Mass concentration and chemical composition of submicron particulate matter (PM$_1$) in the Polish urban areas

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Abstract. Samples of PM$_1$ were collected at two urban background sites, one in Zabrze and one in Warsaw (Poland), in summer (24 June - 22 August 2014) and winter (8 January - 8 March 2015) in parallel. At each site, in each season, 60 diurnal (24-h) samples were taken. The samples were analyzed for carbon (organic OC, and elemental, EC) and water soluble ions ($\text{Na}^+$, $\text{NH}_4^+$, $\text{Cl}^-$, $\text{SO}_4^{2-}$ and $\text{NO}_3^-$) by using, respectively, a Sunset Laboratory carbon analyzer and a Herisau Metrohm AG ion chromatograph. Each 24-h sample of PM$_1$ was checked for mass closure using the categorization of the chemical components into: primary (POM) and secondary (SOM) organic matter, secondary inorganic matter (SIM), elemental carbon (EC), and sodium and chlorine ($\text{Na}$, $\text{Cl}$). The seasonal effects, typical of urban areas in Poland, are clearly seen in the whole series of the 24-hour PM$_1$ concentrations. Most of the PM$_1$ components and PM$_1$ had their mean concentrations higher in winter than in summer. Secondary aerosol (SOM+SIM) accounts for approx. 55% of the PM$_1$ mass in Warsaw and 40% in Zabrze.

1. Introduction

The health effects of submicron particles (PM$_1$) in humans and the PM$_1$ influence on the environment and climate are evident and are widely discussed in scientific literature [1-4]. However, among all available analyses of the physicochemical properties of ambient particulate matter (PM) in Europe [3-7], one can notice a striking lack of chemical characteristics of the most hazardous PM fractions: the fine ones, including PM$_1$.

The primary factor to shape the properties of PM and its precursors is their sources. In other words, the physical and chemical properties of PM at a given location depend on where the PM comes from [3,6,8]. If local sources (industry, transports etc.) contribute to the structure and concentrations of PM most, as it is normal in the centres of large metropolitan areas, the chemical composition and concentrations of PM will change dynamically with the fluctuations in the intensity of emissions from these sources. If there are no local sources, or they are weak, fluctuations in the chemical composition and concentrations of PM will not be easy to predict and interpret, as they will strongly depend on emissions brought by air masses from distant sources.
There are three main sources of PM and its gaseous precursors in Polish urban areas: so-called low or communal sources (in fact, household stoves and local boiler plants combusting poor quality coal, its derivatives, and (illegally) plastic rubbish, solid and organic wastes), the power and industry sectors (power stations, coking plants, foundries, steel mills, cement plants, etc.), and transportation or road traffic in a broad sense [5,9,10]. Dense road networks, growing traffic intensity, great number of vehicles with faulty exhaust systems (damaged or missing catalytic converters or diesel particulate filters) contribute largely to fine PM concentrations in urban areas in Poland [10-13]. Moreover, in some locations, combustion of natural gas contributes vast amounts of ambient inorganic matter precursors, and combustion of heating oil - particles rich in Ni and V[9].

The goal of the work is to present and analyse the concentrations and chemical composition of PM$_1$ at two urban background sites [14] in Poland.

2. Material and Method

2.1. PM$_1$ sampling

PM$_1$ was sampled at two urban sites, one in Warsaw and one in Zabrze (Poland, Figure 1). Zabrze (area of 80 km$^2$) is one of the most industrialized cities in Upper Silesia (southern Poland, area of 1200 km$^2$). There are a big power station, coking plant, and some number of iron, glass, and chemical works in Zabrze. The air quality at the sampling site, which was located in the Zabrze center, represents the air pollution state in the central part of Upper Silesia. The industry, road traffic, and municipal emissions (fossil fuel combustion) maintain ambient PM concentrations at high levels in this region all year round [10].

Warsaw (area of 517 km$^2$) is the capital and biggest city in Poland; it is the ninth-biggest city in Europe. The PM$_1$ sampling site was located in the southern part of Warsaw, in its district Ursynów. The topography of the place, the meteorological conditions, and the structure and amounts of PM emissions and of PM gaseous precursors (coming mainly from road traffic and coal combustion in the power station) at the sampling site are typical of the whole Warsaw Agglomeration (area of 2932 km$^2$) [9].

The heads of the samplers were at about 4 m above the ground level at both sites. In Warsaw, the distance to the nearest residential buildings, where coal and biomass might be combusted for heating, was 850 m; in Zabrze, it was 300 m. The nearest coal-fired power stations were about 4 km and the nearest busy roads about 200 m away from the sampling sites both in Warsaw and in Zabrze.

The 24-h PM$_1$ samples were taken on the same days (in parallel) at both sites; At each site, 60 samples were taken in summer (24 June - 22 August 2014) and 60 in winter (8 January - 8 March 2015).
Figure 1. Location of the sampling sites in Warsaw and Zabrze.

The samples of PM$_1$ were collected using two MVS6 PM samplers (the one used in Warsaw was made by ATMO SERVICE, the one used in Zabrze - by LECKEL) equipped with identical sampling heads (made by TSI) which aspired PM$_1$ at an air flow of 2.3 m$^3$/h. PM$_1$ was collected on quartz fiber filters (Quartz Air Sampling Filter, Grade QMA, Ø47 mm; GE Healthcare Life Sciences Corp.). The mass of the PM$_1$ was determined by weighing the filters before and after exposure on a MYA 5.3Y.F micro balance (RADWAG, 1-µg resolution). Before each weighing, the filters were conditioned for 48 h in the weighing room (relative air humidity 45±5%, air temperature 20±2°C).

After determining the PM$_1$ concentrations, a 1x1 cm square fragment was cut out from each exposed filter in which the organic (OC) and elemental (EC) carbon contents were determined. The rest of the filter was then used to determine the 5 water-soluble ions in PM$_1$.

2.2. Chemical analysis

The OC and EC contents of PM$_1$ were determined using a Lab OC-EC Aerosol Analyzer (Sunset Laboratories Inc.) and the EUSAAR2 protocol[15]. The method performance was controlled by systematic calibrating of the analyser within the range proper for the determined concentrations and by analysing standards with certified carbon content (RM 8785 and RM 8786, NIST) and blank samples. The detection limit for total carbon (TC; TC=OC+EC), computed from the measurements of 10 blanks, was 0.91 µgC/cm$^2$ (0.79 and 0.12 µgC/cm$^2$ for OC and EC, respectively). The standard recovery was from 98% to 122% of the certified value for OC and from 95% to 116% for EC (the certified values were taken from the IMPROVE protocol).

The water extracts of PM$_1$ were made by ultrasonizing each filter containing the sample in 25 cm$^3$ of de-ionized water for 60 min at the temperature 15°C and then shaking the extracts for about 12 h (18°C, 60 r/min). The ion content of the extracts was determined using an ion chromatograph (Metrohm AG). The method was validated against the CRM Fluka product nos. 89316 and 89886; the standard recoveries were from 92% (Na$^+$) to 109% (Cl$^-$) of the certified values. The detection limits were as follows: 10 ng/cm$^3$ for NH$_4^+$, 18ng/cm$^3$ for Cl$^-$ and SO$_4^{2-}$, and 27 ng/cm$^3$ for NO$_3^-$ and Na$^+$. 
2.3. **Analysis of results**

The methodology of evaluation of the shares of primary and secondary matter in the mass of PM$_1$ was based on the following assumptions:

- Primary matter is a sum of elemental carbon (EC), primary organic matter (POM), and a total of sodium and chlorine (Na$_2$Cl).
- Secondary matter is a sum of secondary inorganic matter (SIM) and secondary organic matter (SOM).

The analytically determined mass [EC]$_A$ of elemental carbon was assumed as the mass of EC: [EC] = [EC]$_A$. The mass of OM (OM, all PM-bound organic compounds) was assumed to be 1.4 of the analytically determined mass [OC]$_A$ of OC: [OM] = 1.4[OC]$_A$; [16]. SIA consisted of SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$, so [SIA] = [SO$_4^{2-}$]$_A$+[NO$_3^-$]$_A$+[NH$_4^+$]$_A$; [Na$_2$Cl] = [Cl]$_A$+[Na]$_A$.

The division of OM into secondary (SOM) and primary (POM) matter was based on the assumptions that [OM]=[POM]+[SOM] and that [POM]=1.4[POC] (POC, primary organic carbon; [16]), and [POC]=[EC]$_A$·([OC]/[EC])$_{min}$, where ([OC]/[EC])$_{min}$ is the minimum among all the 24-h proportions of [OC]$_A$/[EC]$_A$; [17].

3. **Results and discussion**

The 24-h concentrations of PM$_1$ are very much lower in the warm July-August period than in the cold January-March one (Figure 2). Such a sharp leap of the PM$_1$ concentrations at the start and their high levels in the course of a heating season are typical of Polish urban areas[5]. Here, this effect is clearly seen in Warsaw and even dramatic in Zabrze: while the summer concentrations were comparable and below 25 µg/m$^3$ at both sites, the winter ones reached 200 µg/m$^3$ in Zabrze and 40 µg/m$^3$ in Warsaw.

The mean concentration of PM$_1$ in Warsaw, both over the whole measuring period and its winter part, were much lower than in Zabrze, and also than in other southern Poland cities and periods (Katowice, Racibórz, Wrocław [5,18,19]), but higher than in northern Poland (Gdynia [20]) (Table 1). They, and the summer ones too, did not differ much from the PM$_1$ concentrations in other European cities, and were much lower than in Asia [21-25]. The mean seasonal PM$_1$ concentrations in Zabrze, especially the winter one, were high, and being close to the concentrations in most polluted areas in Asia, were higher than in other European localities [19-25].

The mean winter PM$_1$ concentration in Zabrze (47.1 µg/m$^3$) was almost 4 times the summer one (12.1 µg/m$^3$) and more than 2 times the winter one in Warsaw (17.4 µg/m$^3$), although the summer means at both sites were very close to each other (Table 1). The means of the ambient concentrations of PM$_1$-bound OC and EC behaved similarly. The mean winter concentration of PM$_1$-bound Cl in Zabrze was 5 times higher than in Warsaw, although the summer ones were identical (Table 1). All this, and also earlier works of the authors [5,10,18], proves tremendous and prevailing influence of local municipal coal combustion on winter PM$_1$ concentrations in Zabrze. PM coming from local sources appears to be a serious problem in southern Poland cities in summer as well [5].

In Warsaw, there is no such intense local coal combustion, the elevated winter levels of PM$_1$ and its components are not due to it (Table 1). The effects from coal combustion in PM$_1$ in winter, not so apparent as in Zabrze, are due to the emissions from big coal fired power stations [26]. They are active all the year everywhere in Poland [10,18], more intensely in winter, but in Zabrze, emissions from household ovens combusting coal, biomass, and plastics can in average add even 35 µg/m$^3$ to the 24-h concentrations of PM$_1$ due to power stations and road traffic in winter (Table 1).
Figure 2. Time series of 24-hour PM$_1$ concentrations in the measuring period in Warsaw and Zabrze.
Table 1. Average concentrations of PM$_1$ and of its main components (µg/m$^3$) in Warsaw and Zabrze in summer and in winter.

|          | Zabrze | Warszawa |
|----------|--------|----------|
|          | summer | winter   | summer | winter |
|          | Ave    | St.dev.  | Ave    | St.dev. | Ave    | St.dev. |
| PM$_1$   | 12.1   | 3.5      | 47.1   | 11.1    | 3.3    | 17.4    | 8.4   |
| OC       | 3.4    | 1.0      | 17.0   | 3.0     | 1.0    | 6.6     | 3.7   |
| EC       | 1.0    | 0.4      | 3.4    | 2.1     | 0.9    | 1.4     | 0.6   |
| Cl$^-$   | 0.1    | 0.1      | 2.8    | 2.2     | 0.1    | 1.6     | 0.2   |
| NO$_3^-$ | 0.5    | 0.3      | 2.7    | 1.9     | 0.3    | 2.4     | 1.7   |
| SO$_4^{2-}$ | 2.5   | 0.8      | 3.1    | 1.4     | 2.2    | 1.2     | 2.4   |
| Na$^+$   | 0.7    | 0.3      | 1.2    | 0.3     | 0.3    | 0.7     | 0.3   |
| NH$_4^+$ | 0.6    | 0.3      | 2.6    | 1.6     | 0.6    | 1.6     | 0.9   |
| SOM$^e$  | 1.6    | 0.9      | 10.1   | 10.5    | 1.9    | 5.6     | 4.0   |
| POM$^f$  | 2.4    | 1.0      | 10.2   | 6.3     | 1.4    | 0.5     | 1.1   |
| SIM$^g$  | 3.5    | 1.0      | 8.3    | 4.7     | 3.1    | 1.4     | 6.4   |

$^a$24 June to 22 August 2014  
$^b$8 January to 8 March 2015  
$^c$Arithmetic mean  
$^d$Standard deviation  
$^e$Secondary organic matter  
$^f$Primary organic matter  
$^g$Secondary inorganic matter

The above is very well confirmed by the mutual correlations between the time series of 24-h concentrations of all the measured substances (Table 2). While almost all pairwise Pearson correlations between the concentration series in Zabrze are high (p<0.05, r>0.6; Table 2), the 24-h concentrations of POM were not correlated or correlated only weakly with the concentrations of PM$_1$-bound OC, Cl$^-$, NO$_3^-$, SO$_4^{2-}$, Na$^+$, NH$_4^+$ and SOM in Warsaw. Moreover, the 24-h concentrations of PM$_1$-bound SO$_4^{2-}$, weakly correlated with POM, were also weakly correlated with the concentrations of OC, EC, Cl$^-$, and NO$_3^-$. All this means that: 1. The origins of SOM and POM differ in Warsaw, i.e. the Warsaw SOM precursors and POM have different sources, 2. The SIM precursors, i.e. nitrogen and sulfur oxides, in Warsaw also have different sources.

It agrees with conclusions of the earlier works of the authors, where road traffic was proved to be the predominant source of fine PM in Warsaw, and PM emissions from coal and biomass combustion, mainly in power stations and local boiler plants, exist, but are of lesser significance and manifest clearer their presence in winter [9,26,27]. In Warsaw, individual flats are heated by natural gas combusting, also many oil/gas fired boiler plants are working. There are relations confirmed between inflow of polluted air from south of Poland (Zabrze) and ambient concentrations of sulfur in Warsaw [9].

In Zabrze, the PM$_1$ composition and concentrations are formed mainly by emissions from combustion of coal, biomass, and their derivatives (coal dust, wasted furniture, etc.) in household stoves and local boiler plants. Additionally, the intensity of road traffic is much less in Zabrze than in Warsaw, therefore the effects of traffic emissions in PM$_1$ are drowned out by emissions from combustion, although these effects can be observed very clearly in some periods of the year [11-13].
Table 2. Coefficients of Pearson correlation between diurnal concentrations of PM1 and PM1 components at measuring sites in Warsaw and Zabrze.

| Warsaw | PM1  | OC   | EC   | Cl−  | NO3− | SO42− | Na+  | NH4+ | SOM  | POM  | SIM  |
|--------|------|------|------|------|------|-------|------|------|------|------|------|
| Zabrze |      |      |      |      |      |       |      |      |      |      |      |
| PM1    | 1.00 | 0.95 | 0.82 | 0.72 | 0.85 | 0.57  | 0.53 | 0.91 | 0.94 | 0.42 | 0.90 |
| OC     | 0.97 | 1.00 | 0.87 | 0.76 | 0.86 | 0.39  | 0.51 | 0.89 | 0.99 | 0.38 | 0.84 |
| EC     | 0.92 | 0.96 | 1.00 | 0.70 | 0.74 | 0.22  | 0.33 | 0.74 | 0.82 | 0.64 | 0.68 |
| Cl−    | 0.91 | 0.88 | 0.86 | 1.00 | 0.82 | 0.33  | 0.64 | 0.80 | 0.74 | 0.01 | 0.78 |
| NO3−   | 0.84 | 0.82 | 0.78 | 0.78 | 1.00 | 0.37  | 0.60 | 0.94 | 0.85 | 0.16 | 0.93 |
| SO42−  | 0.66 | 0.64 | 0.58 | 0.60 | 0.68 | 1.00  | 0.52 | 0.53 | 0.42 | 0.08 | 0.69 |
| Na+    | 0.59 | 0.56 | 0.54 | 0.64 | 0.57 | 0.42  | 1.00 | 0.59 | 0.52 | -0.21 | 0.66 |
| NH4+   | 0.92 | 0.90 | 0.86 | 0.90 | 0.93 | 0.79  | 0.56 | 1.00 | 0.88 | 0.19 | 0.97 |
| SOM    | 0.95 | 0.98 | 0.88 | 0.85 | 0.81 | 0.66  | 0.53 | 0.88 | 1.00 | 0.32 | 0.85 |
| POM    | 0.93 | 0.96 | 1.00 | 0.87 | 0.79 | 0.58  | 0.56 | 0.86 | 0.88 | 1.00 | 0.17 |
| SIM    | 0.87 | 0.85 | 0.81 | 0.83 | 0.95 | 0.86  | 0.56 | 0.98 | 0.85 | 0.81 | 1.00 |

*a Statistically significant correlations are written in red*

The share of secondary matter in PM1 is greater in Warsaw than in Zabrze, especially the difference between the SIM shares are significant. In Zabrze, SIM and SOM make in average 30% and 17% of the PM1 mass in summer, in Warsaw – 28% and 17%, respectively. In winter it is 19 and 18% in Zabrze and 37 and 29% in Warsaw, respectively. The mass contribution of EC to PM1 at both sites and in both seasons is about 8%, despite the differences in the PM1-bound EC ambient concentrations. In Zabrze, the share of POM in the PM1 mass is 20% in summer and 24% in winter; in Warsaw – 13% and 7%, respectively. Also, Na_Cl has a greater share in the PM1 mass in Zabrze than in Warsaw (Figure 3).

Such chemical contents of PM1 at sampling sites can point to the PM1 sources at these sites. The nitrogen oxides and volatile organic compounds from gas combustion are the cause of the winter elevation in SIM and SOM shares in PM1 in Warsaw. In Zabrze, the SIM share in PM1 grows when road traffic, the main source of the SIM precursors, gains its importance in summer. Beyond this period, coal and trash combustion, the source of POM, sulfates in SIM, and Na_Cl, decides on the air quality in Zabrze.

The strength of dominating PM1 sources at the sites can also be compared by comparing the differences between the proportions of the concentrations of the PM1 components at these sites [5,8]. Higher proportion [EC]/[OC] in Warsaw than in Zabrze in both seasons evidences higher contribution of road traffic to PM1 concentrations in both seasons in Warsaw, higher proportion [SO42−]/[NH4+] in Zabrze than in Warsaw suggests that emissions from fossil fuel combustion have contribute more to PM1 concentrations in Zabrze [5].
Figure 3. Diurnal variability of the PM$_1$ chemical composition in the measuring period in Warsaw (a) and Zabrze (b).

4. Conclusions
The shares of primary and secondary matter in the mass of PM$_1$ were determined for each of 120 days of observations at the two sites. In average, secondary aerosol (inorganic and organic together) accounted for about 55% of the PM$_1$ mass in Warsaw and 40% in Zabrze. Seasonally, it was 66% and 38% in winter and 45% and 43% in summer, respectively. This means that on winter days almost 12 μg/m$^3$ of PM$_1$ in Warsaw and 18 μg/m$^3$ in Zabrze came from transformations of ambient volatile
organic compounds, ammonia, and sulphur and nitrogen oxides; in summer it was 5 μg/m³ of PM₁ in both Warsaw and Zabrze.

In Warsaw, main sources of secondary aerosol are road traffic and natural gas combustion. In Zabrze, road traffic is the main source of secondary inorganic aerosol precursors; the precursors of secondary organic aerosol come from municipal sources as in Warsaw, but besides natural gas also coal is combusted. Therefore in Zabrze, great, much greater than in Warsaw, amounts of primary organic matter, containing sodium and chlorine, are released to the air.

Therefore, limiting emissions of primary PM (soot and primary organic carbon from diesel powered engines and household furnaces, road dust, metallic particles from industrial plants, etc.) will not result in a dramatic reduction of PM₁ concentrations in Warsaw. In Zabrze, instead, reducing PM₁ concentrations should consist just in the reduction of primary PM₁ emissions, especially those coming from heating in dwelling houses.

To the best knowledge of the authors, such long time series as presented here of daily concentrations of PM₁ and its main components, measured in two distant cities (Warsaw and Zabrze, Poland) in parallel, allowing for comparison of the efficiencies of the main PM₁ sources in these cities, are considered for the first time. The approach presented here is unique in the length of the measuring period (120 measuring days) and extensiveness of the investigations. It allowed to compare comprehensively the behaviour of PM₁ and the PM₁ sources at two urban background sites.

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