

# SOURCE APPORTIONMENT OF THE ATMOSPHERIC AEROSOL IN THE METROPOLITAN AREA OF SÃO PAULO: A CASE OF STUDY FOR WINTER OF 2003.

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## ABSTRACT

The main purpose of this study was to determine the characteristics of the atmospheric aerosol in the central area of Sao Paulo city, part of the Metropolitan Area of Sao Paulo (MASP), classified as a mega-city. The results presented here are concentrated in the source identification and apportion of fine and coarse mode. Size resolved aerosol mass and chemical characterization were performed in an aerosol sampling campaign during the winter of 2003 (Jul, 07 – Sep, 05) in downtown MASP, characterized by vehicular emissions of light duty fleet (LDV) and heavy duty fleet (HDV) compose of buses and trucks. The concentration of PM<sub>10</sub> (particles with aerodynamic diameter less than 10 µm) usually presents values that are higher than the national standard for average 24-h PM<sub>10</sub> concentration for the air quality stations localized in the metropolitan area. The size distribution concentration was determined by samplings with a cascade impactor type MOUDI, with nine stages and a MiniVol sampler that collects the coarse and fine mode aerosol. The material collected by the two equipments was analyzed through gravimetric procedures, reflectance (for Black Carbon determination), and the PIXE (Particle Induced X-ray Emission) technique (for elemental composition determination). As the processes associated to aerosol are mainly related to its size, an important property to be discussed is the Size Distribution of Particle Number (SDN) and Size Distribution of Mass (SDM) functions. Using the data collected by MD, it was possible to characterize the SDM, and a SMPS (Scanning Mobility Particle Sizer, manufactured by TSI) was used to obtain the SDN, which provides the particle number concentration for size intervals. This paper reports quantitative source apportionment calculations for winter aerosols collected in the downtown area of the MASP. Ambient samples of particles were influenced by local emissions more than by long-range transport pollution. Various sources were identified by a sequential principal component analysis as the resuspended soil particles, oil combustion and emissions from traffic. The resuspended soil dust accounted for a large fraction of the coarse mode aerosol mass. Vehicular associated emissions and urban soil dust are the main air pollution sources in São Paulo Mega city area.

Keyword(s): São Paulo atmospheric aerosol; source apportionment.

## 1. Introduction

Aerosol plays a critical role in many processes which impact on our lives either indirectly (climate) or directly (health). However, our ability to assess these possible impacts is constrained by our limited knowledge of the physical and chemical properties of aerosols, both anthropogenic and natural (Prospero et al., 1983; Schwartz, 1994; Spektor et al., 1991). Aerosols can be derived from several sources, like human activities, photochemical processes, gas-to-particle conversion, eolic erosion, oceans, etc. Therefore, there is a considerable variation in the composition and physical properties of the particles, depending on the sources, and also on the meteorological conditions and geographical localization (Miranda et al., 2002).

The Metropolitan Area of São Paulo (MASP) is the third largest metropolitan conglomerate of the world, with about 17million inhabitants. It is

the most industrialized and developed region of South America, representing 18.5% of the Brazilian GNP (Gross National Product). São Paulo city is home of one of the largest urban motor vehicles fleet (7 millions automobiles) of the world (CETESB, 2003). It is located in a plateau at about 860m above sea level and is surrounded by hills of about 1200m in height. Frequent subsidence and thermal inversion layers provide unfavorable conditions for the dispersion of pollutants during most of the winter season. The strong emissions of trace gases and aerosol particles by vehicles, industry and resuspended soil dust, together with secondary aerosol, coupled with the unfavorable natural conditions of dispersion, are responsible for the high concentrations of pollutants observed in the São Paulo metropolitan area (Artaxo and Castanho, 1998).

During the winter (dry) season, average concentrations of inhalable particles in the MASP can be as high as 150 µg/m<sup>3</sup> (the national standard for average 24-h PM<sub>10</sub> concentration). The main source of pollutants is the fleet of light

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and heavy-duty vehicles, and is responsible for the direct emission of 40% of inhalable particulate matter less than 10 $\mu$ m in diameter (PM<sub>10</sub>) found in the urban atmosphere. Other major sources are secondary reactions, i.e. gas-to-particle conversion, condensation and coagulation, which contribute with 25% of local PM<sub>10</sub> emissions. Resuspension of soil and dust generates 25% of these particles and the other 10% is attributable to industrial sources (CETESB, 2003). Meteorological characteristics related to the height of boundary layer and ozone concentrations on days with high heatstroke play an important role in the occurrence of high PM<sub>10</sub> concentrations during the winter season.

Interpretations of results on metal chemical forms are more difficult than interpretation on their total content. These difficulties are softened by using multivariate analysis with the data of many chemical form concentrations, a good approach to identify sources of atmospheric pollution in urban cities (Espinosa et al., 2004). Several previous works have been done studying air pollution in São Paulo, where the particulate matter has been characterized and sources of pollutants have been identified by receptor models (Andrade et al., 1994; Artaxo et al., 1995; Artaxo and Castanho, 1998; Castanho and Artaxo, 2001; Orsini et al., 1984, 1986).

This paper reports quantitative source apportionment calculations for winter aerosols collected in the downtown area of the MASP. Fine and coarse mode aerosol particles were collected and analyzed for mass, black carbon (BC) and up to 20 trace elements using particle induced X-ray emission analysis. Sources from the sampling site were influenced by local emissions more than by long-range transport pollution. Various sources were identified by a sequential principal component analysis as the resuspended soil particles, oil combustion and emissions from traffic.

## 2. Experimental

An aerosol sampling campaign was performed during the winter of 2003 (Jul 07 – Sep 05) in the Metropolitan Area of São Paulo (MASP). The following equipments were used: a cascade impactor type MOUDI (Micro Orifice Uniform Deposit Impactor, Marple et al., 1986), which allows the determination of aerosol mass concentration deposited in different sizes distributions ( $d_{50\%}$ : 18, 10, 5.6, 3.2, 1.8, 1.0, 0.56, 0.32, 0.10  $\mu$ m), and a Mini Vol (MV), which

collects the coarse ( $2.5 < d_a < 10 \mu$ m) and fine mode aerosol ( $d_a < 2.5 \mu$ m).

The collected samples and appropriate blanks were analyzed by gravimetric procedures, reflectance (for Black Carbon determination), and PIXE at the Physics Institute, University of São Paulo. The filters were irradiated with a 2.4MeV proton beam, and the current was typically 20nA. X-ray spectra were accumulated for 600 s. The following elements were determined: Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Zn, Ga, Br, Zr and Pb. The Cr concentrations rarely exceeded the background level of the Nuclepore filters. The spectra generated by PIXE were reduced with the software AXIL (Van Espen et al., 1986).

This paper reports quantitative source apportionment calculations for winter aerosols collected in the downtown area of the MASP. Sources from the sampling site were influenced by local emissions more than by transferred pollution. Various sources were identified by a sequential principal component analysis as the resuspended soil particles, oil combustion and emissions from traffic.

The sampling sites and meteorological station locations in the city are indicated in the figure 1. The region is characterized by heavy traffic and the local activity is mainly commercial. In this figure is also indicated the climatological station from the Institute of Astronomy, Geophysics and Atmospheric Sciences (IAG-AF) that provided the meteorological data for the analysis.

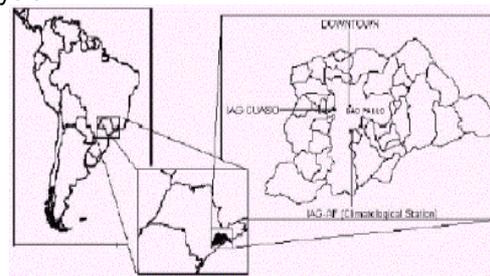


Figure 1 – Metropolitan Area of São Paulo (MASP), aerosol sampling site (Downtown) and the climatological station (IAG-AF).

## 3. Statistical methods

### 3.1. Factor analysis

In this work factor analysis (FA) was applied to evaluate and to identify fine and coarse polluting sources in the MASP in continuation to the work developed by other investigators (Orsini et al., 1986; Andrade et al., 1994; Ccyollo and

Andrade, 2002; Castanho and Artaxo, 2001). FA is a statistical technique that can be applied to a set of variables to reduce its dimensionality. That is, to replace a large set of inter-related variables with a small number of independent variables. This reduction is achieved by transforming the data set into a new set of variables, the factors, which are orthogonal (non-correlated) and are arranged in decreasing order of importance (Poissant et al., 1996). These factors are later associated to the sources of pollutants. The factor analysis model may be written explicitly for the value of variable  $j$  for the value of individual  $i$  as follows:

$$Z_{ji} = \sum_{p=1}^m a_{jp} F_{pi} + d_j U_{ji}$$

$(i = 1, 2, \dots, N; j = 1, 2, \dots, n; m = 1, 2, \dots, m)$

In this expression  $Z_{ji}$  is called a variable  $Z_j$  in standard form, being the variance of  $Z_j$  unity.  $F_{pi}$  is the value of a common factor  $p$  for an individual  $i$  (in this case a sampling of particulate),  $N$  is the total number of individuals (samples),  $n$  is the number of observed variables (pollutants) and  $m$  is the number of latent variables. Each of the  $m$  terms  $a_{jp}F_{pi}$  represents the contribution of the corresponding factor to the linear composition, while  $d_j U_{ji}$  is the "residual error" in the theoretical representation of the observed measurement  $z_{ji}$ . Typically, factor analysis is used when the number of latent variables ( $m$ ) is less than the number of observed variables ( $n$ ) (Harman, 1976; Kessler et al., 1992).

Receptor model methodologies are based on statistical multivariate analysis to infer source contributions at the receptor sites (Henry, 1991; Puri et al., 1996). Absolute principal factor analysis (APFA) was applied to identify the absolute source profiles and quantify the source apportionment (Thurston and Spengler, 1985). The APFA model is based on the eigenvalue and eigenvector analysis of the correlation matrix of the normalized elemental concentration. The aim is to obtain a reduced number of factors that are independent and uncorrelated that could explain most of the original data variability. The factor-loading matrix represents the association between the trace elements and each factor obtained. The factor-scores are an indication of the strength of each sample to each retained factor. The percentage of variance explained for each variable can be used to test the effectiveness of the factor model

to that particular variable. Also the total variance explained by the model can be used as a check if the number of factors and their composition is adequate to the original database (Hopke, 1991).

### 3.2. Source contributions

After the application of factor analysis to the species concentrations and the meteorological data, the absolute profiles for each component for each considered event from July 7 to September 5 of 2003 were computed using the method described by Harman (1976) and Thurston and Spengler (1985). These absolute rotated principal components scores, which are measures for the contributions from each source to each aerosol sample, may be written as follows:

$$g = (B' B)^{-1} B' Z$$

Where  $g$  is the absolute rotated principal component scores,  $B$  is the final matrix for the rotated component solution,  $B'$  is the inverse final matrix for the rotated component solution and  $Z$  is the standard form matrix. The impact of the meteorological parameters on the pollution sources have been evaluated by means of the correlation analysis between the absolute principal component scores of the trace species and meteorological rotated factor matrix.

### 4. Results and discussion

The results of Factor analysis application to the elemental data concentration were generated through the use of STATISTICA package. The trace elements were determined by the PIXE analysis. The VARIMAX rotation technique was applied to analyze the trace-elements concentration variance structure. This is a wide-used methodology to extract information regarding the air pollution sources (Astolfo, 2000; Watson et al., 2002; Espinosa et al., 2004; Zhou et al., 2004).

The results of the Factor Analysis with VARIMAX rotation applied to the Fine and Coarse mode, collected with the MiniVol sampler, are presented in Table 1 and 2. In these tables are presented the factor loadings, the eigenvalues of the factors, the total explained variance and the communality of each element, indicating the total of variability explained by each factor (Andrade, 1993). A descriptive statistics is presented related to the elements analyzed. The analysis was performed

in 69 fine and coarse samples considering the day and night samples together, with the objective of improving the quality of the statistical analysis.

The results for the fine mode analyzed by the Minivol sampler are presented in table 1. Four factors explained 86.4% of data variance, with high communality for all the factors. The elements in red are indicating which are more significant and can be considered tracers of the sources.

The first factor is related to the vehicular emission due to the high loadings of Cu, BC and FP. BC is the product of burning of fossil fuel and is the main tracer of diesel emission (Castanho, 1999). The presence in this factor, also of K and Ti indicate the possibility that this factor is related to the burning of biomass.

The second factor with high loadings for S, V and Ni elements is related to the residual oil burning emissions, as pointed out by many authors, Kowalczyk et al. (1982), Ito et al. (1986), Andrade et al. (1994), Miranda et al. (1996), Ccyollo (2002), Espinosa et al. (2004).

Most of the variability in Factor 3 is explained by Zn, Se and Pb, associated to the industrial emissions and to the burning of wood, associated to the presence of Se, in this factor.

The elements representative of soil dust emissions, Ca, Mn and Fe presented high loadings in Factor 4, allowing the association of this factor with the resuspension of soil dust.

The results of the same analysis performed to the coarse mode concentrations are presented in Table 2. Three factor were retained which explained 84.1% of the data variance. All the elements in this analysis presented high communality. The first factor is related to the soil resuspension due to the presence in this factor of high loadings for Si, Ca, Ti, Mn, Fe, Sr and Z, and with some tracers of burning fuel and biomass (K and V).

The second factor represents the industrial sources (Ni, Zn, Pb), vehicular emission sources (Cu e BC) and the burning of biomass (Se). In the third component are presented the elements that are marine spray emission representative (as Br, S and Cl).

Table 1: Factor Analysis with VARIMAX rotation, descriptive statistics and communality ( $h^2$ ) related to the fine mode elemental composition.

Variables	Average	Minimum	Maximum	Standard Deviation	Factor 1	Factor 2	Factor 3	Factor 4	$h^2$
Si	169,4	0,0	554,3	106,5	0,46	-0,02	0,00	0,67	0,79
S	1116,3	143,7	3573,2	795,9	0,03	<b>0,93</b>	0,00	0,11	0,81
K	275,6	0,0	889,3	193,8	<b>0,81</b>	0,15	0,17	0,47	0,92
Ca	58,8	0,0	153,9	37,8	0,26	-0,04	0,29	<b>0,84</b>	0,83
Ti	12,0	0,0	51,5	9,5	<b>0,71</b>	-0,07	0,07	0,59	0,84
V	2,9	0,0	17,2	3,7	0,23	<b>0,89</b>	0,23	-0,01	0,90
Mn	8,7	0,0	28,0	5,9	0,09	0,33	0,35	<b>0,81</b>	0,83
Fe	247,8	0,0	709,4	145,1	0,46	0,31	0,14	<b>0,73</b>	0,87
Ni	3,0	0,0	14,2	3,3	0,11	<b>0,79</b>	0,49	0,16	0,91
Cu	22,1	0,0	138,3	25,7	<b>0,78</b>	0,13	0,45	0,08	0,86
Zn	121,8	5,1	563,5	98,4	0,28	0,28	<b>0,73</b>	0,37	0,76
Se	5,1	0,0	34,8	7,1	0,13	0,12	<b>0,90</b>	0,19	0,82
Pb	51,0	0,0	232,5	52,8	0,53	0,26	<b>0,75</b>	0,12	0,92
BC *	5,4	1,2	14,9	3,2	<b>0,72</b>	0,17	0,37	0,45	0,93
MPF *	30,5	6,5	71,0	12,8	<b>0,72</b>	0,45	0,19	0,35	0,92
EigenValue					<b>3,73</b>	<b>2,91</b>	<b>2,85</b>	<b>3,46</b>	<b>TOTAL</b>
Explained Variance (%)					<b>24,89</b>	<b>19,41</b>	<b>18,99</b>	<b>23,10</b>	<b>86,38</b>

- Concentration of trace elements in  $ng.m^{-3}$ ; BC and MPF in  $\mu g.m^{-3}$ .

Table 2: Factor Analysis with VARIMAX rotation, descriptive statistics and communality ( $h^2$ ) related to the coarse mode elemental composition.

Variables	Average	Minimum	Maximum	Stand. Dev.	Factor 1	Factor 2	Factor 3	$h^2$
Si	1221,8	105,6	3630,3	878,5	<b>0,93</b>	0,28	0,10	0,97
P	13,4	0,0	146,5	22,3	0,55	0,28	0,49	0,73
S	356,1	57,1	1291,1	258,6	0,28	0,50	<b>0,68</b>	0,81
Cl	441,6	0,0	14550,8	1746,7	-0,10	-0,08	<b>0,93</b>	0,84
K	238,2	0,0	1004,6	225,8	<b>0,72</b>	0,56	0,36	0,99
Ca	721,5	52,7	1765,4	485,7	<b>0,92</b>	0,10	0,08	0,90
Ti	104,1	0,0	330,5	84,2	<b>0,93</b>	0,17	0,00	0,97
V	3,7	0,0	18,9	4,2	<b>0,77</b>	<b>0,54</b>	0,09	0,90
Mn	20,6	0,0	79,7	15,8	<b>0,74</b>	0,56	0,11	0,94
Fe	1085,1	0,0	3826,4	856,0	<b>0,88</b>	0,43	0,07	0,99
Ni	6,7	0,0	42,7	9,3	0,31	<b>0,83</b>	0,09	0,83
Cu	31,6	0,0	186,2	39,7	0,44	<b>0,84</b>	0,11	0,93
Zn	129,2	0,0	662,2	140,3	0,39	<b>0,71</b>	0,04	0,67
Se	1,4	0,0	15,3	3,2	0,05	<b>0,89</b>	0,08	0,83
Br	5,3	0,0	92,7	13,2	0,19	0,63	<b>0,55</b>	0,84
Sr	6,4	0,0	17,3	4,3	<b>0,85</b>	0,22	0,14	0,88
Zr	3,3	0,0	16,0	3,6	<b>0,78</b>	0,30	-0,02	0,72
Pb	27,8	0,0	259,1	48,4	0,34	<b>0,89</b>	0,12	0,95
BC *	0,8	0,1	4,1	0,8	0,56	<b>0,75</b>	0,15	0,97
MPG *	32,6	4,9	112,7	21,8	<b>0,78</b>	0,55	0,09	0,96
<b>Eigenvalue</b>					<b>8,26</b>	<b>6,44</b>	<b>2,13</b>	<b>TOTAL</b>
<b>Explained Variance (%)</b>					<b>41,28</b>	<b>32,21</b>	<b>10,63</b>	<b>84,12</b>

- Concentration of trace-elements in  $\text{ng.m}^{-3}$ ; BC and CP in  $\mu\text{g.m}^{-3}$ .

#### 4.1 Absolute Principal Component Analysis

Applying the methodology described in Yamasoe (1994), the absolute source profile was calculated to the fine and coarse concentration data. The results are presented in Figure 2. The vehicular emission source explained 30% of the total fine mode concentration as shown in Figure 2a. This result corroborates other results for the same area, as Ynoue (2004) and Ccyollo (2002). The urban soil dust resuspension is responsible for 22% of the mass concentration and the residual burning is associated to 19% of the fine mode concentration.

The industrial source is responsible for 1% of the mass. The unexplained mass accounted for 29% of the fine mode concentration. In the coarse mode (Figure 2b) the soil resuspension and the vehicular emission represented 68% of the total mass, the industrial source represented 10% and marine spray (3%). Only 19% of the total coarse mode concentration was not explained. These results showed that the application of this technique is more reliability to the coarse mode.

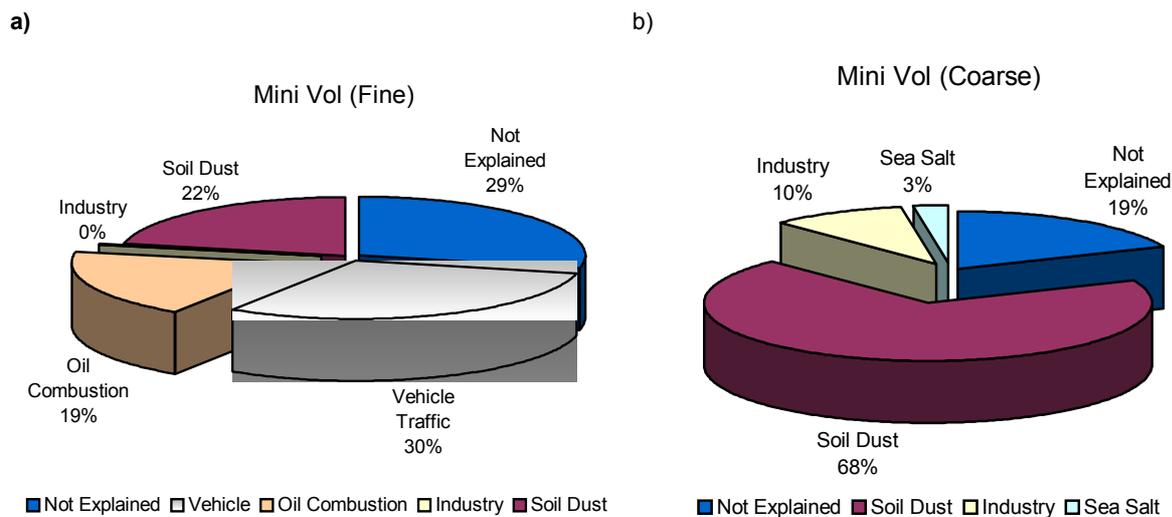


Figure 2: Participation of each source identified to the total fine (a) and coarse (b) mode concentration sampled with the stacked filter unit -MiniVol.

An alternative way of retrieving the absolute profiles of the identified source is the methodology developed by Keiding et al. (1986), and applied by Ccoyllo (2002). With this methodology all the mass is explained by the identified factors.

The results are presented in Table 3, in fraction of mass (ng/ $\mu$ g) of each element for the identified source to the fine and coarse mode. The air pollution sources in São Paulo are mostly associated to the vehicular emissions. It is worthy to point out the importance of Sulfur in the identified sources.

Table 3. Source profiles (in ng/ $\mu$ g) for the identified sources for each trace elements.

Trace Elements	Mini Vol - Fine				Mini Vol - Coarse			
	Vehicle	Oil Combustion	Industry	Soil Dust	Soil Dust	Industry	Sea Salt	
Si	2,79	-0,09	0,01	1,95	29,34	6,08	0,36	
S	1,49	26,05	-0,02	2,48	0,44	0,16	0,04	
K	8,82	1,01	0,48	2,47	2,57	3,22	0,69	
Ca	0,55	-0,06	0,16	0,86	-6,48	-3,54	6,43	
Ti	0,38	-0,02	0,01	0,15	5,87	3,14	0,32	
V	0,05	0,12	0,01	0,00	16,08	1,26	0,15	
Mn	0,03	0,07	0,03	0,13	2,82	0,36	0,00	
Fe	3,77	1,61	0,31	2,89	0,12	0,06	0,00	
Ni	0,02	0,09	0,02	0,01	0,42	0,22	0,01	
Cu	1,13	0,12	0,17	0,06	26,95	9,23	0,24	
Zn	1,53	0,97	1,08	0,99	0,10	0,19	0,00	
Se	0,05	0,03	0,10	0,04	0,62	0,83	0,02	
Pb	1,59	0,48	0,60	0,18	1,94	2,51	0,02	
BC *	0,13	0,02	0,02	0,04	0,01	0,07	0,00	
Sum	22,33	30,39	2,98	12,23	0,09	0,21	0,03	
Percentage %	32,86	44,74	4,39	18,01	0,13	0,02	0,00	
					0,10	0,03	0,00	
					0,59	1,08	0,02	
					0,02	0,02	0,00	
					81,73	25,15	8,34	
					Percentage %	70,93	21,83	7,24

## 5. Conclusions

The principal motivation to this work was to analyze the source profile from fine and coarse concentration measured in downtown São Paulo. Most of the aerosol characterization studies in São Paulo had been performed in the São Paulo University campus, which is not characteristic of a central area.

The sampling site considered in this study was the University Mackenzie Campus (coordinates 23° 32,35' S, 46° 39,08' W), in the period from July 7 to September 5, 2003.

In this abstract it was presented only the results of factor analysis that showed good agreement with data previously analyzed, although the difference in the sampling sites. The results presented here are only a part of the project that considered the determination of size distributions of mass and number of aerosols. The information collected in this experiment was summed to previous works.

According to this work the main source of air pollution in São Paulo is the burning of fuel and other related sources, as the resuspension of soil.

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