4. ASSESSMENT OF PCDD/Fs, PCBs, AND HCB POLLUTION IN THE EMEP REGION

This chapter presents results of the assessment of environmental pollution by PCDD/Fs, PCBs, and HCB performed on the basis of model predictions and measurements. Model simulations were carried out on the basis of officially reported emission data for the new EMEP grid using the multi-media GLEMOS modelling system. Spatial trends of air concentrations and deposition fluxes in 2016 on regional and global scales were characterized. Transboundary transport of pollution between the EMEP countries as well as transport from non-EMEP emission sources was evaluated. Results of model simulations were compared with measurements of the EMEP monitoring sites and data of national monitoring networks. Main emphasis in these activities was given to the evaluation of PCDD/F pollution levels in the EMEP countries. Brief overview of progress and outcome of this work is presented below. Detailed information on modelling results and their analysis is available in the internet on the MSC-E web site (*www.msceast.org*).

4.1. Emission data for model assessment

Assessment of PCDD/F and HCB pollution in the new EMEP domain was made on the basis of gridded emission data with spatial resolution 0.1°x0.1° provided by CEIP. Similar to PAHs, pollution levels of PCDD/Fs and HCB in 2016 were evaluated on the basis of emissions reported for the previous year 2015 due to availability of necessary gridded emission data[7]. Detailed description of estimated PCDD/F and HCB emissions in the EMEP countries, gap-filling methods, and expert estimates applied for preparation of emission inventory, can be found in the Technical report of CEIP [*Tista et al.,* 2017].

Estimates of PCDD/F emissions officially reported by the EMEP countries are most likely subject to considerable uncertainties due to underestimation of releases for some of the source categories (e.g. 'Residential combustion', 'Open burning of wastes') and incomplete coverage of all potential sources [*Breivik et al.*, 2004; *Fiedler*, 2007; *Pulles et al.*, 2005; *Pulles et al.*, 2006]. For this reason two emission datasets were used in model simulations, namely, officially reported gridded emissions and scenario emissions, representing maximum level of PCDD/F releases to the atmosphere. The maximum emission scenario was prepared on the basis of the uncertainty range reported by 12 EMEP countries in their inventory information reports (namely, Belarus, Belgium, Croatia, Cyprus, Denmark, Estonia, Finland, France, Latvia, Poland, Sweden, and the UK). Difference between the maximum and average estimates of PCDD/F emissions in these countries varied from a factor of 1.5 for the UK up to a factor of 4.1 for Croatia. For other EMEP countries, which did not report uncertainty range in their inventories, the maximum level of national PCDD/F emissions was assumed to be 3-fold higher comparing to the officially reported emissions in accordance with the expert estimates [*Pulles et al.*, 2006; *Bogdal et al.*, 2014]. Thus, total PCDD/F emission in the EMEP countries according to the maximum emission scenario exceeded reported data in the inventories by a factor of 3.5 on average.

Gridded emission data for PCB modelling were based on the available expert estimates and officially reported data of the EMEP countries. Model assessment of PCB pollution levels (total and congener

[7] *Update of the modelling results based on the new emission data for 2016 is available at the MSC-E web site [www.msceast.org].*

specific) requires definition of emissions of particular PCB congeners. However, currently reported national inventories of PCB emissions provide total releases of PCBs without distribution by particular congeners. Therefore, to evaluate transport and fate of individual PCB congeners, congener specific emission inventory of *Breivik et al.* [2007] was used for modelling. The indicator congener PCB-153 was selected to characterize transboundary transport and pollution by PCBs. Spatial distribution of PCB-153 emissions was constructed on the basis of gridded PCB emissions officially provided by the EMEP countries. For other EMEP countries, which did not report gridded emission data, gridded population density was used for allocation of emissions.

Maps illustrating spatial distributions of PCDD/F, HCB, and PCB-153 emission fluxes from anthropogenic sources in the EMEP region, used in the model simulations for 2016, are presented in Figs 4.1.

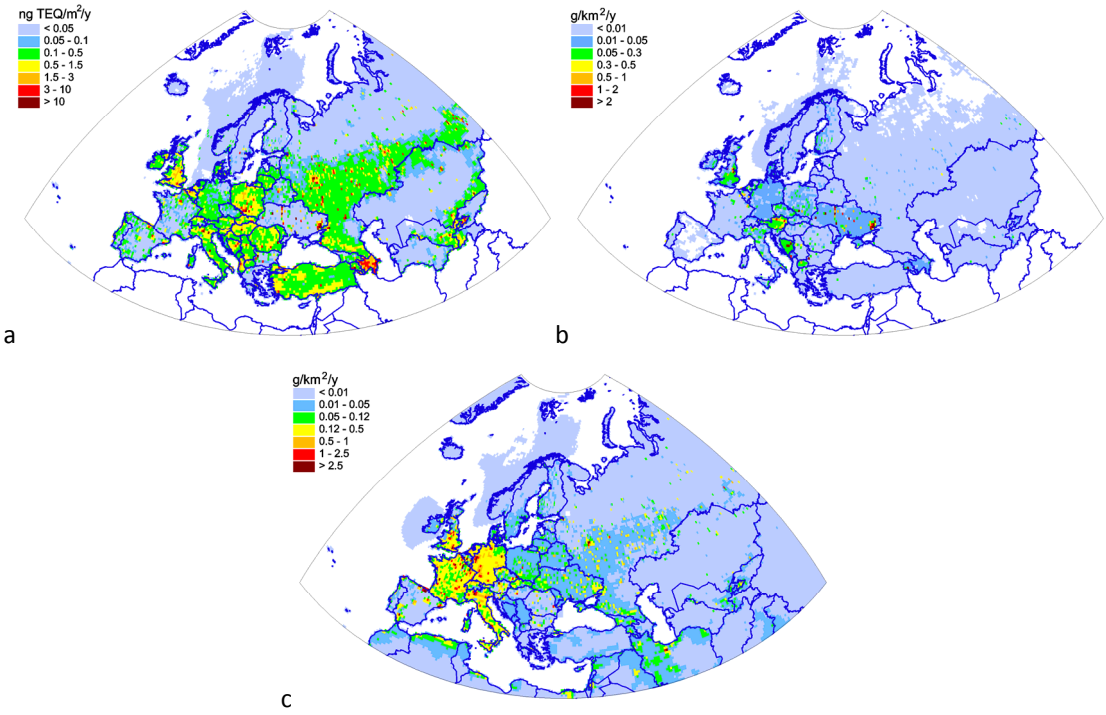


Fig. 4.1. *Spatial distribution of PCDD/F, ng TEQ/m^2/y (a), HCB, g/km^2/y (b), and PCB-153, g/km^2/y (c) emissions in the EMEP region used in model simulations for 2016.*

For the evaluation of global-scale transport and fate of PCDD/Fs, HCB, and PCBs expert estimates of global emissions were applied. In particular, global gridded emissions of PCDD/Fs to the atmosphere and soil were prepared using the national emission inventories reported by countries to the Stockholm Convention [*Gusev et al.*, 2014; *Shatalov et al.*, 2014]. Model simulations of HCB global-scale transport were carried out on the basis of experimental emission scenario of historical HCB releases during the period covering several recent decades [*Shatalov et al.*, 2010]. For PCB-153 modelling, data on global emissions were derived from the inventory of *Breivik et al.* [2007]. Spatial distributions of PCDD/F, HCB, and PCB-153 emissions, used in the global-scale model simulations for 2016, are shown in Fig.4.2.

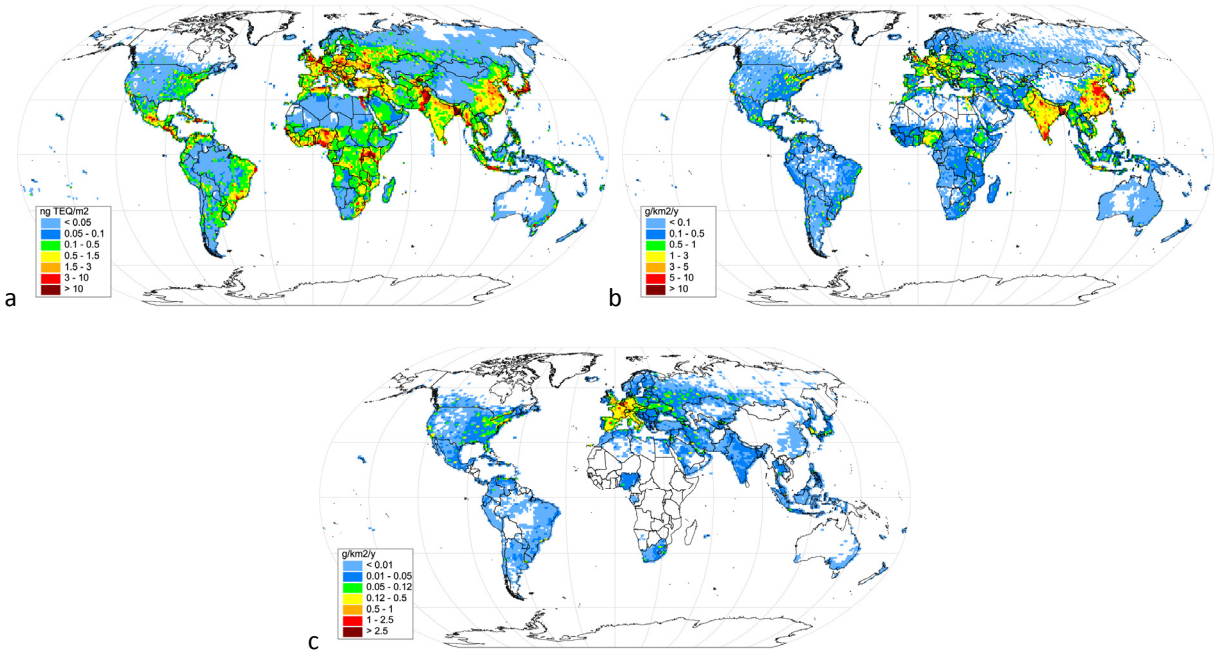


Fig. 4.2. *Spatial distribution of global annual emissions of PCDD/Fs, ng TEQ/m^2/y (a), HCB, g/km^2/y (b), and PCB-153, g/km2/y (c) with spatial resolution 1°x1°, used in model simulations for 2016.*

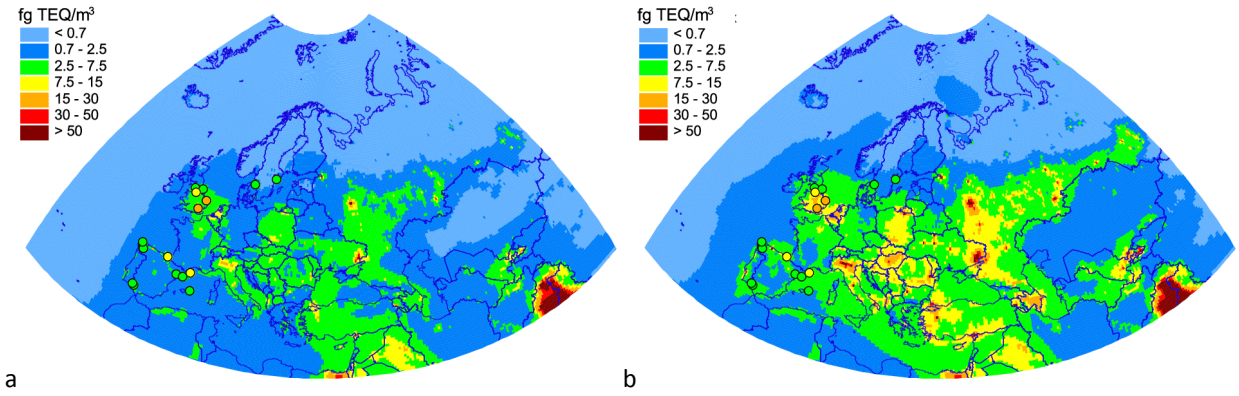
4.2. Pollution levels in the EMEP region

Assessment of PCDD/F, PCB-153, and HCB pollution levels in the new EMEP domain was performed using nested modelling approach. Model simulations were carried out for the global and regional EMEP domains. Results of global scale model runs for 2016 were used to derive lateral boundary conditions for regional model runs. Initial conditions for the evaluation of pollution levels in the EMEP region were prepared on the basis long-term spin-up global model runs. To characterize the spatial variability of PCDD/F pollution levels within the EMEP domain, analysis of modelling results and measurements of national monitoring networks has been carried out.

Regular monitoring of PCDD/F air concentrations is not currently performed at the EMEP network stations. At the same time, long-term measurements of PCDD/F content in air are carried by national monitoring networks in some of the EMEP countries (e.g. in the UK, Spain, and Portugal). In particular, continuous measurements of PCDD/F air concentrations as well as concentrations of some other POPs in the UK are carried out using the Toxic Organic Micro-pollutants Monitoring Network (TOMPs) starting from 1991 up to the present time. The network comprises 6 rural and urban monitoring sites in different parts of the UK. The Spanish Monitoring Program for several POPs including PCDD/Fs was established in 2008 in Spain to explore the effectiveness of existing regulations. Spatial and temporal trends in the observed PCDD/F air concentrations were analyzed using data of national background rural and urban monitoring sites for the period 2008-2015 [*Muñoz-Arnanz et al.*, 2018]. Studies of long-term trends in PCDD/F air concentrations were also carried out in Portugal for the period 2001-2014 [*Coutinho et al.*, 2015]. Along with long-term monitoring activities, concentrations of PCDD/Fs in

ambient air were analysed in the framework of various measurement campaigns in several EMEP countries (e.g. in Sweden, Denmark, and Italy).

In Figure 4.3 model predictions of annual mean PCDD/F air concentrations, calculated using officially reported emissions and maximum emission scenario, are illustrated. The latest available data on the observed PCDD/F air concentrations, namely, measurements for 2015 in Spain, and for 2016 in the UK and Sweden, are shown on the maps (Fig. 4.3).



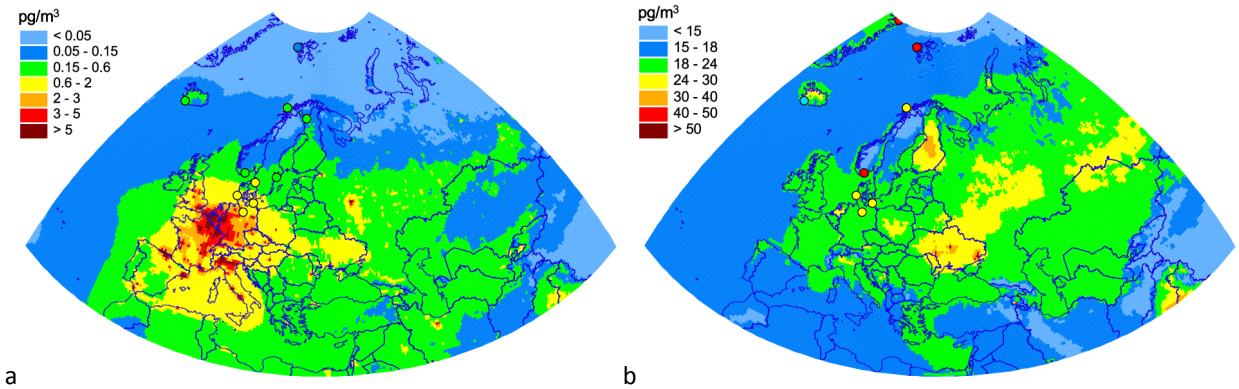
a b

Fig. 4.3. *Spatial distribution of modelled and observed annual mean air concentrations of PCDD/Fs (fg TEQ/m^3) calculated using official emissions (a) and maximum scenario emissions (b) in the EMEP domain for 2016.*

Model predictions with maximum emission scenario (Fig. 4.3b) provided higher levels of pollution in the EMEP countries comparing to the results of simulations with official PCDD/F emissions (Fig. 4.3a). It is seen that maximum scenario results have better agreement with measurements. Model simulations indicate elevated levels of dioxins and furans air concentrations (about 15 - 50 fg TEQ/m^3) for the UK, northern Italy, countries of Central and Eastern Europe as well as in the EECCA countries (e.g. the Russian Federation, Ukraine, Azerbaijan). Lower levels of pollution were estimated for France, Spain, Portugal, and countries of Northern Europe (about 1 - 7 fg TEQ/m^3).

Model estimates of annual mean PCB-153 and HCB air concentrations for 2016 are illustrated in Fig. 4.4a and 4.4b, respectively. Similar to PCDD/Fs, the maps include overlaid observed annual mean PCB-153 and HCB air concentrations, reported by the EMEP monitoring stations.

The highest levels of modelled PCB-153 annual mean air concentrations (3 - 5 pg/m^3 and higher) were estimated for the countries in Western Europe (e.g. Germany, France, Belgium). Simulated PCB-153 concentrations in Northern Europe were generally below 1 pg/m^3 that corresponded to the observed concentrations. Similar levels of pollution were also obtained by the model for Eastern Europe and the EECCA countries.



a b

Fig. 4.4. *Spatial distribution of modelled and observed annual mean air concentrations (pg/m^3) of PCB-153 (a) and HCB (b) in the EMEP domain for 2016.*

Modelling results for HCB showed low spatial variability of annual mean air concentrations, which can be explained by longer residence time in the atmosphere comparing to other considered POPs. Relatively higher levels of pollution were estimated for countries of Central and Eastern Europe (about 20 - 30 pg/m^3). For other areas model estimates were below 20 pg/m^3. Contrary to this, measurements of EMEP monitoring sites indicated more significant variability of HCB air concentrations. In particular, high annual mean concentrations (about 60 - 80 pg/m^3) were observed in Northern Europe and in the Arctic. It is seen that model simulations did not reproduce these high levels of HCB concentrations. Possible reasons of these differences are discussed below in the section related to the comparison of model predictions with measurements (Section 4.3).

4.3. Comparison of modelling results with measurements

Evaluation of PCB-153 and HCB modelling results for 2016 was carried out using measurements of air concentrations of the EMEP monitoring network. Model predictions of PCDD/F air concentrations were evaluated against available measurements of the national monitoring networks in the UK, Spain, and Sweden.

In Figure 4.5 scatter plots of annual mean modelled PCDD/F air concentrations, calculated using official emissions and maximum emission scenario, and measurements of national monitoring sites are presented. It is seen that model simulations with official emissions underestimated observed PCDD/F air concentrations with average bias equal to -51% (Fig. 4.5a). Significant part of model predictions deviated from measurements by more than a factor of 2 (about 60%) and a factor of 3 (about 50%).

Model predictions on the basis of maximum emission scenario showed better agreement with measurements. In particular, the value of average bias (NMB) improved significantly (from 51% to 7%) and values of F2 and F3 indicators increased. For both sets of modelling results the spatial correlation between the modelled and observed PCDD/F air concentrations was about 0.6.

The use of maximum levels of PCDD/F emissions led to improvement of agreement between the modelling results and measurements that could be considered as an indication of possible

underestimation of officially reported PCDD/F emissions. At the same time, amount of available observational data for the evaluation of PCDD/F model predictions was limited to several countries. Therefore, model predictions for other regions in the EMEP domain require further analysis with application of additional PCDD/F monitoring data.

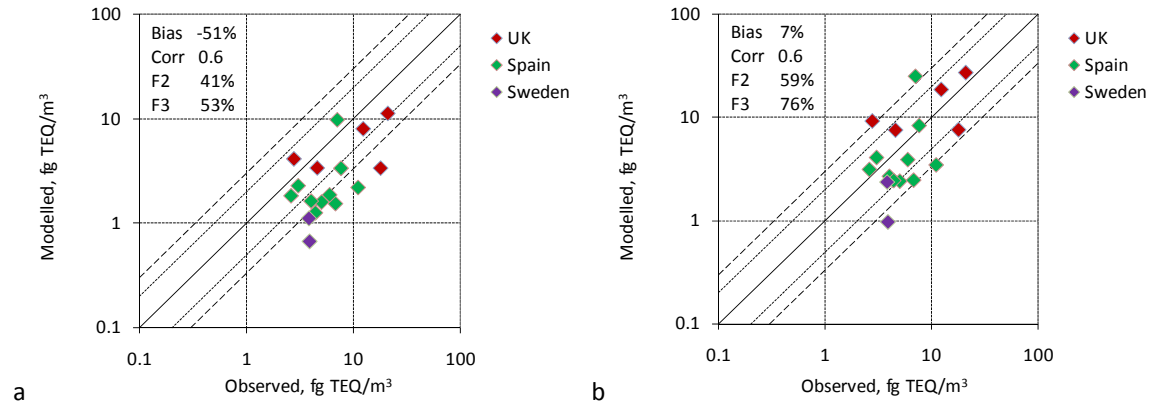


Fig. 4.5. *Comparison of annual mean modelled PCDD/F air concentrations, calculated using official emissions (a) and maximum scenario emissions (b), with measurements of national monitoring sites. Dashed lines indicate the area of agreement between the modelled and observed values within a factor of 2 and 3. NMB is normalized mean bias; R is the spatial correlation; F2 and F3 are fractions of sites for which deviation between modelled and observed values are within a factor of 2 and 3, respectively.*

Annual mean modelled PCB-153 air concentrations are compared with data of EMEP measurements in Fig. 4.6a. Monitoring of PCB-153 air concentrations in 2016 was carried out at 10 EMEP sites, located in Germany, Finland, Norway, Iceland, and Sweden (Chapter 1). ***Model predictions reasonably reproduced spatial distribution of observed PCB-153 levels in ambient air (spatial correlation coefficient was 0.75). For 60% of the monitoring sites the difference between measured and modelled concentrations was within a factor of 2. At the same time, the model tended to under-predict observed concentrations with average bias -23%.*** The most significant under-prediction of measured concentrations was obtained for the monitoring sites NO0042R, NO0090R, IS0091R, and SE0014R.

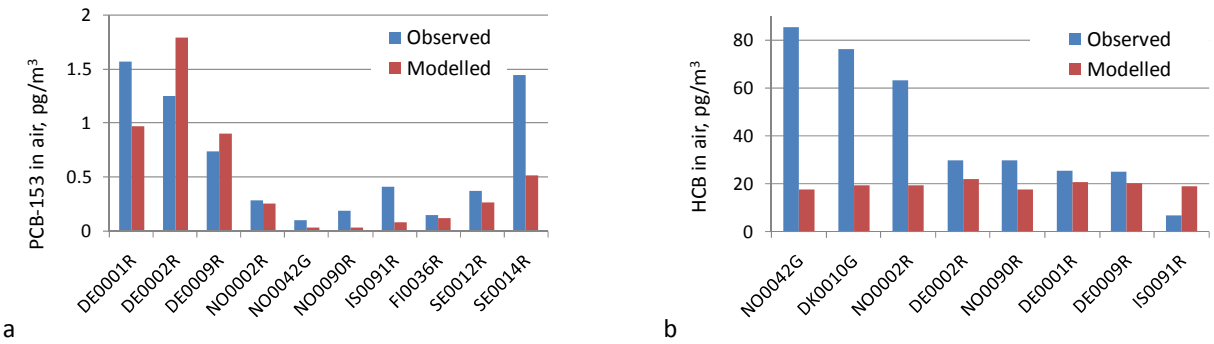


Fig. 4.6. *Comparison of annual mean modelled PCB-153 (a) and HCB (b) air concentrations with measurements of the EMEP monitoring sites for 2016.*

These deviations might be explained by the use of emission expert estimates in model simulations that might underestimate PCB emissions in some of the areas outside the EMEP domain. For instance, global scale model simulations of PCB-153 in this study were based on the emission inventory of *Breivik et al.* [2007], which estimated atmospheric emissions of intentionally produced PCBs. Refinement of emissions of intentionally produced PCBs in China, carried out by *Cui et al.* [2015], resulted in greater values of emissions for recent two decades comparing to the estimates of *Breivik et al.* [2007] which showed gradual decrease after 1970s. Furthermore, the study indicated importance of contributions of unintentionally produced PCBs from various industrial activities as well as from e-waste re-cycling in China. Thus, refinement of global scale model simulations for PCBs is needed taking into account new information on non-EMEP emissions in different regions of the globe. In addition to this, the deviations might be partly attributed to possible uncertainties of the model parameterizations of air-surface exchange and degradation in soil and seawater compartments. Hence, further analysis of obtained under-prediction of observed PCB concentrations is required in co-operation with national experts.

In Figure 4.6b annual mean modelled HCB air concentrations for 2016 are compared with the observed concentrations. Measurements of HCB air concentrations for 2016 were reported by 8 EMEP sites in Germany, Norway, Denmark, and Iceland. ***For most of the monitoring sites the model tended to under-predict observed HCB concentrations in air*** with the exception for IS0091R (for which observed concentrations were over-predicted). The most significant under-prediction (more than a factor of 3) is seen for the sites NO0042R and DK0010R in the Arctic region, and NO0002R in the southern part of Norway. Similar differences between the modelled and measured air concentrations for these sites were discussed in the previous Status Report [*Gusev et al.*, 2017].

In particular, ***under-prediction of observed HCB concentrations by the model was attributed to i) incomplete information on HCB emission sources in the inventories reported by the EMEP countries, ii) possible underestimation of HCB emissions in the regions outside the EMEP domain applied in model simulations, and iii) uncertainties of the model parameterizations for HCB degradation in media and air-surface exchange.*** Besides, measurements of HCB concentrations in ambient air can be subject to considerable uncertainties due to its high volatility ("break-through" effect).

Uncertainties in the HCB emissions, officially reported by the EMEP countries, were discussed during the recent meeting of the Task Force on Emission Inventories and Projections held in 2018 in Sofia (Bulgaria). It was stressed that uncertainties in the reported HCB emissions were still high due to the gaps in coverage of HCB emission sources in national inventories as well as inconsistencies in the methodologies and emission factors used for the evaluation of emissions by different countries. Furthermore, only minor amount of the countries reported HCB emissions due to the application of pesticides containing impurities of HCB (NFR sector 3Df). Thus, the need of further refinement of methodology for estimation of HCB emissions from this source category in the EMEP/EEA Guidebook was highlighted.

4.4. Transboundary transport of pollution

Transboundary transport and source apportionment of PCDD/F, PCB-153, and HCB deposition in the new EMEP domain for 2016 was estimated taking into account the following groups of emission sources, namely, anthropogenic emissions of the EMEP countries, non-EMEP emissions, and secondary emissions. The influence of emissions, located outside the EMEP domain, was evaluated using global-scale model simulations.

Relative contributions of these three groups of emission sources to annual PCDD/F, PCB-153, and HCB deposition in the EMEP countries are shown in Fig. 4.7. The highest contribution of the EMEP anthropogenic emissions was estimated for PCDD/Fs (46%), followed by PCB-153 (36%), and HCB (2%). In the particular EMEP countries these estimates varied for PCDD/Fs from 18% to 66%, for PCB-153 from 14% to 50%, and for HCB from less than 1% to 10%.

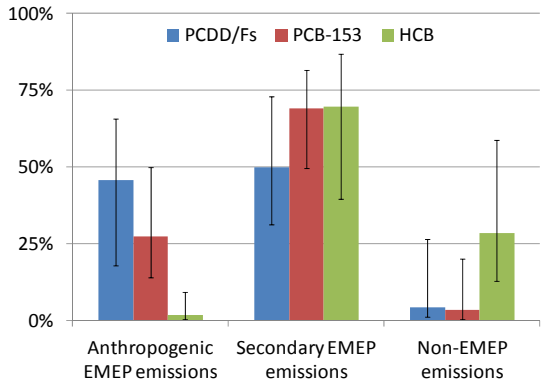


Fig. 4.7. *Contributions of three source groups (anthropogenic EMEP, non-EMEP and secondary emission sources) to deposition over the EMEP countries in 2016 for PCDD/Fs, PCB-153, and HCB. Whiskers denote the range of contributions across the EMEP countries.*

Secondary emission sources of PCDD/Fs, PCB-153, and HCB contributed to deposition 50%, 69%, and 70%, respectively. The contribution of non-EMEP emission sources of PCDD/Fs and PCB-153 was relatively low (about 3 - 4%), whereas for HCB it was estimated to almost 30%, varying from 13% to 59% in the particular EMEP countries.

Results of source apportionment of POP deposition in the EMEP countries are exemplified by PCDD/F modelling results for 2016, calculated using the maximum emission scenario (defined in the Section 4.1). Annual PCDD/F deposition fluxes to the EMEP countries are shown in Fig. 4.8. Elevated levels of PCDD/F deposition fluxes in the EMEP countries (above 5 ng TEQ/m^2/y) were estimated for Azerbaijan, Armenia, and Belgium. Significant deposition fluxes (about 4 ng TEQ/m^2/y) were also characteristic of the Netherlands, Poland, Albania, Turkey, and Slovakia. Relatively low levels of deposition (below 1 ng TEQ/m^2/y) were obtained for the Scandinavian countries as well as Iceland, Estonia, the Russian Federation, Kazakhstan, and Malta.

Long-range transport of PCDD/Fs from the emission sources of a particular country contributes to the deposition over the country itself and to the deposition to the territories of other countries. The ratio of PCDD/Fs emitted from anthropogenic emission sources and deposited within and outside the country's boundaries is shown in Fig. 4.9. ***Model evaluation of PCDD/F distribution in the EMEP domain indicated that long-range transport between the EMEP countries is an important source of pollution.*** As follows from the Figure, ***for 15 EMEP countries (29% of the countries) the fraction of PCDD/Fs, deposited to other EMEP countries is higher than the fraction, deposited to the country itself.***

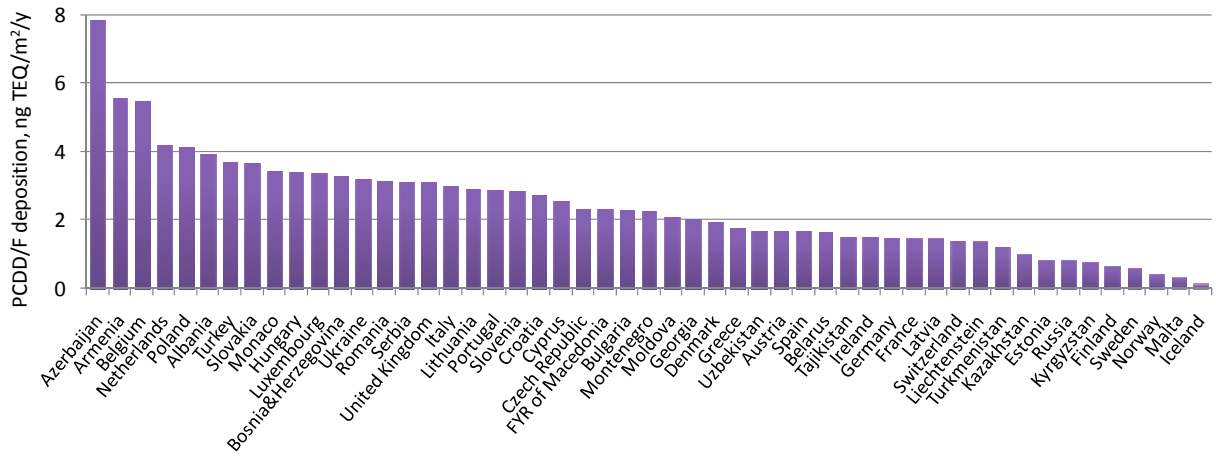


Fig. 4.8. *Annual deposition fluxes of PCDD/Fs in the EMEP countries calculated for 2016, ng TEQ/m^2/y.*

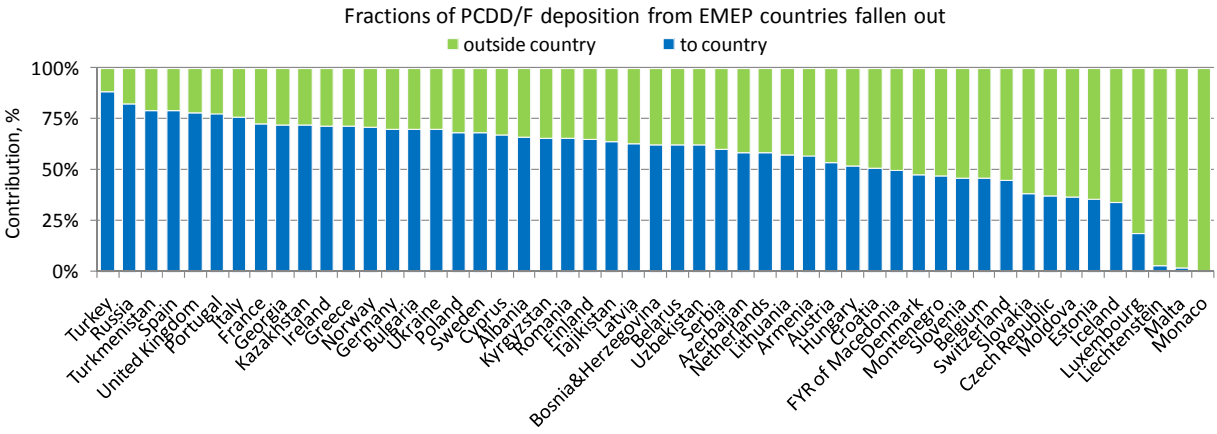


Fig. 4.9. *Fractions of PCDD/F deposition, originated from national emissions of the EMEP countries, fallen out to their own territories and outside their boundaries in 2016.*

According to model estimates of PCDD/F transboundary pollution, the contributions of emission sources, located outside the boundaries of a particular country, to deposition over this country were higher than the contribution of its own national emissions for 28 EMEP countries (55% of the countries) (Fig. 4.10). Results of nested model simulations provided also information on contributions of non-EMEP emission sources to the pollution levels in the EMEP region. These contributions for particular EMEP countries were in the range from 3% to 69% with the highest contribution estimated for Iceland.

Results of model simulations for 2016 pointed out that the largest contributions in absolute values to the PCDD/F deposition in the EMEP countries due to transboundary transport from anthropogenic emission sources were made by the Russian Federation followed by Ukraine, Azerbaijan, Kazakhstan, and Romania.

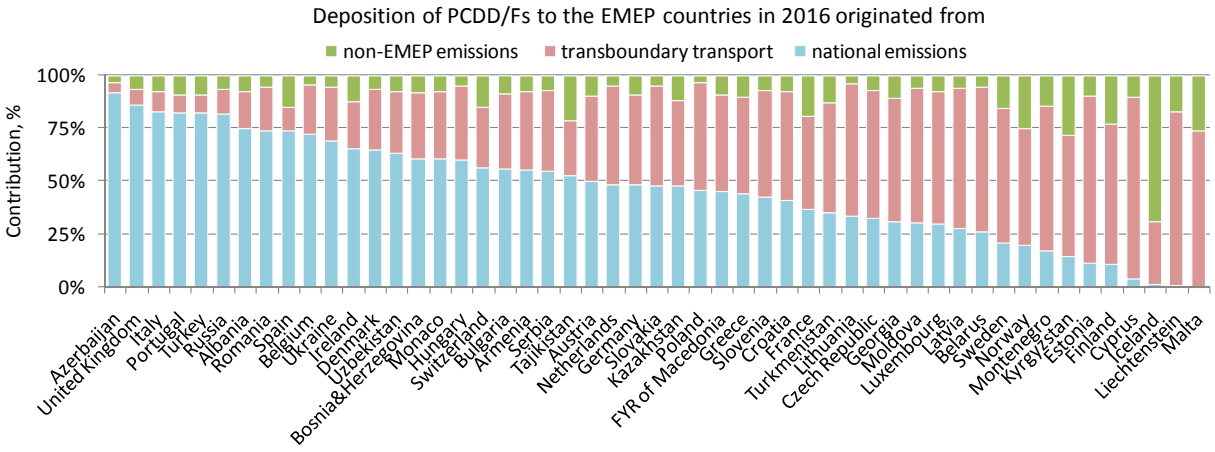


Fig. 4.10. *Relative contributions of national emissions, transboundary transport, and non-EMEP emissions to deposition of PCDD/Fs from anthropogenic sources in the EMEP countries in 2016.*

4.5. Pollution on global scale and in the Arctic region

Evaluation of global scale transport and pollution of PCDD/Fs, PCB-153, and HCB for 2016 was carried out using the GLEMOS modelling system with application of emission expert estimates (Section 4.1). Results of global scale model simulations were used to estimate lateral boundary concentrations for regional EMEP modelling. Besides, these model predictions provide information on the intercontinental transport and pollution of remote areas like the Arctic region.

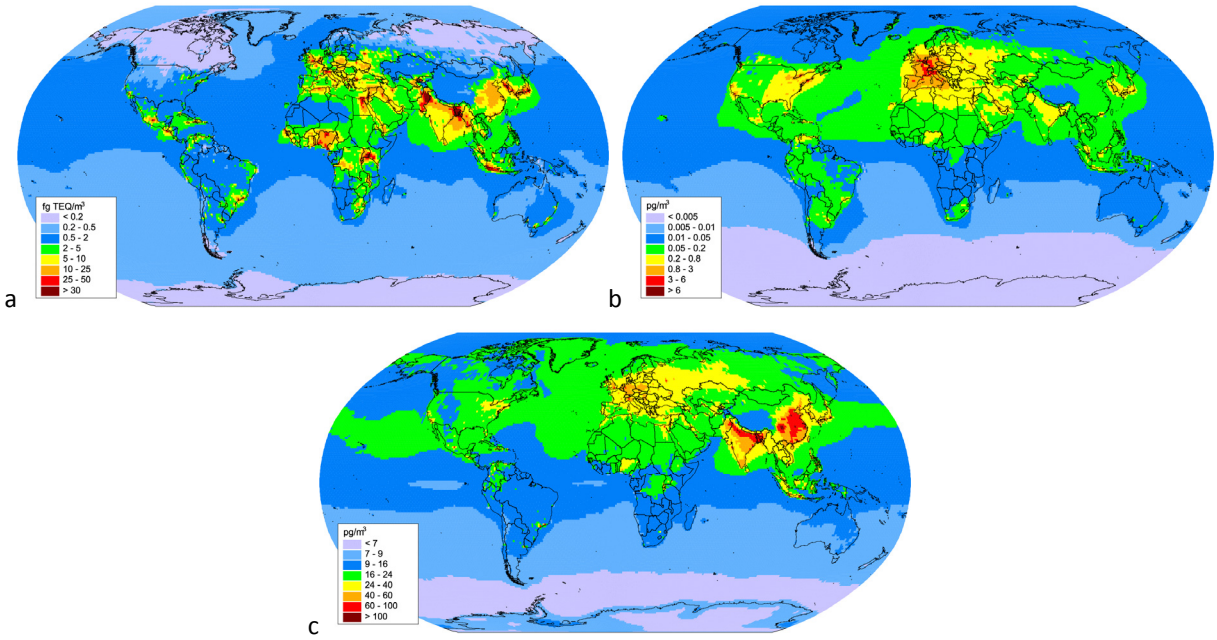


Fig. 4.11. *Spatial distribution of global scale annual mean air concentrations of PCDD/Fs, fg TEQ/m^3 (a), PCB-153, pg/m^3 (b), and HCB, pg/m^3 (c) simulated for 2016.*

Spatial distributions of global-scale annual mean PCDD/F, PCB-153, and HCB air concentrations simulated for 2016 are presented in Fig. 4.11. The highest levels of PCDD/F air concentrations were estimated for Africa and South Asia (25 - 50 fg TEQ/m^3), while levels of pollution in Europe, North and South America, and Australia were lower (1 - 10 fg TEQ/m^3). For PCB-153 elevated air concentrations were obtained for the European region (0.6 - 6 pg/m^3). For other regions less significant air concentrations were estimated (below 0.2 pg/m^3). Model predicted high annual mean HCB air concentrations for Eastern and Southern Asia (50 - 100 pg/m^3), and low concentrations for the European countries (about 20 - 40 pg/m^3). Simulated annual mean air concentrations in the Arctic region varied mostly within the range of 0.1-1 fg TEQ/m^3 for PCDD/Fs, 0.01 - 0.1 pg/m^3 for PCB-153, and 8 - 24 pg/m^3 for HCB. However, it should be noted that the model tended to underestimate levels of PCB-153 and HCB concentrations in the Arctic region (Section 4.3).

Results of regional scale model simulations for the Arctic region are illustrated in the Fig. 4.12a on the example of modelled PCDD/F deposition. As seen from the Figure, relatively high deposition fluxes were estimated for Iceland, and northern parts Scandinavian countries and Russia while lower fluxes were obtained for other areas in the Arctic region.

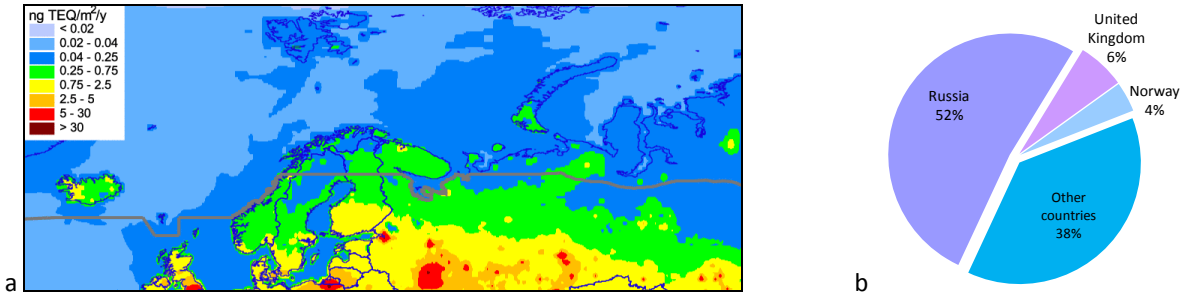


Fig. 4.12. *Annual total PCDD/F deposition fluxes in 2016 (a) and relative contributions of anthropogenic emission sources of the EMEP countries to deposition (b) over the Arctic region, covered by the EMEP domain. Grey line denotes the boundary of the AMAP domain*

Source apportionment of PCDD/F pollution in the Arctic was carried out taking into account contributions of EMEP anthropogenic, EMEP secondary and non-EMEP sources. According to model estimates, EMEP anthropogenic sources of PCDD/Fs contributed to deposition levels in the Arctic 27%. Substantial contributions were also made by non-EMEP emissions (30%) and secondary emissions (42%).

Results of source apportionment of PCDD/F deposition from anthropogenic emissions of the EMEP countries is illustrated in Fig. 4.12b. It is seen that the largest contribution (52%) was made by Russian emission sources followed by the United Kingdom (6%) and Norway (4%). The other EMEP countries contributed about 38%.