# $PM_{10}$ SOURCE APPORTIONMENT FOR NON-ATTAINMENT AREAS BASED ON ROUTINELY AVAILABLE DATA

*Jana Krajčovičová, Jana Matejovičová, Gabriel Szabó*

Slovak Hydrometeorological Institute (SHMU), Bratislava, Slovakia

**Abstract**: Directive 2008-50/EC allows for the postponement of attainment deadlines and an exemption from the obligation to apply limit values of PM10. Member states must notify the Commission about the postponement. Exemption can be granted in case of adverse dispersion characteristics, climatic conditions or transboundary contributions. In any case, a detailed source apportionment must be provided for each limit value exceedance case.
The presentation covers the method used for the source apportionment in 16 air quality management zones and 2 agglomerations in Slovakia. In order to get the transboundary contributions to the concentrations measured at monitoring sites, we run EMEP model with Slovak emissions subtracted from the input. The problem with local $PM_{10}$ emissions is that only part of them can be reasonably quantified with sufficient resolution: large and medium point stack emissions and traffic emissions. Their contributions were modelled using high resolution CEMOD model. The remaining parts of measured concentrations needed to be apportioned to so-called "unknown" sources – local household heating systems fuelled by wood and coal, re-suspended dust from roads, construction sites, uncovered soil, surface mines, waste dumps, industrial sites, etc. In order to do so, a method was used which, at each monitoring site, combined the results of EMEP and CEMOD models with statistical analysis of the correlation of hourly concentrations with local meteorological conditions (wind speed, wind direction) and spatial distribution of all potential PM10 sources around each particular monitoring station. In order to determine a spatial impact extent of different types of "unknown" sources, a series of modeling tests were performed for the conditions of each relevant geographical siting and respective meteorological conditions. This method gave reasonable results especially for the sites with mean annual wind speed higher than 1.5 m/s. For the sites with lower winds and large percentage of calms the resuspension plays minor role and concentration exceedances can be attributed to local and regional stack, chimney and exhaust emissions.

***Key words:*** *Directive 2008-50/EC, $PM_{10}$, atmospheric modeling*

## INTRODUCTION

EU in the Directive 2008-50/EC and respective implementation documents states firm conditions under which an exception from the obligation to apply limit values of $PM_{10}$ until 2011 can be granted to a member state. Basically, a member state asking for the exception is required to demonstrate for each of the concerned non-attainment areas that:

- one of the three conditions (adverse climatic conditions, adverse dispersion characteristics, or transboundary contributions) apply,
- the proper measures, based on detailed source apportionment, has been taken to decrease the $PM_{10}$ concentrations before the initial deadline,
- adequate measures, based on detailed source apportionment, are being applied in order to achieve the compliance by the new deadline (i.e., 2011),
- $PM_{10}$ concentrations will comply with the limit values by the new deadline.

In order to demonstrate that, a detailed information needs to be provided by filling out extensive Excel forms for each exceedance situation, including detailed source apportionment, quantitative impact of each measure taken to each particular relevant source, future quantitative projections of measures currently under implementation and future measures in the form of numerical contributions to particular mean annual concentrations and/or number of exceedances of daily limit values.

In the reference year of 2005 there were 17 non-attainment areas exceeding the allowed daily limit value exceedances, a few of which also exceeded annual limit value. Most of them can demonstrate adverse climatic conditions characterized by low annual wind speeds (less than 1.5 m/s), other, better ventilated are subject to transboundary transport of $PM_{10}$. In order to satisfy the requirement of EU legislation, the procedures summarized above needed to be carried out for each particular non-attainment area, including a detailed source apportionment of $PM_{10}$ concentrations measured at the monitoring stations (1 to 3 in each non-attainment area). Clearly, such an effort requires in each case extensive high-resolution mathematical modeling with the assumption of having input emission data of adequate quality. As we did not dispose of neither time nor money for an additional emission data refinement, we had to make use of what was routinely available at that time.

## SUBTRACTION OF TRANSBOUNDARY CONTRIBUTION

As everybody knows, $PM_{10}$ is a pollutant subject to rather efficient long-range transport, especially the fine particulate portion of the spectra. Another unpleasant property of $PM_{10}$ is the multitude and variety of its emission sources, most of which is of fugitive nature and their quantification is complicated and associated with huge uncertainty, if possible at all. However, many of the sources are possible to assess on larger scales, such as district and national, using various activity statistics and emission factors. These data are then used in long-range chemical transport models such as, e.g., EMEP. According to EMEP publication Nyíri and kol (2007, 2008, 2009), only about 10% of $PM_{coarse}$ and 15% of $PM_{2.5}$ measured in Slovakia is contributed by sources on the territory of the country. Fig. 1 shows the mean annual $PM_{10}$ concentrations (red) and transboundary contributions (blue) at model gridpoints as computed by EMEP model at the reference year of 2005.

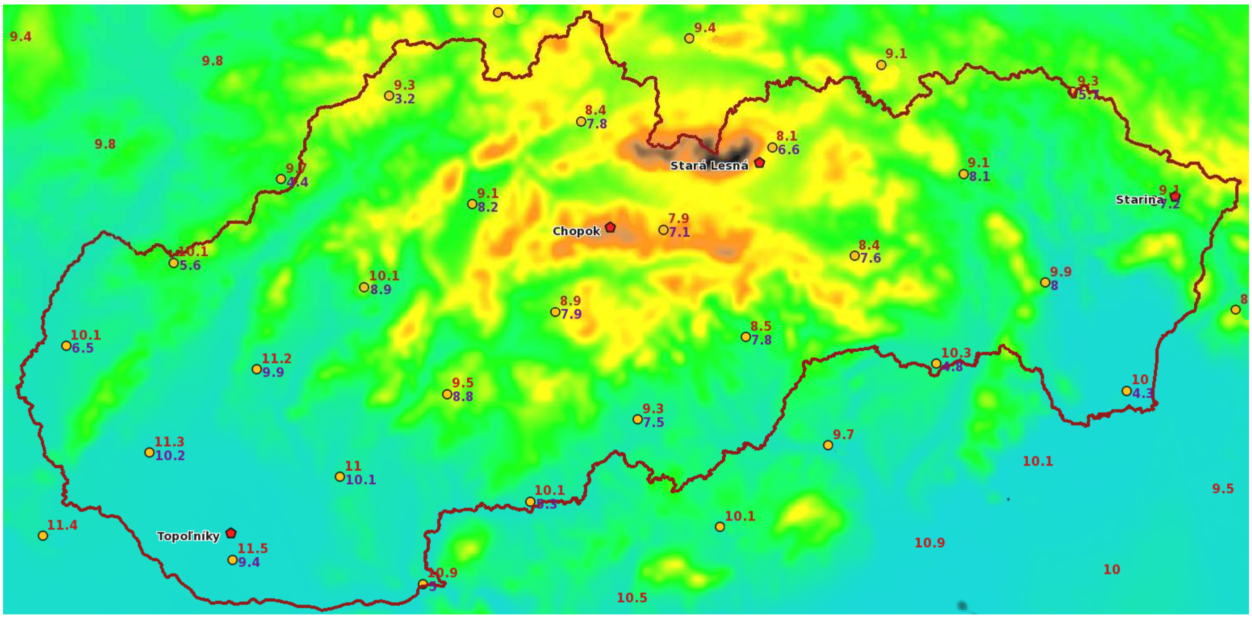



Figure 1.
EMEP model gridpoints (yellow circles) over geographic map of Slovakia, with the total mean annual $PM_{10}$ concentrations (red values, in $\mu g/m^3$) and mean annual transboundary contributions (blue values, in $\mu g/m^3$). Positions of EMEP monitoring stations are marked red (no adjustment to the negative biases has been made).

Unfortunately, the EMEP model underestimates the $PM_{10}$ concentrations by about 45% (according to EMEP Status Report 4/2008, EMEP station in Illmitz, AT, which may be nearest representative station in our geographic area included in model validation). The underestimation is probably caused by the nature and uncertainties in gridded emission data, and cannot be used directly for the estimation of the transboundary contribution at each particular AMS station. However, using EMEP – derived data with caution, it can be used for determination of relative importance of the transboundary vs. local/regional origin of $PM_{10}$ concentrations.

Transboudary contributions do not play a major role in the valley-situated non-attainment areas with low mean annual wind speeds. However, it is important in lowland and wide-valley non-attainment areas.
There are two issues needed to be solved in order to subtract the transboundary portion of the concentration from measured concentration:

1. prove that the $PM_{10}$ measured at the day of an exceedance originated outside the territory of Slovakia
2. determine the portion of $PM_{10}$ concentration which is to be extracted from the measured concentration

The first issue was solved using Hysplit model for obtaining ensemble backward trajectories calculated for the period of 48 hours for each of the AMS station in question. For the cases of high variability in ensemble members, also the EMEP trajectory model was run for a closest EMEP station.

The transboundary portion of $PM_{10}$ concentration to be extracted from the measured concentration was determined in the following way:

EMEP model was run for the whole year of 2005 in two modes: first the complete set of emission data was used, and second the Slovak emissions were set to zero. The ratio of the resulting concentrations computed from the two runs can be considered as the the ratio of regional and transboundary concentration contributions on the territory of Slovakia. However, for the abovementioned purposes, an absolute value of transboundary contribution is needed. As it was mentioned earlier in this chapter, EMEP model has rather large negative bias, which we need to eliminate. This was done by comparing EMEP model concentrations to the concentrations measured at EMEP station in Topoľníky. Model errors obtained this way were used for the correction of the EMEP-computed transboudary contributions in each concerned AMS station and each relevant exceedance day. The corrected transboundary contributions were then subtracted from the concentrations measured at the AMS stations, which reduced the number of exceedances under the limit value of 35, as is shown in Fig. 2.

| Number of exceedances | | | | |
|---|---|---|---|---|
| **AMS station** | **Before subtraction** | **After subtraction** | **Excluded*** | **Final** |
| BA, Kamenné nám. | 45 | 12 | 2 | 14 |
| BA, Mamateyova | 71 | 11 | 1 | 12 |
| BA, Trnavské Mýto | 101 | 34 | | 34 |
| Prievidza | 127 | 56 | 1 | 57 |
| Handlová | 40 | 10 | | 10 |
| Bystričany | 144 | 61 | 1 | 62 |
| Košice, Strojárska | 45 | 21 | 1 | 22 |
| Košice, Štúrova | 75 | 34 | 1 | 35 |
| Nitra | 125 | 44 | 1 | 45 |
| Prešov, Solivarská | 55 | 14 | | 14 |
| Prešov, Levočská | 66 | 26 | 1 | 27 |
| Trenčín | 51 | 11 | | 11 |
| Trnava | 109 | 34 | | 34 |
| Veľká Ida | 198 | 115 | 3 | 118 |

**Excluded based on the trajectory analysis for the sake of conservativeness*

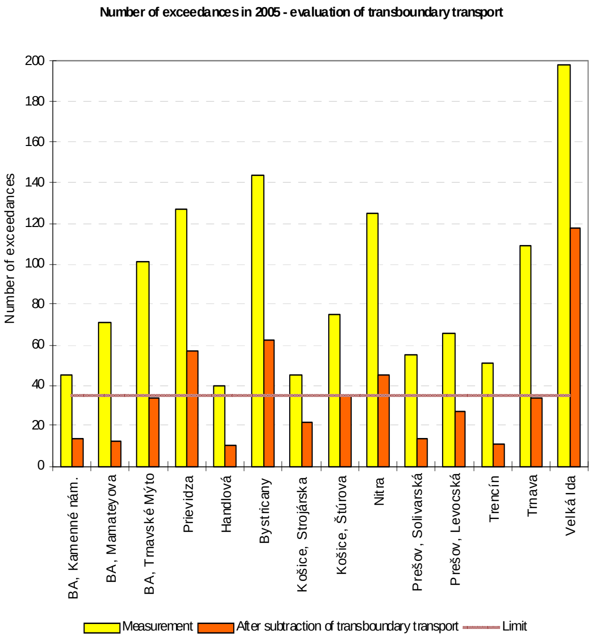



Figure 2
Number of $PM_{10}$ LV exceedances before and after substraction of the transboudary contributions

## $PM_{10}$ SOURCE APPORTIONMENT - ANNUAL

The following paragraph describes a method used in *Air Quality Assessment* yearbooks for the source apportionment of mean annual concentrations measured at the automatic monitoring stations:

The method is based on the fact, that the only $PM_{10}$ emission sources which are quantified in detail sufficient for an application of local air quality model are large and medium pollution sources from national NEIS database, and traffic-related exhaust and non-exhaust emissions based on transportation statistics data, road network and known emission factors. These known emissions were input into CEMOD model (Szabo, 2003) to obtain an annual $PM_{10}$ concentration contribution from these sources. Afterwards, concentration measurements from EMEP background stations were used for the assessment of the regional contribution to mean annual $PM_{10}$ concentrations. As the four EMEP stations represent different terrain elevations from lowlands to mountains (up to 2000 m), an empirical profile has been calculated and used to relate the regional concentration to particular elevations of different AMS stations in AQMA. Finally, the modeled contributions from the large and medium stationary sources and traffic, and the EMEP-based regional contributions were subtracted from the mean annual concentrations measured at AMS stations; resulting concentration residuals were assigned to the remaining *other* sources of $PM_{10}$, which are numerous and as such they are responsible for about 30 to 70% of annual $PM_{10}$ concentrations. These *other* sources include individual local heating systems burning coal and wood, dust from roads including winter sanding material, construction sites, resuspension of dusty material from open surfaces, agricultural soil, seasonal farming activities (harvest, dry plowing, autumn burning of plant remnants), and probably more. The presence of these sources is known in most cases, but their quantification is problematic due to their transient character. However, some of them can be located using detailed photomap of a site.

## $PM_{10}$ SOURCE APPORTIONMENT - DAILY

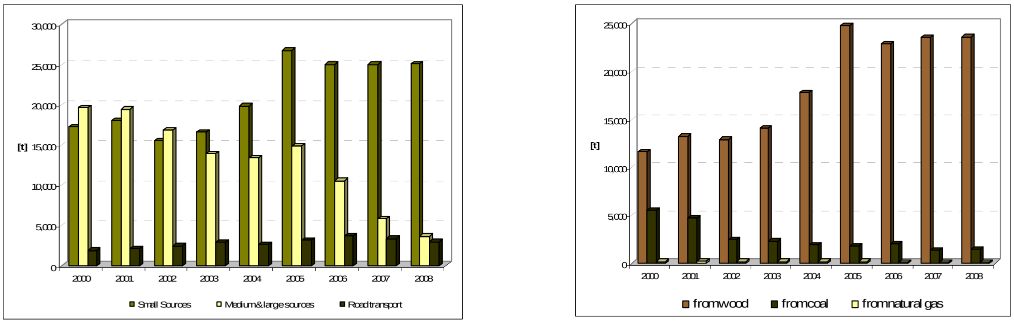
The method described above cannot be used for the source apportionment during the daily exceedance cases in the reference year of 2005 – the reason is that the gravimetric method used for the measurement of $PM_{10}$ concentrations at EMEP stations used 7-day $PM_{10}$ collection period at that time. However, it is rather useful for the first approximation on the $PM_{10}$ balance at each particular AMS station.

To obtain more detail on the reasons of daily exceedances, another complementary method was used, which combines correlated hourly wind and concentration statistics and spatial distribution of potential $PM_{10}$ sources around each monitoring station. It also allows for the determination of the relative importance of the *other* sources and so provides the local authorities with the information necessary for more efficient application of the abatement strategies and measures for the improvement of the air quality in their territories.

Although the $PM_{10}$ limit values apply to daily means, it was found out that a consistent statistical analysis of hourly wind and concentration data is more useful for the kind of analysis required. The wind and respective concentration data has been divided into 6 statistical bins based on the directional sector, and these bins were further divided according the wind speed into 15 bins each, in order to reflect not only the dependence of the concentration on the wind direction but also the wind speed. This distinction is particularly important in case of $PM_{10}$, in order to distinguish between 2 important source groups: local heating and transportation – causing high concentrations during low wind speeds, and local sources of re-suspended dust which occurs during higher wind speeds. To include sufficient number of higher wind speed cases, especially in the mountainous zones, complete set of hourly data for the last 4 years (2005-2008) was used.

## OTHER SOURCES – QUANTIFICATION OF EMISSIONS

The semi-qualitative method of daily $PM_{10}$ source apportionment described in the previous paragraph is a basic solution. It identifies local heating systems as an important potential emission source. It is in accordance with the total national emission assessments for different sectors as illustrated in Fig. 3. It shows the increasing trend in the emissions of small sources since 2002 associated with the increasing prices of natural gas and switching to wood fuel. This is especially true in mountainous regions with adverse climatic conditions, where cheap wood is readily available substitute for cleaner fuels.




a) b)

Figure 3
$PM_{10}$ emissions in Slovakia: a) according to individual sectors, b) small sources according to type of fuel
Note: Included are stack emissions from small, medium and large sources, exhaust and abrasive emissions from mobile sources

Therefore, we focused on determination of $PM_{10}$ emissions from domestic heating in best possible resolution in order to use it as input into CALPUFF air dispersion model in terms of area sources.

After thorough analysis of base data available for Slovakia, it was found out that the best achievable resolution is the spatial resolution of CORINE landuse data (100m).
Following is the list of data we used for the computation of emission fluxes from domestic heating:

- statistical data available down to smallest administrative units – municipalities:
  - number and habitable area of individual family houses - $N_h$, $A_h$
  - number and habitable area of apartments in apartment houses - $N_a$, $A_a$
  - population - $P$
  - number households connected to natural gas supply - $N_{gas}$
  - number of households equipped with a bathroom - $N_b$
  - annual consumption of natural gas in households - $C_{gas}$
- statistical data aggregated on district levels:
  - total amounts of coal and coal products sold in each district area (there are 77 districts in Slovakia) - $C_{co}$
  - population - $P_d$
- statistical data aggregated on national scale:
  - total electrical energy used for heating of households - $E_{ee}$
  - population - $P_n$
- climatologic data related to each individual municipality (from the nearest climatologic station):
  - duration of heating period in winter season (specific for each year) – *d (days)*
  - average temperature of heating period (specific for each year) – $T_{es}$ *(º C)*
  - minimum of the minimum temperatures of three consecutive days in winter season (as required by technical standards in construction industry) – $T_e$ *(º C)*

- GIS data
  - vector map of municipalities
  - raster map of CORINE landuse data

As no data are available on the amount of wood used as fuel for household heating, a method has been used based on balance between the amounts of energy necessary for production on heat in individual households in a given municipality and the amount of energy produced using known amounts of natural gas and coal. The difference between these energies is supposed to be supplied by combustion of wood. The following assumptions are made:

- blocks of apartment housing use central heating system if in cities, or natural gas if situated in villages
- all households having a natural gas connection installed use it also for cooking
- all households having a natural gas connection and are equipped with a bathroom use natural gas for heating of water

The energy needed for heating the households $Q_h$ $(d, T_{es}, T_e, A_h, A_a)$ and for heating water $Q_w$ $(N_h, N_a, P)$ is computed for each municipality using formulas from technical standards in construction industry; Production of energy for heating of households and water using gas $Q_g$ $(C_{gas}, N_{gas})$, coal $Q_{co}$ $(C_{co}, P_d)$, and electricity $Q_{ee}$ $(E_{ee}, P_n)$ are then subtracted. The standard efficiencies of different fuel-to-energy conversions (gas, coal, wood, electricity) are considered during computations, and the resulting mass of wood necessary to supply the energy deficit is multiplied by $PM_{10}$ emission factor for wood, as stated in MZP SR (2008).

Resulting $PM_{10}$ seasonal emissions are then imported to GIS system (we are using GRASS: http://grass.osgeo.org/) and attached to vector layer of municipalities. The vector layer is then converted to raster with required resolution, with pixel values set to $PM_{10}$ emission. Using mask created from CORINE raster of selected classes (1.1.1 and 1.1.2, i.e., continuous and discontinuous residential area) we get geographical locations of residential area sources with attached $PM_{10}$ emissions. Fig. 4 shows the distribution of $PM_{10}$ annual emissions per $m^2$ of household in order to normalize the results. It shows clear correlation of usage of wood fuel with the geographical location in the vicinity of woods in the mountains.

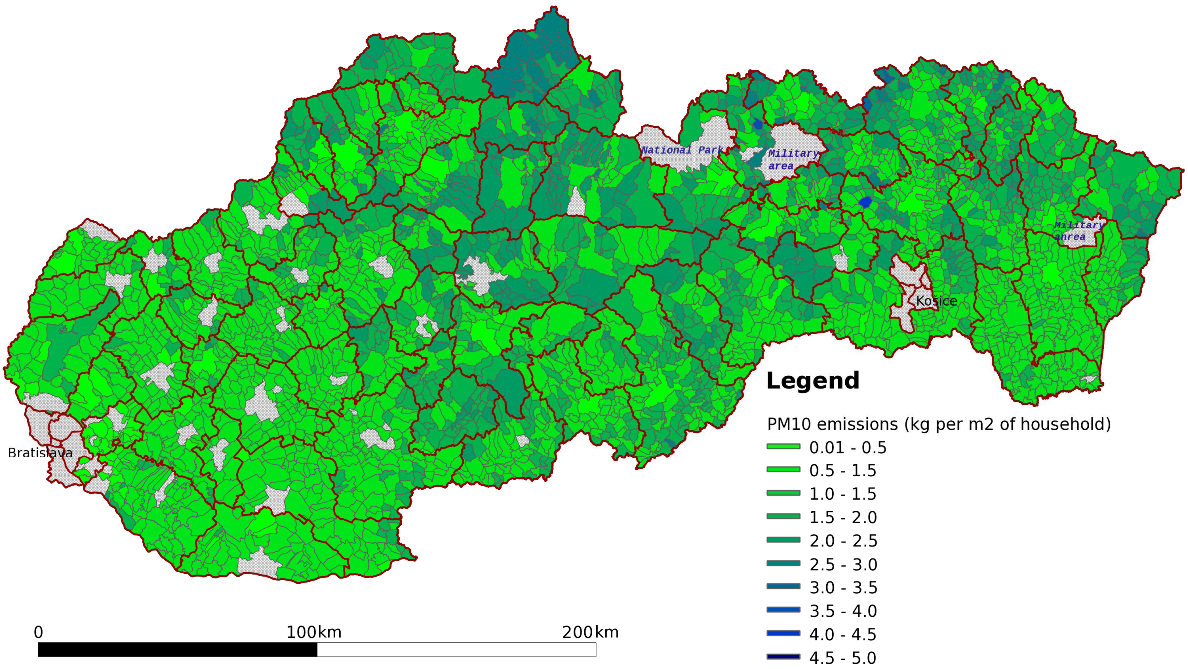



Figure 4.
Map of $PM_{10}$ annual emissions from wood burning (kg per $m^2$ of household)

Although the resulting geographical distribution of wood combustion gives reasonable picture, there are still some problems which bring increased uncertainties to the results. As an example, you can see some of the municipalities grayed. One of them is the area of TANAP national park with sparse permanent population; other two are non-inhabited military areas. The remaining grey municipalities are mostly cities, with Bratislava and Košice being the largest of them. The application of the method to larger cities brings negative values of energy deficit. There are multiple reasons for that: first that the wood combustion is actually very low there compared to gas heating. Second, the municipal gas consumption values which are currently available from the SPP gas vending company include not only households, but also the commercial consumption (small businesses, offices, schools, shops, etc.), which, in case of cities, can be quite numerous, but they are not taken into account when computing the “energy needed” side of the balance equation. Although this fact is taken into account globally by multiplying the gas consumption and connections by an arbitrary factor (0.95), evidently, this is not applicable for large municipalities. The same is true for the number of gas connections used for subtracting the gas used for cooking, leading to the overestimation of this factor. This is a specialty of some cities and towns, actually 40 out of 2922 municipalities, which are affected in such a way. Although they represent a population of almost 1.9 million, they are not supposed to contribute to wood combustion too significantly. In any case, clearly all cities and towns must be treated individually when it comes to a modeling of $PM_{10}$ emissions especially on local scales.

## CONCLUSION AND FUTURE WORK

The paper described a method used for source apportionment of $PM_{10}$ as a response to the Air Quality Directive requirements for the Notification. It is a complicated task when many non-attainment areas are in question and good detailed emission data are not available, while results are laden with huge uncertainties. Therefore, mathematical modeling was only partially used for the emissions which are routinely quantified. The analysis showed that domestic wood combustion is one of the most important seasonal sources hugely contributing to the number of daily limit value exceedances, and the need for quantification of these emissions was identified. Method used for the emission quantification was designed. The first results are promising, but there still issues to be solved, namely:

- Introduce regionally and population-varying building thermal loss factor,
- Research the physical basis of the technical formulas used in civil engineering with the aim of determining their potential bias in a scale of situations,

- Investigate the most appropriate value for the $PM_{10}$ emission factor for wood combustion (the one used in Slovakia is relatively high compared to factors used in different countries,
- Obtain newer housing and household-associated data (new population census in 2011) (number and age of apartments in family houses vs. apartment blocks, household equipment statistics),
- Obtain more disaggregated data on the gas consumption and the number of gas connections (household vs. commercial), especially in cities,
- Search the options for better disaggregation of the use of electricity and fossil fuels for heating.

After refinement, the spatial emission data on wood combustion will be ready as input into mathematical models on local to regional scale and will shed more light into the complicated task of $PM_{10}$ source apportionment in Slovakia.

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**Acknowledgment**

*The authors gratefully acknowledge the EMEP/MSC-W for the provision of the Unified EMEP model and the Norwegian Meteorological Institute, Oslo for provision of meteorological input data for model.*
*The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and/or READY website (http://www.arl.noaa.gov/ready.php) used in this publication.*