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RESEARCH ARTICLE

Comparative Receptor Models Using Principal Component Analysis/Absolute Principal Component Scores and Positive Matrix Factorization to Assess Source Apportionment of PM2.5-10 and PM2.5 in Urban Cities

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Abstract

Objectives: Source apportionment of $PM_{2.5-10}$ and $PM_{2.5}$ was conducted using two receptor models: the principal component analysis/absolute principal component scores (PCA/APCS) and the positive matrix factorization (PMF). **Methods:** The PCA/APCS model resolved four sources, namely, mineral dust, sea salt/secondary sulfur, a mixed source, and traffic and industry emissions. PMF model also resolved four sources of the same origin based on the proportion of each component from the analysis. All models identified the main sources that contribute to $PM_{2.5-10}$ and $PM_{2.5}$ emissions and reconfirmed that the potential sources were the dominants in particulate matter in the Capital city. **Findings:** The first four extracted component accounted for nearly 88.3% of the variability of the data set. The running matrix elements used for data processing within collected air particulate matter was BC, Na, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Rb, Sr, Ba, and Pb. This discrimination of the area of Dakar cites according to the PCA/APCS results confirms the impact that the major activities have on the environment in the most appropriate way, and it verifies the results of previous studies about the influence of Industry emissions, mineral dust, traffic emissions and sea salt/secondary sulfur on the surrounding urban areas. **Novelty:** Continued collection of speciation data at the urban areas will enhance the understanding of local versus regional source contributions for air quality index in sub-Sahara region.

Keywords: Aerosol; Particulate Matter; Source apportionment; receptor Modelling; XRF; heavy metals

1 Introduction

Air pollutants cause adverse effects on human health as demonstrated by epidemiological studies [1–5].In developing countries, particulate matter (PM) has a negative impact on urban ambient air. PM10, with aerodynamic diameters ranging from 2.5 to 10 μm, plays an important role in cloud acidification and visibility [6–9].Notably, epidemiological studies have correlated high PM10 with conditions such as mortality and morbidity [10–12].

In addition, PM2.5, having an aerodynamic diameter of 2.5 μm, causes health effects such as allergies and respiratory, cardiopulmonary, and cardiovascular diseases and even premature mortality [13–20].

The Dichotomous [21], Gent Stacked Filter Unit sampler manufactured by the International Atomic Energy Agency (IAEA) under contract with the University of Gent [22], offers the advantage of collecting fine (PM2.5) and coarse (PM10) fractions simultaneously. In this study, particulate samples were collected from two sites, HLM (industrial area) and Yoff (near the road) in Dakar, using a Gent stacked filter sampler.

Nowadays, the main objectives of atmospheric research are the identification of different sources of particles and their impact on the environment. Many multivariate statistical models, such as chemical mass balance (CMB), factor analysis (FA), principal component analysis (PCA)/absolute principal component scores (APCS), positive matrix factorization (PMF), and UNMIX for source apportionment, are widely used [23–25]. The FA/PCA model can determine the types of pollution sources for each principal component. In this study, multivariate linear regression (MLR) of daily PM concentrations sampled at HLM and Yoff sites and the APCS methods were used to calculate the contribution of each pollution source to the variables. The PCA/APCS model was used to determine the contributions of sources to each pollutant [26]. This model receptor provides quantitative information on source profiles and their contributions. The PCA/APCS model was developed [27–30] for the assessment of water quality indicators.

This study aims to determine the source identification by PCA followed by varimax rotation and quantify the contributions of each PM source by the APCS technique.

2 Methodology

2.1 Sampling and analysis

The sampling was performed in two sites Yoff and Hlm. Yoff (latitude 14°45'14"N, longitude 17°28'04"W) accounts for 89442 inhabitants on a surface of approximately 15 km^2. The sampler is settled on the roadside facing one printer of a factory and 2 km close to the sea. The sampling at Hlm site (latitude 14°42'53"N, longitude 17°26'41"W) was conducted in an area gathering together many small factories. It is considered as an industrial city with 39126 inhabitants in an area of 200 hectares. Figure 1 shows the locations of the sampling sites.

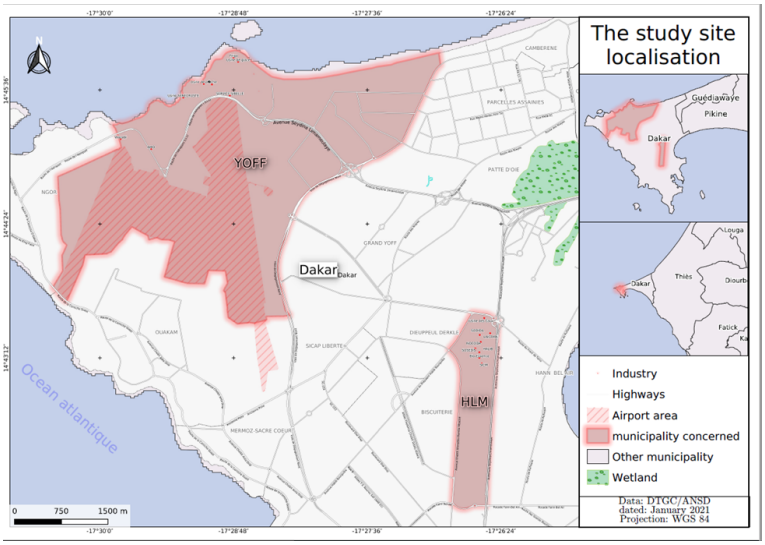

Fig 1. Location of the Hlm and Yoff sampling sites

The sampler was installed in the area on the post made by wood upright in the ground with 2.5 meter high. Air mass could reach the sampler station from all directions undisturbed around, and therefore, the PM collected would be representative in the overall urban area.

The applied sampling protocol and sample analysis methods were based on a study on source apportionment using the PMF model[31]. After sampling, chemical species determination was performed using energy dispersive X-ray fluorescence spectroscopy (ED-XRF, Epsilon 5 from PANalytical, Netherlands)[32].

Table 1. The average concentrations and standard deviation (Stdev) of the different chemical species analysed (μg m^{-3}) in $PM_{2.5-10}$ and $PM_{2.5}$

	BC	Na	Mg	Al	Si	P	S	Cl	K	Ca	Ti	Pb
Mean	0.029	0.828	0.91	1.383	3.334	0.096	0.47	2.227	0.393	5.705	0.22	0.027
Stdev	0.006	0.586	1.344	0.228	0.549	0.029	0.044	0.37	0.037	0.612	0.022	0.014
	V	**Cr**	**Mn**	**Fe**	**Co**	**Ni**	**Cu**	**Zn**	**Br**	**Rb**	**Sr**	**Ba**
Mean	0.008	0.008	0.043	2.228	0.001	0.009	0.027	0.057	0.017	0.006	0.027	0.052
Stdev	0.004	0.003	0.006	0.239	0.0008	0.006	0.006	0.007	0.007	0.007	0.006	0.063

2.2 Principal component analysis (PCA)

The statistical analysis of the collected data was performed using the R studio software with FactomineR and Psych packages. Missing values and values below the detection limit (BDL) of the variables were replaced using the method described by Polissar (1998)[33]. Kaiser's criterion was used to determine the exact number of principal components to maintain[34]. PCA with Varimax rotation was performed to identify the composition of the sources and redistribute the variance to the factors. Typically, the first step of PCA is to normalize the elemental concentrations of the data using the following equation:

$$z_{ij} = \frac{x_{ij} - \bar{x}_i}{s_i} \tag{1}$$

Where:

x_{ij} is the measured concentration of the ith element (i =1... n) in the jth sample (j =1... m);

$\bar{x}_i$ is the mean concentration of element i over all m samples;

s_i is the standard deviation of concentration of element I;

z_{ij} is the normalized concentration of element i in sample j.

The basic mass balance equation that is used by PCA can be written as matrix form:

$$Z = G.F \tag{2}$$

Where p is the number of principal components (aerosol sources);

Z is the n × m matrix of normalized concentrations;

G is the n × p principal component loadings matrix;

F is the p × m matrix of principal component scores

2.3 Absolute principal component scores (APCS)

The first step in APCS is the normalization of all elemental concentrations as z_{ij}according to Equation (1). The second step is to consider an artificial sample with a concentration of zero for all variables. The factor scores for this artificial sample are given by the following equation:

$$(z_0)_i = -\frac{\bar{x}_i}{s_i} \tag{3}$$

The APCS was estimated by subtracting the factor scores of the artificial sample from the factor scores of the real sample. It should be noted that there is a correlation between the principal component scores and the pollution sources impacting the site, which has been demonstrated by Henry et al. (1979)[35]. ^{M}LR was applied to daily PM concentrations as dependent variables and APCS as independent variables to determine the contributions of pollutant sources according to the following equation:

$$C_i = a_0 + \sum APCS_p \times a_{pi} \tag{4}$$

Where

a_0 is the constant term of multiple regression;

$APCS_p \times a_{pi}$ stands for the mass contribution of the source p to Ci;

a_{pi} is the coefficient of multiple regression of the source p for pollutant i.

In this study, PCA was carried out using the R software.

2.4 PMF analysis

Source apportionment is described as the process used to identify the PM sources and can be performed using a wide range of methods/models. Positive matrix factorization (PMF) is amongst the factor analysis models. The model is described for the first time by Paatero [36]. In the PMF 5.0 model, a weighted least-squares fit with known error estimates of the elements of the data matrix was used to derive the weights. The constraints of PMF parameters should not be negative for more accuracy, and only very few values are negative. These constraints are a good asset for the model to achieve environmentally reasonable solutions as it is impossible to have sources with negative contributions to PM mass. Details can be found in Kebe et al. [31].

3 Results and Discussion

3.1 Elemental Concentration

The mean and standard deviations of the elemental concentrations of $PM_{2.5-10}$ and $PM_{2.5}$ samples from both sites are shown in Table 1.

Al, Si, Cl, Ca, and Fe are the most abundant elements, as shown in Table 1. Al, Si, Ca, and Fe are compounds of crustal origin [37,38], and Cl originates from sea salt. The outliers were removed before analysis to avoid error propagation [39], and Mg and Co were removed for the PCA. The eigen value of each extracted factor was greater than 1.0. Figure 2 shows the eigen value of each factor.

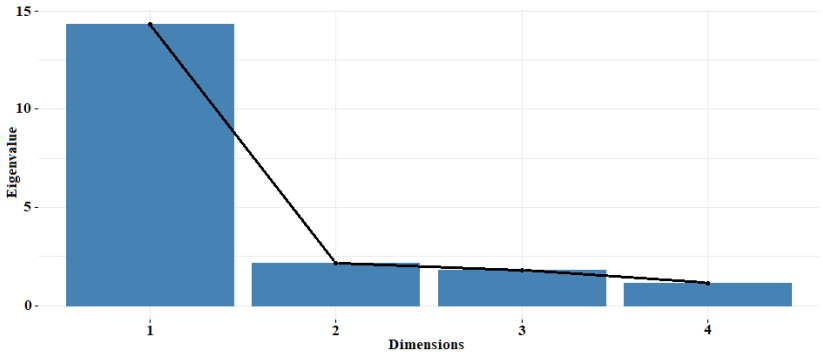

Fig 2. Eigen Values of the four first components

3.2 Source apportionment

In this study, we used a database having 74 samples and 22 elements (BC, Na, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Rb, Sr, Ba, and Pb) with determined elemental concentrations submitted to PCA. With more common low-resolution detectors, such as Si (Li), the data treatment process is less accurate and more time consuming [40]. This limits the use of such algorithms [41] for the analysis of complex matrices. To avoid this limitation, the concentrations for the measured filters were processed using least-square regression analysis and statistical evaluation.

The linear equation Y=a+b.x was used to compute the concentration [42].

The four components considered explained 88.3% of the total variance in the database. The PCA analysis extracted the factor loadings, as shown in Table 2.

Sources obtained based on species factor loading are identified as follows:

Factor component 1 (FC_1) accounted for 34% of the variance in the database. It has high loadings for Al, Si, K, Ti, Mn, Fe, and Rb. This factorial component has been described as a mixture of elements from the Earth's crust (Al, Si, K, and Ti) and from

Table 2. Principal Component Analysis factor loadings with Varimax Rotation Method

Chemical Species	Major Principal Components			
	FC1	FC2	FC3	FC4
BC	0.254	0.213	0.762	0.11
Na	0.469	0.673	0.133	-0.054
Al	0.847	0.349	-0.084	-0.13
Si	0.874	0.377	-0.055	-0.114
P	0.322	0.914	0.003	0.056
S	0.391	0.838	0.353	-0.035
Cl	0.194	0.555	0.699	-0.144
K	0.863	0.31	0.325	-0.122
Ca	0.628	0.609	0.384	-0.011
Ti	0.827	0.435	0.316	-0.034
V	0.556	0.702	0.207	0.117
Cr	0.523	0.808	0.201	0.076
Mn	0.78	0.547	0.221	-0.041
Fe	0.757	0.53	0.333	-0.006
Ni	0.125	0.251	0.182	0.883
Cu	-0.343	-0.186	-0.424	0.711
Zn	0.328	0.824	0.259	0.312
Br	0.199	0.548	0.709	-0.077
Rb	0.841	0.119	0.233	0.255
Sr	0.643	0.603	0.408	-0.014
Ba	0.664	0.547	0.029	0.322
Pb	0.013	0.06	-0.841	-0.017

the steel industries (Mn, Fe, and Rb).

Factorial component 2 (FC_2) was identified as the emissions from the industry. This is confirmed by the high loadings of P, S, V, Cr, and Zn which indicate heavy industry, primary refinery, and/or coal mines [43,44] withan accounted variance of 30.7%.

Factor component 3 (FC_3), with a variance of 16%, has high levels of Cl, Br, and BC concentration and is considered a mixture of elements from sea salt (Cl and Br) and biomass or fossil fuel combustion (BC). The mixing of the two sources could be attributed to synchronous transport from the sea or coastal region to the sampling point.

The factorial component 4 (FC_4) is characterized by high Ni and Cu loading indicating traffic emissions with a variance of 7.6%. Cu comes from pavement abrasion [45].

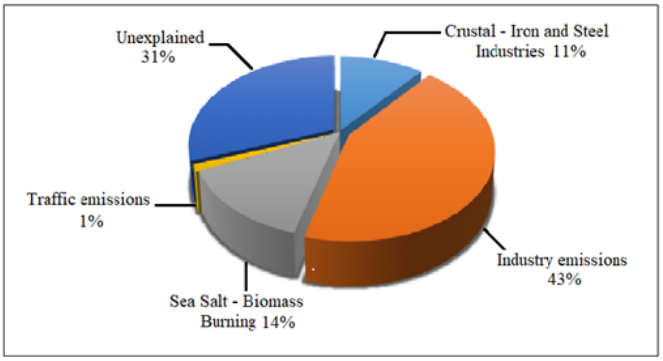

Fig 3. Quantitative source contribution

4 Conclusion

Principal component analysis/absolute principal component scores (PCA/APCS) and positive matrix factorization (PMF), an advanced robust statistics technique was performed to apportion the sources influencing the $PM_{2.5-10}$ and $PM_{2.5}$ levels, a yearlong measurement campaign, during the years 2018 - 2019, twice per week (one working day and one during weekend. Several samples were collected throughout 2018 - 2019, and 71 of them were kept, to perform XRF analysis. Obtained results can be understood from the model used to compute elemental concentration, black carbon and PM. The Factor components highlighted the statistical distance between air particulate matter samples and elemental concentration in a multidimensional space. The first four extracted component accounted for nearly 88.3% of the variability of the data set. The running matrix elements used for data processing within collected air particulate matter was BC, Na, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Rb, Sr, Ba, and Pb. This discrimination of the area of Dakar cites according to the PCA/APCS results confirms the impact that the major activities have on the environment in the most appropriate way, and it verifies the results of previous studies about the influence of Industry emissions, mineral dust, traffic emissions and sea salt/secondary sulfur on the surrounding urban areas. Even if the analysis method is unsupervised, it was capable of sorting the collected air particulate samples in correct groups in compliance with the description in the literature except for the remains factor components which account for 11.7%. The complexity of the performance may be improved by choosing a higher truncation point by normalizing the data or by concatenation with concentration data related to organic and elemental compounds and photo chemically aged aerosols.

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