Three-year long source apportionment study of airborne particles in Ulaanbaatar using X-ray fluorescence and Positive Matrix Factorization

G. Gunchin¹, ²*, M. Manousakas³, J. Osan², ⁴, A.G. Karydas⁵, K. Eleftheriadis³, S. Lodoysamba⁶, D. Shagjjamba¹, A. Migliori², R. Padilla-Alvarez², C. Streli⁷, and I.G. Darby², ⁸

¹ Nuclear Research Center, National University of Mongolia, Ulaanbaatar, Mongolia
² Nuclear Science and Instrumentation Laboratory, Physics section, IAEA, Seibersdorf, Austria
³ Institute of Nuclear & Radiological Sciences & Technology, Energy & Safety, N.C.S.R. Demokritos, Greece
⁴ Environmental Physics Department, Hungarian Academy of Sciences Centre for Energy Research, Budapest, Hungary
⁵ Institute of Nuclear and Particle Physics, N.C.S.R "Demokritos", Athens, Greece
⁶ Faculty of Engineering, German-Mongolian Institute for Resources and Technology, Ulaanbaatar, Mongolia
⁷ Vienna University of Technology, Atominstitut, Vienna, Austria
⁸ SUPA, School of Physics and Astronomy, University of Glasgow, Glasgow, United Kingdom

Abstract

The capital city of Mongolia, Ulaanbaatar, suffers from high levels of pollution due to excessive airborne particulate matter (APM). A lack of systematic data from the region results in great interest for investigations on the kind and origin of the contributing pollution sources, seasonal variations, the effect of meteorological conditions or even the time-dependence of anthropogenic sources. This work reports source apportionment results from a large data set of 184 fine (PM₂.₅) and 184 coarse (PM₂.₅-10) fraction atmospheric PM samples, collected over a three-year period (2014-2016) in Ulaanbaatar, Mongolia. The concentrations of 16 elements measured by an energy dispersive X-ray fluorescence spectrometer together with the black-carbon content measured by the reflectometry method were used as input data for the application of the Positive Matrix Factorization (PMF) method. The PMF results revealed that the contribution of mixed factors dominates the coarse fraction. Interestingly, soil and traffic sources were identified as principle contributors of the fine fraction. Examination of the source profiles and the seasonal variation of their contributions indicates that fly ash emanating from coal combustion mixes with traffic emissions and soil re-suspensions resulting in variable chemical source profiles. Four sources were identified for both fractions, namely soil, coal combustion, traffic and oil combustion. The contribution of these sources to the coarse fraction were 35%, 16%, 41% and 8% respectively, whereas for the fine fraction they were 31%, 27%, 31% and 11%. Additionally, the concentration-weighted trajectory analysis technique was used to assess the probable source contributions from long range transport events.

Keywords: Airborne Particulate Matter; XRF; PMF; Ulaanbaatar.

*Corresponding author. E-mail address: gerelmaa-g@num.edu.mn
INTRODUCTION

Atmospheric pollution due to particulate matter (PM) is a wide concern for both human health and the climate (Cohen et al., 2004; Ostro et al., 2015). Aerosol particles can be formed either from precursor gases via chemical reactions in the atmosphere or be directly emitted from various sources of anthropogenic and natural origin (Davy et al., 2011). PM is a mixture of organic and inorganic compounds and it is important to be able to identify its sources and quantify their contributions (Hasenkopf et al., 2016; Diapouli et al., 2017).

The process of identification and apportionment of pollutants to their sources is an important step in air quality management, since this can be used as a baseline data for the establishment of air pollution mitigation strategies. Multivariate receptor models are very useful tools as they may be applied directly to the observed PM composition data (Santoso et al., 2008). The dataset of the PM elemental composition is an essential input for the characterization of specific emission sources using statistical apportionment tools such as the positive matrix factorization (PMF) model (Hopke et al., 2006), and can be accompanied by complementary data (e.g. black carbon, organic carbon, water-soluble ions). Generally, major elements such as Mg, Al, Si, Ca, Ti and Fe are primarily associated with windblown soil, S with fossil fuel burning, and K with smoke and biomass burning, whereas Cr, Mn, Fe, Cu and Zn are associated with industrial processes, while Pb and Br derive from motor vehicle exhausts (Manousakas et al., 2015; Cohen et al., 1996). For a comprehensive source apportionment a group of analytical techniques should be applied to determine the concentration of the maximum number of elements being either major, minor or trace constituents of the PM mass. Importantly, the respective uncertainties for the elemental concentrations should be well defined for source apportionment studies, as they are used in the model.
X-ray fluorescence (XRF) analysis is a multi-elemental, sensitive and very versatile analytical method widely used in various scientific fields and amongst the ones best suited for environmental researches and particularly for PM analyses (Calzolai et al., 2008; Shaltout et al., 2017; Prost et al., 2018). In this study, an energy dispersive XRF spectrometer (PAnalytical Epsilon 5, Almelo, the Netherlands) was used for the elemental analysis. The combined result of employing several secondary targets, coupled within a three-dimensional polarizing optical geometry leads to the significant reduction in the measured PM XRF spectrum of the scattered X-rays from the tube emission, thus lowering the continuum counts (noise signal). Such a reduction of the noise signal, in combination with the instrument sensitivity gains achieved by optimizing the excitation conditions for different groups of elements, enables obtaining minimum detection limits in the low ng cm$^{-2}$ range or even below for most of the elements of interest (Shaltout et al., 2018). For the determination of Black Carbon (BC) content, a M43D Smoke stain light reflectometry device from Diffusion Systems Ltd, London, United Kingdom was used.

Herein we present source apportionment results from a large data set of fine (PM$_{2.5}$) and coarse (PM$_{2.5-10}$) fractionated atmospheric PM samples collected over a three year period (2014-2016) in Ulaanbaatar, Mongolia.

**METHODS**

**Sample collection**

Mongolia is located across northeast and central Asia at an average altitude of 1400 m above sea level. Its elevation, relatively high latitude, non-coastal geography and the effects of Siberian anticyclone weather patterns give the country a continental dry climate with long, cold winters and relatively short summers. Average temperatures from November to March drop below zero
degrees centigrade over the whole country. The capital and largest city, Ulaanbaatar is located on Tuul River Valley and surrounded by mountains. The total area of the city is 4735.1 km² and has approximately 1.4 million inhabitants, comprising 45% of the whole country’s population. According to a recent report (UbStat report, 2016), 43.3% of Ulaanbaatar residents live in apartment buildings which are connected to the main central heating grid and the remaining 56.7% of inhabitants live in traditional Mongolian dwelling areas, called Ger khoroolol, where coal, wood and other household materials are used as fuel for domestic heating and cooking purposes. The annual consumption statistics of coal and wood for Ulaanbaatar inhabitants are described elsewhere (Guttikunda, 2007). The regional climate, geographical location and residential features of Ulaanbaatar city intensify a winter air pollution problem. It is for these reasons that Ulaanbaatar is reported as one of the world’s most polluted cities (World Bank report, 2011).

A total of 184 air particulate matter samples, size fractionated according to aerodynamic diameter between 10 microns and 2.5 microns (PM\textsubscript{10-2.5} or coarse fraction) and 2.5 microns or less (PM\textsubscript{2.5} or fine fraction), were collected for two days per week (Monday and Thursday) throughout the period 2014 - 2016. The sampling site was located at the Nuclear Research Center (NRC) of the National University of Mongolia (NUM). The monitoring site is surrounded by a combination of buildings, Ger khoroolol, paved and unpaved roads.

Sampling was undertaken using a GENT instrument (Maenhaut et al., 1993), comprising a PM\textsubscript{10} impactor–type size selective inlet and a stacked filter unit (SFU) assembly connected to a pump and an air flow meter. The SFU is made up of two filters arranged sequentially. The upper filter (8 µm pore size polycarbonate) collects the coarse fraction, and the lower filter (0.4 µm pore size polycarbonate) collects the fine fraction. The average air flow rate was 16.0 l min\textsuperscript{-1} and the average mass loadings for PM\textsubscript{2.5} and PM\textsubscript{10-2.5} were 92 µg m\textsuperscript{-3} and 131 µg m\textsuperscript{-3}, respectively. The
collection of the samples covered a 24-hour period. However, on winter days it was not possible
to collect over the entire 24-hours as high pollution resulted in clogging of the filters. Therefore,
the sampler was operated alternating on and off periods over the course of the 24 hours so as to
provide a representative sample for that day.

Gravimetric masses of the samples were obtained in triplicate measurements using a digital
weighing balance that has an automatic calibration function to ensure accuracy. Gravimetric
masses were subsequently divided by the sampled air volume to calculate average PM mass
concentrations.

The black-carbon (BC) measurement and its quantification model are described elsewhere
(Davy et al., 2011).

**Elemental analysis, XRF**

XRF analysis has been performed in the IAEA Nuclear Science and Instrumentation
Laboratory (NSIL) in Seibersdorf, Austria. The spectrometer features a variety of secondary
targets, such as Al, CaF$_2$, Fe, Ge, Zr, Mo, Ag and Al$_2$O$_3$ (Barkla polarization target) which are
excited by a Sc-W tube. The characteristic X-ray radiation emitted by the sample is detected by a
Ge detector with a measured energy resolution of approximately 150 eV FWHM at Mn-K$_\alpha$ (5.89
keV). Eight measuring conditions were selected to optimize the analytical sensitivity and
precision for the determination of certain groups of elements in aerosol particles on filters as
presented in Table 1. For all secondary and polarization targets, the live time of the
measurements was set to 300 seconds. The analysis was performed in vacuum.

The quantification was based on a calibration performed by measuring thin film single element
and compound reference materials (Micromatter, Surrey, BC, Canada), prepared on a
polycarbonate filter support. The nominal areal densities of the certified materials ranged
between 43.8 µg cm\(^{-2}\) and 62.3 µg cm\(^{-2}\) with relative uncertainties quoted as 5\%. The following standards were used for calibration purposes: NaCl, MgF\(_2\), Al, SiO, KCl, CaF\(_2\), Ti, V, Mn, Fe, Co, Ni, Cu, ZnTe, Nb\(_2\)O\(_3\), MoO\(_3\), CdSe, Sn, Sb, BaF\(_2\), CeF\(_3\), NdF\(_3\), HoF\(_3\), YbF\(_3\), WO\(_3\), Au, TlCl and Pb. A standard reference material of particulate matter on filter media (SRM 2783, NIST, Gaithersburg, MD, USA) was used for validation of the analytical method. The provided software of the Epsilon 5 was used for spectrum deconvolution, whereas the elemental concentrations were calculated using a sensitivity curve determined by the available set of thin single- or two-element standards.

For the estimation of the combined uncertainty of the results the following contributing factors were considered: the peak area statistical uncertainty, the fitting error in the calibration (sensitivity) curve, the quoted uncertainties of the elemental areal densities reported for the standards (5\%), the estimated error due to the omission of the self-attenuation effect for the excitation and detection of light-element characteristic X-rays (the calibration was based on presuming the samples as infinitely thin in the sense of x-ray attenuation), as well as the relative standard deviation of three replicate measurements (reproducibility in operation). The self-attenuation effect occurs significantly within the PM sample components due to the low energy of the exciting and characteristic X-rays used in the measurement of light elements. According to a previous study (Gutknecht et al., 2010), the bias of results due to the omission of self-attenuation correction is estimated to be 16\% for Na, 12\% for Mg and 11\% for Al and Si, whereas for the rest of measured elements is considered as 1\%. The measurement’s precision was calculated as the standard deviation of results from three consecutive measurements for twenty representative samples. Finally, the combined relative uncertainty is calculated as the square root of the sum of squares of the various contributing relative uncertainties (Manousakas et al., 2017; Gutknecht et al., 2010).
The elemental minimum detection limits (MDLs) were obtained by measuring the National Institute for Standards and Technology (NIST) standard reference material (SRM) 2783. In Fig. 1, the MDL’s of the measured elements are presented for a measurement time of 300 s and the employed secondary target for each element is denoted by different symbols. The detection limits were calculated taking three times the square-root of background counts (measured continuum under the characteristic peak) as the minimum detectable peak area.

**Positive Matrix Factorization**

USEPA Positive Matrix Factorization (PMF) (Paatero, 1997) 5.0 version was used for the source identification and apportionment of the both PM$_{2.5}$ and PM$_{10-2.5}$ fractions. Fifteen species of PM$_{10-2.5}$ (BC, Na, Mg, Al, Si, S, Cl, K, Ca, Ti, Mn, Fe, Zn, Pb and Mass) and twelve species of PM$_{2.5}$ (BC, Al, Si, S, K, Ca, Ti, Mn, Fe, Zn, Pb and Mass) were used for the PMF modelling. Na, Mg, and Cl were considered as “weak” or “bad” depending on the uncertainties that were associated with their measurement.

The basic equation that refers to the solution of the mass balance problem is common for all receptor models including PMF. The equation can be written as Eq. (1):

$$X = G \times F + E$$  \hspace{1cm} (1)

Where $X$ is the concentration of species measured on sample, $G$ is the contribution of the source, $F$ the source profile and $E$ the residual. Since the PMF solution is not unique, to obtain the optimum solution $G$ and $F$ are adjusted until a minimum $Q$ for a given number of factors is found. $Q$ is defined as Eq. (2):
\[ Q = \sum_{j=1}^{m} \sum_{i=1}^{n} \frac{E_{ij}^2}{s_{ij}^2} \]  

(2)

Where \( s_{ij} \) is the uncertainty of the \( j_{th} \) species concentration in sample \( i \), \( n \) is the number of samples, and \( m \) is the number of species.

Solutions with a number of factors from 3 to 8 were investigated in order to identify the meaningful solution with the highest number of factors. For both PM fractions the optimum solution was found to be for 4 factors. Data were screened by their uncertainties (signal–to–noise ratio (S/N) in PMF). Variables with very low S/N ratios (\( \leq 0.2 \)) were defined to be bad and excluded from the analysis, while weak variables (\( 0.2 \leq S/N \leq 2 \)) were down-weighted (Paatero and Hopke, 2003). For PM\(_{2.5}\) analysis, the elements Na, Mg and Cl were excluded; V and Cr were defined as “bad” and BC, Cu, Zn, Pb and As were defined as “weak” variables and for PM\(_{10-2.5}\) analysis only Cr defines as “bad” and BC, Na, Mg, Cl, Ni, Zn, Pb and As species were defined as “weak”. The mass was set as total variable for analyses. The robustness of the solutions was investigated using the uncertainty estimation tool that EPA PMF 5.0 offers. According to Bootstrap analysis (Adam et al., 2007) the factors were reproduced at a level higher than 80%, while no factor swaps appeared for the minimum level of Q at Displacement and Bootstrap Displacement analysis. The comparison between predicted (PMF) and gravimetric masses of the both fractions were found to be high (\( R^2_{\text{ coarse}} = 0.82 \) and \( R^2_{\text{ fine}} = 0.86 \)).

**RESULTS**

**Particulate matter concentrations**
The average concentrations of annual and seasonal coarse and fine particulates are shown in Table 2 and 4. The presented values indicated that annual concentrations of the both fractions are much higher than Mongolian National Ambient Air Quality Standards (MNS 4585:2007) set for year average to 50 µg m\(^{-3}\) for PM\(_{10}\) and 25 µg m\(^{-3}\) for PM\(_{2.5}\) as well as for the World Health Organization’s (WHO) guideline (WHO, 2005) corresponding values of 20 µg m\(^{-3}\) and 10 µg m\(^{-3}\). PM10 limit values are used in the text not for direct comparison but as reference to provide a better metric regarding the measured mass concentration levels. Particulate matter concentrations were dominated by the coarse fraction for the whole study period. The maximum concentrations were measured at 580 µg m\(^{-3}\) for the coarse particle and 7% of the total sampling days were above 300 µg m\(^{-3}\), and the annual and seasonal values show that no drastic difference between warm (April – September) and cold (October – March) season averages exist (Fig. 3). The fine particle concentration increases during the cold season and highest values were recorded during winter months while the concentration went often over 100 µg m\(^{-3}\).

**PMF analysis results**

**PM\(_{10-2.5}\) data analysis**

In Table 2, the descriptive statistics regarding PM\(_{10-2.5}\) and elemental mass concentrations are presented. The minimum detection limits (MDLs), number of elemental constituents which were detected above their respective MDLs, as well as average elemental concentrations during warm and cold season are also included in the table.

As presented in Table 2, for the coarse fraction the elements Al, Si, Ca, Ti and Fe originate mainly from a crustal matter source and were higher in the warm period. The Si:Al ratio was found to be 2.6, indicative of a typical soil composition – aluminosilicates (Lide, 1992).
Several factors (3-8) were used in order to obtain the optimum PMF solution, and settling upon four (4) contributing factors to be the solution was choses with regard to the highest number of factors with physical sense. The factors have been identified based on the key elements in the source fingerprint and the correlations between them. The selected sources are traffic, soil, coal combustion and oil and their elemental profiles are presented in Fig. 4.

The first profile has been identified as traffic. Although, elemental profile shows that this factor can be a combination of several combustion sources as it contains the highest percentage of BC and S, as well as elements with crustal and anthropogenic origin. However, the high percentage of elements as Ni, Cu, Zn and Pb indicates that this factor mainly represents a traffic source (Sarigiannis et al., 2017). This source contributes 41% of the total PM$_{10-2.5}$ mass and seasonal variation presents higher contribution in the cold season. The fact that the source has higher contribution during the cold season is another indication that the source is mixed with other combustion sources such as coal, which is used for residential heating and is expected to have a higher contribution during the cold season.

The second source profile was identified as soil since it contains crustal matter elements Mg, Al, Si, Ca, Ti, Mn and Fe. Moreover, this factor has high presence of Na, Cl, K and Zn which indicates that it might be mixed with some other sources such as salt and anthropogenic sources that can contribute to this source by soil resuspension (Eleftheriadis and Colbeck, 2001). The contribution of this source is 35% of the total PM$_{10-2.5}$ mass. Seasonal variations indicated that this source has a higher contribution during the warm period (Fig. 5).

The third source profile, coal combustion, is traced by the high concentration of BC and S. It is not very common that a combustion source affects the coarse PM fraction to that extent. A possible explanation for this can be that as the ashes which remain in the stove after the burning of coal for domestic heating, are in many cases discarded by the populace on the soil, the ashes
may then become resuspended along with soil and other anthropogenic related elements. The contribution of this source is 16%.

The last factor was identified as oil combustion as it contains a significant amount of V and Ni. Interestingly, this source is for the first time identified on this site. In previous studies conducted in the area (Davy et al., 2011; Gunchin et al., 2012) this source profile was not found. This source has a contribution of 8% of the total mass and presents low temporal variation.

The output of PMF was used to calculate the annual contribution of the sources for the 3 years of the study. Table 3 presents average mass contributions of the sources to the PM$_{10-2.5}$ fraction for each year. The results of Table 3 reveal that the sources of the coarse fraction appear to be stable for the three years of the study period. However, the average mass of the year 2015 was relatively low when compared with that of the years 2014 and 2016. This might be affected by the missing data of November and December 2015. As it is presented in Table 3, the variance in the source contributions for the three years the study was conducted is low.

PM$_{2.5}$ data analysis

Table 4 presents PM$_{2.5}$ descriptive statistics. The minimum detection limits (MDLs), number of elemental constituents which were detected above their respective MDLs and average concentrations during warm and cold season are also included in the Table 4. As presented in Table 4, BC, S, Zn and As species significantly dominate the fine fraction. The average concentrations were much higher in cold periods compared to warm seasons. The correlation of BC and S components were 0.96, clearly indicating that they have a common source which is likely coal combustion.
Based on the key contribution of certain elements, the number of sources that were identified for the fine fraction was four (4) as well. These are soil, traffic, coal combustion and oil. The elemental source profiles are presented in Fig. 6.

The elemental profile of soil includes clear contribution of crustal matter elements such as Mg, Al, Si, Ca, Ti, Mn and Fe. The correlation of the concentrations of Al and Si were 0.96 which indicating typical soil origin. A contribution of this source is 31% of the total PM$_{2.5}$ mass and slightly higher mass variation during the warm period than annual average mass concentration (Fig. 7).

The second source profile is identified as traffic. Higher percentage of elements as Ni, Cu, Zn and Pb in this profile indicates that this factor mainly from traffic. This source has a contribution of 31% of the total PM$_{2.5}$ mass. However, this source profile also includes a high presence of BC and this might influence the total contribution of this source. Seasonal variance shows this factor has higher contribution during the cold season.

The third source has been identified as coal combustion since it is characterized by significantly higher concentrations of BC and S. The correlation of these two components was 0.90. This factor represented 27% of the total PM$_{2.5}$ mass. Interestingly, contribution of this source has lower than crustal matter source in a fine fraction. However, this factor also containing different source elements such as V, Zn and Pb which assumed that there might be mixture source of traffic and road dust were included to this profile. Additionally, As appears in this source profile in relatively enhanced concentration. The same feature applies to coal combustion source in the coarse fraction as well. It should be noted that As is a known tracer of coal combustion.
The last factor identified as oil presents the same elemental profile as the coarse fraction, and has a significant amount of V and Ni. A contribution of this source has 11% of the total mass and has higher seasonal trend during the cold months.

The annual contributions were calculated for the fine fraction as well. Table 5 presents average mass contributions of each year of the PM$_{2.5}$ sources. It seems that in 2016 the coal combustion source was increased by 13-15%, whereas oppositely the soil source was decreased. The traffic source was higher in 2014 and during the next two years it was quite constant. The oil source was also relatively stable during the period of study.

Concentration weighted trajectory analysis

The Concentration weighted trajectory (CWT) is a type of statistical trajectory model and it is widely used to locate the regional source areas where potentially affecting to the receptor site by long range transportation (Chandra et al., 2014). In this method, the mean concentration is calculated and used as weight for the residence time of the trajectory in modeled each grid cell and assigned equally to all segments of trajectory (Wang et al., 2009). Back trajectory calculations were done by HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model using a global meteorological dataset GDAS. The trajectories were computed for an altitude of 1500 m above ground level and 120-hour backward for the sampling days. The height was selected to better represent long range transport events. Otherwise, it may be affected by local sources and topography. For the CWTs, Zefir tool was used (Petit et al., 2016).

Fig. 8 shows air mass back trajectories of each source of PM$_{2.5}$ contributions. From the result of the CWT analyses, it seems clearly that northwest and north part contributes dominantly to the sampling area. Due to the breakdown in Siberian High-Pressure zone, wind originates usually from northwest direction to Ulaanbaatar (Davy et al., 2011). However, the combustion
and traffic source trajectories indicate a more local behavior. The Oil source seems affected from long range transportation from northerly direction.

CONCLUSIONS

This study conducted at Ulaanbaatar, Mongolia provides an insight of the APM related pollution in the area. A secondary target ED-XRF commercial system was used for the chemical analysis of PM samples, which found to be very suitable for rapid quantitative analysis. Obtained MDL values demonstrated that determination of low concentrations of an element on filter is possible.

PMF statistical technique was used to identify elemental source fingerprints and their contributions to the total mass of PM samples. The results of the study indicate that coal combustion for domestic heating as primary source as well as the scattering of the dust that is produced in the heating ovens has a significant effect on the air quality of the area. Coal dust is produced in high amounts and it is in many cases discarded directly on soil. Thus, the dust that is resuspended is a mixture natural and anthropogenic emissions. This is confirmed by the the source profiles and the temporal variation of the source contributions which indicate that fly ash originating from coal combustion was mixed with traffic emissions and soil resuspension resulting to mixed chemical source profiles. The following four sources were determined in both fractions: soil, coal combustion, traffic and oil combustion. The contributions of these sources to the coarse fraction were 35%, 16%, 41% and 8%, whereas for the fine fraction 31%, 27%, 31% and 11%, respectively.

Seasonal source mass contribution to the fine fraction shows that combustion related source is much dominant in cold season (Oct - Mar) and crustal matter source is the highest for warm
season (Apr – Sept). For the coarse fraction, traffic (mixed with coal combustion) source is the highest in cold season and soil is the dominant during the warm season.

An analysis of concentration weighted trajectory was applied to investigate the influence of long range transport to the sampling area. The results indicated that Heavy Oil combustion is the source that it is mainly affected from long range transport.

Overall the study highlights the importance of detailed air quality studies in less studied areas. The differences that exist in people habits, environmental conditions and topography when compared to other areas such as Europe and North America, as well as other parts of Asia, creates the opportunity to reevaluate the performance of well-known methods of environmental analysis.

ACKNOWLEDGEMENTS

This project was funded by Mongolian Foundation for Science and Technology and supported in part by the International Atomic Energy Agency (IAEA) through the Regional Cooperation Agreement Program “RAS/7/023”.

Moreover, this work has been supported by the IAEA and the International Centre for Theoretical Physics and through an IAEA-ICTP Sandwich Training Educational Program fellowship C6/MON/15001 granted to Gerelmaa Gunchin.

REFERENCES


Ostro, B., Tobias, A., Karanasiou, A., Samoli, E., Querol, X., Rodopoulou, S., Basagaña, X., Eleftheriadis, K., Diapouli, E., Vratolis, S., Jacquemin, B., Katsouyanni, K., Sunyer, J.,


Wobrauschek, P., Streli, C., (2018), Comparative elemental analysis of fine particulate matter
(PM$_{2.5}$) from industrial and residential areas in Greater Cairo-Egypt by means of a multi-
At Spectrosc.* 145: 29–35.

Ulaanbaatar Statistical report. (2016). Department of statistics, Ulaanbaatar city governor’s
implementation agency.

various trajectory statistical analysis methods to identify potential sources from long-term air


Figure Captions

**Fig. 1.** Nuclear Research Center (NRC) air quality monitoring site, 4 kilometers east of the center of Ulaanbaatar (latitude 47°.55’, longitude 106°.55’, 1300 m above sea level). Source: ©1992 MAGELLAN Geographix™ and https://maps.google.com/

**Fig. 2.** Minimum detection limit values established from the NIST reference material (SRM 2783) air particulate filter

**Fig. 3.** Time series of the both PM$_{10-2.5}$ (upper) and PM$_{2.5}$ (lower) fractions at the sampling site, from January 2014 to January 2017

**Fig. 4.** Elemental source profiles derived by PMF for PM$_{10-2.5}$ samples

**Fig. 5.** Seasonal source mass contribution to PM$_{10-2.5}$

**Fig. 6.** Elemental source profiles derived by PMF for PM$_{2.5}$ samples

**Fig. 7.** Seasonal source mass contribution to PM$_{2.5}$

**Fig. 8.** CWT for the PM$_{2.5}$ source contributions showing potential source locations (concentrations in unit of ng m$^{-3}$)
Fig. 1.
Fig. 2.
Fig. 3.
Fig. 4.
Fig. 5.
Fig. 6.
Fig. 7.
Fig. 8.
Table Captions

Table 1. XRF spectrometer (PANalytical Epsilon-5) conditions for elemental analysis of APM

Table 2. Summary statistics for PM$_{10-2.5}$, BC and elemental concentrations

Table 3. Yearly source contributions to PM$_{10-2.5}$

Table 4. Summary statistics for PM$_{2.5}$, BC and elemental concentrations

Table 5. Yearly source contributions to PM$_{2.5}$ by average mass and percentage
Table 1. XRF spectrometer (PANalytical Epsilon-5) conditions for elemental analysis of APM

<table>
<thead>
<tr>
<th>N r.</th>
<th>Targets</th>
<th>Detected elements</th>
<th>Voltage (kV)</th>
<th>Current (mA)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Al</td>
<td>(Na), Mg</td>
<td>25</td>
<td>24</td>
</tr>
<tr>
<td>2</td>
<td>CaF$_2$</td>
<td>Al, Si, P, S, Cl, K</td>
<td>40</td>
<td>15</td>
</tr>
<tr>
<td>3</td>
<td>Fe</td>
<td>Ca, Sc, Ti, V, Cr</td>
<td>75</td>
<td>8</td>
</tr>
<tr>
<td>4</td>
<td>Ge</td>
<td>Mn, Fe, Co, Ni, Cu, Zn</td>
<td>75</td>
<td>8</td>
</tr>
<tr>
<td>5</td>
<td>Zr</td>
<td>Ga, Ge, As, Se, Br, Pb-L</td>
<td>100</td>
<td>6</td>
</tr>
<tr>
<td>6</td>
<td>Mo</td>
<td>Sr, Y</td>
<td>100</td>
<td>6</td>
</tr>
<tr>
<td>7</td>
<td>Ag</td>
<td>Zr, Nb, Mo, Tc, Ru</td>
<td>100</td>
<td>6</td>
</tr>
<tr>
<td>8</td>
<td>Al$_2$O$_3$</td>
<td>Rh, Pd, Ag, Cd, In, Sn, Sb, Te, I, Cs, Ba, La, Ce</td>
<td>100</td>
<td>6</td>
</tr>
<tr>
<td>Element</td>
<td>ng m$^{-3}$</td>
<td>Average</td>
<td>S.D</td>
<td>Median</td>
</tr>
<tr>
<td>---------</td>
<td>-------------</td>
<td>---------</td>
<td>-----</td>
<td>--------</td>
</tr>
<tr>
<td>Mass</td>
<td>13100</td>
<td>10200</td>
<td>0</td>
<td>101000</td>
</tr>
<tr>
<td>BC</td>
<td>3850</td>
<td>2130</td>
<td>3800</td>
<td>3800</td>
</tr>
<tr>
<td>Na</td>
<td>708</td>
<td>394</td>
<td>625</td>
<td>625</td>
</tr>
<tr>
<td>Mg</td>
<td>604</td>
<td>325</td>
<td>549</td>
<td>549</td>
</tr>
<tr>
<td>Al</td>
<td>4660</td>
<td>3400</td>
<td>4000</td>
<td>4000</td>
</tr>
<tr>
<td>Si</td>
<td>12500</td>
<td>9340</td>
<td>10900</td>
<td>10900</td>
</tr>
<tr>
<td>K</td>
<td>1000</td>
<td>913</td>
<td>662</td>
<td>662</td>
</tr>
<tr>
<td>Ca</td>
<td>338</td>
<td>280</td>
<td>266</td>
<td>266</td>
</tr>
<tr>
<td>K</td>
<td>1830</td>
<td>1390</td>
<td>1550</td>
<td>1550</td>
</tr>
<tr>
<td>Ca</td>
<td>4270</td>
<td>2800</td>
<td>3580</td>
<td>3580</td>
</tr>
<tr>
<td>Ti</td>
<td>368</td>
<td>264</td>
<td>322</td>
<td>322</td>
</tr>
<tr>
<td>V</td>
<td>5.00</td>
<td>4.00</td>
<td>4.00</td>
<td>4.00</td>
</tr>
<tr>
<td>Cr</td>
<td>7.00</td>
<td>4.00</td>
<td>7.00</td>
<td>7.00</td>
</tr>
<tr>
<td>Mn</td>
<td>99.0</td>
<td>67.0</td>
<td>84.0</td>
<td>84.0</td>
</tr>
<tr>
<td>Fe</td>
<td>3930</td>
<td>2840</td>
<td>3250</td>
<td>3250</td>
</tr>
<tr>
<td>Ni</td>
<td>4.00</td>
<td>2.00</td>
<td>4.00</td>
<td>4.00</td>
</tr>
<tr>
<td>Cu</td>
<td>18.0</td>
<td>9.0</td>
<td>16.0</td>
<td>16.0</td>
</tr>
<tr>
<td>Zn</td>
<td>62.0</td>
<td>46.0</td>
<td>52.0</td>
<td>52.0</td>
</tr>
<tr>
<td>Pb</td>
<td>29.0</td>
<td>28.0</td>
<td>23.0</td>
<td>23.0</td>
</tr>
<tr>
<td>As</td>
<td>8.00</td>
<td>8.00</td>
<td>6.00</td>
<td>6.00</td>
</tr>
</tbody>
</table>

Table 2. Summary statistics for PM$_{10-2.5}$, BC and elemental concentrations.
Table 3. Yearly source contributions to PM\textsubscript{10-2.5}

<table>
<thead>
<tr>
<th>Sources</th>
<th>2014</th>
<th></th>
<th>2015</th>
<th></th>
<th>2016</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Average (µg m\textsuperscript{-3})</td>
<td>%</td>
<td>Average (µg m\textsuperscript{-3})</td>
<td>%</td>
<td>Average (µg m\textsuperscript{-3})</td>
<td>%</td>
</tr>
<tr>
<td>Traffic</td>
<td>28</td>
<td>42</td>
<td>14</td>
<td>37</td>
<td>25</td>
<td>43</td>
</tr>
<tr>
<td>Soil</td>
<td>24</td>
<td>36</td>
<td>12</td>
<td>39</td>
<td>19</td>
<td>32</td>
</tr>
<tr>
<td>Coal combustion</td>
<td>10</td>
<td>15</td>
<td>5</td>
<td>17</td>
<td>10</td>
<td>17</td>
</tr>
<tr>
<td>Oil</td>
<td>5</td>
<td>8</td>
<td>3</td>
<td>8</td>
<td>5</td>
<td>8</td>
</tr>
</tbody>
</table>
Table 4. Summary statistics for PM$_{2.5}$, BC and elemental concentrations

<table>
<thead>
<tr>
<th>ng m$^{-3}$</th>
<th>Average</th>
<th>S.D</th>
<th>Median</th>
<th>Max.</th>
<th>Min.</th>
<th># Samples &gt; MDL</th>
<th>Warm</th>
<th>Cold</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mass</td>
<td>92800</td>
<td>9520</td>
<td>72700</td>
<td>70000</td>
<td>2770</td>
<td>184</td>
<td>77300</td>
<td>10800</td>
</tr>
<tr>
<td>BC</td>
<td>6590</td>
<td>6340</td>
<td>4260</td>
<td>50900</td>
<td>1030</td>
<td>184</td>
<td>3120</td>
<td>10100</td>
</tr>
<tr>
<td>Na</td>
<td>391</td>
<td>210</td>
<td>331</td>
<td>1900</td>
<td>69.0</td>
<td>184</td>
<td>304</td>
<td>478</td>
</tr>
<tr>
<td>Mg</td>
<td>251</td>
<td>178</td>
<td>218</td>
<td>1600</td>
<td>91.0</td>
<td>157</td>
<td>210</td>
<td>299</td>
</tr>
<tr>
<td>Al</td>
<td>737</td>
<td>470</td>
<td>588</td>
<td>2292</td>
<td>133</td>
<td>184</td>
<td>739</td>
<td>736</td>
</tr>
<tr>
<td>Si</td>
<td>1490</td>
<td>1110</td>
<td>1130</td>
<td>5570</td>
<td>141</td>
<td>184</td>
<td>1550</td>
<td>1430</td>
</tr>
<tr>
<td>S</td>
<td>1740</td>
<td>2600</td>
<td>684</td>
<td>20600</td>
<td>119</td>
<td>184</td>
<td>458</td>
<td>30490</td>
</tr>
<tr>
<td>Cl</td>
<td>43.0</td>
<td>69.0</td>
<td>28.0</td>
<td>817</td>
<td>5.00</td>
<td>182</td>
<td>30.0</td>
<td>56.0</td>
</tr>
<tr>
<td>K</td>
<td>259</td>
<td>155</td>
<td>214</td>
<td>946</td>
<td>23</td>
<td>184</td>
<td>241</td>
<td>276</td>
</tr>
<tr>
<td>Ca</td>
<td>567</td>
<td>390</td>
<td>449</td>
<td>2690</td>
<td>50.0</td>
<td>184</td>
<td>511</td>
<td>626</td>
</tr>
<tr>
<td>Ti</td>
<td>38.0</td>
<td>28.0</td>
<td>30.0</td>
<td>153</td>
<td>4.00</td>
<td>184</td>
<td>40.0</td>
<td>37.0</td>
</tr>
<tr>
<td>V</td>
<td>2.00</td>
<td>1.00</td>
<td>2.00</td>
<td>6.00</td>
<td>2.00</td>
<td>70</td>
<td>2.00</td>
<td>3.00</td>
</tr>
<tr>
<td>Cr</td>
<td>5.00</td>
<td>5.00</td>
<td>4.00</td>
<td>37.0</td>
<td>2.00</td>
<td>173</td>
<td>3.00</td>
<td>6.00</td>
</tr>
<tr>
<td>Mn</td>
<td>18.0</td>
<td>10.0</td>
<td>15.0</td>
<td>51.0</td>
<td>4.0</td>
<td>183</td>
<td>16.0</td>
<td>21.0</td>
</tr>
<tr>
<td>Fe</td>
<td>455</td>
<td>312</td>
<td>350</td>
<td>1950</td>
<td>34.0</td>
<td>184</td>
<td>431</td>
<td>479</td>
</tr>
<tr>
<td>Ni</td>
<td>3.00</td>
<td>1.00</td>
<td>2.00</td>
<td>8.00</td>
<td>2.00</td>
<td>128</td>
<td>2.00</td>
<td>3.00</td>
</tr>
<tr>
<td>Cu</td>
<td>5.00</td>
<td>10.0</td>
<td>3.00</td>
<td>126</td>
<td>2.00</td>
<td>166</td>
<td>5.00</td>
<td>6.00</td>
</tr>
<tr>
<td>Zn</td>
<td>45.0</td>
<td>41.0</td>
<td>30.0</td>
<td>260</td>
<td>2.00</td>
<td>184</td>
<td>29.0</td>
<td>61.0</td>
</tr>
<tr>
<td>As</td>
<td>28.0</td>
<td>38.0</td>
<td>18.0</td>
<td>394</td>
<td>7.00</td>
<td>183</td>
<td>15.0</td>
<td>42.0</td>
</tr>
<tr>
<td>Pb</td>
<td>11.0</td>
<td>9.00</td>
<td>8.00</td>
<td>53.0</td>
<td>5.00</td>
<td>87</td>
<td>5.00</td>
<td>12.0</td>
</tr>
</tbody>
</table>
Table 5. Yearly source contributions to PM$_{2.5}$ by average mass and percentage

<table>
<thead>
<tr>
<th>Sources</th>
<th>2014 Average ($\mu$g m$^{-3}$)</th>
<th>%</th>
<th>2015 Average ($\mu$g m$^{-3}$)</th>
<th>%</th>
<th>2016 Average ($\mu$g m$^{-3}$)</th>
<th>%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Soil</td>
<td>12</td>
<td>31</td>
<td>16</td>
<td>40</td>
<td>14</td>
<td>28</td>
</tr>
<tr>
<td>Traffic</td>
<td>14</td>
<td>36</td>
<td>11</td>
<td>28</td>
<td>14</td>
<td>29</td>
</tr>
<tr>
<td>Coal combustion</td>
<td>8</td>
<td>21</td>
<td>8</td>
<td>19</td>
<td>17</td>
<td>34</td>
</tr>
<tr>
<td>Oil</td>
<td>4</td>
<td>12</td>
<td>5</td>
<td>14</td>
<td>4</td>
<td>9</td>
</tr>
</tbody>
</table>