

# LOCAL PM<sub>10</sub> SOURCE APPORTIONMENT FOR NON-ATTAINMENT AREAS IN SLOVAKIA

Jana Krajčovičová<sup>1</sup>, Martin Kremlér<sup>1</sup>, Jana Matejovičová<sup>1</sup>

<sup>1</sup> Slovak Hydrometeorological Institute, Jeséniova 17, Bratislava (Slovakia)

**Abstract:** The method used for the source apportionment in 17 air quality management areas (AQMA) in Slovakia is presented, as well as the results of the study. Local PM<sub>10</sub> emissions include traffic emissions, local seasonal heating sources from residential housing, industrial and point sources, and fugitive sources. Mathematical modeling has been performed using CALPUFF model, driven by meteorological fields created by CALMET meteorological model. Domains ranged between 35 - 300 km<sup>2</sup> in size, with the horizontal resolution of 200 – 500m, depending of the complexity of the terrain. Regional and transboundary contributions were computed using EMEP background measurements. For every domain, each source group has been modeled separately (i.e., road segments, continuous housing areas), in order to perform further post-processing aimed at adjusting their original emission rates in such a way as to best satisfy linear regression models fitted to daily mean PM<sub>10</sub> concentrations measured at monitoring stations. In most domains this method yielded encouraging results, consistently showing general overestimation of the original local heating emissions and underestimation of traffic emissions, as had been suspected from the beginning. The apportionment of PM<sub>10</sub> emissions itself will also be presented. In most AQMAs the major contributors are local heating using biomass burning (in winter) and road transport in both seasons, with high contribution from regional and transboundary transfer.

**Key words:** Atmospheric dispersion modeling, CALPUFF, source apportionment, PM<sub>10</sub>

## INTRODUCTION

Slovakia, as many other EU countries, encounters problems with exceeding the daily, and in some cases also the annual concentrations of PM<sub>10</sub>. Directive 2008/50/EC states the conditions under which air quality plans are required to be established. Annex XV contains the list of information to be included in those plans, namely, the origin of the pollution and details of the factors responsible for the exceedance. This is only possible to achieve using mathematical modeling. This paper aims to explain the methodology and results of PM<sub>10</sub> source apportionment as applied to 17 AQMA for which the air quality plans with regard to PM<sub>10</sub> have been established. The source apportionment was used as a basis for measures to be taken in order to combat the high levels of PM<sub>10</sub> concentrations as efficiently as possible in the framework of current valid national legislation and available financial resources.

## RELEVANT SOURCES OF PM<sub>10</sub> EMISSIONS

PM<sub>10</sub> is a pollutant of multiple origin, moreover, part of the emissions is a natural component of living environment. Only small part – large sources – are registered in the National Emission Information System (NEIS). The other sources have to be assessed based on the combination of different statistical and geographic data and respective emission factors. Methods for determination of these emissions are of varying accuracy, which strongly depends on the quality of available inputs.

The following source groups have been included in the simulations:

- Large and medium sources from NEIS database
- Seasonal sources of residential heating
- Road transport

### **Large and medium sources from NEIS database**

They comprise of seasonal point sources (centralized heating), non-seasonal point sources (industrial stacks), and fugitive industrial sources, represented as volume sources in the simulations.

### **Seasonal sources of residential heating**

These sources are associated with relatively high level of uncertainty, as they are not registered in any database. As they emit large amounts of PM<sub>10</sub>, it is necessary to fill this gap in future. However, until that time, it was necessary to elaborate an indirect method for quantifying and geographic allocation of this important emission sector. A method was designed based on energy balance approach (Krajčovičová & Matejovičová, 2010). Based on municipal statistical data such as number of households, technical parameters of houses, and local temperature data for particular heating season it is possible to compute the energy required for heating the households. At the same time, based on statistical data on the number of gas connections and total household gas consumption which is available for each municipality, it is possible to compute the energy supplied by gas burning appliances. From the available data on coal sales for each district, it is possible to compute the energy supplied by coal burning. The hypothesis is that the remaining energy deficit is supplied by wood burning. Using wood burning emission factors from literature, it is possible to assess PM<sub>10</sub> emissions for each municipality as a whole. These emissions are then geographically allocated to residential areas identified using Google Earth.

### **Road transport**

Exhaust and non-exhaust vehicle emissions were calculated using a top-down method from the total national emissions (COPERT 4), based on the ratio of road network length inside the domain to the total road network length. Consequently, the emissions are distributed throughout the roads in the domain based on the road category and length, vehicle counts and categories.

Resuspension of dust was estimated based on the AP 42 (US EPA) methodology using bottom-up approach.

From the resulting emissions per road segment, resuspension to sum of exhaust and non-exhaust emissions ratio is approximately 3:1. Available literature suggests that the ratio should be the opposite or at least 1:1 in most cases. In principle, the method used for allocation of exhaust and non-exhaust emissions tends to underestimate the emissions in cities and overestimate rural emissions. The underestimation of our emissions is also confirmed by the modeling results in most of the AQMA.

### **Regional background**

It forms an important part of total PM<sub>10</sub> concentrations in all AQMA. Regional background can be determined using measurements at EMEP background stations or applying a regional model with a sufficient accuracy. We had tried EMEP model, but the resolution was insufficient and the best option as appeared to be using EMEP background stations data.

## **MODELING TOOLS AND SETUP**

Most of Slovak territory is formed by a rather complex terrain with most of the AQMA situated in mountain valleys, causing generally low average winds and high percentage of calms over the year. This motivated the selection of CALPUFF (Scire a kol., 2000b) as our modeling tool, driven by diagnostic meteorological model CALMET (Scire a kol., 2000a). Modeling domains are between 60km<sup>2</sup> and 400km<sup>2</sup> in size, with the highest level at 3000m over the surface. Horizontal resolutions are 200m to 500m, depending of the complexity of the terrain, with 10 vertical layers. The terrain model (SRTM – Farr et al., 2007) and landuse (CORINE – Bossard et al., 2000) together with meteorological profiles and surface meteorological measurements are input to CALMET model, which calculates high resolution three dimensional wind fields reflecting local orography and circulation systems. CALPUFF is a lagrangian puff model which is capable of treating low wind and calm situations, while it contains basic chemical parametrisations for secondary aerosol formations.

## **SIMULATION AND POSTPROCESSING**

Involving large number of sources and long time period, CALPUFF simulations are computationally demanding. In order to manage the computing times efficiently, in each domain we divided the emission sources into 3 main groups: point sources – treated as stacks and volumes, small (local heating) sources – treated as adjacent volume sources covering continuous areas, and roads – treated as lines consisting of adjacent volumes. Each of the three main groups had several subgroups determined mostly by their geographic integrity. These geographically integral subgroups were simulated separately, keeping in mind a possible future scaling of their emissions.

As it was mentioned above, the estimates of local heating emissions and roads was associated with quite a large uncertainties; one could therefore call it as a „first guess“ estimate. Therefore, the post processing included an application of linear statistical model (LSM) at each receptor point located at the measurements site, in order to determine a kind of scaling coefficient for each emission group in a particular AQMA. As the same emission estimation methods have been used in most of AQMAs, it was supposed that if the scaling coefficients resulting from linear statistical models are consistent among AQMAs, they may reflect the level of under- or overestimation of our first guess values, while background coefficient reflects the geographical variability of regional background (the background stations are not located inside the AQMA domains).

In our case, the LSM is expressed as follows:

$$C_{oi} = k_1 \cdot C_{obi} + k_2 \cdot C_{mvi} + k_3 \cdot C_{mdi} + k_4 \cdot C_{mpi},$$

where  $C_{oi}$  is mean daily concentration measured at day  $i$  at the monitoring station,  
 $C_{obi}$  is mean daily concentration measured at day  $i$  at background monitoring station,  
 $C_{mvi}$  is the contribution of local heating at day  $i$ , modeled at the monitoring station using CALPUFF model,  
 $C_{mdi}$  is the contribution of road transport at day  $i$  modeled at the monitoring station using CALPUFF model,  
 $C_{mpi}$  is the contribution of point sources at day  $i$  modeled at the monitoring station using CALPUFF model,  
 $k_1, k_2, k_3, k_4$  are coefficients of LSM for background, local heating, road transport and point sources.

## RESULTS AND DISCUSSION

The LSM coefficients resulting from the simulations and subsequent postprocessing seem to be relatively consistent among the domains. As can be seen from Table 1, their values suggest a relatively high original overestimation of local heating emissions ( $k_2 = 0.1\text{-}0.5$ ), and underestimation of road transport emissions ( $k_3 = 0.5\text{-}2.5$ ) in most domains. Correlation coefficients between measured and modeled concentrations vary from 0.49 to 0.77. Figure 1 shows the source apportionment of monthly concentration values for different AQMAs. Regional background forms the most relevant contribution to these averages in all stations. The other important sources are local heating by wood and traffic.

However, the partitioning needs to be taken into consideration with care, as the inputs, namely, the emission estimates are associated with high level of uncertainty. Therefore, our future plans are focused to the improvement of the emission database. Improvements in local heating emission model are planned, including up-to-date statistical data on housing and heating sources from the latest census. Moreover, local governments of AQMA are encouraged to acquire relevant data on household heating appliances and consumption of wood in their area of interest. As to the road transport database, a new methodology is going to be applied based on the bottom-up approach in collaboration with the national and local road operators.

Table 1. Basic statistical scores of modeled daily concentrations after LSM coefficients were applied

AQMA monitoring station	LSM coefficients				Rho	RMSE	FB <sub>n</sub>	FB <sub>p</sub>	FB
	$k_1$	$k_2$	$k_3$	$k_4^*$					
Nitra (UB)	1	0.5	2	-	0.77	9.85	0.27	0.06	0.21
Žiar nad Hronom (UB)	1.3	0.2	2	-	0.75	8.94	0.13	0.11	0.02
Malacky (UT)	1.5	0.1	2	-	0.73	10.12	0.14	0.09	0.05
Hnúšťa (SB)	1.5	0.4	2	-	0.66	13.71	0.2	0.11	0.09
Jelšava (UB)	1.5	2.0	2	-	0.62	19.7	0.25	0.16	0.09
Krompachy (UB)	1.5	1	2	-	0.71	12.84	0.18	0.12	0.06
Martin (UT)	1.5	0.16	2	-	0.7	12.58	0.15	0.11	0.04
Prešov (UB)	1.5	0.2	2.3	-	0.63	15.1	0.18	0.14	0.04
Senica (UT)	1.55	0.41	1	-	0.77	8.54	0.14	0.10	0.04
Strážske (UB)	1.1	0.3	0.5	-	0.71	10.36	0.2	0.14	0.06
Trenčín (UT)	1.2	0.1	0.5	-	0.64	10.99	0.15	0.17	-0.03
Trnava (UT)	1.5	0.3	3	-	0.7	13.16	0.19	0.1	0.09
Vranov nad Topľou (UB)	1.5	0.1	2.5	-	0.66	14.02	0.16	0.13	0.03
Žilina (UB)	1.5	0.2	0.9	-	0.71	14.09	0.17	0.14	0.02
Ružomberok (UB)	1.5	0.2	3	-	0.73	13.99	0.14	0.14	0.00
Banská Bystrica (UT)	1.3	0.2	1.7	-	0.54	23.86	0.26	0.14	0.12
Veľká Ida (SI)	1	0.1	2	2	0.47	22.21	0.31	0.08	0.23
Košice – Štúrova (UT)	1	1	1	1	0.7	14.76	0.11	0.24	-0.13
Košice – Strojarska (UB)	1	1	1	1	0.72	13.31	0.15	0.14	0.00

**Rho** – Spearman correlation coefficient, **RMSE** – Root mean square error, **FB<sub>n</sub>**, **FB<sub>p</sub>**, **FB** – Fractional biases: negative, positive and total, Station classification: UB – urban background, UT – urban transport, SB – suburban background, SI – suburban industrial

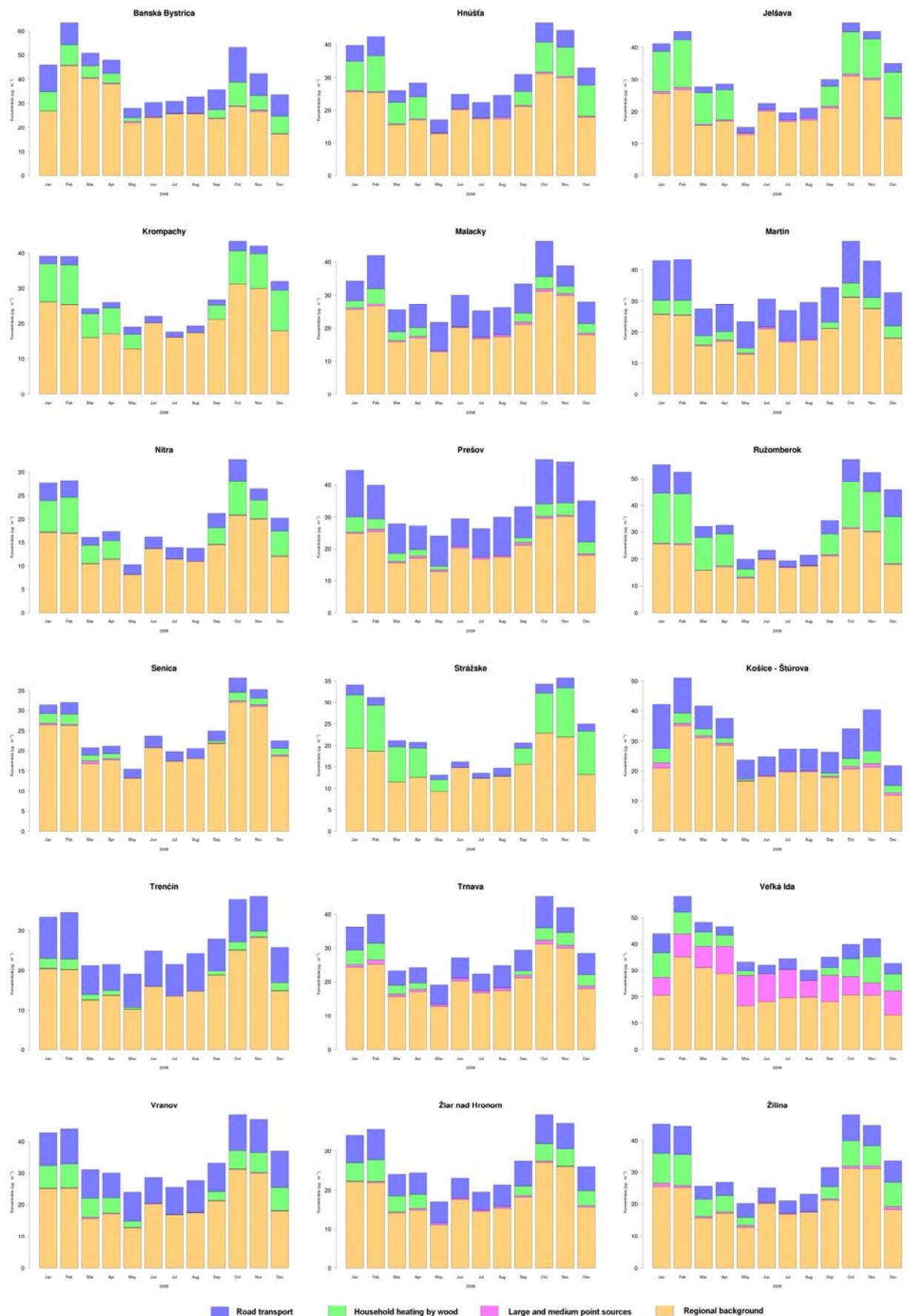


Figure 1. Source apportionment of monthly mean  $\text{PM}_{10}$  concentrations at different monitoring stations. (Note the scale of Y-axis is not constant throughout the graphs).

## **REFERENCE**

- Scire J.S., Robe F.R., Fernau M.E., Yamartino R.J., 2000a: A User's Guide for the CALMET Meteorological Model. Earth Tech, Inc., Concord, MA
- Scire, J.S., Strimaitis, D.G., Yamartino, R.J., 2000b: A User's Guide for the CALPUFF Dispersion Model, Earth Tech, Inc. Concord, MA.
- Krajčovičová J., Matejovičová J. 2010: Modelovanie geografického rozloženia emisií PM<sub>10</sub> z malých zdrojov – emisie z vykurovania drevom. *Ochrana ovzdušia 2010*. Kongres Studio s.r.o., ISBN 978-80-970356-3-1. 77-79
- Farr, T. G., et al., 2007: The Shuttle Radar Topography Mission, *Rev. Geophys.*, **45**, RG2004, doi:10.1029/2005RG000183.
- Bossard, M. et al., 2000: CORINE land cover technical guide – Addendum 2000. European Environment Agency, Copenhagen.