EXECUTIVE SUMMARY

Heavy metals and persistent organic pollutants (POPs) are toxic substances known for their harmful effects on human health and biota. The priority heavy metals and POPs addressed by the Convention on Long-range Transboundary Air Pollution (CLRTAP) include lead (Pb), cadmium (Cd), mercury (Hg), polychlorinated biphenyls (PCBs), polychlorinated dibenzo(p)dioxins and dibenzofurans (PCDD/Fs), hexachlorobenzene (HCB), and polyaromatic hydrocarbons (PAHs). The priority PAHs are 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 (IcdP). Co-operative Programme for Monitoring and Evaluation of Long-range Transmission of Air Pollutants in Europe (EMEP, www.emep.int) performs scientific support of implementation of the Protocols. Centre on Emission Inventories and Projections (CEIP) is responsible for coordinating the emission related work of EMEP. Methodological guidance on monitoring activity in the EMEP region is carried out by Chemical Coordinating Centre (CCC). Information on results of atmospheric transboundary transport modelling is provided by Meteorological Synthesizing Centre – East (MSC-E).

Main activities of the EMEP centres in the field of heavy metals and POPs, carried out in accordance with the bi-annual workplan of the Convention for 2020-2021 [ECE_EB.AIR_144_Add.2], are overviewed in this Status report. The report includes information on the current status of emissions, monitoring, model assessment of pollution levels in the EMEP region, research and model development as well as cooperation with subsidiary bodies to the Convention, national and international organizations. More detailed information is also available in technical reports [*Travnikov et al.*, 2021, *Gusev et al.,* 2021; *Aas et al.*, 2021; *Pinterits et al.,* 2021; *Schindlbacher et al.,* 2021] and Supplementary Data Reports on heavy metals [*Strizhkina et al.*, 2021a] and POPs [*Strizhkina et al.*, 2021b] as well as on the MSC-E websites (www.msceast.org, https://projects.nilu.no/ccc/reports.html, https://www.ceip.at/).

Emissions

Completeness and consistency of submitted emission data have improved significantly since EMEP has been collecting information on emissions. In 2021 44 parties reported data on emissions of priority heavy metals, and 45 countries – on POPs. However, uncertainty of the reported data is considered relatively high. Time series of the reported data for 2000-2019 are analysed separately for the western and eastern parts of the EMEP domain. In the western part the reduction of emissions make up 16% for PAHs, about 60% for PCDD/Fs, 65% for PCB, and 95% for HCB. For heavy metals the reductions are around 40% for Cd, 50% for Hg and 70% for Pb. In the eastern part of the EMEP region time series are inconsistent exhibiting large variations explained by incomplete reporting in particular countries. Emission data for 2018 reported in 2021 were compared with 2018 emissions reported in 2020. For 26 countries, data changed by more than ±15 % for one or several pollutants. In contrast, for six countries the changes due to recalculations are below 1%.

Main emission sectors in the EMEP region are *Industry production*, *Public electricity and heat production*, and *Other stationary combustion* (*Residential combustion*). *Industry production* is the main emission sector for Cd, Pb, Hg and PCB. Most of PAHs are emitted by *Other stationary combustion* sector. Besides, this and *Industry production* sectors are almost equally important for PCDD/Fs. For HCB the main sector is *Public electricity and heat production*.

Sectoral gridded heavy metal and POP emissions were reported by 33 countries. For the remaining areas missing emissions are gap-filled and spatially distributed by expert estimates. Final emissions maps for modelling were generated by MSC-E based on the reported emissions data collected and gap-filled by CEIP and supplemented by additional information on vertical distribution, seasonal variation and chemical speciation of emissions. Global emission maps were also prepared based on data derived from research projects and expert estimates.

Monitoring

Monitoring activity for EMEP is coordinated by CCC. In 2019, there were 37 sites measuring heavy metals in both aerosols and precipitation, and altogether there were 65 measurement sites. 24 sites were measuring mercury in either air or precipitation, 14 of these with concurrent measurements in air and precipitation. The highest concentrations in air and precipitation of the first (Pb, Cd) and second (As, Ni, Cu, Cr) priority metals are in general seen in Eastern Europe. However, there are hotspots for some elements in other parts of Europe (around the English Channel, Cyprus, Italy). The highest Hg concentrations in air take place in Poland followed by sites in Germany and UK, while in precipitation the highest concentrations are seen in the Czechia and in Finland.

Quality of heavy metal measurements is evaluated via EMEP laboratory intercomparisons carried out annually. The majority of EMEP laboratories participating in the intercomparisons report results of good quality, however a few laboratories would benefit from the quality improving. In particular, for the results related to 2019, only 3 laboratories do not satisfy the accepted data quality objective for As and Cu, 2 for Zn and 1 for Pb, whereas all laboratories passed for Cd, Cr and Ni. In order to assess and improve quality of Hg measurements it is recommended to perform a field intercomparison or to include Hg into regular EMEP laboratory intercomparison studies.

In total there are 39 sites reporting 2019 data on POPs whereof 27 sites with measurements in both air and precipitation. Along with active air sampling method, recommended within EMEP, passive air sampling is widely used as a complementary approach. For instance, involving data from comprehensive passive air sampling campaign carried out across European countries in 2016 together with the GLEMOS model simulations allowed establishing the prevailing role of long-range atmospheric transport of a number of POPs in Norway.

The new strategic plan for 2020-2029 includes recommendation for monitoring of POPs other than those listed for Level 2 stations as well as organic contaminants of emerging concern (CECs), for example, polybrominated diphenyl ethers (PBDEs), per- and polyfluorinated alkyl substances (PFAS) and short-chain chlorinated paraffins (SCCPs). In general, the concentrations of PBDEs and PFAS are

low at the EMEP sites but analysis of the spatial pattern for these new POPs is hampered by limited number of stations, lack of laboratory intercomparisons and high uncertainty of data. Concentrations of CECs are higher than those of PAHs and other legacy POPs. This fact shows the importance of including CECs in monitoring programmes.

Status of heavy metal and POP pollution in 2019

Pollution levels of Pb, Cd, Hg, PAHs, PCDD/Fs, HCB and PCB-153 in 2019 are analyzed for the EMEP region. Overall pollution load is the highest in Central Europe where the most significant average deposition fluxes of Pb, Cd, Hg and PAHs take place. The lowest levels of pollution occur in Northern Europe as well as in Caucasus and Central Asia. At the same time, the highest PCB-153, HCB and PCDD/Fs deposition fluxes take place in Western, Eastern and Southern Europe, respectively.

In order to evaluate changes caused by inter-annual meteorological variability between the current (2019) and previous (2018) years, the pollution patterns for these years based on the same emission data were compared. Pollution levels of all considered contaminants declined in Southern Europe, and almost all – in Western Europe. In Central and Northern Europe the changes were insignificant (within ±10%). In Eastern Europe and Caucasus and Central Asia deposition of most pollutants increased. The exception is the changes of Cd and Pb. The levels of Pb and Cd significantly declined in most of sub-regions. Especially large decline (around 30%) is noted for Central Asia due to decrease of contribution of wind re-suspension in 2019.

Verification of the modelling results was carried out by comparing modelled concentrations in air and wet deposition fluxes with the corresponding values measured at stations of the EMEP monitoring network. Reasonably good agreement between modelled and observed concentrations is noted for Pb and Cd. Mean relative bias for Pb and Cd air concentrations is 4% and 23%, respectively, and for majority of stations the deviations between modelled and observed values lie within a factor of two. However, the model tends to underestimate the observed wet deposition fluxes by 30% for Pb and 35% for Cd. Modelling results are in agreement with measured background Hg^0 concentrations, with relative bias ranging from -10% to 20%. Although the model tends to overestimate the observed Hg deposition fluxes, the deviation between modelled and observed values is within a factor of two for most of stations.

Evaluation of modelling results for PAHs against the EMEP measurements showed almost no bias for B(a)P and IcdP, while for B(b)F and B(k)F some overestimation of observed air concentrations was obtained (about 60% and 23%, respectively). For most of selected monitoring sites, the difference between the modelling results and measured concentrations is within a factor of two. For PCB-153, HCB and PCDD/Fs the model predictions are within a factor of 2 compared to measured concentrations.

The deposition fluxes of heavy metals within the EMEP region are formed by EMEP anthropogenic emissions, secondary emissions (wind re-suspension of dust particles containing Pb and Cd,

natural/legacy emissions of Hg), and emissions from sources located outside the EMEP region (non-EMEP sources). For Pb the contributions of the EMEP anthropogenic and secondary emissions are comparable, whereas for Cd the role of EMEP anthropogenic emissions is prevalent. The contribution of non-EMEP sources of Pb and Cd is relatively low for the EMEP countries as a whole, varying from 5% to 25% in Europe and reaching 30-35% in Central Asia and Caucasus. In case of Hg, the main contributor to deposition in the EMEP region is non-EMEP sources followed by EMEP anthropogenic sources. The contribution of secondary emissions within the EMEP region is minor.

The contribution of the EMEP anthropogenic sources to total deposition of PAHs ranges from about 70% in the Caucasus and Central Asia to 80% and higher in Central Europe. The relative contribution of secondary sources and non-EMEP anthropogenic sources is much lower. For other POPs the contribution of the EMEP anthropogenic sources to total deposition in the considered sub-regions is 20-50% for PCDD/Fs, 15-40% for PCB-153, and 1-3% for HCB. The contribution of secondary emissions in the EMEP countries is approximately 60% for PCDD/Fs, 70% for PCB-153 and 75% for HCB.

Deposition fluxes of Pb, Cd and Hg to different types of land cover within the EMEP domain were also calculated to support the Working Group on Effects (WGE) in assessing adverse effects of heavy metals on ecosystems and human health. However, it would be important to update the existing evaluation of the adverse effects on human health and biota.

Model assessment and monitoring data for 2019 indicated high level of annual mean B(a)P air concentrations, exceeding the EU target value, in some of EMEP countries (e.g. Poland, Croatia, Slovakia, Czechia, Hungary, and some EECCA countries). Thus, considerable part of total population of the EMEP countries was exposed to B(a)P air concentrations higher than the air quality guidelines (7% for the EU target level and 63% for the WHO reference level). Besides, importance to consider wider list of toxic PAHs in the assessment of population exposure was highlighted. In particular, model estimates of B(a)P-equivalent air concentrations of 4 PAHs showed higher fractions of population living in the areas of concentrations above the EU target value (15%) and of WHO reference level (more than 70%).

In addition to the pollution assessment for the EMEP countries, atmospheric loads of heavy metals and POPs to remote regions such as the Arctic and the marginal seas (the Baltic, Black, Caspian, Mediterranean and North Seas) is evaluated and discussed in the report. Besides, the regional assessment within the EMEP domain is supported by global-scale simulations to take into account effect of long-range transport of the pollutants from emission sources located in other regions and continents. It is particular relevant for Hg and some POPs (HCB, PCDD/Fs), which are characterized by long residence time in the atmosphere. However, improvement of the global-scale assessment requires additional efforts for development of global emissions inventories for heavy metals and POPs in co-operation with other international bodies (UN Environment, Stockholm Convention, Minamata Convention).

Research and development

Analysis of the factors responsible for temporal changes of Hg and PAH pollution in different regions of the globe was initiated by MSC-E in cooperation with Task Force on Hemispheric Transport of Air Pollution (TF HTAP). Along with anthropogenic emissions, these factors include meteorological conditions, chemical properties of the atmosphere, surface characteristics, etc. Results of the pilot model simulations for the 1990-2018 period indicate gradual decrease in atmospheric concentrations of the considered pollutants in the European region. The long-term dynamics of all three pollutants in Europe is primarily determined by changes in anthropogenic emissions both inside and outside the region. Source apportionment has shown that contribution of regional sources to PAH pollution levels in Europe remain predominant over the whole period. For Hg, prevailing contribution of regional sources decreases over the period and is replaced by increasing contribution of East Asian sources. Other factors additionally contribute to temporal variability of the pollutants. Thus, the proposed approach can be applicable for understanding of long-term pollution dynamics. However, the results are sensitive to uncertainties of the key input parameters

MSC-E continued research activities to improve assessment of PAH pollution levels in the EMEP region and to contribute to the analysis of the effectiveness of measures to reduce unintentional releases of PAHs. In particular, analysis of long-term changes of the observed and modelled PAH concentrations, exemplified by B(a)P, demonstrates that most significant decrease takes place in Western, Central, and Northern Europe (by 65%, 60%, and 40%, respectively). However, the levels in Caucasus and Central Asia increased by 65%. The major contributor to B(a)P concentrations in the EMEP countries is *Residential Combustion* emission sector. Noticeable contribution was also made by the *Industry sector* in the beginning of the 1990s in Northern, Western and Southern Europe. However, later on its share decreased significantly. Besides, countries of Southern Europe are characterized by considerable contribution of the *Agriculture sector*.

In more detail, PAH pollution on a national scale was assessed as a part of a case study for Poland. Current stage of the study is focused on the model assessment of B(a)P pollution in the country. using three different emission data sets, based on national inventories and emission scenarios. The model simulations with the scenario emissions allow improving agreement between the model and measurements and indicate possible underestimation of national B(a)P emissions in Poland. Further steps of the study can include multi-model simulations, application of more detailed temporal and spatial disaggregation of B(a)P emissions, and co-operation with national experts in monitoring, modelling, and emissions.

Gaseous exchange of POPs and Hg between the atmosphere and vegetation is an important process that affects their distribution in the environment. A new model parameterization of the air-vegetation exchange processes for GLEMOS has been developed. The effect of the implementation of updated air-surface gaseous exchange parameterization was evaluated using a one-year simulation of PCB-153. The application of the updated scheme resulted in considerable changes of simulated PCB concentrations in vegetation and soils in different parts of the EMEP region. The

modelling results obtained with the new model parameterization need to be further evaluated against available measurements in the vegetation and soil compartments..

Information on land cover is essential for estimation of ecosystem-dependent deposition fluxes. Long-term changes in the surface conditions are considered as one of the important factors which can possibly affect changes of heavy metal and POP levels in the EMEP and other regions. To study the impact of time variable land cover on the GLEMOS model simulations a new data set based on MODIS satellite observations was analyzed and compared with the currently used dataset. The main differences between the considered datasets are related to fractions of urban and cropland types of land cover. It is important to stress that currently there is still no agreed land cover data to be used within EMEP and the Working Group on Effects (WGE). A harmonized dataset is crucial for assessment of ecosystem-dependent deposition fluxes and evaluation of ecosystem critical loads exceedances.

Both the model parameterisations and input data require periodical revisions and updates to keep the model in line with new findings of the scientific community. This year updates of the GLEMOS modelling system for Hg include utilizing a new Hg global emission data set used for generation of boundary concentrations for the EMEP domain, and new data on atmospheric concentrations of reactants involved into the Hg atmospheric chemistry. The updated version of the model demonstrates better performance than the previous one in terms of comparison with observations. Besides, it should be noted that the Centre continues research activities focused on the study of Hg chemical mechanisms in the atmosphere in collaboration with other scientific groups. The ultimate aim of the research is improvement of the model estimates of Hg pollution levels in the EMEP countries.

Pollution by microplastics is recognized as a global problem affecting all environmental compartments. Once released into the atmosphere, microplastics can be transported over long distances and can affect human health and biota. The adverse health effects of microplastics can be attributed to the presence of toxic constituents. Besides, microplastics can absorb and accumulate other pollutants, including heavy metals and POPs. The main source of microplastic releases to the atmosphere is secondary emission from the land and the ocean. Currently, data on the quantitative characteristics of the emission of microplastics into the atmosphere, as well as information on the processes that determine their atmospheric transport, are limited, and further research is required.

Cooperation

An overview of MSC-E activities in the field of assessment of heavy metal and POP pollution in the EMEP region and on a global scale was presented at the recent virtual TFMM meeting. Much attention was paid to the analysis of long-term changes of heavy metal and POP pollution levels. In particular, the changes of Hg and PAH levels in Europe and other regions of the globe were studied. Besides, the pollution changes were examined in the framework of cooperation with the regional marine conventions (HELCOM and OSPAR). Along with this, main results of a country-specific case

study of PAH pollution in Poland were presented. Finally, directions of future research activities in the field of heavy metals and POPs relevant to TFMM work were discussed during the special sub-session of the meeting devoted to heavy metals, POPs, chemicals of emerging concern, and microplastics.

MSC-E continues collaboration with the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) on Hg and POP pollution assessment. In particular, the Centre and TF HTAP jointly hosted two workshops to identify near-term opportunities and longer-term research needs to improve the scientific basis for assessment of Hg and POP pollution and trends. The workshops examined current work and efforts throughout the international science community aimed at addressing Hg and POP pollution problem on global and regional scales. A program of multi-model assessment and attribution of long-term Hg and POP pollution trends in the EMEP and other regions was proposed by MSC-E. Pilot results of a model assessment of Hg and POP pollution trends and their attribution to various factors (changes in anthropogenic emissions, meteorological conditions, atmospheric chemistry, land cover etc.) were presented to illustrate possible outcome of the study and input data required.

MSC-E took part in the twenty-fourth meeting organized by Task Force on Health. The assessment of PAHs pollution levels in the EMEP region, analysis of the key sources and trends were presented. The assessment was prepared to contribute to the analysis of effectiveness of the Protocol on POPs) in co-operation with Task Force on Techno-economic Issues (TFTEI) and Task Force on Health. The Task Force was informed about the changes of PAH emissions, modelled and observed concentrations over the recent 20 years in the EMEP region, population exposure to B(a)P concentrations exceeding threshold levels, and model experiments on evaluation of joint toxicity of the group of sixteen PAHs.

MSC-E continued cooperation with international organizations such as the Arctic Monitoring and Assessment Programme (AMAP), Minamata Convention, Stockholm Convention, European Commission, HELCOM and OSPAR . Recently, MSC-E participated in the AMAP Assessment of the Arctic pollution by Hg (AMAP Mercury Assessment 2021). The Centre also took part in the Minamata Online Session “Multimedia modelling of global mercury movement” aimed to bridge the scientific community and international policy to better understand abilities of the multimedia mercury modelling for assessing the state of the environment and effectiveness of pollution control measures. In the framework of cooperation with HELCOM, atmospheric deposition of selected heavy metals and POPs to the Baltic Sea were estimated for the period 1990-2018. Finally, the work on assessment of of Pb, Cd and Hg deposition to the sub-regions of the OSPAR maritime area was initiated.

Future activities

Future research activities of the EMEP Centre's will be aimed at improvement of heavy metal and POP pollution assessment in the EMEP region. Detailed assessment of PAH pollution levels will be continued with focus on the analysis of population exposure to PAH and atmospheric aerosol from combustion sources. In particular, the Centre will contribute to a multi-model analysis of B(a)P pollution levels as a part of the TFMM/EuroDelta-Carb intercomparison exercise. Activities aimed at improvement of the modelling approaches for assessment of Hg and POP long-term trends, source-receptor relationships, and future projections will be initiated in collaboration with TF HTAP. They will include a multi-model study of Hg dispersion and cycling on a global scale with focus on air-surface exchange and secondary/natural emissions, global/regional multi-model evaluation of source-receptor relationships for combustion-related POPs, as well as contribution to the TF THAP exploratory workshops on wildfires, chemicals of emerging concern (CEC) and microplastics. The country-scale assessment of heavy metal and POP pollution in a form of case studies will be continued aiming at detailed analysis of pollution levels in selected countries and refinement of the EMEP operational modelling. Ecosystem-related analysis of heavy metal pollution will be performed in co-operation with WGE. Besides, the assessment of atmospheric pollution of the marine environment with heavy metals and POPs will be carried out in collaboration with HELCOM and OSPAR. Finally, the Centre will continue co-operation with Stockholm and Minamata Conventions and other international bodies in relation to assessment of POP and Hg pollution.