.000	0.014	0.000	0.000	0.370	**PT**
RO	0.000	9.525	0.081	1.034	2.061	0.000	0.019	1.702	0.001	0.000	2.714	**RO**
RS	0.000	5.194	0.020	0.494	0.824	0.000	0.016	0.953	0.000	0.000	2.043	**RS**
RU (all)	0.002	62.60	5.853	8.993	28.55	0.000	0.043	1.029	0.012	0.000	8.292	**RU (all)**
SE	0.000	7.522	1.487	1.813	8.096	0.000	0.003	0.106	0.003	0.000	0.826	**SE**
SI	0.000	2.054	0.007	0.255	0.329	0.000	0.001	0.279	0.000	0.000	2.187	**SI**
SK	0.000	3.027	0.025	0.442	0.852	0.000	0.002	2.094	0.000	0.000	0.719	**SK**
TJ	0.000	1.264	0.005	0.077	0.088	0.000	0.001	0.009	0.000	0.000	0.142	**TJ**
TR	0.000	23.39	0.106	1.798	2.766	0.000	0.075	0.372	0.001	0.000	4.978	**TR**
TU	0.000	4.241	0.043	0.344	0.614	0.000	0.004	0.043	0.000	0.000	0.547	**TU**
UA	0.000	13.95	0.342	1.786	4.356	0.000	0.017	1.134	0.002	0.000	2.860	**UA**
UZ	0.000	3.740	0.046	0.309	0.595	0.000	0.003	0.038	0.000	0.000	0.455	**UZ**
	EE	**ES**	**FI**	**FR**	**GB**	**GE**	**GR**	**HU**	**IE**	**IS**	**IT**	

***Table A.4**. Matrix of HCB country-to-country deposition in 2009, kg/y (continued)*

Receptors↓ Emitters ⟶

	KZ	LT	LU	LV	MD	ME	MK	NL	NO	PL	
AL	0.000	0.000	0.000	0.000	0.059	0.000	0.005	0.056	0.044	0.008	**AL**
AM	0.000	0.000	0.000	0.000	0.019	0.000	0.000	0.015	0.022	0.002	**AM**
AT	0.000	0.000	0.000	0.000	0.084	0.000	0.000	0.663	0.278	0.112	**AT**
AZ	0.000	0.000	0.000	0.000	0.043	0.000	0.000	0.038	0.070	0.004	**AZ**
BA	0.000	0.000	0.000	0.000	0.111	0.001	0.001	0.210	0.145	0.038	**BA**
BE	0.000	0.000	0.000	0.000	0.006	0.000	0.000	3.378	0.113	0.004	**BE**
BG	0.000	0.000	0.000	0.000	0.859	0.001	0.005	0.228	0.242	0.041	**BG**
BY	0.000	0.020	0.000	0.003	0.621	0.000	0.000	0.555	1.075	0.124	**BY**
CH	0.000	0.000	0.000	0.000	0.021	0.000	0.000	0.242	0.084	0.008	**CH**
CY	0.000	0.000	0.000	0.000	0.008	0.000	0.000	0.006	0.006	0.001	**CY**
CZ	0.000	0.001	0.000	0.000	0.068	0.000	0.000	0.779	0.321	0.331	**CZ**
DE	0.000	0.002	0.000	0.001	0.145	0.000	0.000	15.12	1.958	0.163	**DE**
DK	0.000	0.000	0.000	0.000	0.012	0.000	0.000	0.592	0.706	0.009	**DK**
EE	0.000	0.002	0.000	0.003	0.025	0.000	0.000	0.146	0.420	0.008	**EE**
ES	0.000	0.000	0.000	0.000	0.066	0.000	0.000	0.883	0.469	0.025	**ES**
FI	0.000	0.003	0.000	0.002	0.098	0.000	0.000	0.668	3.669	0.031	**FI**
FR	0.000	0.001	0.001	0.000	0.126	0.000	0.000	5.195	1.157	0.064	**FR**
GB	0.000	0.000	0.000	0.000	0.026	0.000	0.000	1.782	1.036	0.013	**GB**
GE	0.000	0.000	0.000	0.000	0.087	0.000	0.000	0.051	0.087	0.006	**GE**
GR	0.000	0.000	0.000	0.000	0.304	0.000	0.008	0.169	0.172	0.023	**GR**
HR	0.000	0.000	0.000	0.000	0.087	0.000	0.000	0.195	0.119	0.045	**HR**
HU	0.000	0.000	0.000	0.000	0.221	0.001	0.000	0.376	0.252	0.144	**HU**
IE	0.000	0.000	0.000	0.000	0.005	0.000	0.000	0.206	0.184	0.002	**IE**
IS	0.000	0.000	0.000	0.000	0.008	0.000	0.000	0.113	0.393	0.002	**IS**
IT	0.000	0.001	0.000	0.000	0.277	0.001	0.002	0.846	0.465	0.088	**IT**
KY	0.000	0.000	0.000	0.000	0.030	0.000	0.000	0.045	0.073	0.004	**KY**
KZ	**0.051**	0.003	0.000	0.001	0.583	0.000	0.001	1.032	2.496	0.080	**KZ**
LT	0.000	**0.089**	0.000	0.004	0.083	0.000	0.000	0.271	0.513	0.031	**LT**
LU	0.000	0.000	**0.000**	0.000	0.001	0.000	0.000	0.091	0.011	0.001	**LU**
LV	0.000	0.015	0.000	**0.028**	0.059	0.000	0.000	0.244	0.578	0.018	**LV**
MC	0.000	0.000	0.000	0.000	**0.000**	0.000	0.000	0.000	0.000	0.000	**MC**
MD	0.000	0.000	0.000	0.000	17.06	**0.000**	0.000	0.066	0.121	0.016	**MD**
ME	0.000	0.000	0.000	0.000	0.040	0.001	**0.001**	0.039	0.028	0.007	**ME**
MK	0.000	0.000	0.000	0.000	0.075	0.001	0.036	**0.061**	0.050	0.009	**MK**
MT	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	**0.000**	0.000	**MT**
NL	0.000	0.000	0.000	0.000	0.006	0.000	0.000	21.42	0.173	**0.005**	**NL**
NO	0.000	0.002	0.000	0.001	0.075	0.000	0.000	1.238	72.21	0.033	**NO**
PL	0.000	0.011	0.000	0.002	0.499	0.000	0.000	2.063	1.891	3.942	**PL**
PT	0.000	0.000	0.000	0.000	0.009	0.000	0.000	0.119	0.076	0.003	**PT**
RO	0.000	0.001	0.000	0.000	6.590	0.001	0.002	0.557	0.631	0.152	**RO**
RS	0.000	0.000	0.000	0.000	0.257	0.010	0.006	0.247	0.179	0.048	**RS**
RU (all)	0.027	0.034	0.000	0.014	2.834	0.001	0.002	5.728	18.34	0.424	**RU (all)**
SE	0.000	0.005	0.000	0.002	0.140	0.000	0.000	1.781	17.19	0.066	**SE**
SI	0.000	0.000	0.000	0.000	0.033	0.000	0.000	0.107	0.058	0.024	**SI**
SK	0.000	0.000	0.000	0.000	0.120	0.000	0.000	0.290	0.208	0.305	**SK**
TJ	0.000	0.000	0.000	0.000	0.016	0.000	0.000	0.022	0.033	0.002	**TJ**
TR	0.001	0.002	0.000	0.000	1.437	0.000	0.002	0.648	0.851	0.079	**TR**
TU	0.002	0.000	0.000	0.000	0.100	0.000	0.000	0.134	0.280	0.012	**TU**
UA	0.001	0.007	0.000	0.002	9.164	0.000	0.001	1.082	2.068	0.328	**UA**
UZ	0.001	0.000	0.000	0.000	0.085	0.000	0.000	0.127	0.281	0.011	**UZ**
	KZ	**LT**	**LU**	**LV**	**MD**	**ME**	**MK**	**NL**	**NO**	**PL**	

Table A.4. *Matrix of HCB country-to-country deposition in 2009, kg/y (continued)*

Receptors↓ Emitters ⟶

	PT	RO	RS	RU (all)	SE	SI	SK	TR	UA	Total	
AL	0.004	0.006	0.005	0.001	0.007	0.001	0.000	0.001	0.405	**5.17**	**AL**
AM	0.001	0.001	0.000	0.002	0.004	0.000	0.000	0.005	0.252	**1.24**	**AM**
AT	0.013	0.007	0.001	0.002	0.062	0.025	0.009	0.001	0.890	**38.56**	**AT**
AZ	0.002	0.002	0.000	0.009	0.012	0.000	0.000	0.005	0.668	**2.89**	**AZ**
BA	0.007	0.017	0.011	0.001	0.027	0.004	0.002	0.001	0.880	**12.40**	**BA**
BE	0.006	0.000	0.000	0.000	0.019	0.000	0.000	0.000	0.072	**17.35**	**BE**
BG	0.009	0.128	0.011	0.008	0.043	0.001	0.002	0.015	4.594	**28.14**	**BG**
BY	0.011	0.019	0.001	0.036	0.275	0.001	0.002	0.003	13.70	**28.52**	**BY**
CH	0.009	0.002	0.000	0.001	0.014	0.001	0.000	0.000	0.203	**18.97**	**CH**
CY	0.001	0.000	0.000	0.000	0.001	0.000	0.000	0.002	0.083	**0.72**	**CY**
CZ	0.010	0.006	0.001	0.002	0.078	0.003	0.014	0.001	0.837	**32.73**	**CZ**
DE	0.051	0.009	0.001	0.007	0.448	0.004	0.004	0.002	1.656	**302.42**	**DE**
DK	0.003	0.001	0.000	0.001	0.240	0.000	0.000	0.000	0.134	**10.43**	**DK**
EE	0.002	0.001	0.000	0.009	0.166	0.000	0.000	0.000	0.391	**3.82**	**EE**
ES	1.807	0.006	0.001	0.002	0.072	0.003	0.001	0.001	0.623	**2272.23**	**ES**
FI	0.012	0.005	0.000	0.037	1.097	0.001	0.001	0.002	1.477	**33.28**	**FI**
FR	0.212	0.009	0.001	0.005	0.188	0.005	0.002	0.002	1.340	**345.27**	**FR**
GB	0.029	0.002	0.000	0.002	0.131	0.000	0.000	0.000	0.289	**159.44**	**GB**
GE	0.003	0.004	0.000	0.012	0.015	0.000	0.000	0.010	1.249	**3.99**	**GE**
GR	0.011	0.021	0.003	0.005	0.028	0.001	0.001	0.017	2.291	**16.73**	**GR**
HR	0.007	0.012	0.005	0.001	0.024	0.029	0.003	0.001	0.797	**12.65**	**HR**
HU	0.009	0.059	0.008	0.003	0.051	0.011	0.037	0.001	3.430	**31.15**	**HU**
IE	0.008	0.000	0.000	0.000	0.020	0.000	0.000	0.000	0.053	**6.47**	**IE**
IS	0.005	0.000	0.000	0.001	0.030	0.000	0.000	0.000	0.088	**3.28**	**IS**
IT	0.058	0.026	0.006	0.005	0.081	0.031	0.004	0.006	2.448	**215.34**	**IT**
KY	0.004	0.002	0.000	0.003	0.011	0.000	0.000	0.002	0.356	**3.74**	**KY**
KZ	0.048	0.029	0.002	0.253	0.392	0.003	0.002	0.018	9.083	**51.28**	**KZ**
LT	0.004	0.003	0.000	0.008	0.188	0.000	0.001	0.001	1.385	**7.36**	**LT**
LU	0.001	0.000	0.000	0.000	0.002	0.000	0.000	0.000	0.009	**1.64**	**LU**
LV	0.004	0.003	0.000	0.009	0.227	0.000	0.000	0.001	0.977	**6.48**	**LV**
MC	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	**0.01**	**MC**
MD	0.002	0.056	0.000	0.004	0.023	0.000	0.000	0.002	6.490	**25.95**	**MD**
ME	0.001	0.009	0.016	0.000	0.005	0.000	0.001	0.000	0.253	**2.32**	**ME**
MK	0.003	0.008	0.006	0.001	0.009	0.000	0.000	0.002	0.499	**4.63**	**MK**
MT	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	**0.02**	**MT**
NL	0.005	0.000	0.000	0.000	0.031	0.000	0.000	0.000	0.079	**34.49**	**NL**
NO	0.017	0.004	0.000	0.008	1.211	0.001	0.001	0.001	0.916	**95.85**	**NO**
PL	0.027	0.032	0.002	0.017	0.553	0.005	0.022	0.003	10.61	**64.06**	**PL**
PT	**5.081**	0.001	0.000	0.000	0.012	0.000	0.000	0.000	0.084	**152.88**	**PT**
RO	0.017	**1.432**	0.014	0.015	0.117	0.004	0.008	0.012	16.17	**48.53**	**RO**
RS	0.008	0.059	**0.104**	0.003	0.033	0.002	0.003	0.003	1.616	**14.87**	**RS**
RU (all)	0.160	0.116	0.006	**3.726**	3.343	0.009	0.011	0.057	67.06	**242.42**	**RU (all)**
SE	0.023	0.007	0.001	0.017	**11.65**	0.001	0.001	0.002	1.937	**60.64**	**SE**
SI	0.003	0.003	0.001	0.001	0.013	**0.128**	0.002	0.000	0.319	**7.71**	**SI**
SK	0.006	0.018	0.001	0.002	0.044	0.003	**0.092**	0.001	2.371	**14.22**	**SK**
TJ	0.002	0.001	0.000	0.002	0.005	0.000	0.000	**0.001**	0.181	**1.98**	**TJ**
TR	0.039	0.073	0.004	0.046	0.147	0.003	0.003	0.784	**14.57**	**56.83**	**TR**
TU	0.008	0.005	0.000	0.018	0.044	0.001	0.000	0.006	1.506	**8.67**	**TU**
UA	0.031	0.180	0.004	0.128	0.423	0.004	0.011	0.022	264.5	**309.52**	**UA**
UZ	0.007	0.004	0.000	0.017	0.044	0.000	0.000	0.004	1.250	**7.68**	**UZ**
	PT	**RO**	**RS**	**RU (all)**	**SE**	**SI**	**SK**	**TR**	**UA**	**Total**	