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

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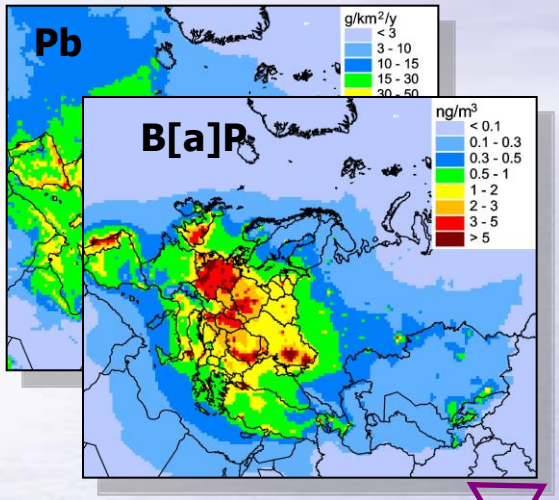
Meteorological Synthesizing Centre East

# EMEP activities on HMs and POPs in 2013

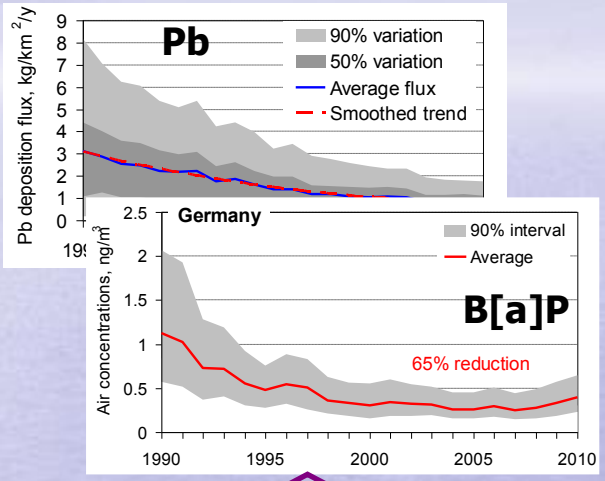
- Monitoring of HMs and POPs within EMEP (CCC)
- Operational modelling of HM and POP transboundary pollution within EMEP region
- National/local scale pollution assessment – Case studies
- Assessment of long-term changes of POPs transboundary pollution (1990-2011)
- Quality analysis of HM and POP assessment 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

# Assessment of HM and POP pollution in EMEP domain

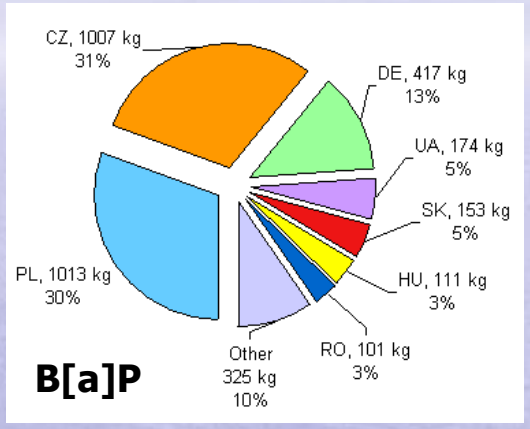
Pollution levels (2011)



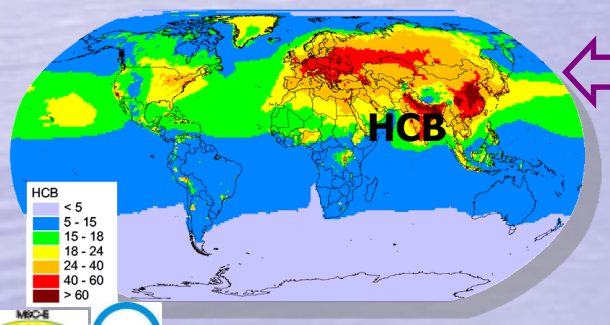

Long-term trends (1990-2011)



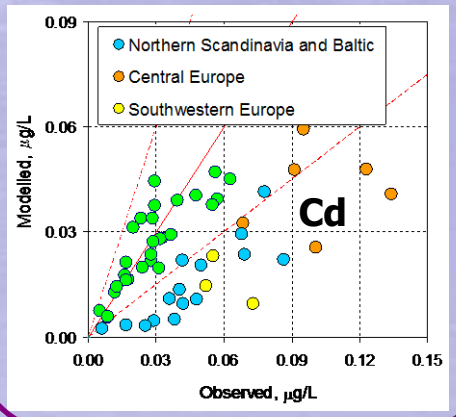

Transboundary transport



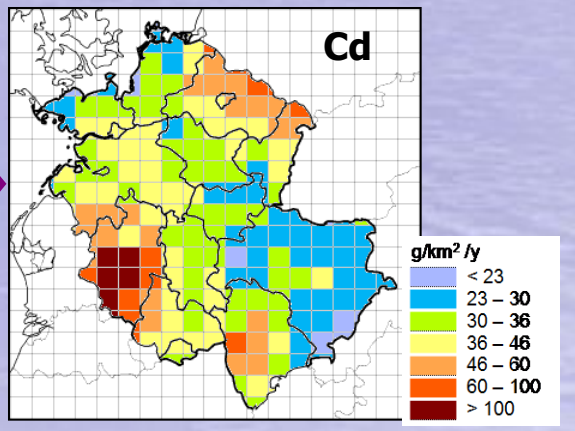

Intercontinental transport




Quality of assessment



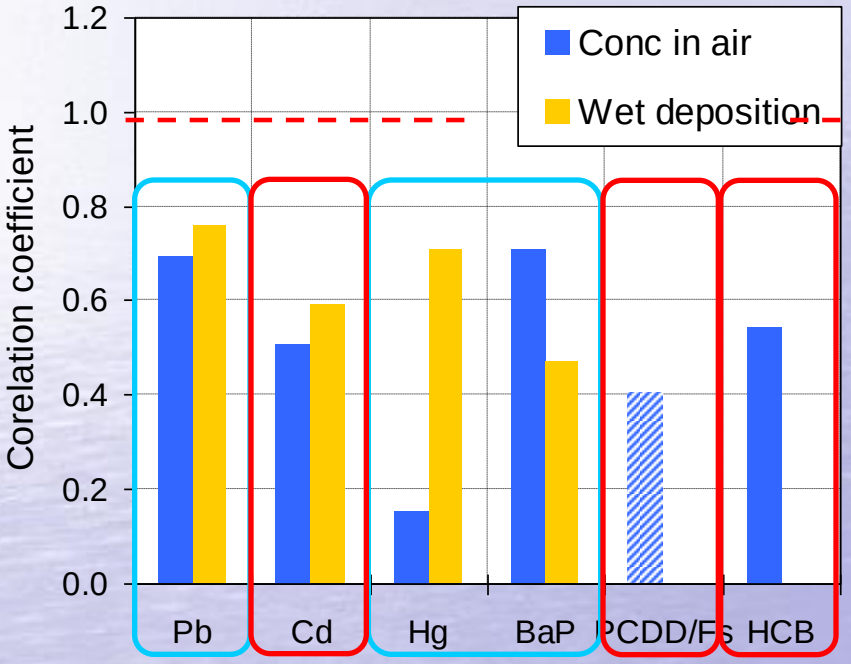

Country-specific data



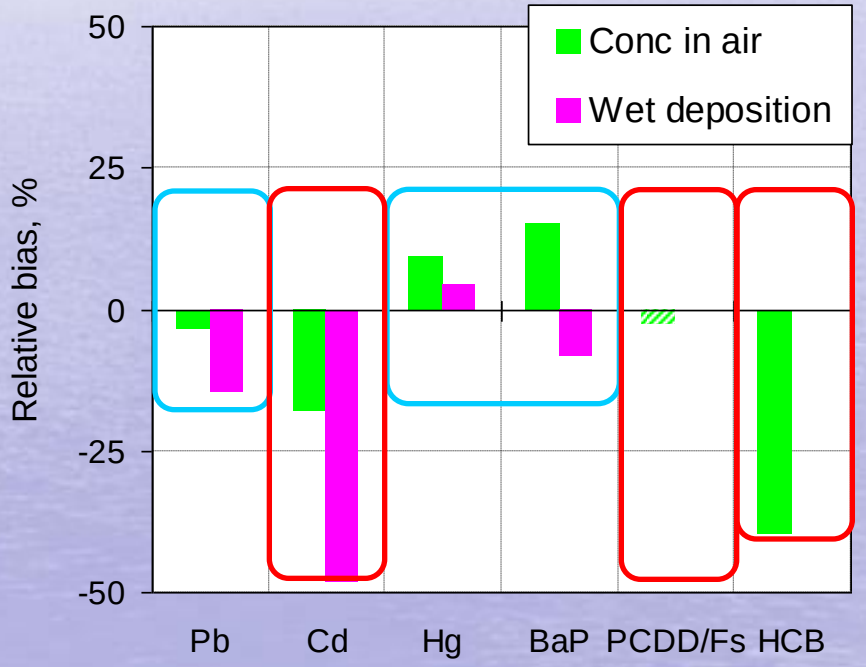
# Quality of assessment

**Multi-year statistics of model results evaluation vs. measurements**

Pearson's correlation




Relative bias

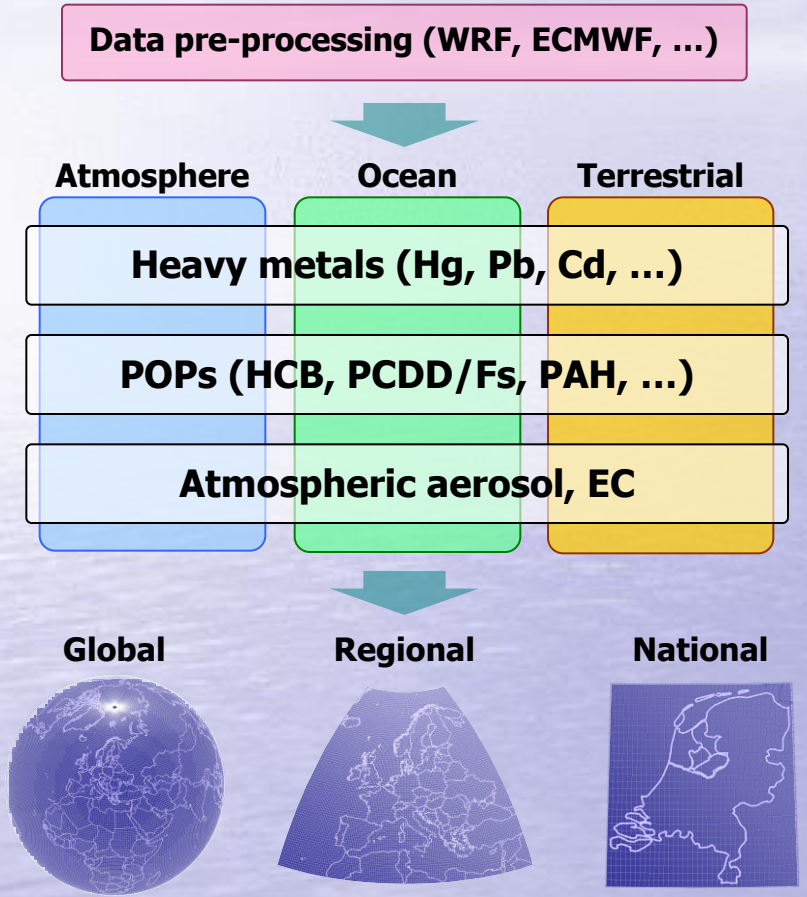



Correlation coefficient > 0.7 **and**
Relative bias < ±20%

Correlation coefficient < 0.7 **or**
Relative bias > ±20%

# Research and development to improve quality of the assessment

GLEMOS model scheme




- 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)

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

Discussed at TFMM, TFEIP and TFHTAP

# 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)

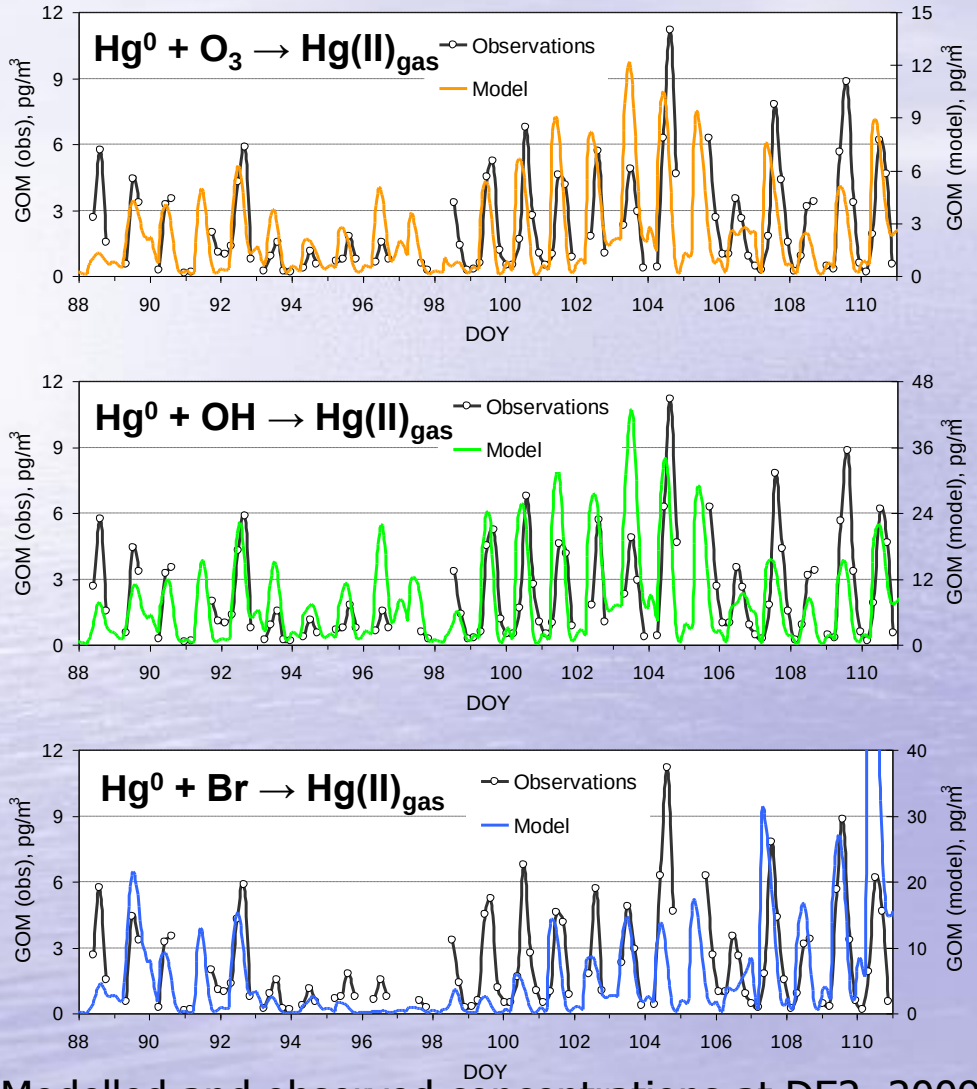
**Method:** Model sensitivity runs with individual chemical mechanisms and evaluation vs. field campaign measurements

**Processes studied:**

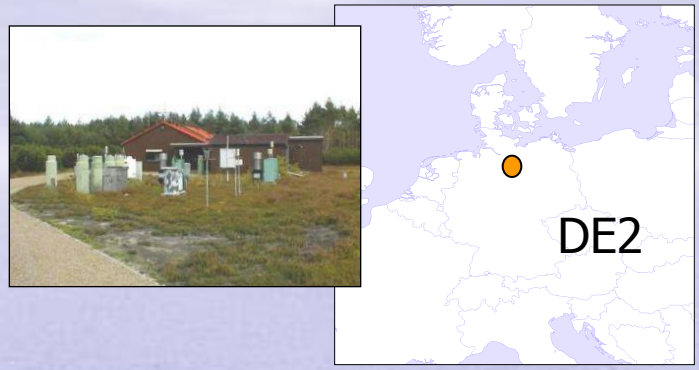
- Speciation of Hg anthropogenic emissions
- Hg oxidation by $O_3$, OH, Br
- Oxidation products (gaseous or particulate)
- Gas-particle partitioning of Hg(II)

# Refinement of atmospheric processes (Hg)

## Model sensitivity runs with different chemical mechanisms




Modelled and observed concentrations at DE2, 2009



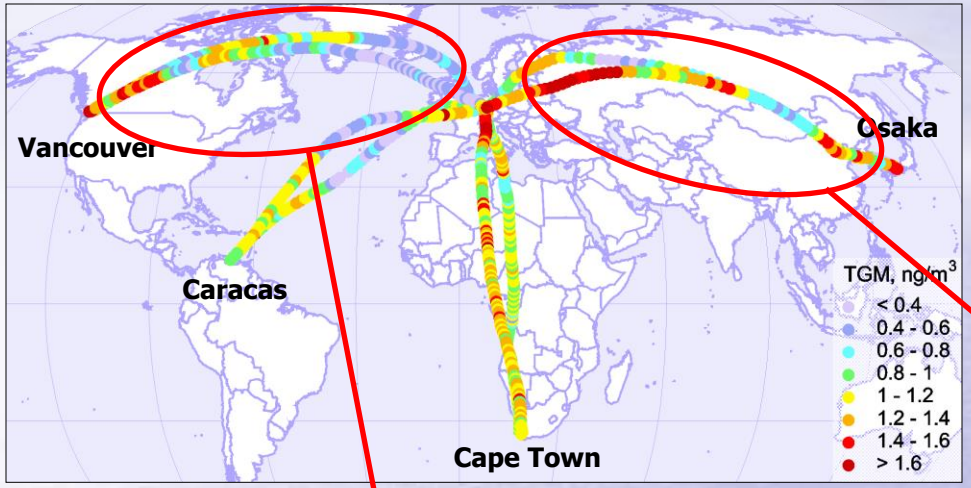

Location of DE2 site

**Preliminary conclusions:**

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

# Refinement of atmospheric processes (Hg)

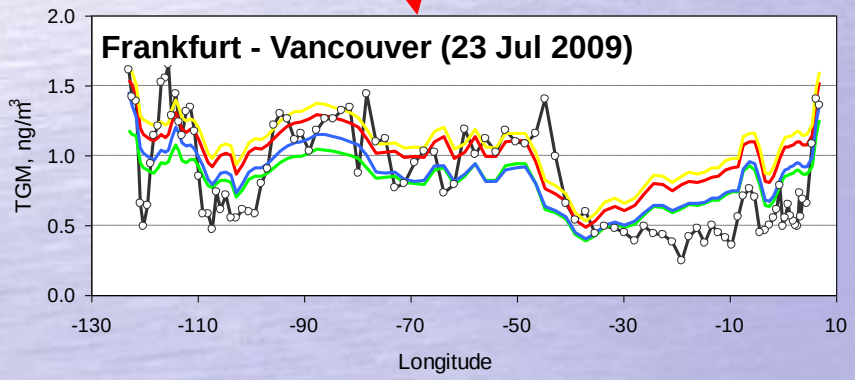
## Selected CARIBIC flights

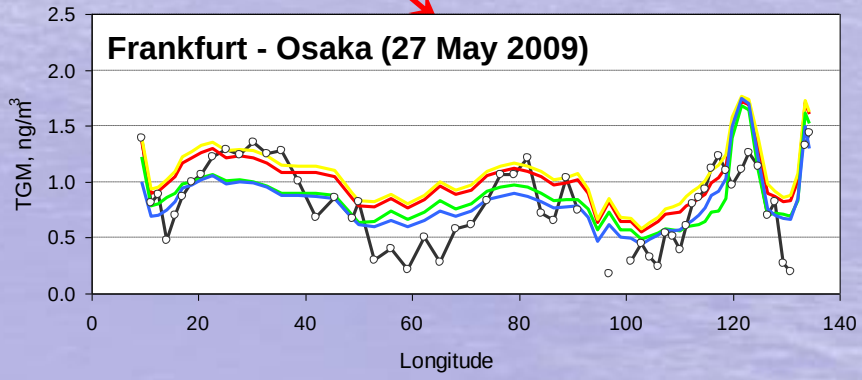



## Preliminary conclusions:

- 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

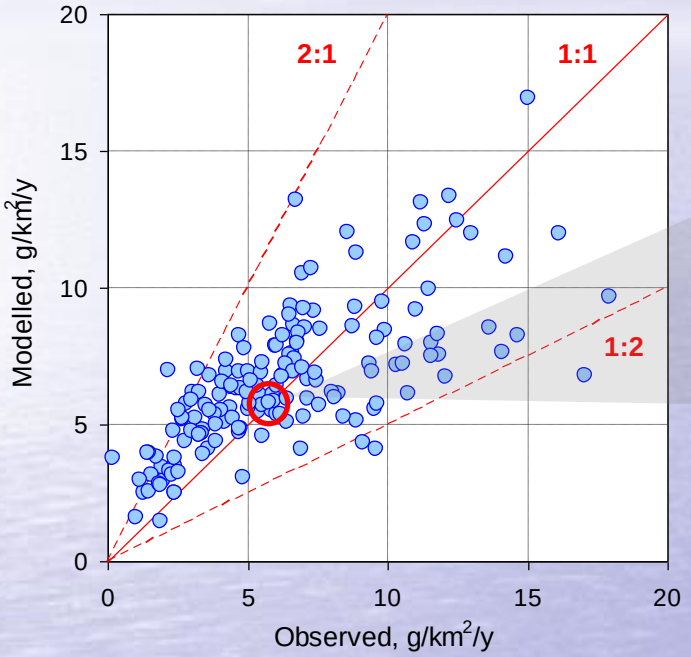


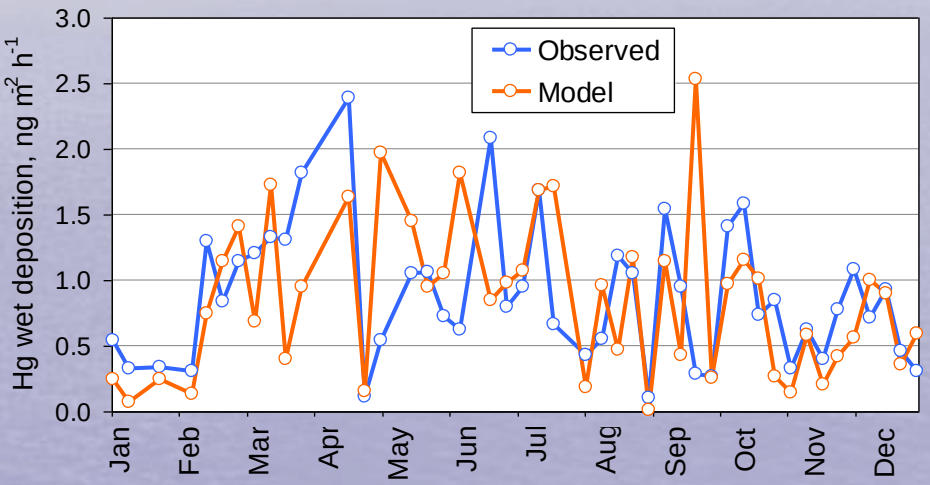

Obs  Base  $O_3$  OH  Br

# Refinement of atmospheric processes: seasonal variation of Hg wet deposition

Hg wet deposition (1990-2011)




Hg wet deposition at site DE08 (Schmücke, Germany), 2009

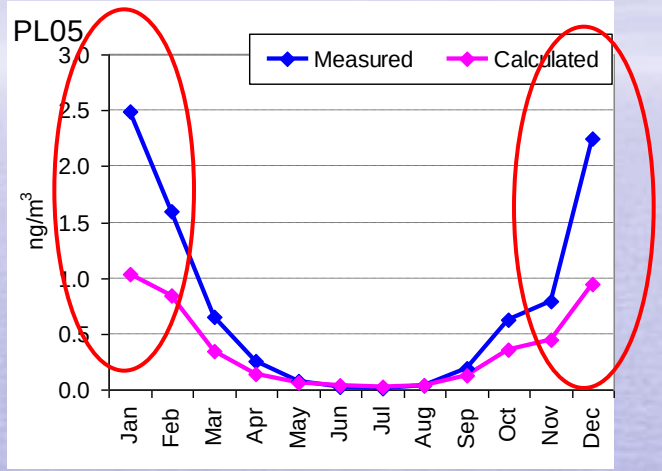



**Future research:**

- 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

BaP air concentration at PL5 in 2011




**Motivation:**

Underestimation of winter time PAH air concentrations

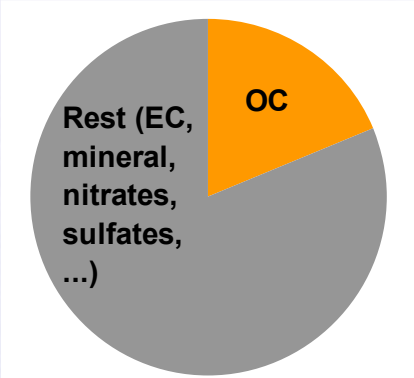
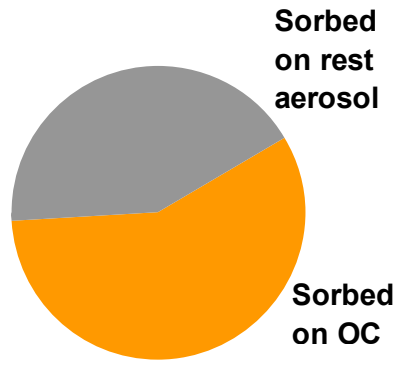
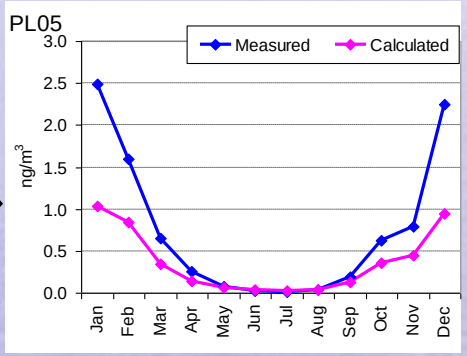
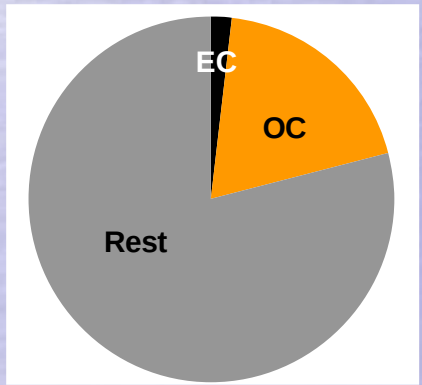
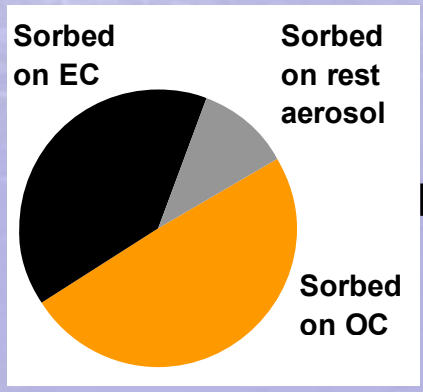
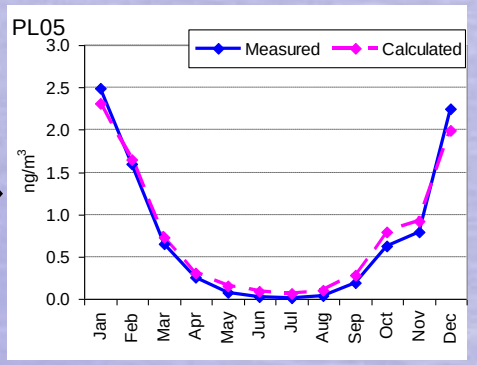
**Ongoing activities:**

- 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

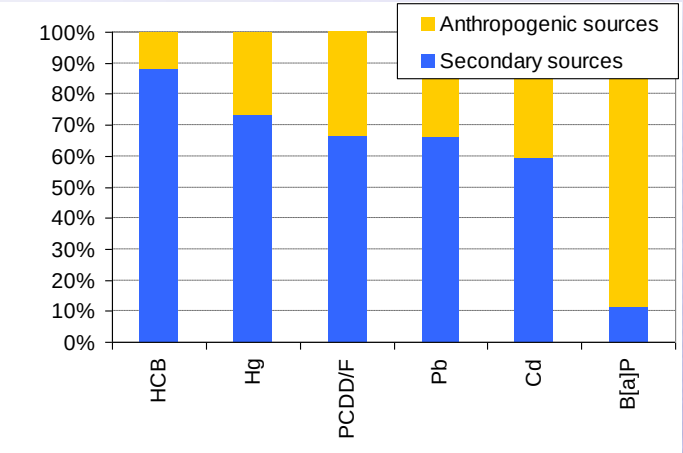
| | Aerosol composition | B[a]P sorption on aerosol components | Model vs measurements |
|---|---|---|---|
| Previous model parameterization |   |   |   |
| Modified model parameterization (including EC) |   |   |   |

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

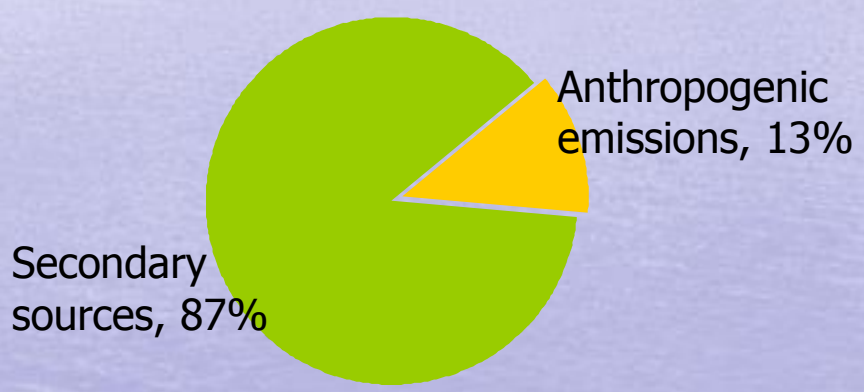
# Long-term accumulation in media (HCB)

**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




**Method**

Updating of expert estimates of global historical HCB emissions

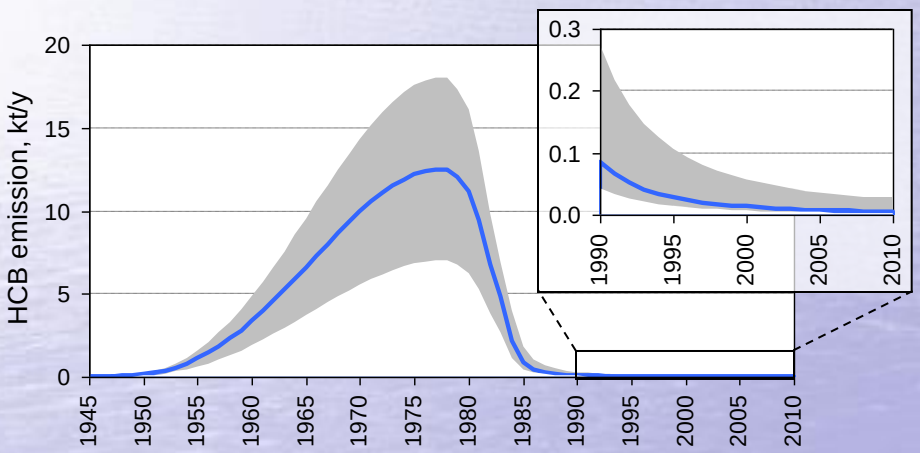
Modelling of HCB long-term accumulation in the environmental media

GLEMOS model runs for refinement of contributions of various source groups (EMEP and non-EMEP anthropogenic and secondary emissions)

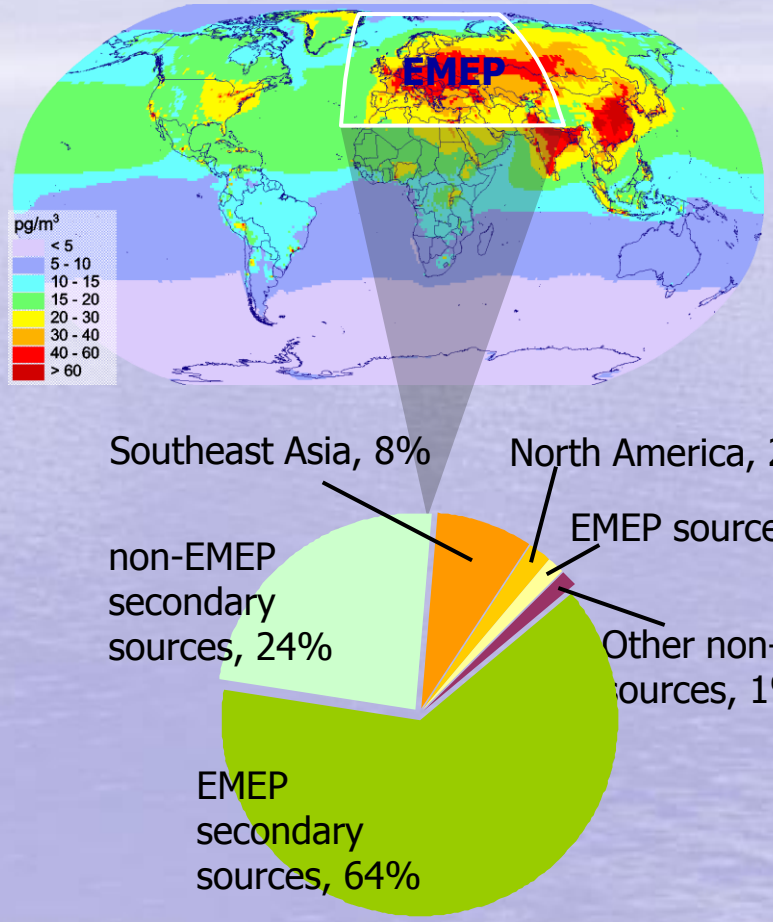
# Long-term accumulation in media (HCB)

**Long-term emission scenario**

HCB global emissions (1945-2011)



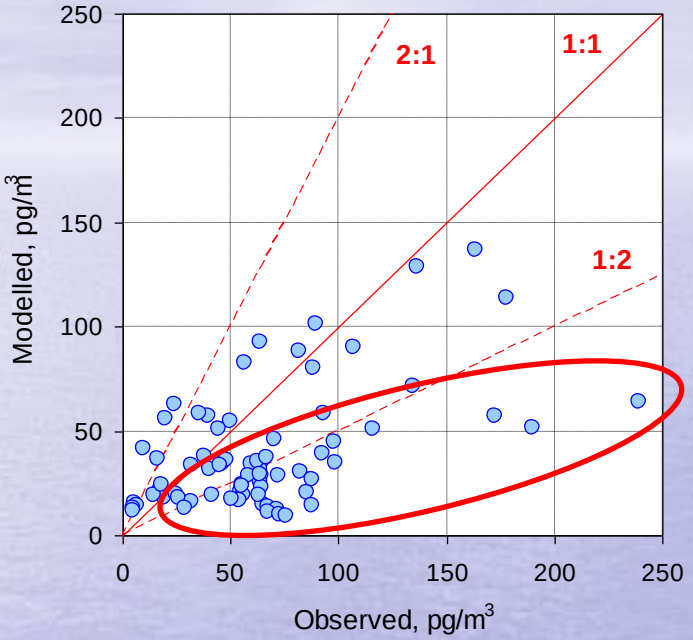

HCB air concentrations (2011)




Contributions of sources to HCB air concentrations in EMEP region

# Long-term accumulation in media (HCB)

Comparison with measurements



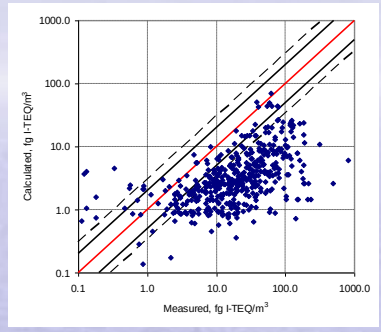

**Future research:**

- 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

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

**Motivation**:

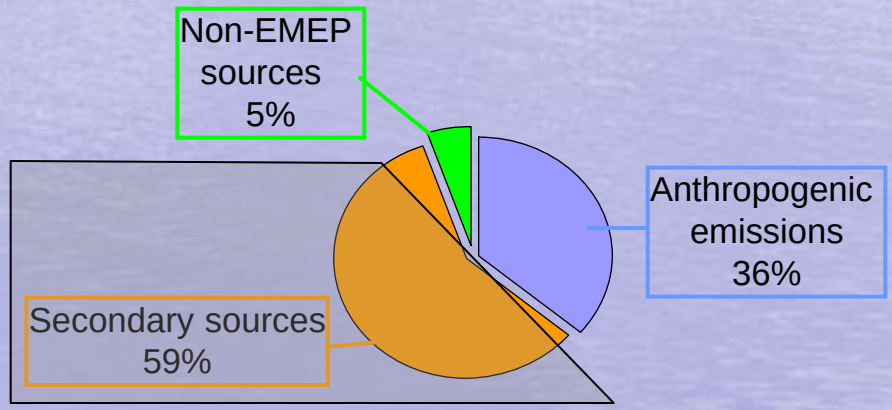
- underestimation of the contribution of PCDD/F secondary sources
- disagreement between model results and measurements




Comparison with measurements

**Method**:

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

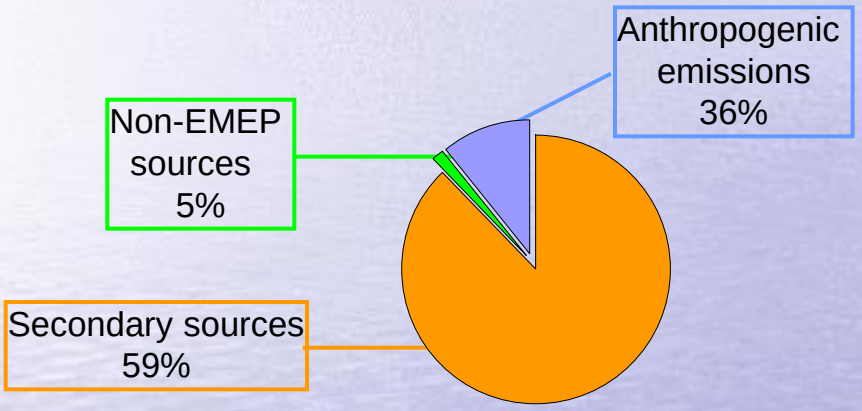



Overall emissions → Emissions to air
Overall emissions → Emissions to soil

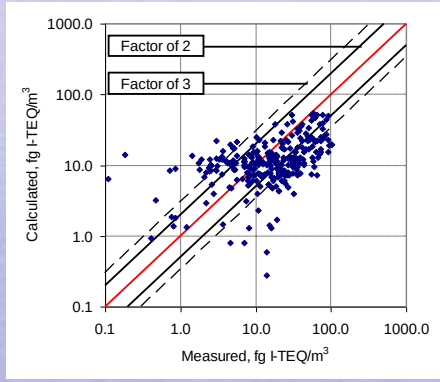
Data on PCDD/F emissions to soil for modellingis based on EU Project "*Releases of Dioxins and Furans to Land and Water in Europe*"

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

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




Comparison of calculated PCDD/F air concentrations with measurements



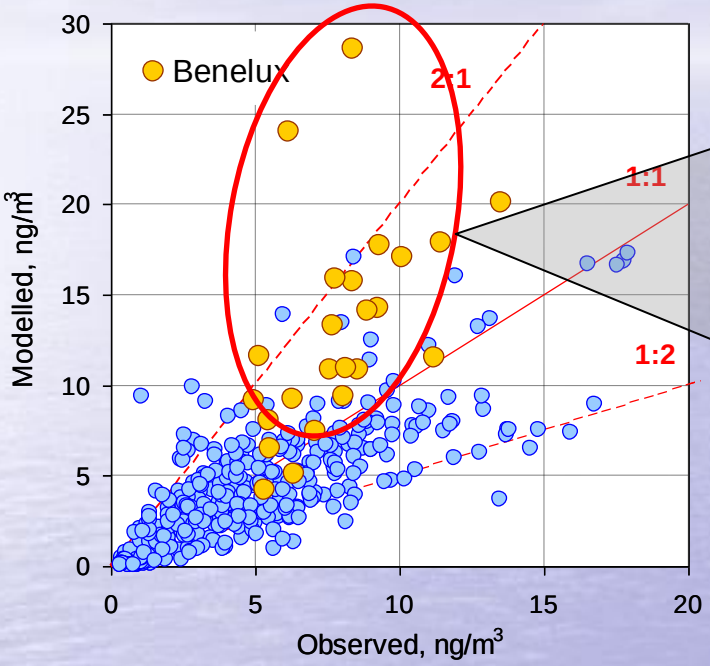

| Comparison of soil concentrations | |
|---|---|
| Calculated | 0.1 – 3 ng TEQ/kg |
| Measured | 0.16 – 10 ng TEQ/kg |

**Future research:**

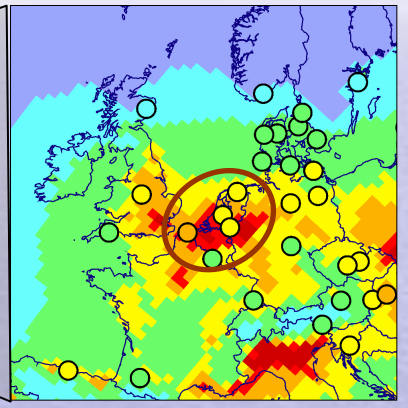
- Continuation of the analysis of PCDD/Fs emissions to the media other than atmosphere and their application in modelling

# Refinement of model parameterization: wind re-suspension (Pb)

Pb in air (2000-2011) model vs measurements




Pb in air (2000-2011)



Motivation:

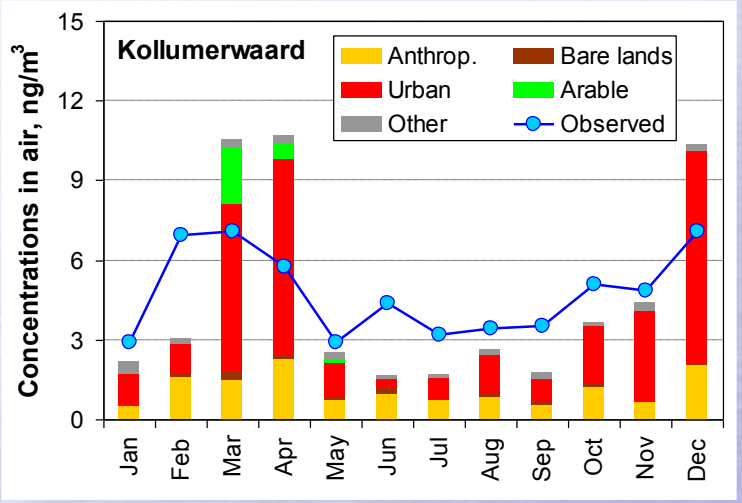
Overestimation of air concentrations in Benelux region

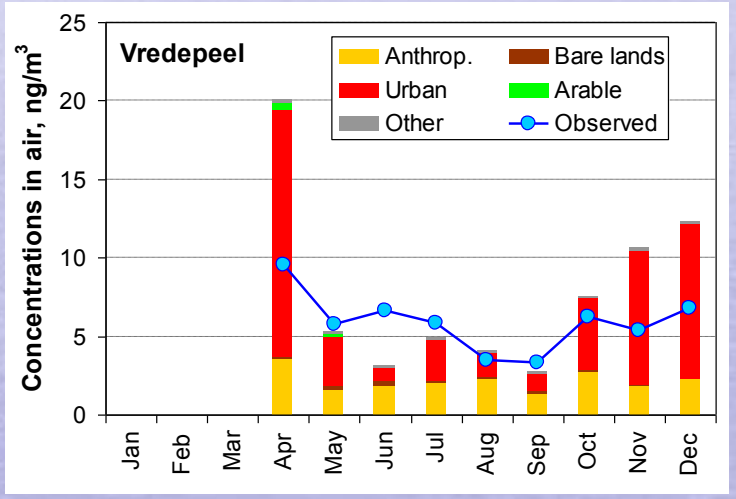
Method:

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



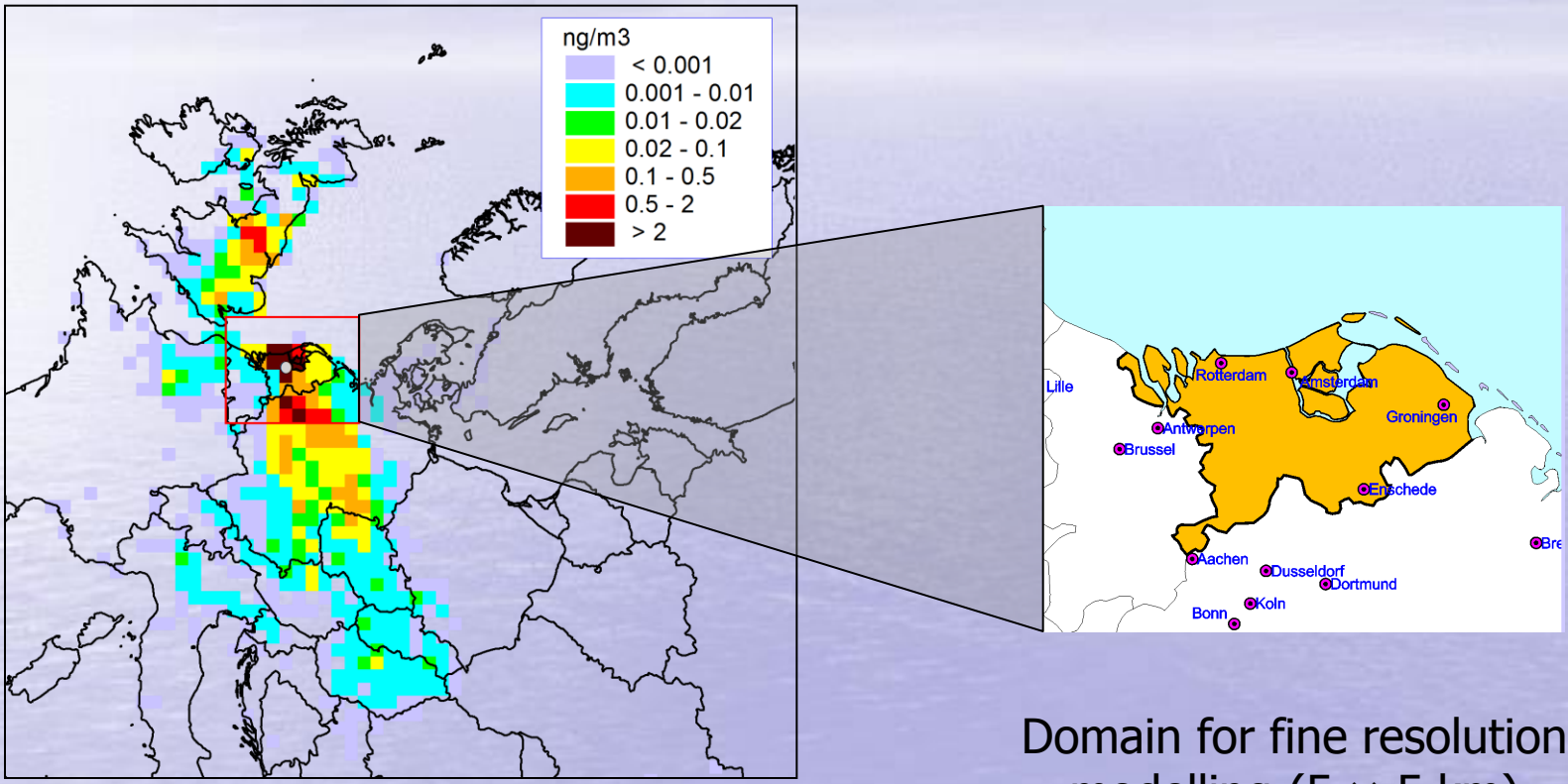




Wind re-suspension from urban areas

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

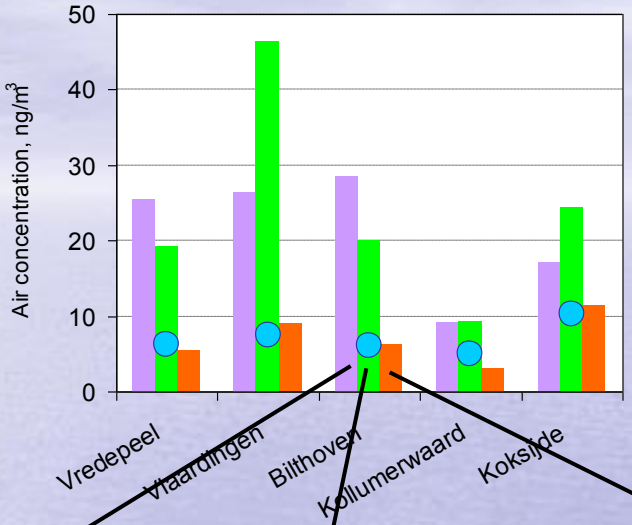



Contributions of re-suspension flux from urban regions to concentrations of lead at Bilthoven site (7 – 19 of April)

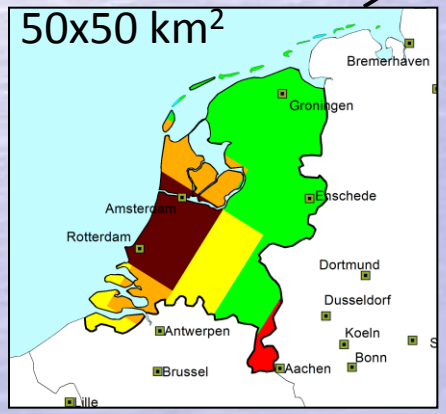
Domain for fine resolution modelling (5 × 5 km)

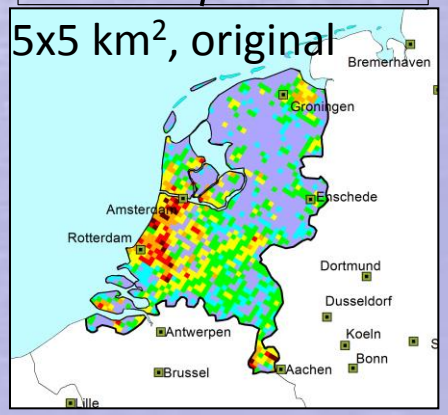
# Refinement of model parameterization: modelling with fine resolution (case study)

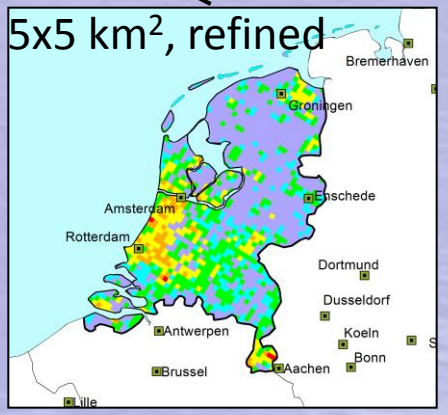
Modelled and observed Pb air concentrations at Dutch and Belgian stations



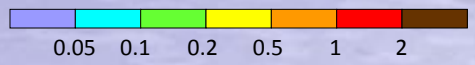

Modelling results




Wind re-suspension of Pb in the Netherlands, kg/km$^2$/y

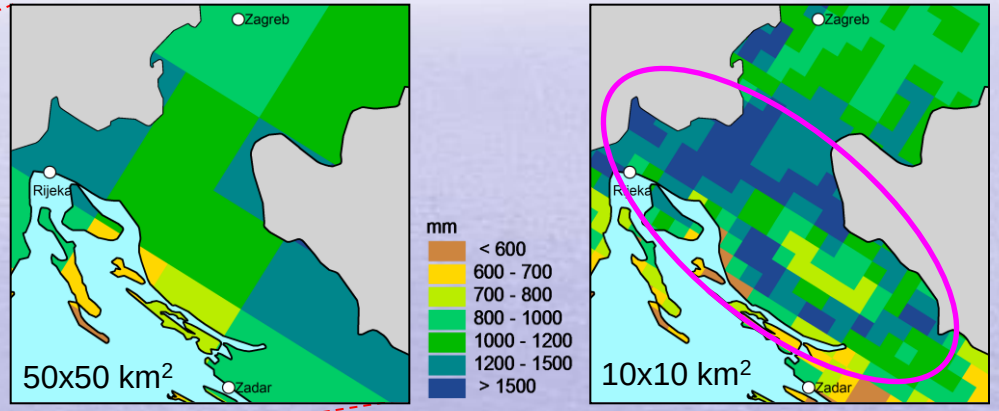


# Fine resolution

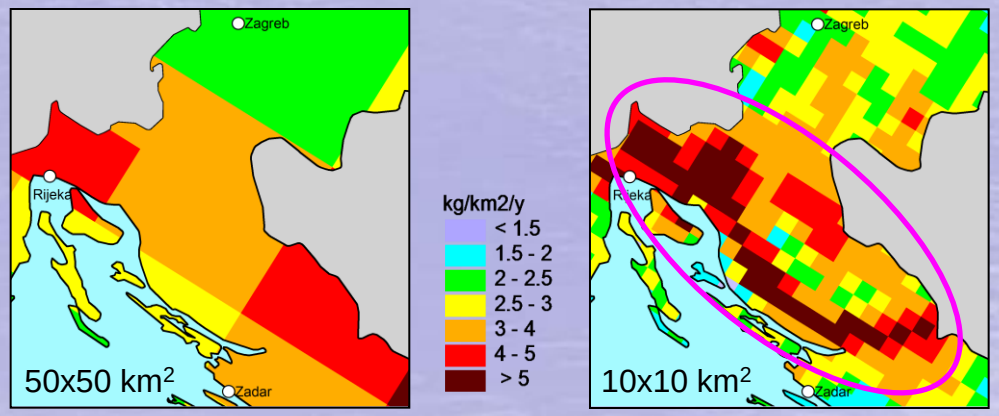
**Effect of use of fine resolution meteorological and geophysical data**

Croatia


Annual precipitation amount (2007)



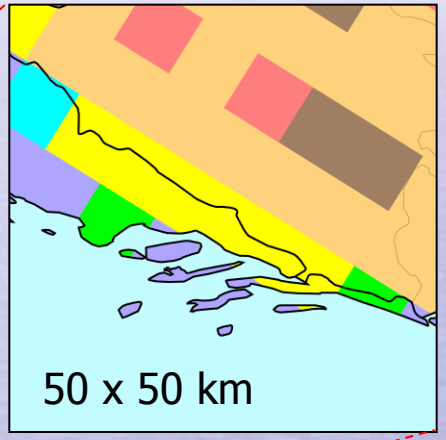

Total Pb deposition (2007)

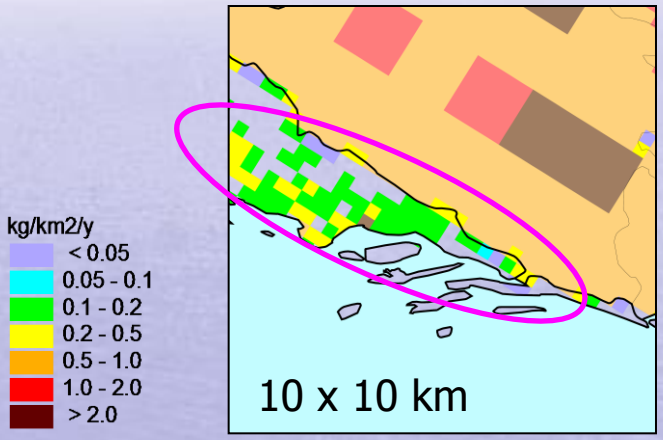


# Fine resolution

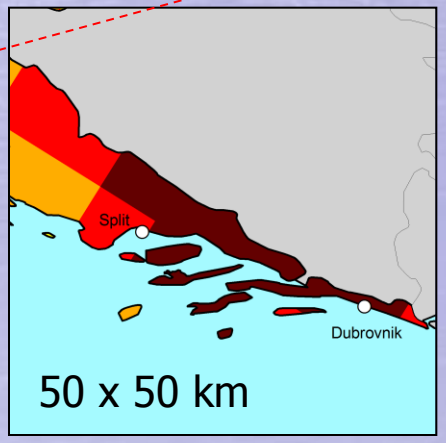
**Refinement of deposition fields in Croatia due to increased resolution of emission data**

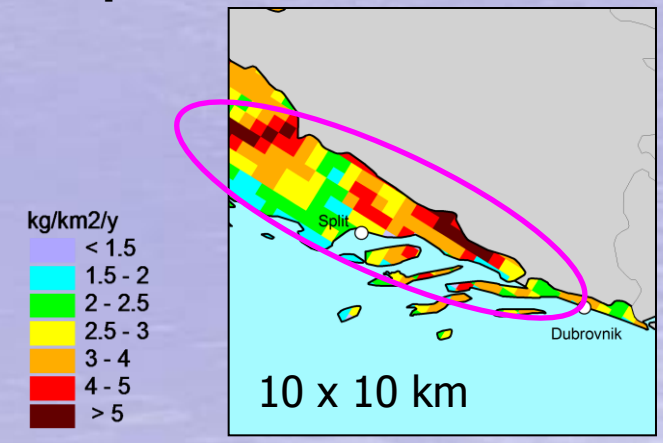

**Pb emission data**



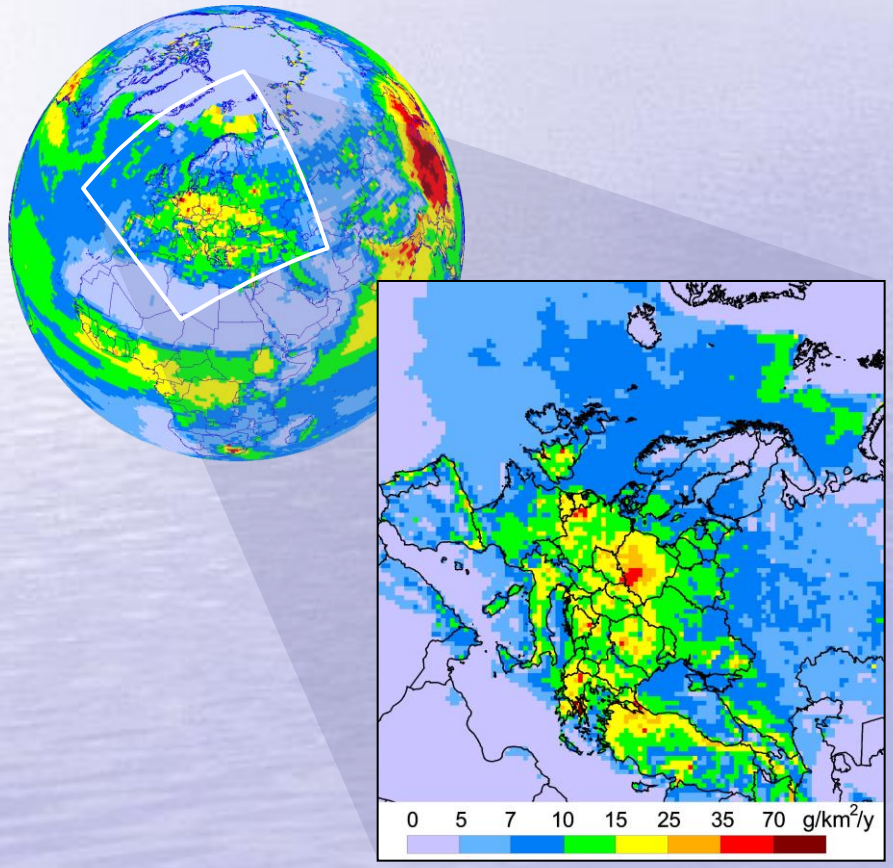

**Pb total deposition**



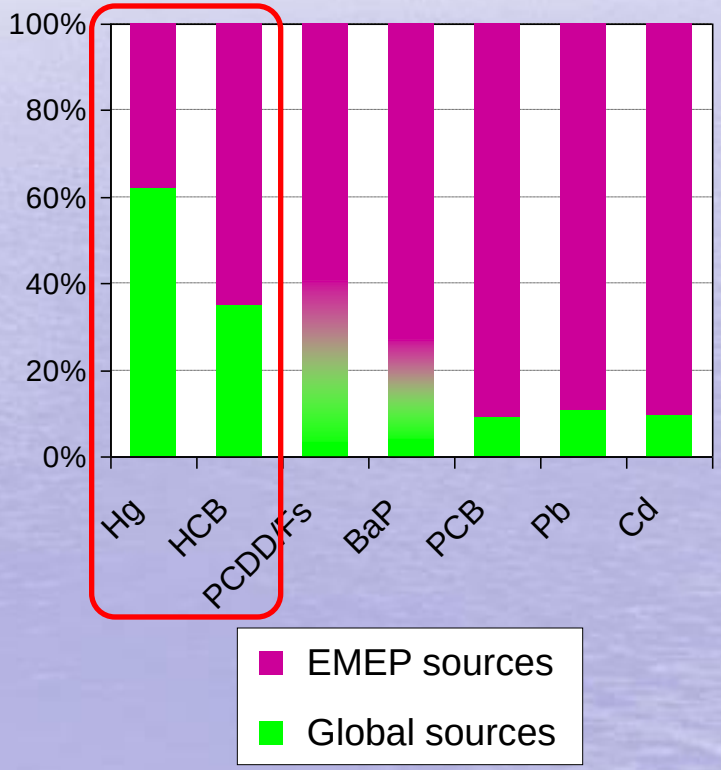



# Global modelling in lat/lon projection

Generation of boundary conditions for regional modelling




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



# Transition to new EMEP grid and finer resolution

Current grid, 50× 50 km

New grid, 0.1º × 0.1º

**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

| *Dataset* | *Chemicals* | *Years* | *Resolution* |
|---|---|---|---|
| AMAP/UNEP | Hg | 2010 | 0.5°×0.5° |
| Tao et al., 2009 | PAHs | 2004 | n/a |
| Breivik et al., 2007 | PCBs | 1930-2100 | 1°×1° |
| Bailey et al., 2001 | HCB | 1995 | n/a |
| NILU/CGEIC | Pb | 1989 | 1°×1° |

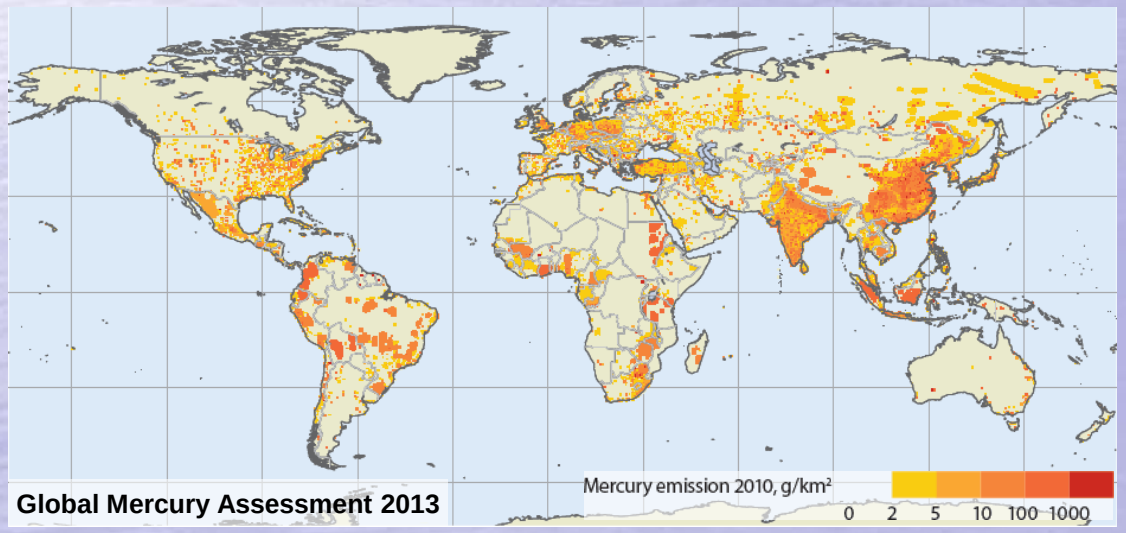
No global data for **Cd** and **PCDD/Fs**

# Global Hg emissions inventory

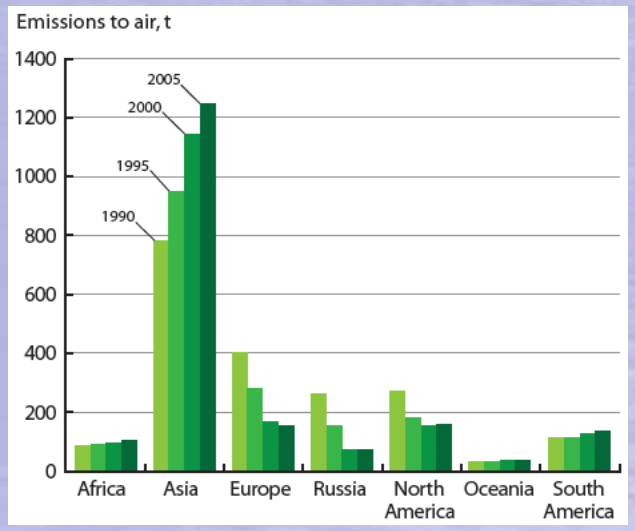
**Assessment of Hg emissions to the atmosphere:**

- 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**



# 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)