MODELLING OF POLYCYCLIC AROMATIC HYDROCARBONS FROM SELECTED POINT AND AREA SOURCES IN CENTRAL EUROPE

Jana Matejovičová [1,2], Christos Efstathiou[2], Gerhard Lammel [2,3], Jana Krajčovičová[1], Martin Kremler[1]

[1] Slovak Hydrometeorological Institute, Jeséniova 17, Bratislava, Slovakia
[2] Masaryk University, Research Centre for Toxic Compounds in Environment, Kamenice 5, Brno, Czech Republic
[3] Max Planck Institute for Chemistry, Hahn-Meitner-Weg 1, Mainz, Germany

Abstract: Atmospheric levels of polycyclic aromatic hydrocarbons (PAH) were established in the local and regional scales with a combination of modelling approaches and tools. The spatial distribution of benzo(a)pyrene B(a)P was modelled for a domain covering Europe using a modified version of the Community Multi-scale Air Quality (CMAQ) modelling system linked with the Weather Research and Forecast (WRF) model. In addition, the Lagrangian model CALPUFF was used to study the significance of point and area sources for B(a)P levels on the local scale.

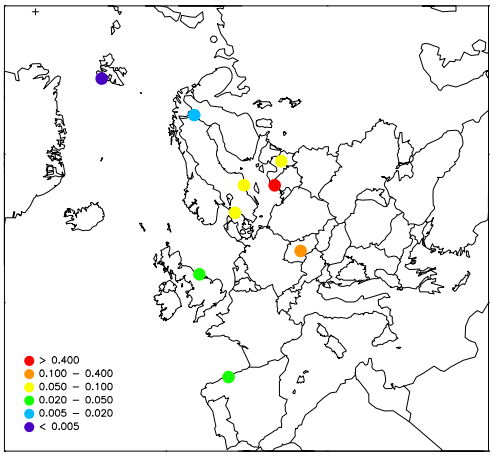
Key words: *Air quality modelling, CMAQ, CALPUFF, PAH emission sources, benzo(a)perene, PM_{10}*

INTRODUCTION

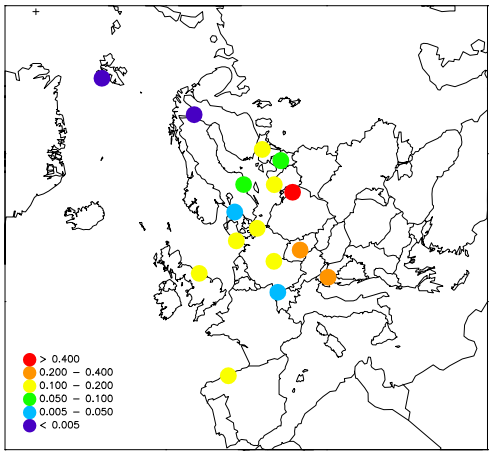
Mean annual concentrations of benzo(a)pyrene (B(a)P) frequently exceed the target value 1 ng m^{-3} at air quality measurement stations across Europe. Air quality modelling can bring additional information on the spatial distribution and the main sources of B(a)P, regional transport and processes, which influence its atmospheric levels. This study consists of two parts – simulations of B(a)P concentrations on the regional and local scale and their linkage with existing monitoring networks.

MEASUREMENTS OF SELECTED PAH

In the framework of the EMEP measurement programme there were 9 sites measuring POPs in two atmospheric phases (air and aerosol) in 2006 and 16 such sites measuring POPs in 2008.



Benzo-a-pyrene (BaP) in air, 2006 (ng.m^{-3})



Benzo-a-pyrene (BaP) in air, 2008 (ng.m^{-3})

Figure 1. Annual mean concentration of benzo-a-pyrene (B(a)P) in air on EMEP stations (source: Aas & Breivik 2008; Aas & Breivik 2010)

REGIONAL STUDY - SPACE DISTRIBUTION OF B(A)P CONCENTRATION

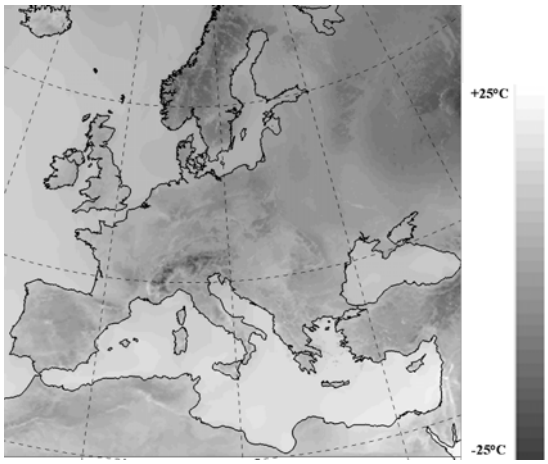
Models used

The Community Multi-scale Air Quality (CMAQ) modelling system was used to simulate air pollution in the regional scale. The computational design involves the preparation of meteorological inputs with the Weather Research and Forecasting (WRF) model for a gridded domain that covers Europe. Detailed emission inventory was created using the SMOKE-EU emissions model developed by the Institute for Coastal Research of Helmholtz Centre Geesthacht (Bieser et al. 2011).This work demonstrates the initial steps towards modelling of the processes that determine the concentrations of POPs in the atmosphere.

Meteorological input data for the regional study

Preliminary daily runs of WRF 3.2.1 have been completed with GFS data (space resolution 0.5° x 0.5°)
Computational domain: 450 x 370 grid cells, spatial resolution 12 x 12km.

Set up of WRF 3.2.1: WSM 3-class simple ice scheme, radiation long wave scheme 'rrtm'; radiation short wave scheme ' Dudhia ', near-surface Monin-Obukhov scheme, 4 soil layers, unified Noah land-surface model.



02.01.2006 06:00 Temperature in 2m

02.01.2006 06:00 wind in 10m

Figure 2. Example of the meteorological fields prepared by WRF 3.2.1

Emissions input data for the regional study

The emission model SMOKE is the official emission model of the Unites States Environmental Protection Agency (US EPA) and is one of the most used emission models worldwide (Houyoux et al. 2000). SMOKE was originally created by the MCNC Environmental Modeling Center (EMC) and developed further by the US EPA. It is the official emission model of the Models-3 Community Modeling and Analysis System (CMAS) and creates emission data suitable for CMAQ (Byun & Ching 1999; Byun & Schere 2006). Although SMOKE is highly specialized for usage with officially reported data in the US, there have been several successful attempts to use it for other regions. A modified version of SMOKE, SMOKE for Europe (SMOKE-EU), was developed by the Institute for Coastal Research of the Helmholtz Centre Geesthacht (Bieser et al. 2011). The emission data set was prepared for CMAQ 4.7.1.

Computational domain for CMAQ and model set up

Computational domain: 357 x 357 cells, space resolution 12 x 12km,
17 (non-hydrostatic sigma-p) vertical layers, chemical mechanism CB V, aerosol module aero5.

Figure 3. Input for CMAQ - emission rate of primary organic carbon, 02.01.2006 18:00

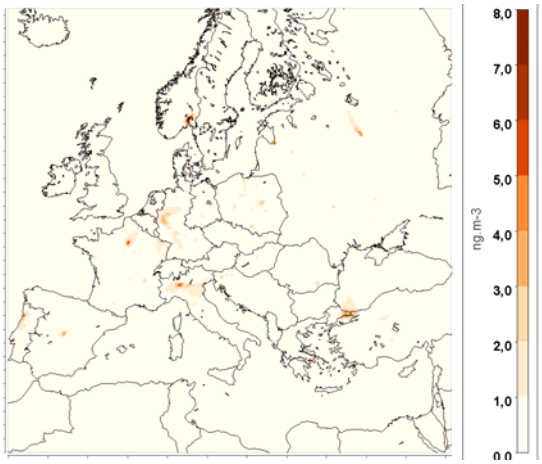


Figure 4. B(a)P 2.1.2006, 18:00 (scaled output of CMAQ)

The model simulation neglects chemical sinks (no degradation in gas or particle phase), while dry and wet deposition remove B(a)P from air.

LOCAL STUDY – COMPARISON OF POINT AND AREA SOURCES OF B(A)P

Model used, computational domain

The Lagrangian puff model CALPUFF (Scire et al., 2000) was used for the local study focused on the comparison of the emission sources. The aim was the source apportionment of PM_{10}. The domain of the size 13.5 x 22 km, with metallurgy and coke production (large industrial point source) and several areas of domestic heating was selected for modelling of B(a)P. The spatial resolution of the domain was 500 x 500 m. Simulation of PM_{10} for the year 2008 was run for each source type separately. No sink of B(a)P was included (dry and wet deposition switched off, no chemical degradation). Estimated annual mean concentration of B(a)P for each source type was obtained from annual mean concentration of PM_{10}, scaled by the ratio of the emission factors, i.e. $f_{B(a)P}/f_{PM10}$. Emission factors used for the reporting to the CLRTAP were used (Slovak Republic informative inventory report under the Convention on long range transboundary air pollution, 2010). In domestic heating, only wood combustion was taken into account, (amount of coal used for residential heating is negligible).
Detailed information on simulation set up and input data acquisition and processing is presented in a companion paper (HARMO 2013 poster No. H15-50; Krajčovičová et al., 2013)

RESULTS AND DISCUSSION

A regional-scale simulation was run for selected days of January 2006. B(a)P concentration was obtained in coherence to the method used for the local study - from the primary carbonaceous pollutants scaled by the ratio of B(a)P and PM_{10} emission factors, $f_{B(a)P}/f_{PM10}$. The dominance of strong area sources (urban areas including strong point sources) for the B(a)P distribution can be recognized (Figure 4). In the next step chemical degradation by O_3 will be included.
Atmospheric concentrations of B(a)P provided by the local-scale study were comparable to the concentrations measured at the air quality station close to the industrial source addressed. Concentrations of B(a)P in the range of 1.2 – 5.3 $ng.m^{-3}$ were measured in January 2008. The local study (Figure 5) confirmed the expectations that a large industrial point source contributes more significantly to B(a)P levels in the vicinity of this source than domestic heating does.

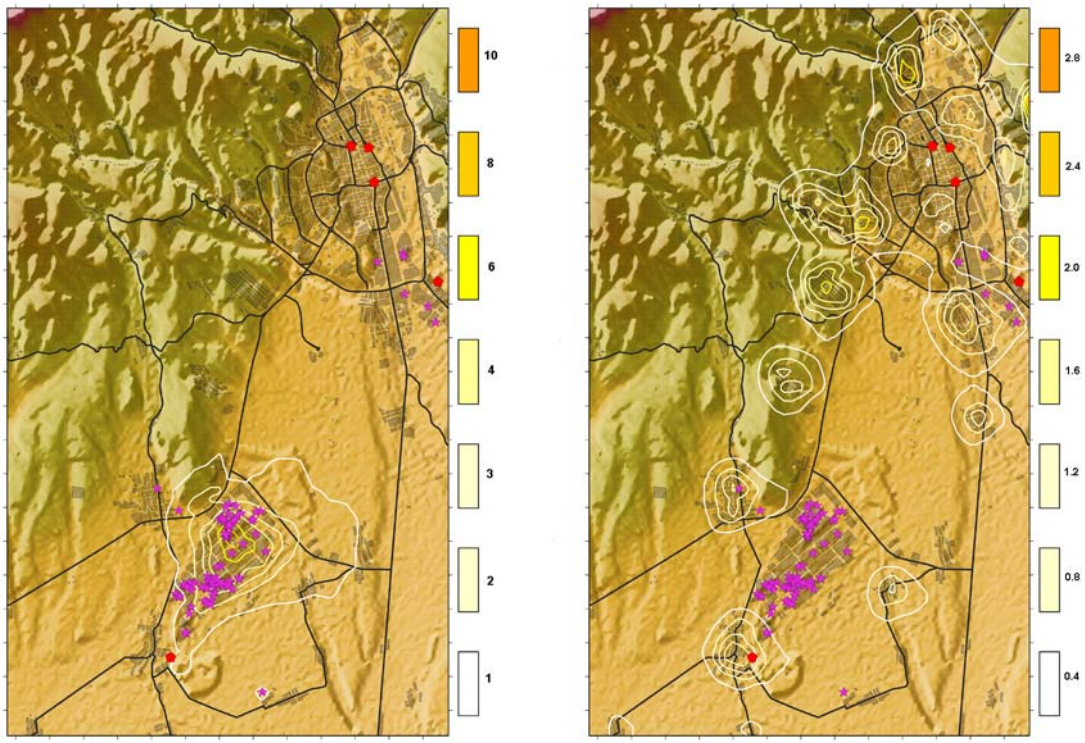


Figure 5. Annual mean concentration of B(a)P (ng m^{-3}) from large point source (metallurgy, on the left) and domestic heating (on the right). *(Legend: pink stars – point sources, red diamonds – air quality stations. B(a)P is measured close to the factory on the south)*

REFERENCES

Aas, K. & Breivik, K., 2008. *Heavy metals and POP measurements*, Kjeller: EMEP/CCC.

Aas, K. & Breivik, K., 2010. *Heavy metals and POP measurements*, Kjeller: EMEP/CCC.

Bieser, J. et al., 2011. SMOKE for Europe – adaptation, modification and evaluation of a comprehensive emission model for Europe. *Geoscientific Model Development*, 4(1), pp.47–68.

Byun, D. & Schere, K.L., 2006. Review of the Governing Equations, Computational Algorithms, and Other Components of the Models-3 Community Multiscale Air Quality (CMAQ) Modeling System. *Applied Mechanics Reviews*, 59(2), p.51.

Byun, D.W. & Ching, J., 1999. *Science algorithms of the EPA Models-3 community multiscale air quality (CMAQ) modeling system*, Washington, DC 20460: Office of Research and Development - USEPA.

Houyoux, M.R. et al., 2000. Emission inventory development and processing for the Seasonal Model for Regional Air Quality(SMRAQ) project. *Journal of Geophysical Research*, 105(D7), pp.9079–9090.

Scire, J.S., Strimaitis, D.G., Yamartino, R.J., 2000: A User's Guide for the CALPUFF Dispersion Model, Earth Tech, Inc. Concord, MA.

Slovak republic informative inventory report under the Convention on long range transboundary air pollution, Bratislava, 2010*, p.14. http://www.ceip.at/status-of-reporting/2010-submissions/*

ACKNOWLEDGEMENT

This research was funded by the CETOCOEN project from the European Regional Development Fund (No.CZ.1.05/2.1.00/01.0001) and the Czech Science Foundation (GACR project No. P503/11/1230)

The access to computing and storage facilities owned by parties and projects contributing to the National Grid Infrastructure MetaCentrum, provided under the programme "Projects of Large Infrastructure for Research, Development, and Innovations" (LM2010005) is highly appreciated.

We would like to acknowledge Johannes Bieser and Armin Aulinger, Institute for Coastal Research, Helmholtz Centre Geesthacht, for preparing of high quality emissions data.