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# Source apportionment of episodic $PM_{10}$ air pollution in Polish urban areas

Magdalena Reizer*, Katarzyna Juda-Rezler, Katarzyna Maciejewska

*Warsaw University of Technology, Faculty of Environmental Engineering, Warsaw, 00653, Poland*

**Abstract**

A combination of five source apportionment methods was applied to distinguish long-range transport (LRT) and regional transport (RT) from local pollution (LP) sources of a severe wintertime $PM_{10}$ episode. The air quality in several urban areas in Poland was investigated using air quality monitoring and meteorological data, air mass back trajectories, correlation analysis and Principal Component Analysis with Multivariate Linear Regression Analysis (PCA-MLRA). Daily patterns of $PM_{10}$ levels show that severe episode, with daily $PM_{10}$ levels up to 480 $\mu g/m^3$ occurred throughout the whole Poland from 22nd to 28th January. The occurrence of high $PM_{10}$ concentrations was associated with extremely unfavourable meteorological conditions caused by the influence of a Siberian High rigde. Backward trajectories indicate that during episode air quality was under the influence of air masses from Eastern Europe (LRT) and from national RT. Moreover, regional industrial sources had the biggest (up to 73%) share in $PM_{10}$ levels, followed by LRT and LP sources. PCA analysis shows that ion-sums of secondary inorganic aerosols account for LRT pollution, As, Cd, Pb and Zn for industrial RT sources, while Cr and Cu represent markers of LP sources of $PM_{10}$. Analysis of correlation between PM and gaseous pollutants allows to distinguish 2 main types of LP sources responsible for high $PM_{10}$ concentrations during episode: road transport and residential sector.

*Keywords: PM episode; $PM_{10}$; Poland; coal combustion; source apportionment; PCA-MLRA; backward trajectories.*

## 1. Introduction

Poor urban air quality due to high concentrations of particulate matter (PM) remains a major public health problem worldwide. Although in Europe during the past decade emissions of primary PM and precursors of secondary PM decreased (EEA, 2013), severe

*Corresponding Author: magdalena.reizer@is.pw.edu.pl

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PM episodes are occurring continuously in many European cities, e.g., in: Cracow and Warsaw (January 2006), Athens (December 2012), London (February 2014) and Paris (March 2014). According to the latest report of the European Environmental Agency (EEA, 2013), up to a third of the European Union's (EU) urban population is exposed to levels of hazardous $PM_{10}$ (particles with an aerodynamic diameter <10 μm) exceeding daily air quality limit value (50 $\mu g/m^3$), while the exposure to $PM_{10}$ levels that do not meet the annual air quality guideline (20 $\mu g/m^3$) set by the World Health Organization (WHO) is significantly higher, comprising over 85% of the EU urban population. Furthermore, most recently the specialized cancer agency of the WHO, the International Agency for Research on Cancer (IARC), has classified outdoor air pollution as carcinogenic to humans. PM as a major component of this pollution was evaluated separately and was also classified as carcinogenic to humans (WHO, 2013).

Therefore, it is crucial to gain knowledge about the types of emission sources responsible for severe PM events. To address this issue different source apportionment (SA) methods are widely used in worldwide studies (e.g., Johnson et al., 2011; Belis et al. 2013). Two main types of approaches are used: (1) bottom-up methods based on numerical dispersion modelling and (2) top-down methods applying receptor modelling (RM) techniques, based on multidimensional statistical analysis of ambient PM concentrations and its chemical composition.

In Poland, hard coal and lignite are still the main energy carriers, amounting to approximately 50% in the structure of primary energy consumption and about 80% in the structure of electricity consumption. Such high coal exploitation is a predominant cause of high PM concentrations, especially during winter.

The main goal of this paper was to distinguish remote emission sources (LRT – Long-Range Transport), regional industrial sources (RT – Regional Transport) and local pollution (LP) as well as to discern different types of coal combustion sources: industrial and domestic, that determine high $PM_{10}$ concentrations recorded in Polish urban areas. For this purpose, the case study of severe wintertime PM episodes that occurred in 2010 has been selected for investigation. The original method of PM source identification (Juda-Rezler et al., 2011), which is based only on data from routine monitoring network and combines five SA techniques, was applied and further developed.

## 2. Materials and methods

In the present work, the air quality was investigated during a severe wintertime $PM_{10}$ episode which occurred in January 2010 in five Polish cities, situated in Northern (Szczecin), Central (Warsaw) and Southern (Cracow, Zabrze, Jelenia Góra) part of the country (see Fig. 1). The selected cities are characterized by different conditions with respect to climate, topography and emission sources.

The influence of specific anthropogenic sources on $PM_{10}$ levels during the episode was investigated by the combination of different apportionment techniques applied to routinely available air quality and meteorological monitoring data, i.e.: (1) analysis of the $PM_{10}$ patterns at both urban and regional background monitoring sites, following the Lenschow approach (Lenschow et al., 2001); (2) analysis of the synoptic situation and variability of the local meteorological parameters; (3) analysis of the air mass back trajectories; (4) receptor modelling using Principal Component Analysis with Multivariate Linear Regression Analysis (PCA-MLRA) and (5) analysis of correlation between $PM_{10}$ and gaseous pollutants.

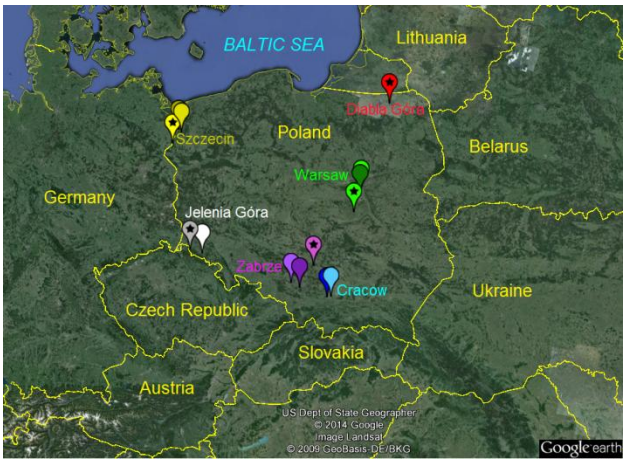


Fig. 1. Location of urban background (light shades), traffic (dark shades) and regional background (marked by stars) air quality monitoring sites.

For each city, air quality monitoring data from urban background (UB), traffic (TRAF) and the nearest regional background (REG) sites were considered, while EMEP (European Monitoring and Evaluation Programme) rural background site in Diabla Góra (PL0005R) was chosen as representative for continental background (Fig. 1). The data of $PM_{10}$ mass concentrations and concentrations of its constituents: trace elements (TEs) and Secondary Inorganic Aerosols (SIA), as well as concentrations of gaseous pollutants from selected sites were extracted from the Voivodeship air pollution networks which belong to the Polish Voivodeship Environment Protection Inspectorates. Data was also extracted from EMEP (http://www.emep.int) and Air-Base, European Air quality dataBase (http://acm.eionet.europa.eu/databases/airbase) databases. The criterion of 75% completeness of the data sets within an analysed period was applied. Depending on the monitoring site, concentrations of $PM_{10}$ were determined by either a reference gravimetric method (Standard EN 12341), Tapered Element Oscillating Microbalance (TEOM®), beta ray attenuation method (MP101M) or Rupprecht & Patashnick Co. Sequential two-channel Dichotomous Partisol® Plus (model 2025) air sampler. None of three latter methods of $PM_{10}$ measurements are considered to be the reference methods regarding the standard EN 12341, hence Poland, as the EU member state is required to demonstrate that their methods yield results equivalent to the reference (EC, 2010). However, until now the procedures for equivalency demonstration have not yet been accustomed in Poland. Therefore, herein we have used the official data as they were reported to EMEP and AirBase by Polish Chief Inspectorate of Environmental Protection. The concentrations of gaseous pollutants, i.e. $SO_2$, CO, and $NO_2$, were determined by UV fluorescence method (Standard EN 14212), non-dispersive infrared spectroscopy method (Standard EN 14626) and chemiluminescence method (Standard EN 14211), respectively. The analysis of TEs content was performed using Inductively Coupled Plasma – Atomic Emission Spectroscopy (ICP–AES), while sulphate and sum of nitric acid and nitrate, as well as sum of ammonia and ammonium were determined by Capillary Electrophoresis (CE) and spectrophotometry, respectively.

Transport of air masses to each city was investigated by the means of back trajectories using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler & Rolph, 2013) from the Air Resources Laboratory (ARL) of the National

Oceanic and Atmospheric Administration (NOAA). The model was forced by the global reanalysis meteorological data produced by the National Weather Service's National Service for Environmental Prediction (NCEP) to compute advection and dispersion of air parcels. 3-day back trajectories for air parcels arriving over selected cities at an altitude of 50, 100, 200, 500 and 700 m were generated. Receptor modelling technique based on PCA-MLRA was applied to both TEs and SIA concentrations measured at Diabla Góra site for source apportionment analyses following the methodology proposed by Thurston and Spengler (1985). Correlations between levels of $PM_{10}$ and gaseous pollutants were calculated using Pearson's coefficient. All statistical analyses were performed with STATISTICA software.

## 3. Results and discussion

### *3.1 Daily patterns of $PM_{10}$ concentrations*

For each selected city, daily patterns of $PM_{10}$ concentrations observed during January 2010 at both UB and REG air quality monitoring sites indicate that the investigated $PM_{10}$ episode of 22-28$^{th}$ January was preceded by 2 episodes with lower PM concentrations: the 2-10$^{th}$ and the 11-20$^{th}$ of January (Fig. 2). During all these periods, $PM_{10}$ concentrations at UB sites exceeded the daily EU air quality limit value (50 µg/m$^3$). The episode of 22-28$^{th}$ January was characterized by the highest $PM_{10}$ levels, up to 480 µg/m$^3$ on 27$^{th}$ January in the southern city of Jelenia Góra. During this time 2 peaks of $PM_{10}$ concentrations were observed at both UB and REG sites as well as at rural background site in Diabla Góra, however the peak concentration at the latter site was recorded 2 days before the maximum $PM_{10}$ levels registered at the rest of sites.

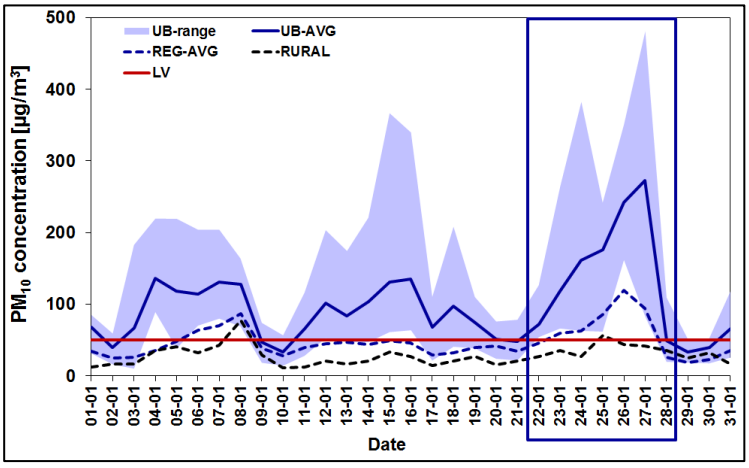


Fig. 2. Daily patterns of $PM_{10}$ levels [µg/m$^3$] during January 2010 averaged for five urban background (UB-AVG, blue solid line) and five regional (REG-AVG, blue dashed line) monitoring sites and at rural background EMEP site in Diabla Góra (RURAL, black dashed line). Blue light background indicates the range of $PM_{10}$ concentrations at UB sites, blue frame – the period of PM episode and red solid line – EU limit value for $PM_{10}$ (50 µg/m$^3$).

The course of daily patterns of $PM_{10}$ concentrations at UB and REG sites clearly shows that during the episode, high pollution was linked to urban anthropogenic sources, however the regional background was also quite high. The ratio of REG/UB levels in the selected

cities increased from southern (more industrialized) to northern part of Poland and ranged from 0.15 (in Jelenia Góra), trough 0.5-0.6 (in Cracow, Zabrze and Warsaw) to 1.1 (in Szczecin).

It should be pointed out that during the episode unfavourable local meteorological conditions, that foster the occurrence of increased $PM_{10}$ pollution events, were observed. The whole country was under the influence of the Siberian High ridge, that brought extremely cold and stable air mass. Low air temperature was observed with daily minimum below -17 °C in southern cities (on 24th January), -19 °C in central city of Warsaw (on 25th January) and below -16 °C in northern city of Szczecin (on 26th January). Low wind speed – not exceeding 5 m/s – and thermal inversion were also recorded. In such meteorological conditions a significant rise in coal and biomass combustion in the residential sector in Poland is regularly noted (Wojdyga, 2008). In all investigated cities, the peak $PM_{10}$ concentrations occurred 1-3 days after the maximum air pressure and minimum air temperature. As an example, Fig. 3 presents daily $PM_{10}$ concentrations during episode in southern city of Jelenia Góra in comparison with daily mean air temperature and wind speed.

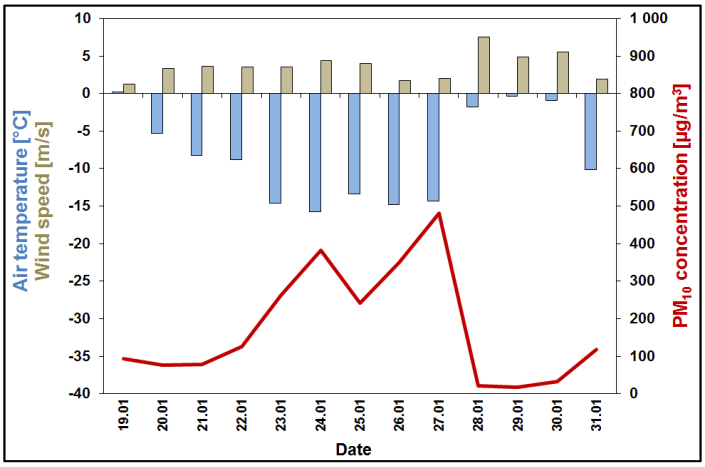


Fig. 3. Daily pattern of $PM_{10}$ concentrations [µg/m$^3$] (red solid line) during the period of 19 – 31th January 2010 at urban background monitoring sites in Jelenia Góra in comparison with daily mean of meteorological parameters: air temperature [°C] (blue bars) and wind speed [m/s] (beige bars).

### *3.2 Air mass back trajectories*

Transport of air masses to pairs of UB and REG sites of each city was examined by means of an atmospheric 3-days back-trajectory. Due to the fact that the air mass trajectories calculated for each day of the episode demonstrate similar pattern and that the trajectories determined at all altitudes were almost identical for each city, only back trajectories calculated at 50 m of altitude are presented (Fig. 4).

Analysis of air mass back trajectories shows that during the episode all pairs of UB and REG sites were under the influence of the same air masses moving along the high-pressure system edges. These air masses, coming mainly from the European part of Russia, were transported through Ukraine, Belarus and further over Poland, thus both remote sources and regional industrial ones impacted the air quality during episode.

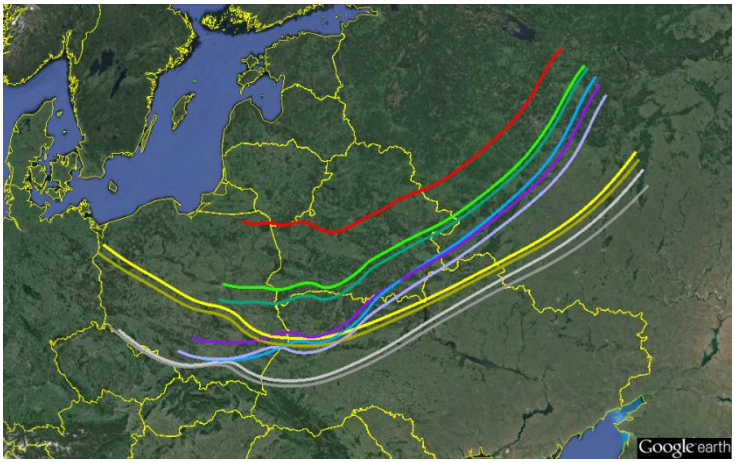


Fig. 4. 3-day air mass back trajectories at the altitude of 50 m a.g.l. at urban (light colours) and regional (shaded colours) monitoring sites of the five Polish cities: Szczecin (yellow); Warsaw (green); Cracow (blue); Zabrze (purple); Jelenia Góra (white), and at rural the background EMEP site in Diabla Góra (red), calculated by HYSPLIT model at 00 UTC on January 25$^{th}$ 2010.

### *3.3 The PCA-MLRA analysis*

PCA with Varimax rotation was applied to trace elements and SIA concentrations, measured at rural background. The interpretation of the principal components (PCs) was based on the variables with factor loadings with absolute values greater than 0.6.

Three PCs were extracted explaining up to 81% of the total variance (Table 1). A major group of TEs that includes As, Cd, Pb and Zn was collocated in the PC1 which explains around 40% of the total variance. The second set (PC2 with 25.5% of explained variance) is characterized by the group of SIA components, however lacking sulphate, while the third one (PC3 with almost 16% of explained variance) is represented mostly by Cr and Cu.

Following Juda-Rezler et al. (2011), As, Cd, Pb and Zn were associated with industrial sources: coal combustion in power generation utilities and metallurgy industry. Moreover, analysis of the physical and morphological properties of TEs and of their enrichment level on the way from the power plants' boiler to the stack, carried out by Juda-Rezler & Kowalczyk (2013), confirmed that the group of As, Cd, Pb and Zn is mostly representative for industrial pollution. In contrary, Cr and Cu were found to indicate coal combustion in small scale installations, i.e. domestic stoves/local boiler houses (Juda-Rezler et al., 2013) and brake abrasion from road transport (e.g., Querol et al., 2007), respectively, and thus both were considered as originating mainly from LP sources. As SIA are commonly related with an 'external' source of pollution (e.g., Aarnio et al., 2008), this group was identified as a marker of the LRT process involved in the overall $PM_{10}$ state of pollution.

Table 1. Factor loadings determined in the PCA analysis with Varimax rotation of trace elements and SIA measured at rural background EMEP site in Diabla Góra during January – March 2010. Factor loadings >0.6 are represented in bold type.

| **$PM_{10}$ component** | **PC1** | **PC2** | **PC3** |
|---|---|---|---|
| As [ng/m$^3$] | **0.82** | 0.37 | 0.05 |
| Cd [ng/m$^3$] | **0.86** | 0.09 | 0.37 |
| Cr [ng/m$^3$] | 0.44 | 0.27 | **0.62** |
| Cu [ng/m$^3$] | 0.30 | 0.30 | **0.75** |
| Ni [ng/m$^3$] | 0.53 | **0.68** | 0.09 |
| Pb [ng/m$^3$] | **0.91** | 0.28 | 0.21 |
| Zn [ng/m$^3$] | **0.91** | 0.20 | 0.27 |
| $SO_4^{2-}$ [µg(S)/m$^3$] | 0.49 | 0.32 | 0.52 |
| $NH_3 + NH_4^+$ [µg(N)/m$^3$] | 0.31 | **0.85** | 0.17 |
| $HNO_3 + NO_3^-$ [µg(N)/m$^3$] | 0.11 | **0.91** | 0.25 |
| % Variance | 39.7 | 25.5 | 15.7 |
| *PM source* | ***RT*** | ***LRT*** | ***LP*** |

The contribution of each source to the PM burden was further quantitatively assessed by means of MLRA (Fig. 5).

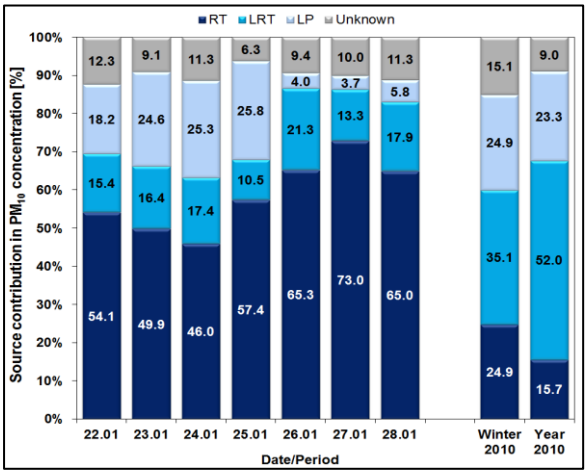


Fig. 5. Contributions of $PM_{10}$ emission sources: Long-Range Transport (LRT, blue), Regional Transport (RT, dark blue), Local Pollution (LP, light blue), unknown source (grey), determined in the PCA-MLRA analysis for each day of PM episode, winter (January – March) and the whole year 2010.

During the episode the regional industrial sources had the biggest (up to 73%) share in $PM_{10}$ levels, being 2-3 times higher over this period in comparison to winter average (25%) and 3-4 times higher than the whole year average (16%). The contribution of the LRT sources ranged during the episode from 10% to over 21%, while of the LP sources – from 4% to 25%. The share of LRT during episode was 2-3 times lower in comparison to both winter average (35%) and the whole year average (52%). The share of the LP sources in

$PM_{10}$ concentrations at the beginning of the episode was at the same level as for both winter and the whole year averages (around 25%), while during the last days of the episode – 5 times lower (5%). The share of sources unidentified by PCA-MLRA was relatively small for all analysed periods, accounting for 10 – 15%.

*3.4 The correlation analysis*

To determine the type of LP sources responsible for high PM concentrations during the episode, the analysis of correlations between $PM_{10}$ concentrations and selected gaseous pollutants ($SO_2$, $NO_2$, CO) was applied. In this analysis $NO_2$, CO and $SO_2$ were considered to be specific markers of traffic pollution and of coal combustion, respectively. Road transport was identified as the main source of LP in Szczecin ($r = 0.90$ for $NO_2$, $p < 0.05$), Warsaw ($r = 0.96$ for $NO_2$, $p < 0.05$; $r = 0.97$ for CO, $p < 0.05$) and Cracow ($r = 0.99$ for $NO_2$, $p < 0.05$; $r = 0.99$ for CO, $p < 0.05$). Residential sector appeared the main LP source in southern cities of Zabrze ($r = 0.81$ for $SO_2$, $p < 0.05$) and Jelenia Góra (r = 0.998 for $SO_2$, $p < 0.05$).

## 4. Summary and conclusions

The combination of five source apportionment techniques was applied for determining sources involved in the build-up of high $PM_{10}$ concentration event of January 2010 in Polish urban areas. The whole procedure demonstrated that however the severe winter episodes of $PM_{10}$ are for a large part caused by a number of anthropogenic emission processes, unfavourable synoptic- and local-scale meteorological conditions are also responsible for the occurrence of air pollution events. One of the main findings concerning meteorological influences on PM levels is that the peak concentrations occur usually 1-3 days after the maximum atmospheric pressure and minimum air temperature observed, which are related to the strongest expression of the high pressure system impact. The application of the methodology allowed to identify and discern the possible sources of $PM_{10}$ episodic pollution: long-range transport (LRT), regional transport (RT) and local sources (LP) as well as their contributions to the bulk PM mass. At the rural background site, representative for continental background pollution, $PM_{10}$ levels were dominated by RT sources, followed by LRT and LP sources. High correlation between $PM_{10}$, $NO_2$ and CO confirmed that traffic emission represented a common and an important LP source of urban pollution in 3 Polish cities (Szczecin, Warsaw, Cracow), while $PM_{10}$ highly correlated with $SO_2$ indicated that coal combustion for domestic heating or industrial activities was an important LP source of $PM_{10}$ in 2 southern cities (Zabrze, Jelenia Góra). Finally, the group of trace elements: As, Cd, Pb and Zn, and Cr with Cu were found as representative markers of industrial RT pollution and domestic LP sources, respectively, while SIA were identified as a marker of the LRT sources. The results show that proposed methodology is efficient for the identification of PM emission sources in urban areas, thus provides the key information to the development and implementation of policies aiming at human health protection.

## 5. Acknowledgements

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