Project closing report for postdoctoral project

PD 125086 - Investigation of sources of atmospheric aerosol at urban environment

01.12. 2017- 30.11. 2020

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1. Scope of project

Atmospheric particle matter (APM) is one of the most hazardous components of the air pollution in densely populated urban environments. Anthropogenic perturbations are concentrated in these areas, thus the negative impact on human health is mainly identifiable in these regions. In order to understand the effect of APM, it is essential to identify aerosol sources, their diurnal contribution and their spatial origin which is a relevant task not only for researchers but for governmental agencies as well. In addition, temporal and spatial variation of particle number concentration is also essential in studies of human exposure of pollutants.

Analysing atmospheric aerosol, the bulk elemental analytical techniques, like macro-PIXE, are very extensively used. However, more detailed source identification and information about human exposure would be realized if particle number concentration and BC could be analysed too.

Source apportionment (SA) studies applied to quantify the effect of individual sources of air pollutant on the ambient concentration through receptor modelling. The classical approach in these studies is based upon the collections and chemical characterization of 24h samples. Even though this methodology has proven to be very effective, it cannot provide information on the diurnal patterns of source contributions due to its low time resolution. Nevertheless, the database on city source apportionment is not well documented in numerous Eastern European cities.

In the frame of the OTKA-PD17 (125086) project the main goal was to learn about source apportionment and properties of atmospheric particle matter sources in an urban background location of Debrecen. My aim was to create a large data set with high time resolution including major, minor and trace components of the PM_{fine} and PM_{coarse} fraction, containing black carbon (BC) in fine fraction. Simultaneously, several thousands of particle size distribution data were examined in 2020 to determine those sources being the most harmful for human health. This complex database is an important milestone in accessing knowledge about PM pollution in Eastern Europe.

2. Experimental methods and data analysis

2.1 Sampling

In the first year of the project, 7-10 days sampling campaigns were carried out under my coordination during different meteorological conditions and in different seasons (26 January - 1 February, 16 - 23 April, 28 August - 4 September, 15-24 October, 2018) by a sequential PIXE International streaker at an urban background station. Nano-MOUDI impactor was also used during the following campaigns: 30 August - 1 September, 17-20 October, 2018.

In the second year of the project sampling campaigns were continued applying streaker and nanoMOUDI impactor (21 February – 1 March, 24 April – 2 May, 5–13 September, 28 October- 7 November, 2019). At the end of the year there was an opportunity to carry out a more complex analysis in the frame of the GINOP-2.3.2-15-2016-00009 (IKER) project by procuring and installing a state-of-the art aerosol monitoring device, an optical particle sizer (OPS).

In the third year of the project we extended our infrastructure by an aethalometer. The sampling campaigns were carried out with streaker and nanoMOUDI (7-17 February 1-6, April, 12-18 August, 6-13 November, 2020). Furthermore, from April an OPS and an aethalometer were also applied.

2.2 Analysis

Proton induced X-ray Emission (PIXE) analysis was used to determine the elemental composition ($Z \geq 11$) on the collected aerosol samples. The ion beam measurements were carried out at PIXE chamber of the VdG accelerator of Atomki up to the autumn of 2019. Due to the COVID-19 pandemic the measurement at the accelerator was cancelled in the first semester of 2020. Since November we have measured at the new external beam setup of the Tandetron accelerator (http://iba.atomki.hu/index_en.html). Besides PIXE analysis the concentration of elemental carbon was measured with a smoke strain reflectometer on the same samples until the end of 2019.

2.3 Receptor model calculations

The EPA PMF v. 5.0 receptor model was applied (Paatero et. al. 1994, Norris et al. 2014) for the source apportionment studies. To relate the source contribution to sources and source areas, two additional methods were used: conditional probability function (CPF) and possible source contribution function (PSCF). CPF was used to examine the relationship of local source impacts with wind direction. Furthermore, PSCF could be interpreted as a conditional probability describing the spatial distribution of probable geographical source locations inferred by using trajectories arriving at the sampling site. Several backward trajectories were calculated using a HYSPLIT model with archived gridded meteorological data.

3. Results

3.1 Source apportionment

3.1.1 Source apportionment study during heating seasons

The main purpose of these campaigns was to characterize and identify the local and regional PM_{fine} sources and to determine their contribution to local ambient pollution level in an urban background station during heating seasons.

In the course of PMF analysis six sources were identified: secondary sulphates originating from combustion and regional background, biomass burning as domestic heating, two soil types: sand and loess, traffic, as well as a source traced by Cl in the fine fraction. The CPF and PSCF analysis (Fig. 1) indicated that all soil factors originated from the northeast and northern directions. The CPF plot of traffic primarily pointed from the south towards the city centre. The highest contribution from the secondary sulphate factor came from the northwest where an industrial park and a suburban area are located. Meanwhile the regional contribution was also relevant. The highest probability of the biomass burning location was the nearest to the sampling site, suggesting that this was the most probable local source while the source traced by Cl had the highest long-range transport potential. It could be identified as sea salt.

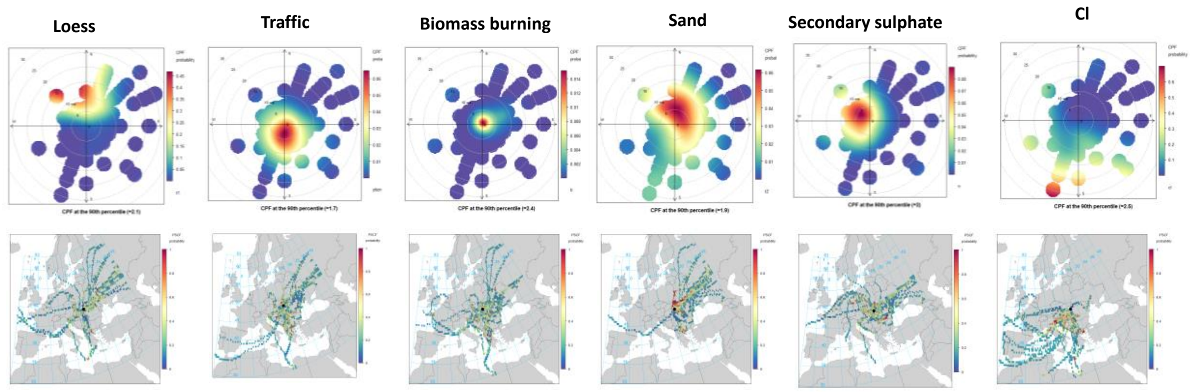

Fig. 1 CPF and PSCF results for the PMF-modelled source contributions.

The results have been presented at the European Aerosol Conference (poster session) in Göteborg, Sweden, in August 2019 and at the Hungarian Aerosol Conference (oral presentation) in Visegrád, Hungary in October 2019.

3.1.2 Source apportionment study during smog episodes

In this study, two successive long-drawn smog episodes caused by an anticyclone were chosen to characterize the APM pollution and provide a unique snapshot of the contributing sources for such periods. The dominant sources in the $PM_{2.5}$ fraction during these smog events in Debrecen, Hungary, were biomass burning and traffic, with a combined contribution of 70%. In the coarse fraction, combustion sources were also present, although the dominant factors were soil (32%) and tramline construction (30%) (Fig. 2). Nevertheless, PBL calculations were conducted by WRF to investigate the effects of a high-pressure blocking condition on the city. The highest accumulated pollution was observed under the lowest PBL height on 17 November. Waste incineration and biomass burning contributed the most to fine aerosol mass while traffic, soil, coal combustion and Cl to coarse aerosol mass. Thus, the feedback effect between the development of PBL and the increased air pollution concentrations was apparent. Nevertheless, the evolution of the sources strongly related to the time course of the routine activities in the city.

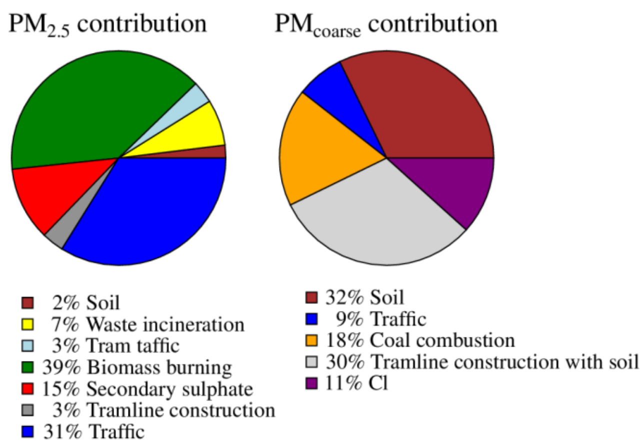

Fig. 2 The relative source contributions to $PM_{2.5}$ and PM_{coarse} by source category.

These results are submitted for publication in Air Quality, Atmosphere and Health.

3.1.3 Source apportionment study in the frame of intercomparison organized by FAIRMODE WG3 (forum for air quality modelling in Europe, Working Group 3).

The dataset of measurements used for the receptor model (RM) intercomparison was produced in the framework of the CARA project directed by the French reference laboratory for air quality monitoring (LCSQA). This dataset contained 116 PM10 daily concentrations collected every third day between March 2011 and March 2012. The concentration of 98 chemical species and their uncertainties were provided for every sample including: ions, organic and elemental carbon (OC/EC), trace elements, polycyclic aromatic hydrocarbons (PAHs), anhydrosugars, hopanes, n- alkanes, primary organic aerosol (POA) markers, and the total PM10 gravimetric mass concentrations. The main goal was to evaluate the receptor models (RMs) and the chemical transport models (CTMs) for PM10 source apportionment. In this project, I was one of the two participants from Hungary. My task was to carry out the source apportionment on the obtained data and to interpret my result. RM results were obtained using the US-EPA version 5 of the positive matrix factorisation tool. I also contributed as co-author and report writer on this project. In this study, a high number of RM results (38) were evaluated which gave a unique opportunity to analyse the conditions that influenced the performance of the results. To that aim, their RMSEu and z-scores were compared with the self-declared experience of the RM practitioners and the number of candidates in the reported results. At the Lens site, the performance of RMs as well as those of CTMs compared with RMs was also reported. Thus the intercomparison offered the possibility for their cross-validation. The results have been published in Atmospheric Environment.

In the frame of the FAIRMODE project I was the only contributor from Hungary to the JRC Technical Report which was entitled: Source apportionment to support air quality management practices: A fitness-for-purposes guide.

3.1.4 Related activity for source apportionment

A special field trip for a group of secondary school students was held under my instruction in November 2018 to introduce the importance of source apportionment studies.

Participation in the Regional Training Course on Recent Advances in Receptor Models for Source Identification and Apportionment of Air Particulate Matter, Ljubjana, Slovenia in May, 2019.

3.2 Indoor aerosol sources

3.2.1 Source identification in indoor environments (passive houses)

Although not mentioned in my research plan, this topic was relevant for my ongoing postdoctoral project. Thus, to estimate personal PM exposures in a building of interest and in trams, it is important to know what types of connection exist between the indoor and outdoor environment as well as how many outdoor sources can get through to the buildings and trams.

In the case of passive houses, my task was to collect and analyse the aerosol samples using the PIXE technique as well as to evaluate the obtained data. The elemental composition was determined at each sampling site. Furthermore, the mass size distribution of some elements indoors and outdoors were examined. As a result, the sources of the fine fraction aerosol were identified as the outdoor air. Supporting this claim, the indoor/outdoor elemental ratios and the mass size distribution data (Fig. 3) indicated that the PM_{coarse} was sufficiently filtrated in the passive houses while the PM_{fine} fraction could get through the filters without hindrance. Furthermore, the PM_{fine} levels were independent of the ventilation modes.

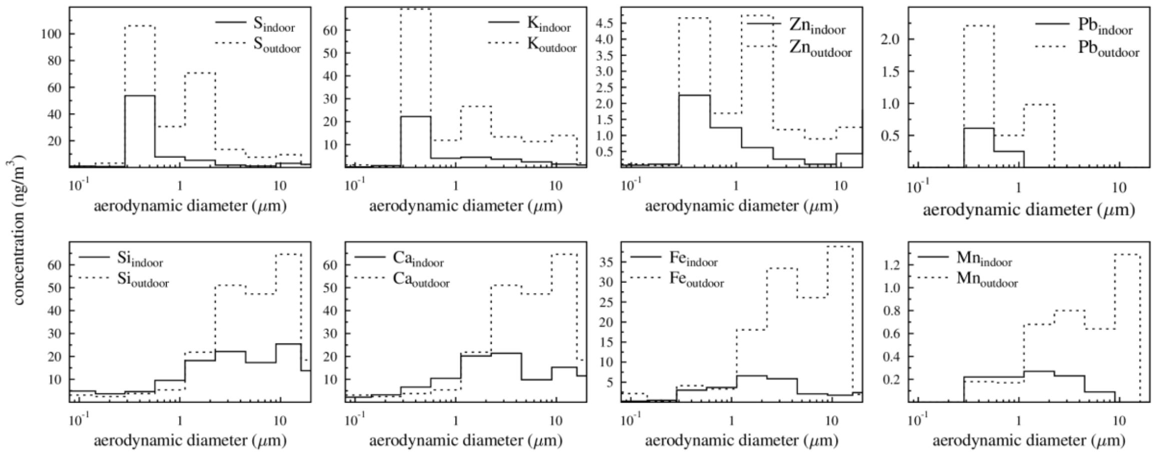

Fig. 3 Mass size distribution of elemental concentration indoors and outdoors.

The results have been published in AAQR.

3.2.2 Source identification in indoor environments (trams)

In the frame of the tram's project, fine (PMfine) and coarse (PMcoarse) particulate matter concentrations and elemental compositions were investigated inside trams during travelling in heating and non-heating seasons. New CAF and older KCSV tramcars were involved in the study. I have participated in the analysing process of the composition using the PIXE method as well as in the identifying of the sources. The origin of the elevated pollution was the re-suspension of the mineral and road dust. A high amount of iron was found inside the trams which could be originated from the rails. The abrasion of the wires could be the reason of the elevated Cu, Zn concentrations. The abrasion of the furniture and the vehicle also can cause elevated pollution levels and contribute to indoor sources. The exposure of the passengers and drivers could be further reduced by using modern air technology and sufficient maintenance. The results have been published in NIMB.

4. Conclusions

The main objective of the project was to characterize and identify the local and regional sources and to determine their contribution to the local ambient pollution level in an urban background station, in Debrecen, Hungary. My goal was to create a large dataset based on the simultaneous use of different aerosol monitoring devices. With the help of the extended infrastructure it can be possible to identify and characterize the variation of APM sources, including the harmful ones, both on a short and a long time scale.

I created a large data set including major, minor and trace components of the PM_{fine} and PM_{coarse} fraction, containing black carbon (BC) in fine fraction in Debrecen. I succeeded to identify the regional and local sources in the urban background stations of Debrecen, Hungary and in Lens, France. During the smog periods in Debrecen I observed the feedback effect between the development of planetary boundary layer (PBL) and of increased air pollution concentrations. Nevertheless, the evolution of the sources strongly related to the time course of the routine activities in the city. In the case of 2020 campaigns, changes in particle size distribution, particle number concentration and BC concentration was observed simultaneously. The meteorological data (HR, T, wind) was compared with a group of wide range results (aerosol size distribution and aerosol number concentration) which together helped to study aerosol transport, transformation and mixing processes. This complex database can serve as a basis for source assessment studies. Due to the COVID 19 situation, as it has been mentioned earlier in the "*Analysis"* section the outcome of the 2020 campaigns will be published next year.

In addition, with my participation in various indoor measurement projects in passive houses and trams, the relationship between the indoor and outdoor sources has been estimated, as well as it was studied how different ventilation rates and modes affected the indoor particulate matter (PM) contamination.

Thanks to the improvements carried out in the frame of the the GINOP-2.3.2-15-2016-00009 (IKER) project I was enabled to execute a more complex research using state-of-the art mobile aerosol monitoring devices.

Three papers were published in scientific journals and conference proceedings, and one more is submitted for publication in AQAH. The results were presented in international (EAC2019) and national (Hungarian Aerosol Conference, 2019) conferences.

Publication in the frame of the PD125086

Scientific papers

*K. Szirtesi, **A. Angyal**, Z. Szoboszlai, E. Furu, Z. Török, T. Igaz, Z. Kertész Airborne particulate matter: An investigation of buildings with passive house technology in Hungary AEROSOL AND AIR QUALITY RESEARCH 18 : 5 pp. 1282-1293. , 12 p. (2018)*

*E. Papp, D. Nagy, Z. Szoboszlai, **A. Angyal**, Zs. Török, Á. Csepregi, E. Furu, Zs. Kertész, Investigation of aerosol pollution inside trams in Debrecen, Hungary NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH SECTION B-BEAM INTERACTIONS WITH MATERIALS AND ATOMS 477 pp. 138-143. , 6 p. (2020)*

C.A. Belis, D. Pernigotti, G. Pirovano, O. Favez, J.L. Jaffrezo, J. Kuenen, H. Denier van Der Gon, M. Reizer, V. Riffault, L.Y. Alleman, et al. Evaluation of receptor and chemical transport models for PM10 source apportionment ATMOSPHERIC ENVIRONMENT: X 5 Paper: 100053 (2020)

Report (contributor)

*P. Thunis, A. Clappier, G. Pirovano, E. Pisoni, C. Guerreiro, A. Monteiro, H. Dupont, V. Riffaut, E. Waersted, S. Hellebust, J. Stocker, S. Gilardoni, A. Eriksson, **A. Angyal**, G. Bonafe G, F. Montanari, J. Matejovica, J. Bartzis Source apportionment to support air quality management practices Luxembourg, Luxemburg : European Commission, Office for Official Publications of the European Union (2020) ISBN: 9789276197430*

Conference abstracts, talks, posters

***A. Angyal**, E. Furu, E. Papp, Zs. Török, Z. Szikszai, Zs. Kertész Identification of Urban Aerosol Sources During Heating Seasons in Debrecen, Hungary. In: European Aerosol Conference (2019) Paper: P1-066 (poster presentation and abstract)*

***A. Angyal**, E. Furu, E. Papp, Zs. Török, Z. Szikszai, Zs. Kertész Identification of atmospheric aerosol derived from local sources, from long- and short-range transport (in Hungarian). (2019) XIV. HUNGARIAN AEROSOL CONFERENCE in Visegrád, Hungary in October 2019. (oral presentation and abstract)*

*Zs. Török, **A. Angyal**, E. Furu, E. Papp, I. Borbélyné-Kiss, Zs. Kertész Sources of sulphate aerosol in Debrecen (in Hungarian). (2019) XIV. HUNGARIAN AEROSOL CONFERENCE in Visegrád, Hungary in October 2019. (oral presentation and abstract)*

*Zs. Török, **A. Angyal**, E. Furu, E. Papp, K. Rostislav, S. Mikhail, Zs. Kertész Long-range transport of aerosol particles (PM2.5 and sulphur) in Debrecen, Hungary In: European Aerosol Conference (2019) Paper: P1-069 , 1 p. (poster presentation and abstract)*

*Zs. Török, **A. Angyal**, E. Furu, E. Papp, K. Rostislav, S. Mikhail, Zs. Kertész Long-range Transport of Fine Aerosol Particles in Debrecen, Hungary (2019) CAMS 4thGeneral Assembly and User Day Budapest, 2019. szeptember 16-20. (oral presentation)*

E. Papp, D. Nagy, Z. Szoboszlai, **A. Angyal***, Zs. Török, Á. Csepregi, E. Furu, Zs. Kertész Investigation of aerosol pollution in trams in Debrecen (in Hungarian). (2019) XIV. Magyar Aeroszol Konferencia, Visegrád, 2019. október 2-4. (poster presentation)*

Zs. Kertész, Z. Szoboszlai, I. Major, T. Varga, **A. Angyal***, Zs. Török, E. Papp, M. Molnár Investigation of aerosol pollution during smog episode by radiocarbon and PIXE analysis (in Hungarian). (2019) XIV. Magyar Aeroszol Konferencia, Visegrád, 2019. október 2-4. (oral presentation and abstract)*

W*orks under publication:*

A. Angyal*, Z. Ferenczi, M. Manousakas, E. Furu, Z. Szoboszlai, Zs. Török, E. Papp, Z. Szikszai, Zs. Kertész Source identification of fine and coarse aerosol during smog episodes in Debrecen, Hungary*

Submitted for publication in Air Quality, Atmosphere and Health.