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# AEROSOL TRANSPORT MODELLING OVER DEBRECEN, HUNGARY

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**Abstract**: Backward trajectory modelling (NOAA HYSPLIT) together with statistical techniques like Positive Matrix Factorization (EPA PMF) can be used to determine the relative contributions of local and distant sources of aerosol pollution. These methods were completed with the observational footprints computed with System for Integrated modeLling of Atmospheric coMposition (SILAM). In addition, the variations of emission from different areas in Europe were followed with seasonal- and yearly- averaged footprints, whereas the distribution of aerosol pollution were determined with daily average footprints.

***Key words:*** *source identification, long range transport*

## INTRODUCTION

Aerosol pollution is a major environmental problem in urban areas due to negative impact of PM on health and on the built environment. In cities, aerosol particles can originate from local (e.g. biomass burning for residential heating, local industrial emissions or traffic) or from distant sources (e.g. sea salt, desert dust or regional industrial emissions). Sometimes local emissions alone cannot explain high air pollution episodes at a receptor site, i.e. long-range transport processes of natural and anthropogenic aerosols can have a significant influence on the local PM concentration levels. In order to get a picture about the geographical distribution of main pollution sources affecting a specific place, receptor modelling combined with backward trajectory calculation is applied (Borbély-Kiss et al., 1999). However, backward trajectory usually indicates only the main directions of the transport and the subjective analysis of the paths is not always conclusive. A more comprehensive methodology for quantitative analysis of the observed footprints is adjoint dispersion modelling.

In the present work, distant source areas have been determined for a Hungarian city, Debrecen. At an urban background station, concentration and elemental composition of $PM_{10}$ and $PM_{2.5}$ have been monitored regularly twice a week during 24 hours for more than 20 years (Borbély-Kiss et al., 1996). NOAA-HYSPLIT backward trajectory model was used to define the movement of air masses, which reached Debrecen. Correlation analysis (Spearman's correlation) and a receptor model (PMF) were applyied to identify elemental fingerprints. This research was completed with source identification via footprints using the SILAM dispersion model, which provided integrated map of distant sources affecting the observation site in each particular day.

## MATERIALS AND METHODS

### Sampling and analysing

The samples were collected in the garden of Institute for Nuclear Research regulary in two size fractions: PM10-$PM_{2.5}$ (particles with aerodynamic diameter between 2.5 and 10 μm) and $PM_{2.5}$ (particles with aerodynamic diameter < 2.5μm) using a Gent type stacked filter unit since 1993. The aerosol concentration is measured by gravimetry and the elemental composition (for Z >13) is determined by

Proton Induced X-ray Emission (PIXE) method at the macro-PIXE chamber in the IBA Laboratory of Atomki (Borbély-Kiss et al., 1985).

**Modelling tools**

Positive Matrix Factorization (EPA PMF)

Source apportionment was carried out with the positive matrix factorisation receptor model developed for aerosol source characterization, provided by US EPA (Paatero and Tappert, 1994). Mass of species apportioned to factor, percentage of species apportioned to factors and contributions associated with factors were determined for the coarse and fine fractions separately. Variability of source contributions by year, season, working day/weekend and hours of the day were also studied as well as the dependence on meteorological parameters, such as wind directions. The source types are identified by comparing them to measured profiles. Source contributions are used to determine how much each source contributed to a sample.

Backward trajectory model (NOAA HYSPLIT)

HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) model developed by NOAA's Air Resources Laboratory (ARL) (Draxler et al., 2012) was was used to determine the movement of air masses which reached Debrecen. The trajectories were calculated for 72 h periods at three different heights (200, 500 and 1000 m a.g.l.). The meteorological data were obtained from NCEP/NCAR Reanalysis project (Kalnay et al., 1996). The global data are on a latitude-longitude grid (2.5 degree) from 1948 through the end of the previous year. Model vertical velocity was used for simulations, which use the vertical velocity field from the meteorological data.

Dispersion model (SILAM)

In this study SILAM (System for Integrated modeLling of Atmospheric coMposition) dispersion modelling by the Finnish Meteorological Institute was used with Eulerian transport algorithm (Sofiev et al., 2015). The meteorological data were obtained from ECMWF (European Centre for Medium-Range Weather Forecast) with 3-h time interval. The model included 10 vertical layers up to height of 10.225 Km, the near-surface layer being 25 m thick. The investigated period ranged from 1993 to 2013 with 5 elements chosen for the analysis: S, Si, Pb, Zn and Cr. These species were considered to be fingerprints of four different source types. A model time step was 15 min, horizontal resolution was 0.15 x 0.15 degree and output averaged to 1 h.

**RESULTS**

The investigated period (1993-2013) contains more than 1400 sampling days and ~ 65.000 concentration data. Four elements were chosen for the footprint analysis: Si – tracer for soil/dust, Pb and Zn – may originate from incineration and industry, Cr – comes from traffic, S - tracer for industry and present in the aerosols in the form of sulphate. As described Viana et al. (Viana, 2008), sulphate possess a high complicity, and although it was grouped in a single category but separated as two individual sources. The first combination: $SO_4$, V and Ni and the second: $SO_4$, $NO_3$ and $NH_4$. The V/Ni/$SO_4$ source was occasionally found in combination with trace elements such as Pb or Cu (interpreted as regional-scale pollution), OC and K, or Zn and Pb (long-range transport or anthropogenic pollution). On the other hand, the second combination was interpreted as secondary aerosols, regional background or long-range transport. Debrecen has no heavy industry and the residence time of $SO_4$ in the atmosphere is a few days, therefore it is associated with long range transport.

Receptor model (PMF) were applyied to identify the PM sources in Debrecen. This way we could separate local (e.g. road dust, biomass burning, soil, heavy oil combustion) and remote sources (e.g. Saharan dust, metallurgy from Southern Europe). PMF is resulted some factors with S and other elements: S/V/Ni, S/Cu and S/Cr.

For the present study two years were chosen in order to compared the sources of sulphate: 2000 was a "dirty" year while 2011 was "clear". The $PM_{2.5}$ concnetrations can be seen on Figure1 for this two years. The average $PM_{2.5}$ concentration in 2000 was 19.2 $\mu g/m^3$ and in 2011 was 15.3 $\mu g/m^3$.

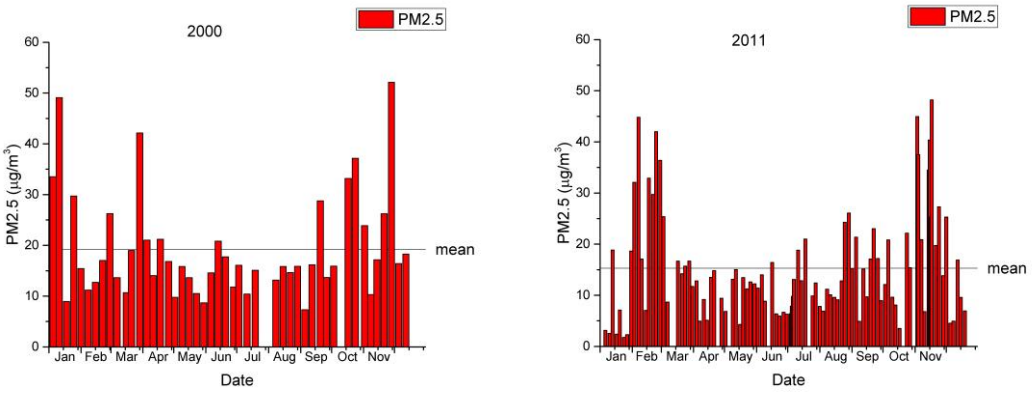


**Figure 1**. The $PM_{2.5}$ concnetrations of 2000 and 2011. 2011 was more clear in spring and autumn.

The average S concentration was higher in 2011 (1288±998 ng/m$^3$) namely caracteristically in winter (november and december: 2137±1347 ng/m$^3$). In 2000 the average concentration was 1103±618 ng/m$^3$ and it topped from january to march (1235±942 ng/m$^3$). The results of SILAM (Fig.2) present the potential sources of sulphate for these periods: in 2000 it can be originated from regional-scale pollution, while in 2011 from long-range transport pollution. The main emission source was Romania.

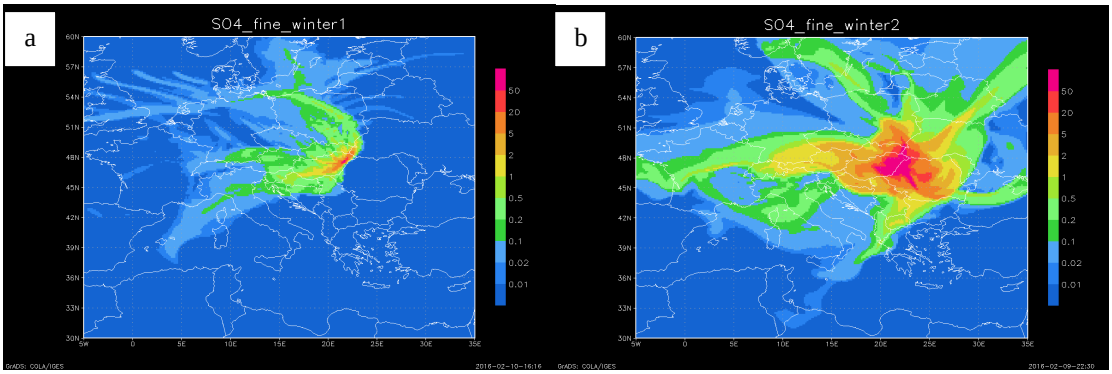


**Figure 2.** Seasonal source apportionment of sulphate using SILAM model. In 2000 (a) sulphate had regional and in 2011(b) had long-range transport source.

In the investigated period there were some days when the $PM_{2.5}$ concentration was more than 50 µg/m$^3$ and the elemental concentration of S was higher than 4000 ng/m$^3$; like in 01.11.1993 when air masses arrived from East and Northeast Europe. The backward trajectories can be seen on Figure 3.

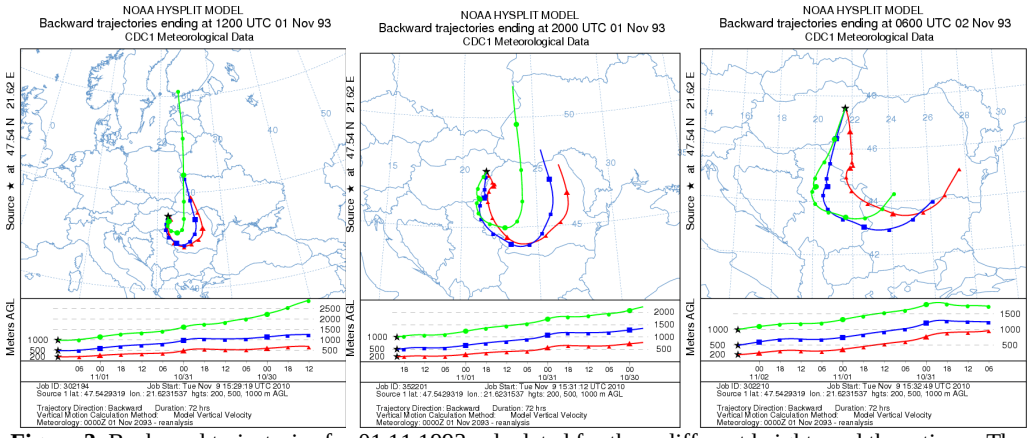


**Figure 3.** Backward trajectories for 01.11.1993 calculated for three different heights and three times. The source areas were East and Northeast Europe.

The SILAM model validated the source apportionment of this pollution episode. On Figure 4 the powerful contribution of Romania can be clearly seen. The computed periods were 5 days for $PM_{2.5}$ (a) and 3 days for $SO_4$ (b).

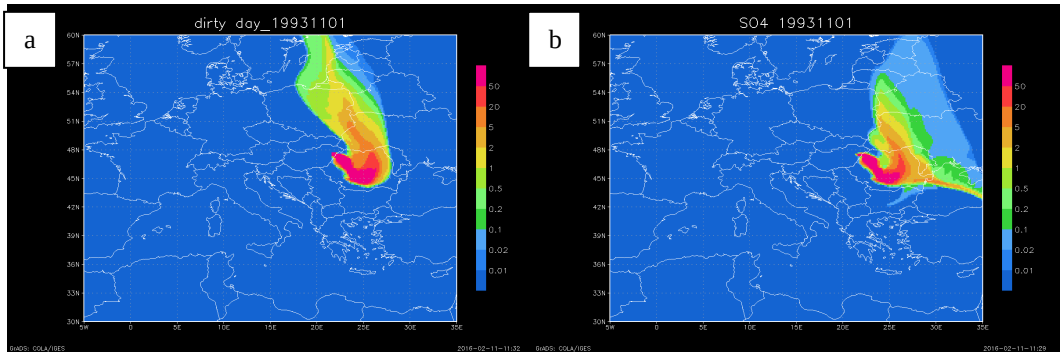


**Figure 4.** Results of SILAM model. The footprints of $PM_{2.5}$ (a) and $SO_4$ (b) calculated 5 and 3 days back.

## CONCLUSIONS

The present study demonstrated source apportionment using two models: HYSPLIT in combination with PMF and SILAM. Our huge database can be helpful to identify regional and long range transport aerosols. The applied statistical techniques (Spearman correlation and positive matrix factorization) can determine the type of sources (soil, traffic, industry, etc.) and then the applied dispersion models can define the origin of air pollution over the observation site.

Over Debrecen the aerosol pollution arrives mainly from Eastern and Southern Europe. The heavy industry in the surrounding countries emitted S, Ni, V, Cu, Zn and Pb which could be measured sometimes in high concentrations at our receptor site.

## ACKNOWLEDGEMENT

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## REFERENCES

Borbély-Kiss, I., Koltay, E., László, S., Szabó, Gy., Zolnai, L., 1985: Experimental and theoretical calibration of a PIXE setup for K and L X-rays, *Nucl. Instr. Meth*. B, **12**, 496-504

Borbély-Kiss, I., Koltay, E., Szabó, Gy.,1996: Elemental composition of urban aerosol collected in Debrecen, Hungary, *Nucl. Instr. Meth*. B, **109/110**, 445-449

Borbély-Kiss, I., Koltay, E., Szabó, Gy., Bozó, L., Tar, K., 1999: Composition and sources of urban and rural atmospheric aerosol in Eastern Hungary. *Journal of Aerosol Science*, **30**, 369-391.

Draxler, R. R., Stunder, B., Rolph, G., Stein, A., Taylor, A., 2012: HYSPLIT_4. User's Guide, via NOAA ARL. http://www.arl.noaa.gov/documents/reports/HYSPLIT_user_guide.pdf. NOAA Air Resources Laboratory, Silver Spring, MD, Dec., 1997 revised March, 2012.

Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha, S., White, G., Woollen, J., Zhu, Y., Chelliah, M., Ebisuzaki, W., Higgins, W., Janowiak, J., Mo, K. C., Ropelewski, C., Wang, J., Leetmaa, A., Reynolds, R., Jenne, R., Joseph, D., 1996: The NCEP/NCAR 40-Year Reanalysis Project., *Bull. Amer. Meeort. Soc*. **77**, 437–471.

Paatero, P., Tappert, U., 1994: Positive matrix factorization: a non-negative factor model with optimal utilization of error stimates od data values, *Environmetrics*, **5**, 111–126.

Sofiev, M., Vira, J., Kouznetsov, R., Prank, M., Soares, J., Genikhovich, E., 2015: Construction of the SILAM Eulerian atmospheric dispersion model based on the advection algorithm of Michael Galperin, *Geosci. Model Dev*., **8**, 3497-3522.

Viana, M., Kuhlbusch, T.A.J., Querol, X., Alastuey, A., Harrison, R.M., Hopke, P.K., Winiwarter, W., Vallius, M., Szidat, S., Prévôt, A.S.H., Hueglin, C., Bloemen, H., Wåhlin. P., Vecchi, R., Miranda, A.I., Kasper-Giebl, A., Maenhaut, W., Hitzenberger, R., 2008: Source apportionment of particulate matter in Europe: A review of methods and results, *Aerosol Science*, **39**, 827-849.