EXECUTIVE SUMMARY

Heavy metals and persistent organic pollutants (POPs) are toxic substances targeted by national and international environmental organizations and regulated by international agreements including the UNECE Convention on Long-range Transboundary Air Pollution (hereafter, CLRTAP or the Convention). Protocol on Heavy metals and Protocol on POPs to the Convention, aimed at reduction of emissions of these pollutants to the atmosphere, were adopted in 1998 and amended in 2012 and 2009, respectively. According to the Protocols, the priority heavy metals and POPs are lead (Pb), cadmium (Cd) and mercury (Hg), polychlorinated biphenyls (PCBs), polychlorinated dibenzo(p)dioxins and dibenzofurans (PCDD/Fs), hexachlorobenzene (HCB) and polyaromatic hydrocarbons (PAHs). The considered PAHs comprise 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 (I(cd)P). According to the amendments made in 2009 a number of Contaminants of Emerging Concern (CECs) were also included to the POP Protocol.

Scientific support of the implementation of the Protocols is carried out by the co-operative Programme for Monitoring and Evaluation of Long-range Transmission of Air Pollutants in Europe (EMEP, www.emep.int). EMEP activities on emission data, results of monitoring activity, model assessment of heavy metal and POP pollution, research and model development as well as cooperation with subsidiary bodies to the Convention, national and international organizations is described in the report. More detailed information is available in the Technical reports [*Ilyin et al.*, 2022; *Aas et al.*, 2022; *Pinterits et al.,* 2021; *Schindlbacher et al.*, 2021] and the Supplementary Data Reports on heavy metals [*Strizhkina et al.,* 2022a] and POPs [*Strizhkina et al.*, 2022b] as well as on the websites (www.msceast.org; https://projects.nilu.no/ccc/reports.html; https://www.ceip.at/).

Emissions

Completeness and consistency of submitted data have improved significantly since EMEP has been collecting information on emissions. In 2022 the emission data on heavy metals and POPs were submitted by 47 (92%) Parties. The quality of submitted data across countries differs quite significantly. Uncertainty of reported data (national totals, sectoral data) is considered to be relatively high. The completeness of reported data is not satisfactory for all pollutants and sectors either.

Analysis of the long-term heavy metal and POP emission trends from 2000 to 2020 demonstrated that temporal variability of the emissions differs significantly in the western and eastern parts of the EMEP domain. In the western part the reduction of PCDD/Fs, HCB , PCB and PAHs made up 61, 96, 59 and 16%, respectively. The reduction of heavy metal emissions was 47% (Cd), 59% (Hg) and 76% (Pb). The trends of heavy metal and POP emissions in the eastern part of the EMEP domain are inconsistent, mainly due to incomplete reporting.

“Industry” sector is the main contributor of Cd, Hg, Pb and PCB in the EMEP region, making up 39 – 83% to total emission. For PAHs (76%) and PCDD/Fs (47%) most of emissions are presented by the sector “Other stationary combustion”. For HCB the sectors “Public electricity and heat production”, “Industry”, “Other stationary combustion” and “Other emissions from agriculture” have comparable proportions (22 – 28%) to the total emissions. For most of the considered pollutants emissions in the eastern part of the EMEP domain are characterized by higher contribution of sector “Public electricity and heat production” compared to the emissions in the western part.

Emission data for modelling were gridded over latitude-longitude grid based on the gridding system developed by CEIP. If no reported gridded data is available, data of Global Unintentional POPs Emissions data from the Emission Database for Global Atmospheric Research or particulate matter data from the Copernicus Atmospheric Monitoring Service were used as proxies. Final emission data sets for modelling were generated by MSC-E based on the inventories provided by CEIP and additional information on temporal variability, vertical distribution and chemical speciation of emissions. Global emission data were also prepared by MSC-E using the results of the related research projects and expert estimates.

Emission data for modelling were gridded over latitude-longitude grid based on the gridding system developed by CEIP. If no reported gridded data is available, relevant data from various research projects were used as proxies. Final emission data sets for modelling were generated by MSC-E based on the inventories provided by CEIP and additional information on temporal variability, vertical distribution and chemical speciation of emissions. Global emission data were also prepared by MSC-E using the results of the related research projects and expert estimates.

Monitoring

In 2020, there were 40 sites measuring heavy metals of the first and the second priority in both aerosols and precipitation, and altogether there were 67 measurement sites. Mercury in air or precipitation was measured at 27 sites. POP monitoring data were reported by 39 stations, and measurements in both air and precipitation were available at 25 stations. The data on heavy metals were reported by 21 Parties to the Convention, and the data on POPs by 17 Parties. The highest concentrations of heavy metals in air are mainly seen along the coast and especially around the English Channel. Relatively high levels of the concentrations in precipitation generally occur in Eastern Europe. For POPs higher levels are noted for the central and eastern parts of Europe while lower levels take place in the northern part.

Analysis of measured long-term trends of HCB revealed that air concentrations of HCB demonstrate very slow decline or even increase. The reasons for this remain poorly understood. Various factors possibly affecting the levels and long-term tendencies such as re-volatilization from previously contaminated surfaces, seasonal variability, peculiarities of atmospheric transport patterns and differences in sampling and analytical methodologies were discussed.

Status of heavy metal and POP pollution in 2020

Heavy metal (Pb, Cd, Hg) and POP (PAHs, PCDD/Fs, HCB, PCB-153) atmospheric pollution, in 2020 was analyzed for the whole the EMEP region, sub-regions and individual countries. The largest pollution levels of most contaminants are found in Central Europe. This region is characterized by the highest mean deposition fluxes of Pb, Cd, Hg and PAHs and significant pollution levels for PCDD/Fs, PCB-153 and HCB. Northern Europe and Caucasus and Central Asia are the regions with the lowest average pollution levels. According to the estimates, pollution changes between 2019 and 2020 caused by meteorological variability did not exceed ±15% on average for all considered contaminants in all the sub-regions.

Evaluation of the modelling results against observations was carried out for air concentrations and wet deposition fluxes available from the EMEP monitoring network. At most of the stations the modelled air concentrations of Pb and Cd and wet deposition fluxes of Pb, Cd and Hg agree with the observed levels within a factor of two. The model tends to overestimate air concentrations of Pb and Cd and underestimate their wet deposition fluxes. Modelled air concentrations of Hg match the observed levels within ±15%, and somewhat overestimate wet deposition fluxes. Evaluation of PAH, HCB and PCB-153 estimates demonstrate that the absolute value of mean bias between modelled and observed concentrations is below 14%. At the majority of stations the modelled and observed concentrations of PAHs agree within a factor of two. The model matches observed air concentrations of HCB, PCB-153 and PCDD/Fs within a factor of two at about half of the stations.

Deposition fluxes of heavy metals and POPs within the EMEP region are formed by the EMEP anthropogenic emissions, secondary emissions (wind re-suspension of dust particles containing Pb and Cd, natural/legacy emissions of Hg, re-emission of POPs), and emissions from sources located outside the EMEP region (non-EMEP sources). Contributions of anthropogenic and secondary sources to Pb deposition in the EMEP region are comparable, while in case of Cd the anthropogenic component prevails. The contribution of non-EMEP sources to Pb and Cd deposition is generally relatively low, except for Southern Europe and Caucasus and Central Asia. For Hg the contribution of non-EMEP sources exceeds that of the EMEP anthropogenic sources, while the contribution of secondary emissions from the EMEP domain is minor. Secondary sources are the main contributors to deposition of PCDD/Fs and PCB-153, and are the predominant source of HCB pollution in the EMEP region. The contribution of non-EMEP sources of these POP pollutants is relatively low.

In order to support the activities of the Working Group on Effects (WGE) on evaluation of adverse effects of heavy metal pollution on human health and biota, ecosystem-specific deposition fluxes of Pb, Cd and Hg were calculated. It was shown that deposition flux to high vegetation land-cover types (e.g., forests) is several-fold higher than that to low-vegetation types. Besides, the contribution of dry deposition to total deposition in forest is higher than that in other ecosystems. It is worth to note that an update of the critical loads estimates is needed to evaluate the present-day effect of the pollutants on the environment.

Results of the model simulations and measurements of PAH pollution levels in the EMEP domain were used to evaluate exposure of population to high levels of concentrations exceeding air quality guidelines. Model estimates show that about 8% of the EMEP countries population in 2020 resided in areas with annual mean B(a)P air concentrations exceeded the EU target level. The upper assessment thresholds (UAT) and lower assessment thresholds (LAT) values were exceeded in the areas with about 15% and 25% of the population, respectively. The WHO Reference level was exceeded for about 66% of population of the EMEP countries.

However, if the mixture of the four priority PAHs is considered, the percentage of population resided in areas, where the threshold levels are exceeded, is higher (e.g., 11% for the EU target value and 75% for the WHO reference level).

Atmospheric loads to the Arctic and to the marginal seas (the North, Baltic, Black, Mediterranean and Caspian) were also assessed. The assessment includes information on deposition levels and source attribution of heavy metals and POPs in 2020. Besides, results of the model simulations of the pollution on a global scale are shown. These simulations are used to estimate contribution of non-EMEP sources to heavy metal and POP pollution levels in the EMEP countries. However, inventories of heavy metals and POPs global scale emissions are subject of considerable uncertainties. Thus, improvement of the global-scale assessment requires additional efforts on development of global emissions inventories for heavy metals and POPs in co-operation with other international bodies (UN Environment, Stockholm Convention, Minamata Convention, etc.).

Research and development

Multi-model analysis of B(a)P pollution levels in Europe is being performed as a part of the joint TFMM/EuroDelta-Carb intercomparison exercise of national experts and MSC-E. Main objectives of the study on B(a)P are to explore the model performance, analyze sources of uncertainties of modelling results, and to contribute to the refinement of B(a)P emissions from the combustion of fossil fuel and biomass burning. Preliminary analysis of modelling results demonstrates generally reasonable level of agreement of model predictions with observed B(a)P concentrations. At the same time, for some of the stations modelled concentrations significantly deviated from the observed values indicating possible effect of uncertainties in emission estimates, modelling approaches, and measurements. It is planned to perform more detailed analysis to explore the reasons of the differences between the output of participating models, the deviations of the model predictions from observed B(a)P concentrations, and to compare other characteristics and parameters (e.g. B(a)P concentrations in precipitation and deposition fluxes, concentrations of species affecting B(a)P chemical transformations in the atmosphere).

Contaminants of emerging concern (CECs) comprise a wide range of substances having potential to adversely affect wildlife and human health. Some of the CECs are receiving increasing attention in many international and national environmental organizations. Selected CECs were added to the CLRTAP POP Protocol for regulation of their production and use, namely, hexachlorobutadiene (HCBD), octabromodiphenyl ether (octa-BDE), pentachlorobenzene (PeCB), pentabromodiphenyl ethers (PBDEs), perfluorooctane sulfonates (PFOS), polychlorinated naphthalenes (PCN) and short-chain chlorinated paraffins (SCCPs). Ambient concentrations of CECs (e.g. of PBDEs, SCCPs, and PFAS (per- and polyfluoroalkyl substances)) are measured at the EMEP monitoring stations and at a number of sites of national monitoring networks. Furthermore, preparatory work for evaluation of CEC pollution levels, transport and fate in the environment is carried out.

Global character of Hg dispersion leads to contamination of terrestrial and aquatic ecosystems of the high latitude regions and the northern countries. A detailed study of Hg pollution on a country scale was performed for Norway as a part of the Norwegian Mercury Assessment 2022 (NMA 2022). The study was carried out in close collaboration with national experts and involving detailed national data on observations and emissions. Elevated Hg deposition levels were found in the southern part of the country and along the

coast including the most northern areas, whereas low levels occur in the inland territories of the country. Mercury deposition to the country underwent relatively small reduction over past 30 years because of significant contribution of global emissions, which have been slowly changing. The study could useful for the effectiveness evaluation of national and international pollution reduction measures. Besides, further analysis of deviations between the modelling results and observations can help further improving the modelling approaches.

PAH pollution assessment on a national scale was continued in the framework of a country-scale study for Poland. The second phase of the study was focused on the model assessment of B(b)F, B(k)F, and I(cd)P. In particular, analysis and update of model parameterizations for these compounds was carried out. The GLEMOS model was used to evaluate previous and updated national emission inventories of these PAHs. Results of model simulations showed that updated national PAH emission inventory (reported to EMEP) allowed to achieve improvement of pollution assessment accuracy for B(b)F and B(k)F. At the same time, no improvements were seen in case of I(cd)P. On the whole, the study showed that the modelling results still tend to underpredict observed air concentrations of PAHs in the country. Thus, further activities to refine emission estimates and to reduce uncertainties of the model parameterizations for PAHs are of importance.

In order to evaluate the effect of wildfires on heavy metal and POP pollution levels, development of a model parameterization for wildfire emissions was initiated. Available databases on wildfire parameters were analyzed and pilot estimates Pb and B(a)P emissions from wildfires in the EMEP region were carried out. Analysis of the preliminary results showed that the annual mean contribution of wildfires to overall emissions and pollution levels of Pb and B(a)P is relatively low. Nevertheless, in particular countries and in certain months the wildfires can be significant contributors to the Pb and B(a)P pollution levels in the EMEP region. Further research will include sensitivity studies, involvement of wider range of measurement data for verification purposes and testing of the proposed approach to other heavy metals and POPs.

Cooperation

Main results of MSC-E in the field of research and model development were discussed at the virtual meeting of the Task Force on Measurements and Modelling (TFMM). In particular, participation of MSC-E in the assessment of Hg pollution of the Arctic and the results of the Hg case study for Norway were discussed. Besides, the parameterization of heavy metal and POP emissions from wildfires and preliminary results of the model simulations using the wildfire emissions were demonstrated. State-of-the-art information about emissions, monitoring and modelling of CEC was overviewed. Finally, current results of the EuroDelta-Carb project and the country-scale study of PAH levels in Poland were presented.

MSC-E continues co-operation with the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) on assessment of Hg and POP pollution. In particular, the Centre took part in the TF HTAP virtual meetings focused on global emissions and modelling of Hg (May 18, 2022) and POPs/CEC (May 25, 2022). The meetings were aimed at discussion of practical programs for fulfilment of the near-term plans of Hg and POP activities formulated at the TF HTAP Workshops (April 2021). It was agreed that near-term TF HTAP activities on Hg would benefit, if formulated in line with the on-going effectiveness evaluation procedure performed under the Minamata Convention on Mercury (MCM). It was concurred to consider

opportunities of multi-pollutant studies of combustion-related POPs (e.g. PAHs, PCDD/Fs) and PM, and to continue reviewing the progress in the studies of CECs and microplastics on global and regional scales.

Long-term changes of heavy metal and POP deposition in comparison with observed concentrations of these pollutants in mosses and measured levels at the EMEP monitoring stations was analyzed in cooperation with ICP-Vegetation of WGE. The presented analysis showed that combined usage of the results of biomonitoring, the EMEP atmospheric monitoring and the modelling improves investigating long-term heavy metal and POP pollution changes in particular countries as well as at local scale. Results of the modelling and biomonitoring demonstrate similar rates of Pb, Cd, Hg and Cu reduction in most of EMEP countries. The moss measurements, observations at the EMEP monitoring stations and the modelling results demonstrate declining trends of BDE-99 levels and minor changes of PCB-153. Results of the moss surveys are important for evaluation of long-term trends of heavy metals and POPs in the EMEP region. Further cooperation between EMEP and ICP-Vegetation is highly appreciated.

MSC-E completed assessment of atmospheric loads of Pb, Cd and Hg to the marine regions of the North-East Atlantic under support of the OSPAR Commission. Long-term deposition trends for the period from 1990 to 2019 were calculated for each OSPAR maritime region and source-receptor relationships were estimated for 1995, 2005 and 2015. The modelling results were evaluated against observed data from the OSPAR Comprehensive Atmospheric Monitoring Programme (CAMP). It was shown that current estimates of long-term trends were comparable with the results of the previous studies. The results of this work were summarized in the Technical Report [*Ilyin et al*., 2022].

Furthermore, MSC-E continued cooperation with Task Force on Technical and Economic Issues (TFTEI), HELCOM and Stockholm Convention. MSC-E took part in the 7th TFTEI annual meeting. Updated results on long-term changes in measured and modelled B(a)P air concentrations had been demonstrated with emphasis on key emission source categories and exceedances of B(a)P air quality guidelines and population exposure. MSC-E took part in the preparation of the 3rd Global Monitoring Report of the Stockholm Convention, which compiled the most recent outcomes of research activities on monitoring and modelling of pollution levels and trends for legacy POPs as well as for CECs. In the framework of cooperation with HELCOM, the data on atmospheric emissions and modelled deposition of copper, HCB, and BDE-99 for the period 1990-2019 were prepared. In addition to this, a review of information on regulation, emissions, monitoring, and model assessment of SCCPs and PFOS was performed. Results of this work are available in the joint report of the EMEP Centres for HELCOM [*Gauss et al.*, 2021].

Future activities

Future activities of the EMEP Centres are aimed at further improvement of the assessment of heavy metal and POP pollution levels in the EMEP countries. Detailed evaluation of PAH pollution in the EMEP region will be continued as a part of the TFMM/EuroDelta-Carb multi-model intercomparison study. Collection and evaluation of global emission inventories of Hg and POPs, multi-model studies of Hg air-surface exchange as well as source apportionment of combustion-related POPs will be continued in cooperation with TF HTAP, Minamata and Stockholm Conventions and AMAP. Preparatory work for assessment of CECs long-range transport and fate will be further conducted. Case studies of heavy metal and POP pollution on a country scale aimed at detailed analysis in co-operation with national experts will be initiated in accordance with demands from EMEP countries. MSC-E will proceed with long-term co-operation with WGE focusing on

joint analysis of heavy metal measurements in moss in co-operation with ICP Vegetation, data support of ICP Integrated Monitoring and ICP Forests as well as data exchange with TF Health on PAH pollution levels and exceedances of air quality guidelines. It is planned to perform assessment of atmospheric pollution of the Baltic Sea and North-East Atlantic with heavy metals and POPs in co-operation with HELCOM and OSPAR Commissions.