PILOT MODELLING OF PCDD/F TRANSPORT AND FATE ON GLOBAL SCALE AND WITHIN THE EUROPEAN REGION

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Introduction

Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) are highly toxic pollutants characterized by significant persistence in the environment. Reduction of pollution by PCDD/Fs is in the scope of the activities of many international organizations (UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP), UNEP Stockholm Convention (SC), Arctic Monitoring and Assessment Program (AMAP), OSPAR Convention, and Helsinki Commission (HELCOM)). Dioxins and furans can be released to the environment by various emission sources including anthropogenic and secondary emissions (re-volatilization from environmental media). They can remain to be hazardous to human health and ecosystems even at low concentrations long after the moment they were originally emitted. Characterization of spatial and temporal variations of their levels in the environment is of importance for the evaluation of health risks. Assessment of pollution levels of PCDD/Fs requires taking into account their distribution between the environment compartments and application of multi-media modeling approach.

The aim of this study was to provide preliminary estimates of global scale dispersion and cycling of dioxins and furans in the environment and to characterize their intercontinental transport with emphasis to the European region. Pilot model simulations were performed using the Global EMEP Multi-media Modeling System (GLEMOS) and emission scenario constructed on the basis of the UNEP SC inventory of dioxins and furans emissions. Modeling results were evaluated against available measurements of PCDD/F air concentrations in rural and remote regions.

Materials and methods

Description of the GLEMOS modeling system. The GLEMOS is a multi-scale multi-pollutant simulation platform that allows evaluation of dispersion and cycling of different pollutants (e.g. heavy metals (HMs), POPs) in the environment with a flexible choice of the simulation domain (from global to local scale) and spatial resolution. This modeling system is being developed within the EMEP program under LRTAP Convention for the evaluation of HM and POP pollution. Development of the modeling system is documented in a series of reports[1,2,3,4,5]. A modular architecture of the GLEMOS allows flexible configuration of the model set-up for particular research task and pollutant properties. The set of modules related to modeling of POPs is largely based on the previously developed and tested hemispheric/regional chemistry transport model MSCE-POP[6,7]. Model parameterization of physical-chemical properties for dioxins and furans is described in technical report[7]. Current version of the GLEMOS considers the cycling of POPs between the atmosphere, soil, seawater, and vegetation including description of the following processes: three-dimensional transport in the atmosphere and ocean water, vertical transport in upper soil layer, sedimentation in seawater, phase partitioning and degradation in media as well as inter-media exchange.

Scenario of global PCDD/F emissions. Global PCDD/F emission scenario for modeling was constructed on the basis of the information on dioxins and furans releases compiled under UNEP SC[8,9]. National inventories of annual PCDD/F emissions were available for 68 countries representing the level of emissions during the recent decade. Analysis of this information[10] permitted to reveal correlation between the intensity of PCDD/F emissions in the countries and their economic status. Obtained regression relationship between the national PCDD/F emissions and the data on GDP and total population of the countries was applied to estimate emissions

for the other countries, for which the information on emissions was not available. Compiled inventories covered main modes of entry of emissions to the environment (the atmosphere, land, water, residues, and products), among which major releases were indicated to the atmosphere (45%). Releases of PCDD/Fs to the atmosphere and land compartments were taken into account in the constructed scenario for modeling, while other vectors were neglected. Spatial distribution of PCDD/F emissions was generated using the population density as a surrogate (Fig. 1).

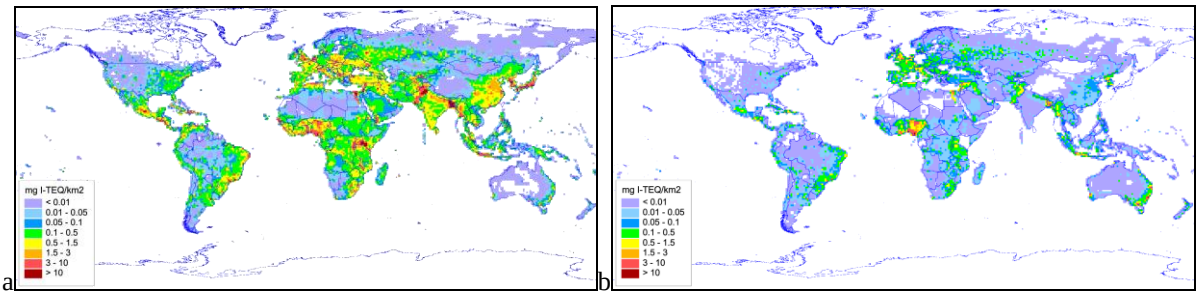


Fig. 1. *Spatial distribution of annual PCDD/F emissions (mg I-TEQ/m^2/y) to the atmosphere (a) and to soil (b) constructed on the basis of UNEP global PCDD/F emission inventory.*

Pilot model simulations were performed using the meteorological data for 2012 and spatial resolution 1° x 1°. In order to set up initial conditions the spin-up model run for the period of four decades was carried out to achieve observed levels of concentrations in rural and remote regions.

Results and discussion

Global transport and fate of dioxins and furans was evaluated using the GLEMOS modeling system and constructed emission scenario based on the PCDD/F emission inventory of the UNEP SC. Spatial distribution of PCDD/F pollution levels is presented in Fig. 2. Elevated air concentrations were characteristic of Europe, Southern and Eastern Asia, and Africa, while relatively lower levels of pollution were indicated for North and South America and Australia.

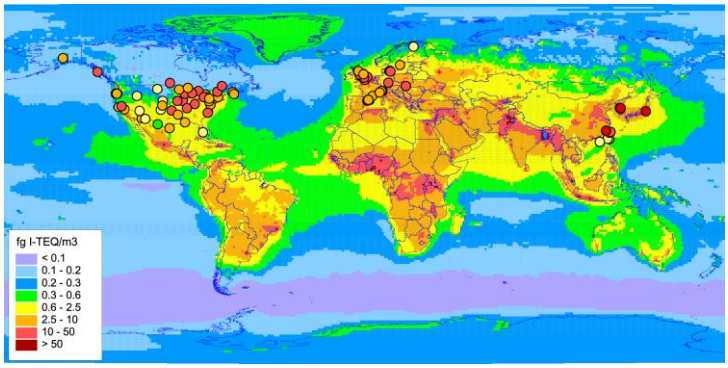
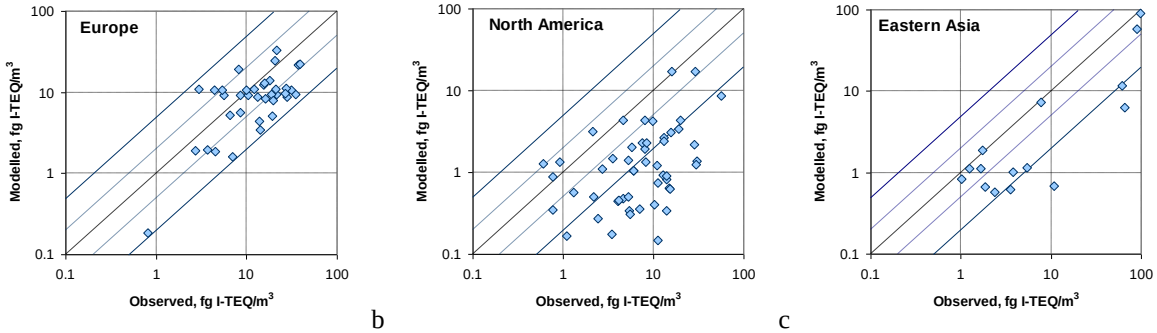


Fig. 2. *Spatial distribution of modelled annual mean PCDD/F air concentrations (fg I-TEQ/m^3) in comparison with measurements made in rural and remote regions (locations of measurements are illustrated as circles).*

Results of pilot model simulations were evaluated against available measurements of PCDD/F air concentrations performed in rural and remote areas performed during the recent decade (Fig. 2 and Fig. 3). Compiled information on observed PCDD/F air concentrations includes results of measurements undertaken in European countries (Denmark, France, Germany, the United Kingdom, Italy, Sweden, Finland, the Netherlands, and Spain), in North America, and Eastern Asia[11]. In addition, measurements made by NDAMN network[12,13] and in South Korea[14] and around Taiwan[15] were included. Though this dataset comprises the measurements made using

different equipment and methodology, it is used for the preliminary evaluation of modeling results to provide initial estimates of the agreement between the modeled and observed PCDD/F concentrations.



a b c

Fig. 3. *Comparison of modelled annual mean PCDD/F air concentrations (fg I-TEQ/m^3) with measurements performed in Europe, North America, and Eastern Asia. Dashed lines denote the area of agreement within a factor of 2, solid lines – a factor of 5.*

Comparison of pilot modeling results with measurements indicated reasonable agreement between the modeled and observed PCDD/F air concentrations for European countries. More than a half of modeled values were within a factor of 2 with measurements, and about 80% were within a factor of 3. For other regions level of agreement was somewhat lower. In general, the model tended to underestimate observed air concentrations with more significant deviations for rural areas of North America, which was likely caused by the uncertainties of the constructed emission scenario for this region. At the same time, better agreement of model predictions was found with measurements made in remote areas in North America.

Pilot model simulations with the experimental scenario of global PCDD/F emissions permitted to estimate relative contributions of emissions of different source groups to the pollution of the European region (Fig. 4).

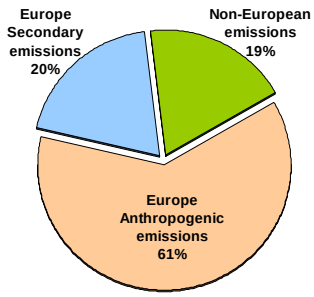


Fig. 4. *Relative contributions of emissions of different source groups to the pollution of the European region.*

According to preliminary modeling results levels of PCDD/F air concentrations in Europe were largely formed by anthropogenic emissions and secondary releases (re-volatilization from environmental compartments) of the European countries. Contributions of these sources were estimated to approximately 60% and 20%, correspondingly. At the same time, significant contribution to the pollution levels belonged to the intercontinental transport (about 20%). Thus, distant emission sources located outside the European region can essentially contribute to the pollution of European countries by dioxins and furans. To improve evaluation of PCDD/F pollution in Europe and on the global scale further refinement of global emission inventory for dioxins and furans is needed.

References:

1. Tarrasón L, Gusev A. (2008);EMEP/MSC-W Technical Report 1/2008. (emep.int/publ/reports/2008/emep_technical_1_2008.pdf)

2. Travnikov O, Jonson JE, Andersen AS, Gauss M, Gusev A, Rozovskaya O, Simpson D, Sokovyh V, Valiyaveetil S, Wind P. (2009); EMEP/MSC-E Technical Report 7/2009. (www.msceast.org/reports/7_2009.pdf).
3. Jonson JE and Travnikov O. (2010); EMEP/MSC-W Technical Report 1/2010. (emep.int/publ/reports/2010/emep_technical_1_2010.pdf)
4. Travnikov O, Jonson JE. (2011); EMEP/MSC-E Technical Report 1/2011. (www.msceast.org/reports/1_2011.pdf)
5. Jonson JE and Travnikov O. (2012); EMEP/MSC-W Technical Report 1/2012. (emep.int/publ/reports/2012/emep_technical_1_2012.pdf)
6. Malanichev A, Mantseva E, Shatalov V, Strukov B, Vulykh N. (2004); *Environ Poll*. 128(1-2): 279-289.
7. Gusev et al., (2005); EMEP/MSC-E Technical Report 5/2005. (www.msceast.org/reports/5_2005.pdf)
8. Fiedler, 2007; *Chemosphere* 67: S96–S108
9. Fiedler H, Cao Z, Huang J, Wang B, Deng S, Yu G. (2012); *Organohalogen Comp.* 74: 1521-1524.
10. Cao Z, Fiedler H, Wang B, Zhang T, Yu G, Huang J, Deng S. (2013); *Chemosphere* 91: 328-335
11. Shatalov V, Ilyin I, Gusev A, Rozovskaya O, Sokovykh V, Travnikov O, Vulykh N, Wiberg K, Cousins I. (2012); EMEP/MSC-E Technical Report 4/2012.
12. Larber M, Ferrario J, Byrne C. (2013) ; *Atmospheric Environment* 77: 311-317.
13. U.S. Environmental Protection Agency (EPA), (2013); EPA/600/R-13/183F. (epa.gov/ncea)
14. Min Y, Lee M, Kim D, Heo J. (2013) ; *Atmospheric Environment* 77: 222-230.
15. Chi KH, Lin CY, Wang SH, Lin NH, Sheu GR, Lee CT. (2013) ; *Atmospheric Environment* 78 : 203-210.