Quality of assessment of HM/POP pollution. Research and development

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Meteorological Synthesizing Centre East
EMEP activities on HMs and POPs in 2013

- Monitoring of HMs and POPs within EMEP (CCC)
- Operational modelling of HM and POP transboundary pollution within EMEP region
- National/local scale pollution assessment – Case studies
- Assessment of long-term changes of POPs transboundary pollution (1990-2011)
- Quality analysis of HM and POP assessment results
- Research and development:
  - Further development of GLEMOS multi-scale modelling system
  - Model study of major Hg oxidation processes in the atmosphere
  - Assessment of the role secondary emissions in POP pollution
  - Application of inverse modelling for evaluation of primary and secondary emission sources
- Co-operation with national experts, international organizations and programmes

Steering Body to EMEP, September 9 – 11, 2013
Assessment of HM and POP pollution in EMEP domain

Pollution levels (2011)

Long-term trends (1990-2011)

Transboundary transport

Intercontinental transport

Quality of assessment

Country-specific data

Pb deposition flux, kg/km²/y

90% variation
50% variation
Average flux
Smoothed trend

Air concentrations, ng/m³

90% interval
Average

Pb deposition flux, kg/km²/y

Germany

65% reduction

B[a]P

65% reduction

Cd

HCB

B[a]P

B[a]P

Pb

Cd

HCB

CZ, 1,007 kg
31%

DE, 417 kg
13%

UA, 174 kg
5%

SK, 153 kg
5%

HU, 111 kg
3%

CZ, 325 kg
10%

Other

RO, 101 kg
3%

< 23

23 – 30

30 – 36

36 – 46

46 – 60

60 – 100

> 100

Northern Scandinavia and Baltic

Central Europe

Southwestern Europe

Modelled, µg/L

Observed, µg/L

Steering Body to EMEP, September 9 – 11, 2013
Quality of assessment

Multi-year statistics of model results evaluation vs. measurements

Pearson's correlation

Relative bias

Correlation coefficient > 0.7 and Relative bias < ±20%

Correlation coefficient < 0.7 or Relative bias > ±20%
Research and development to improve quality of the assessment

GLEMOS model scheme

Data pre-processing (WRF, ECMWF, ...)

Atmosphere

Ocean

Terrestrial

Heavy metals (Hg, Pb, Cd, ...)

POPs (HCB, PCDD/Fs, PAH, ...)

Atmospheric aerosol, EC

Global

Regional

National

- Refinement of atmospheric processes
  - Atmospheric speciation and chemistry (Hg)
  - Interaction with atmospheric aerosols (PAHs)

- Secondary sources and multi-media modelling
  - Refinement of wind re-suspension and other secondary sources (Pb, Cd, Hg, PAHs, HCB)
  - Analysis of emissions to the media other than atmosphere (PCDD/Fs)

- Further development and application of inverse modelling
  - Analysis and refinement of spatial distribution and temporal variation of anthropogenic emissions (Cd, PCDD/Fs, PAHs)

- Transition to lat-lon grid and modelling with fine resolution
- Input data for modelling

Discussed at TFMM, TFEIP and TFHTAP

Steering Body to EMEP, September 9 – 11, 2013
Refinement of atmospheric processes: mercury speciation and chemistry

**Motivation:** refine knowledge on major Hg oxidation mechanisms in the atmosphere to improve deposition to underlying surfaces (aquatic surfaces)

**Method:** Model sensitivity runs with individual chemical mechanisms and evaluation vs. field campaign measurements

**Processes studied:**
- Speciation of Hg anthropogenic emissions
- Hg oxidation by $O_3$, OH, Br
- Oxidation products (gaseous or particulate)
- Gas-particle partitioning of Hg(II)
Refinement of atmospheric processes (Hg)
Model sensitivity runs with different chemical mechanisms

**Preliminary conclusions:**

- All oxidation mechanisms reproduce diurnal variation but lead to overestimation of measurements.
- $\text{Hg(II)}_{\text{gas}}$ concentration in is more affected by atmospheric chemistry, whereas $\text{Hg(II)}_{\text{part}}$ by direct anthropogenic emissions.

**Modelled and observed concentrations at DE2, 2009**

*Steering Body to EMEP, September 9 – 11, 2013*
Refinement of atmospheric processes (Hg)

Selected CARIBIC flights

Preliminary conclusions:

- Model successfully reproduce spatio-temporal variation of Hg in the upper troposphere
- The highest correlation was for the runs governed by the $O_3$ chemistry; the lowest for the OH chemistry

Evaluation of model results vs. observations of TGM

Frankfurt - Vancouver (23 Jul 2009)

Frankfurt - Osaka (27 May 2009)
Refinement of atmospheric processes: seasonal variation of Hg wet deposition

Future research:
- Improvement of model parameterization of Hg atmospheric chemistry
- Development of multi-media approach to simulation of Hg dispersion in the environment
Refinement of atmospheric processes: PAH interaction with aerosol

Motivation:
Underestimation of winter time PAH air concentrations

Ongoing activities:
- Improvement of process parameterizations related to interaction of PAHs with atmospheric aerosols (sorption, heterogeneous chemistry)

Processes studied:
- gas-particle partitioning
- degradation in particulate phase
Refinement of model parameterization of PAH: sorption/degradation

Aerosol composition

B[a]P sorption on aerosol components

Model vs measurements

Previous model parameterization

Modified model parameterization (including EC)

Low degradation rates of PAH sorbed on EC lead to higher concentrations in winter
Refinement of model parameterization: secondary sources

**Processes**: re-suspension / re-volatilization from environmental media to the atmosphere.

Contributions of secondary sources to contamination of the EMEP region are essential for most of HMs and POPs

Relation between anthropogenic and secondary sources

**Processes affecting secondary emission fluxes**:  
- Long-term accumulation in media other than the atmosphere  
- Accumulation due to direct emissions to environmental media (soil, seawater)  
- Wind re-suspension
Long-term accumulation in media (HCB)

Motivation
Contemporary levels of HCB in air are mostly controlled by secondary emissions (re-volatilization) [Barber et al., 2005]
Re-volatilization of HCB from soils essentially exceeds contemporary anthropogenic emissions [Franke et al., 1996]

Contributions of different sources to HCB concentrations in the EMEP region
- Anthropogenic emissions, 13%
- Secondary sources, 87%

Method
Updating of expert estimates of global historical HCB emissions
Modelling of HCB long-term accumulation in the environmental media
GLEMOS model runs for refinement of contributions of various source groups (EMEP and non-EMEP anthropogenic and secondary emissions)
Long-term accumulation in media (HCB)

Long-term emission scenario

HCB global emissions (1945-2011)

HCB air concentrations (2011)

Contributions of sources to HCB air concentrations in EMEP region

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Long-term accumulation in media (HCB)

Comparison with measurements

Future research:

- Evaluation of HCB media content
- Refinement of parameterization of the behaviour of HCB in media (particulatly, degradation process)
- Refinement of contribution of HCB secondary emission and global sources to pollution levels in the EMEP domain

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Accumulation due to direct emissions to environmental media (PCDD/Fs)

Motivation:

- underestimation of the contribution of PCDD/F secondary sources
- disagreement between model results and measurements

Method:

refinement of model description of secondary sources taking into account direct emissions to environmental media

Comparison with measurements

Data on PCDD/F emissions to soil for modelling is based on EU Project “Releases of Dioxins and Furans to Land and Water in Europe”

Steering Body to EMEP, September 9 – 11, 2013
Accumulation due to direct emissions to environmental media (PCDD/Fs)

Contributions of sources to PCDD/F air concentrations in EMEP region

- Non-EMEP sources 5%
- Secondary sources 59%
- Anthropogenic emissions 36%

Comparison of calculated PCDD/F air concentrations with measurements

<table>
<thead>
<tr>
<th>Factor of 2</th>
<th>Factor of 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1 – 0.3 ng TEQ/m³</td>
<td>0.01 – 0.1 ng TEQ/m³</td>
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</tbody>
</table>

Comparison of soil concentrations

<table>
<thead>
<tr>
<th>Calculated</th>
<th>0.1 – 3 ng TEQ/kg</th>
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<tbody>
<tr>
<td>Measured</td>
<td>0.16 – 10 ng TEQ/kg</td>
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Future research:

- Continuation of the analysis of PCDD/Fs emissions to the media other than atmosphere and their application in modelling

Steering Body to EMEP, September 9 – 11, 2013
Refinement of model parameterization: wind re-suspension (Pb)

Pb in air (2000-2011) model vs measurements

Motivation:
Overestimation of air concentrations in Benelux region

Method:
Improvement of wind re-suspension scheme in the region using inverse modelling approach and fine resolution modelling
Refinement of model parameterization: wind re-suspension (Pb)

Monthly mean modelled and observed concentrations of lead at the Dutch stations

Primary evaluation of the results: wind re-suspension from urban areas is overestimated
Refinement of model parameterization: wind re-suspension (Pb)

Examination of re-suspension flux from urban areas using inverse modelling approach

Contributions of re-suspension flux from urban regions to concentrations of lead at Bilthoven site (7 – 19 of April)

Domain for fine resolution modelling (5 × 5 km)
Refinement of model parameterization: modelling with fine resolution (case study)

Modelled and observed Pb air concentrations at Dutch and Belgian stations

Wind re-suspension of Pb in the Netherlands, kg/km²/y
Fine resolution

Effect of use of fine resolution meteorological and geophysical data

Annual precipitation amount (2007)

Total Pb deposition (2007)
Refinement of deposition fields in Croatia due to increased resolution of emission data

Pb total deposition

Pb emission data

50 x 50 km

10 x 10 km
Global modelling in lat/lon projection

Generation of boundary conditions for regional modelling

Contribution of regional vs. global sources to HM and POP pollution

- EMEP sources
- Global sources

Global modelling in lat/lon projection

Steering Body to EMEP, September 9 – 11, 2013
Transition to new EMEP grid and finer resolution

Refinement of input data for modelling:
- Anthropogenic emissions (EMEP and global)
- Meteorological input
- Atmospheric reactants
- Geophysical data (land use, ocean currents, soil and vegetation characteristics, ...)
- ...

Updating and testing of modelling approach:
- Adapting of model parameterizations
- Evaluation of modelling results on new grid
Available expert estimates of global HM and POP emissions

<table>
<thead>
<tr>
<th>Dataset</th>
<th>Chemicals</th>
<th>Years</th>
<th>Resolution</th>
</tr>
</thead>
<tbody>
<tr>
<td>AMAP/UNEP</td>
<td>Hg</td>
<td>2010</td>
<td>0.5°×0.5°</td>
</tr>
<tr>
<td>Tao et al., 2009</td>
<td>PAHs</td>
<td>2004</td>
<td>n/a</td>
</tr>
<tr>
<td>Breivik et al., 2007</td>
<td>PCBs</td>
<td>1930-2100</td>
<td>1°×1°</td>
</tr>
<tr>
<td>Bailey et al., 2001</td>
<td>HCB</td>
<td>1995</td>
<td>n/a</td>
</tr>
<tr>
<td>NILU/CGEIC</td>
<td>Pb</td>
<td>1989</td>
<td>1°×1°</td>
</tr>
</tbody>
</table>

No global data for Cd and PCDD/Fs
Global Hg emissions inventory

Assessment of Hg emissions to the atmosphere:

- Update of existing inventories for current anthropogenic emissions for Hg
- Evaluation of natural sources and re-emission
- Development of future emission scenarios

Hg anthropogenic emissions (2010)

Hg emission trends

Global Mercury Assessment 2013
Main directions of research to improve quality of assessment (2014-2015 work plan)

- Refinement of parameterizations of key processes affecting HM and POP air pollution to support the implementation of the Protocols on POPs and HMs
- Assessment of pollution levels of HMs in selected countries with fine resolution
- Evaluate results of the EMEP models in the new grid: model performance, trends and source receptor relationships
- Assess contribution of intercontinental transport and secondary sources to HM and POP pollution (in cooperation with TFHTAP, AMAP and UNEP)
- Make annual release of GLEMOS open source codes
- Input data preparation for modelling: emissions and meteorological data for years 2012 and 2013 for the new EMEP grid and global modelling (in cooperation with TFEIP and TFMM)