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Particle size distributions in an office environment: An experimental approach and a source apportionment analysis through positive matrix factorization.

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ABSTRACT

The identification of particulate matter sources is an important step in the development of indoor air quality control strategies. Advanced modelling techniques, termed as receptor modelling, are used to identify the presence, determine the sources of aerosols and most importantly quantify the source contributions. This study presents a source apportionment analysis, supported by targeted experimental campaign, to identify the main pollution sources in an office indoor environment. For the purpose of the experimental campaign, continuous monitoring of particle (0.3 - > 20.0 μm in aerodynamic diameter) number concentrations took place during the period between 16th and 27th July 2007 in two office environments, in Athens, Greece. A portable dust monitor (GRIMM model 1.108) was used to measure in real-time the particles size distribution in 16 different size channels. Two adjacent offices -with similar characteristics except for the presence of smokers- were used. Additionally, the contribution of other possible sources (e.g. photocopy machine, air conditioner, penetration and outdoor environment) was investigated. The indoor temperature, relative humidity and light intensity levels were also recorded in a continuous basis. Finally, all activities were daily recorded in detail on a logbook. The positive matrix factorization (PMF) method has been applied on the data collected during the experimental campaign for source apportionment analysis. The output of the analysis has been related with source information provided by the occupants and other source profiles by bibliography. This study aims to elicit indoor source characteristics based on particles size distribution data, especially in situations where the indoor sources are unknown beforehand.

Keywords: particulate matter; size distribution; positive matrix factorization.

1. INTRODUCTION

Several studies underline the role of good indoor air quality, since an average person spends more than 80% of the day in an indoor environment (Robinson and Nelson, 1995) Indoor air quality depends on several factors e.g. outdoor concentrations, indoor sources, ventilation, building design etc. These factors play different roles, depending on the kind of the indoor environment (Hameri et al., 2003). For an occupied building, indoor particle sources such as smoking and resuspension from clothes etc. are often prevalent while outdoor particles are often from both mobile and stationary (natural or anthropogenic) sources. Each of these particles sources produces different size distributions that may result

in studying the mortality and morbidity effects of airborne particulate matter (PM) because they will have different penetrability and deposition patterns.

The investigation of chemical pollutants in the air of public utility buildings, including offices has been the aim of a lot of studies for several years (Brown 1999, Jankowska et al., 2004). Smoking seems to be an important determinant of personal exposures in such an indoor environment as PM and Volatile Organic Compounds (VOC) levels have been found even 2 times higher among a tobacco-smoke exposed working group compared to a non-smoke exposed group (Lai et al., 2004). Factors related to the use of printers and photocopy machines (Guo et al., 2003), to the building and constructing materials (Pellizari et al., 1987), to outdoor sources etc have been examined in the atmosphere of an office environment. Fine particles which were found in office buildings and other non-residential buildings were most of outdoor origin (Ogulei et al., 2006 [a]). When low or no activity was present in the building, such as when the house was unoccupied, Kopperud et al., 2004 observed that up to 60 % of the observed PM_{2.5} originated indoors.

The identification of air pollution sources is an important step in the development of air quality control strategies. Abatement strategies may significantly improve the air quality after identifying the main aerosol sources. *Receptor modelling* has been used to determine sources of aerosols in ambient or indoor environments. Receptor models *identify the presence and quantify of source contributions* to the receptor.

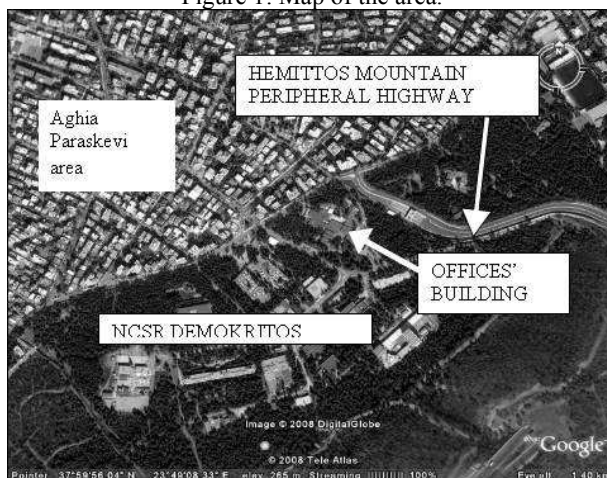
The fundamental principle of receptor modelling is based on the assumption that *mass is conserved* and that on this basis a mass balance analysis can be used to identify and apportion sources in the atmosphere. All source apportionment (receptor modelling) techniques are based on assumptions regarding the source the chemical or physical characteristics (e.g. chemical composition and size distribution) and the measurement methodology. Some of the methods require a certain degree of knowledge about the number of sources, the sources profiles and the source strength regardless the origin of the source (outdoors or indoors).

It is the purpose of this study to present the results of Positive Matrix Factorization (PMF) method application on the data collected during an experimental campaign for source apportionment analysis. The output of the analysis has been related with source information provided by the occupants and other source profiles by bibliography. This study aims to elicit indoor source characteristics based on particles size distribution data, especially in situations where the indoor sources are unknown beforehand.

2. DESCRIPTION OF THE FIELD CAMPAIGN

For the purpose of the study, two offices with similar characteristics were employed. The two offices are located in the same building, in NCSR "Demokritos" which is situated in Aghia Paraskevi, a north-eastern suburb of Athens (fig.1). The experimental campaign took place between 16th and 27th July 2007. As it has been reported, the kind of building and furniture materials, the equipment and the external environment is similar and the major factor which differentiates the two environments is smoking activity. In the first office, where the smoking is permitted, there are two employees; a smoker and a passive smoker. The second office is used by two, non-smoker employees and smoking activity is not allowed. Both offices have an air conditioning system, two computers, a printer and a photocopy machine. It has to be mentioned that both offices are daily open to public, while during weekend no activity is occurred. Important information is also that all employees were following the same working time schedule, as recorded in questionnaires.

Figure 1: Map of the area.



2.1 Experimental results

For the particle number measurements/size range an automatic portable aerosol spectrometer (GRIMM 1.108) was used in the offices. This unit is based on the principle of light scattering and can give the size distribution of dust particles in $\mu\text{g}/\text{m}^3$ or in counts/L.

Regarding particle concentration (counts/L) measurements, a different picture is observed between the two offices. For the smokers' office, the number of smaller particles (with diameter less than $0.40 \mu\text{m}$) reached the value of 6,439,100 particles per litre when smoking activity occurred and windows were kept closed. During hours with open windows and the same intensity of smoking activity, the number of these particles was lower (1,084,730 particles/L). Furthermore, in the early morning hours (before 8.00am) the average concentration was 63,111 particles/L while in the afternoon hours -and after the occupants' leaving- the concentration was 94,287, following a decreasing trend during night. During weekend, the size distribution of the number particles concentration presents a different picture, possibly due to the absence of the main anthropogenic sources. In particular, the concentration of the smaller particles ($<0.40 \mu\text{m}$) presented a peak two orders of magnitude lower than this in weekdays (87,510 particles/L) and did not follow a variation because of the stable activity and ventilation conditions.

In non-smokers' office, smaller particles' number concentration during occupants' absence was in average, 90,920 particles/L (background). During working hours, concentration reached the value of 449,379 particles/L but significantly decreased when windows were open (190,150 particles/L). The same picture as in smokers' office was noticed for larger in size particles which were much fewer in number, reaching the order of 10 particles/L (for particles $>20 \mu\text{m}$).

3 PMF & data handling

All data obtained by GRIMM were averaged to 5 min resolution so as to eliminate wide changes in concentrations that could severely distort results of any apportionment study. As size distributions from a specific source are expected to remain reasonably stable over a short time interval, averaging the number or volume concentrations in this way was not expected to considerably affect the outcome of this study (Ogulei et al., 2006).

In this study, PMF was used with the GRIMM data. PMF is a new variant factor analysis method and is described in detail by Paatero (1997). Only a brief description of the technique is given here. PMF uses a weighted least-squares fit with the known error estimates of the elements of the data matrix used to derive the weights. The factor model (PMF2) can be written as

$$X=GF+E, \quad (1)$$

where X is the known $n \times m$ matrix of the m measured chemical species in n samples. G is an $n \times p$ matrix of source contributions to the samples (time variations). F is a $p \times m$ matrix of source compositions (source profiles). Both G and F are factor matrices to be determined. E is defined as a residual matrix, i.e., the difference between the measurement X and the model Y as a function of factors G and F .

$$e_{ij} = x_{ij} - y_{ij} = x_{ij} - \sum_{k=1}^p g_{ik} f_{kj} \quad (i = 1, \dots, n; j = 1, \dots, m; k = 1, \dots, p). \quad (2)$$

The objective of PMF is to minimize the sum of the squares of the residuals weighted inversely with error estimates of the data points. Furthermore, PMF constrains all of the elements of G and F to be non-negative; meaning that sources cannot have negative species concentration ($f_{kj} \geq 0$) and sample cannot have a negative source contribution ($g_{ik} \geq 0$). The task of PMF analysis can thus be described as to minimize Q , which is defined as

$$Q(E) = \sum_{i=1}^n \sum_{j=1}^m (e_{ij}/s_{ij})^2 \quad (3)$$

with $f_{kj} \geq 0$; $g_{ik} \geq 0$ and s_{ij} is the error estimate for x_{ij} . The solution of Eq. (3) is obtained by a unique algorithm PMF2 in which both matrices, G and F , are adjusted in each iteration step. The process continues until convergence (Paatero 1997).

A critical step in PMF analysis is determination of the number of factors. It is a fact that choosing too few factors may lead to non-well separated sources, whereas too many factors may essentially lead to split up of a true source into two or more non-existing sources.

The primary consideration is basically to obtain a good fit of the model to the original data. The theoretical Q -value should be approximately equal to the number of degrees of freedom, or approximately equal to the number of entries of data array, provided that correct values of s_{ij} have been used in equation 3. Often there is no reliable information on s_{ij} and the presence of outliers (even in the robust mode) complicates the situation. Then it may be impossible to determine whether the observed value of Q is normal or too large. It is helpful to examine the distributions of scaled residuals (e_{ij}/s_{ij}). In a well-fit model, the residuals e_{ij} and the error estimates s_{ij} should be about equal and the ratios e_{ij}/s_{ij} should fluctuate between ± 2 (Juntto and Paatero, 1994).

4. RESULTS AND DISCUSSION

In the frame of this study, one day's PMF application results are presented as six factors were successfully identified. The selected day was Thursday, 19th July 2007 where the measurements were conducted in smokers' office. However, information from PMF application results from other days of the experimental campaign was used. In this work, the number of factors that was examined ranged from four to seven, although a number between five and six seems more appropriate. The calculated Q -values obtained from trials with five or six factors did not show that either was much better than the other.

The final number of factors, and therefore existing sources, were identified by relating the obtained source contributions to the source information provided by the occupants (logbook with detailed activities). The subjective analysis together with the application of the robust PMF mode and with rotation resulted in six factors for all experimental days. The *source contributions* and *time series concentrations* are presented in Table1 and figures 2a- f, respectively.

Table 1. Percentage contribution of particle channel to each Factor

Channel	Size range (μm)	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6	sum
1	0.30-0.40	4.6	10.2	9.8	5.7	1.1	68.6	100%
2	0.40-0.50	5.4	11.5	5.1	25.3	0.5	52.4	100%
3	0.50-0.65	4.0	19.0	1.8	39.4	1.8	33.9	100%
4	0.65-0.80	0.9	35.0	5.1	29.8	6.8	22.3	100%
5	0.80-1.0	0.0	44.7	13.0	8.9	13.1	20.3	100%
6	1.0-1.6	25.9	33.8	27.6	0.6	0.0	12.1	100%
7	1.6-2.0	0.0	24.4	45.1	1.6	21.1	7.8	100%
8	2.0-3.0	23.0	6.0	47.3	0.0	21.5	2.2	100%
9	3.0-4.0	29.4	0.6	28.8	1.1	40.0	0.0	100%
10	4.0-5.0	24.8	0.0	20.5	0.0	53.4	1.4	100%
11	5.0-7.5	21.0	0.5	8.8	1.9	67.4	0.5	100%
12	7.5-10	72.5	0.0	0.0	0.0	24.4	3.1	100%
13	10.0-15.0	84.0	1.3	12.8	0.0	0.0	1.9	100%
14	15.0-20.0	68.8	0.0	0.0	11.8	19.4	0.0	100%

Factor 6 has been identified as **tobacco smoking**. This factor has its strongest contribution at smaller particles (0.30-0.40 μm and 0.40-0.50 μm), that is in ultra fine particles as expected (He et al., 2004). Time series concentration shows that this factor exists only during working hours and presents picks which match with the periods that smoking activity actually occurred, as recorded in the logbook. Additionally, concentration picks seem to decrease with low rate because cigarette smoke tends to remain even for few hours after cigarette smoking (Halios et al., 2005).

Factor 3 has a number mode at 2.0-3.0 μm and is suggested to represent **primary PM emissions from office equipment and furniture**. It is remarkable that this factor remains almost constant; presenting a slight fluctuation during day and night, a fact that amplifies the above suggestion. As it can be noticed, a more intense fluctuation can be observed during working hours, where human activities occurred and air streams could affect concentration at the receptor site.

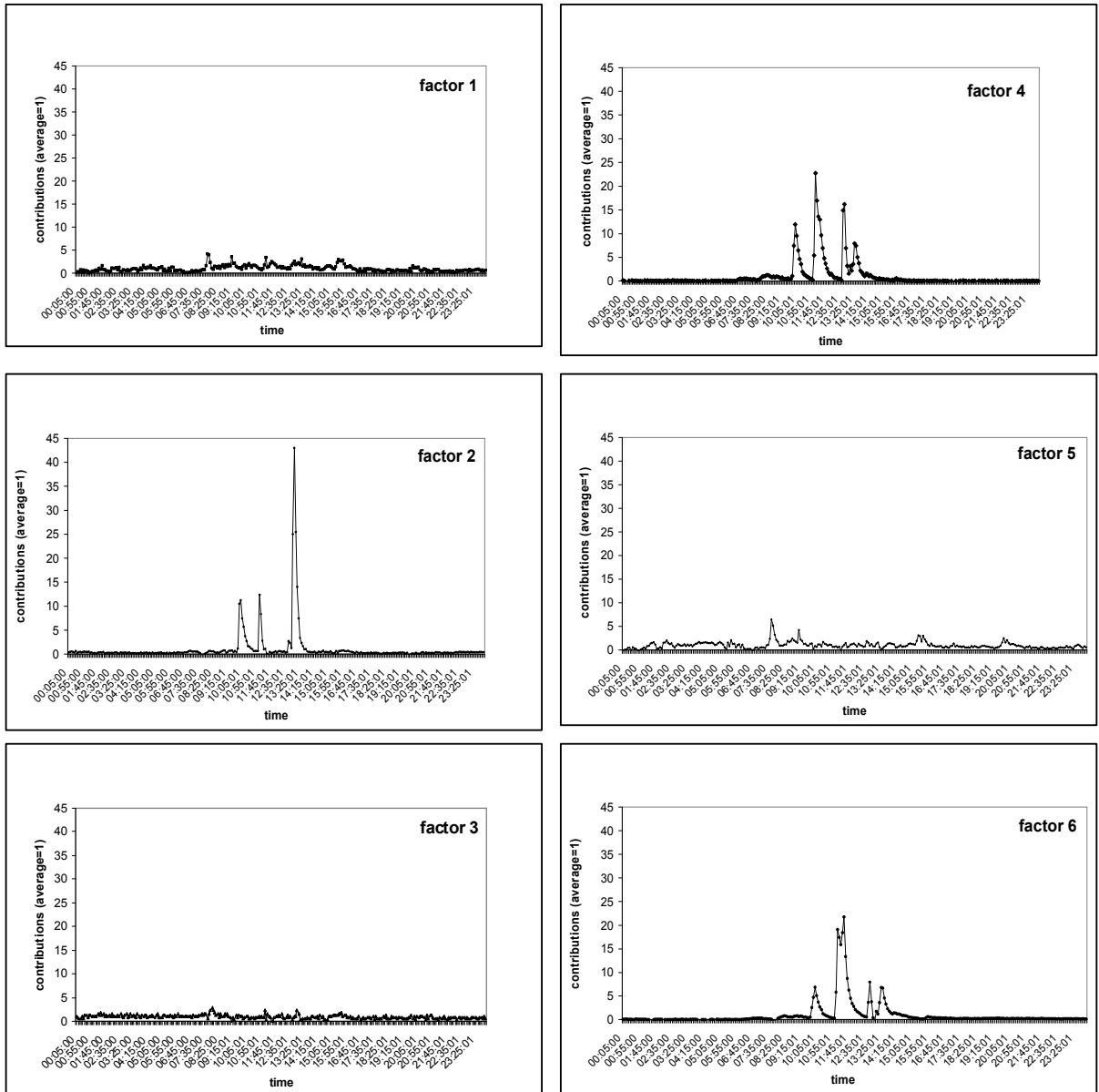
Factor 2 has its strongest contribution at particle size range of 0.80-1.0 μm . Time series concentration presents picks during working hours that do not match in number and time with the real cigarettes smoked. This fact, in combination with information received from the other experimental days should lead to another factor. Actually, in the same office, during weekend (when no human activity occurred) a test of air conditioner 12-hour function took place. PMF results (not shown here) presented a new factor, which has a number mode at 0.80-1.0 μm , too. Furthermore, similar picks were found during this 12-hour interval. Taking all the above into consideration, we can assume that Factor 2 is associated with **air conditioner emissions**, as its presence match in time with the air conditioner function as recorded in questionnaires.

Factors 1 and 5 are suggested to represent **outdoor sources** e.g. vehicles exhausts emissions, construction activities, pine trees etc as they are associated with larger particles, 10-15.0 μm for factor 1 and 5.0-7.5 μm for factor 2. For both factors, time series present small fluctuations during all day except for the period of working hours, when opening of windows occurred several times and air exchange took place. Outdoor environment contribution could possibly happen either by air exchanging through the window or by infiltration (cracks, small openings in doors and windows etc) or by the combination of all these. It is noticeable that these factors appeared in all days as expected, since outdoor environment is a constant source. Especially for factor 1 (major contribution at the size

range of 10-15 μm), an additional observation amplifies the assumption that it is associated with vehicles emissions, possibly from Hemittos mountain peripheral highway (see fig. 1), an avenue in short distance to the building (100m). To be more specific, time series of this factor during all days suggests a frequent activity from 6.30am to 11.00pm that is the hours that vehicles circulation occurs. Furthermore, peaks are presented during rush-hours, even on evening indicating the external origin of the source.

Factor 4 presents its major contribution at the size range of 0.50-0.65 μm and can attributed to **penetration**. In particular, it can be associated with particles originated outdoors with have been transferred and remained indoors. Penetration sources can mainly associated with accumulation mode particles probably because those particles can persist in the air since they are too small for inertial deposition and too big for diffusive removal processes and, hence, are capable of penetrating into buildings and remaining airborne for longer periods of time. Otherwise, this factor can be associated with the **unapportioned** PM fraction which represents the fraction that could not be resolved by the model. It is possible that this fraction may represent background levels and/or other unresolved sources.

Figures 2 a- f Time series concentration (19/7/2007)



5. CONCLUSIONS

PMF method has been successfully applied on the PM data collected during the experimental campaign in an office environment for source apportionment analysis. Six size distribution profiles were resolved, using information kept in log books by the occupants. In general, the profiles identified are in agreement with literature references. The six profiles included tobacco smoking, office equipment emissions, air conditioner emissions, penetration, vehicle exhausts emissions, other outdoor sources.

In a source apportionment study one should take under consideration some errors that can be the result of several critical problems. In a study like this, someone must be very careful about the information collected on the logbook because either participants can very subjectively estimate the duration of smoking/other activities or minutes of smoking can not be a good indicator for PM emission from smoking (Yakovleva et al., 1999).

Finally, another source apportionment method (e.g. chemical mass balance method) using results from the chemical analysis of particles collected in such an experimental campaign could provide an additional aspect of the study. As reported (Ogulei et al., 2006 [b]), once particles are emitted from a given source, their size, number, and chemical composition change by several mechanisms until they are ultimately removed by natural processes. However, the particle size distribution would be expected to remain approximately stationary at some appropriate distance from the emission source. Thus, properties of the observed size distribution at the receptor site can be used along with chemical composition information to identify the emission sources. This approach has been recently explored by Zhou et al., 2005.

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