Available online at http://www.journalijdr.com

International Journal of
DEVELOPMENT RESEARCH

ISSN: 2230-9926

International Journal of Development Research
Vol. 06, Issue, 06, pp. 8085-8091, June, 2016

Full Length Review Article

SOURCES APPORTIONMENT OF TOTAL SUSPENDED PARTICLES AT A COASTAL CITY IN FRANCE

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ARTICLE INFO

Article History:
Received 22nd March, 2016
Received in revised form
14th April, 2016
Accepted 10th May, 2016
Published online 30th June, 2016

Key Words:
TSP, NMF,
Source,
Composition

ABSTRACT

The elemental composition of TSP collected upon different wind sectors in the highly industrialized harbor of Dunkirk (France) were measured and the data was interpreted using a Non Negative Matrix Factorization (NMF) receptor model. Our objective was to identify the possible sources of TSP and to determine their relative contribution at the sampling site. We decided to carry out a daily sampling campaign at a fixed site and a sufficiently long period, i.e. 3 months, from February to May 2008. Different weather conditions have been met during this period and samples have been collected under marine influences, industrial and urban. The physicochemical analysis was focused on the inorganic fraction of the particles. TSP were collected in order to analyze 19 elements: Al, Ca, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, Sn, Ti, V, Zn Cl, NO_3^-, SO_4^{2-}, NH_4^+. Ion chromatography (IC) and inductively coupled plasma-atomic emission spectroscopy (ICP-AES) were used to obtain the concentration of different water soluble anions and elements respectively. Nine sources factor were determined by NMF for all wind sectors. It showed that 9 sources contributed to the ambient particulate pollution at the sampling site: 4 background sources and 5 industrial sources.

INTRODUCTION

In recent years, considerable amount of concerns have arisen on the extent of pollution, particularly the contribution of industrial particulate matter (PM) emissions and metal levels in the neighboring residential areas (Moreno *et al*., 2004 and Dall'Osto *et al*., 2008). The town of Dunkirk (51°04′N; 2°38′E) was chosen as the study site because some of its specificities; it is located on the sea-side of Nord Pas de Calais. Several industries were established of various fields: petrochemical, steel, cement, metallurgy. Therefore, the atmosphere of this site may well suffer gaseous and particulate inputs from various origins: marine, industrial and vehicular.

The various sources of pollutants and the meteorological changing conditions make of Dunkirk a representative study of the effect of industrial and urban development on air quality under marine influence. In recent years, several studies have been conducted in order to characterize the particles and to understand their behaviors in the atmosphere of the Nord-Pas de Calais region and especially Dunkirk (Ledoux *et al*., 2003 ; Laversin *et al*., 2006; Choël *et al*., 2006 and Lamaison *et al*., 2006). Iron is the most anticipated element in particulate emissions of a steel plant. However, such a site cannot be considered as a unique source from which a single type of particles would be emitted. A steel plant includes all necessary facilities for the production of steel (raw materials ...) and each of these units should be seen as a source of particles in itself. Thus, each site unit may emit air particles whose physicochemical characteristics is different and depend on the materials used and the applied treatment. The origin tracers are not identical for each of the emission sources. Many receptor and dispersion models are still being used for source identification (Viana *et al*., 20008). With some studies pushing for the use of models which consider a prior knowledge on the sources of emissions. In this study, we have used a constrained receptor model which appear of particular interest in the attempt to separate sources characterized by similar elements in their chemical profiles, as in the industrial profiles related to integrated steelworks emissions (Hleis *et al*., 2013). This work discusses the determination of the different source profiles contributing to the suspended particles emissions in Dunkirk application of a constrained weighted non negative matrix factorization model (NMF).

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Experimental

TSP were sampled at the top of a 3 m mast implanted on the roof of the Research Institute that makes the sampling height of about 10 m above ground level. The sampling point is directly under the influence of the industrial sector emissions when the wind blows from direction 260 degrees to 320 degrees (Figure 1). TSP were collected by global filtration on cleaned Whatmans 41 cellulose filters and mounted in a PVC holder, for a period of 12 h using a high capacity air aspiration device at a flow rate of 5 m^3/h, to which is connected a calibrated dry gas volume meter with a precision superior to 1%.

After sampling, filters were dried under laminar flow hood until constant mass and weighed with a precision of 0.01 mg. Meteorological conditions (wind direction and speed, rain falls, temperature, humidity, atmospheric pressure) have been recorded for each sample (Figure 2)

Analytical

Ion chromatography (IC) and inductively coupled plasma-atomic emission spectroscopy (ICP-AES) were used to obtain the concentration of different water soluble anions and elements respectively.

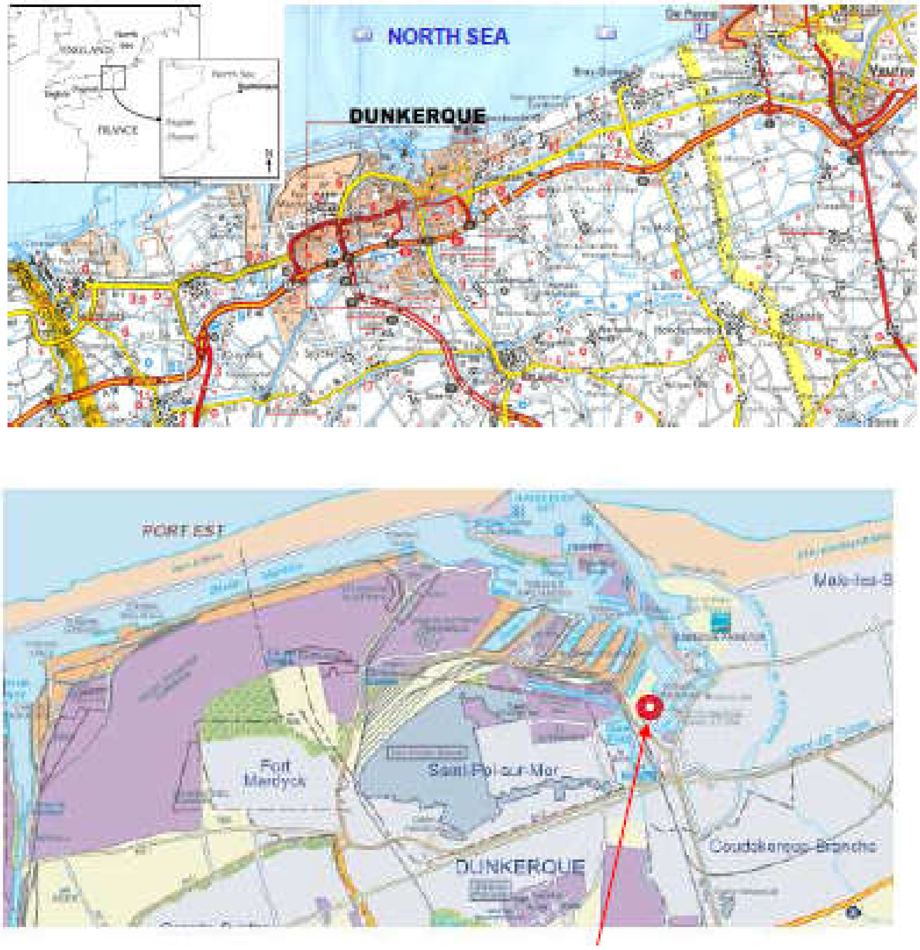

Figure 1. Sampling site

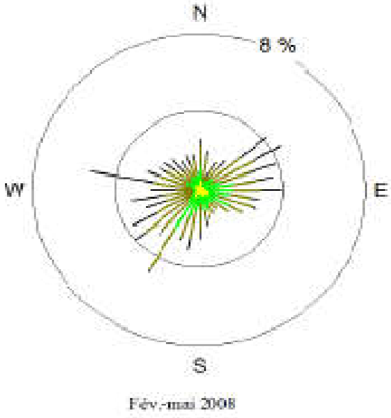

Figure 2. Wind rose recorded during sampling period

A Dionex DX100 chromatograph was used for the analysis of water soluble anions (Cl , NO_3 , SO_4^{2}) and a Dionex ICS900 for the analysis of ammonium ions (NH_4^+). Water soluble anions were extracted (about 5 mg of particles) by leaching using an ultrasonic treatment for 30 min in 10 mL of ultra-pure water (MilliQ®, Millipore; resistivity = 18.2 M.cm). The leachate was filtered on a cellulose acetate membrane (porosity 0.45 m) in a polyethylene flask and the volume adjusted to 20 mL, in each flask, by adding ultrapure water. The final solutions were conserved at 4 C before injection in the analytical column. Element analysis was conducted using a Varian® Vista-Pro spectrometer. The list of elements that were analyzed was restricted to: Na, Mg, Al, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Cd, Sn, Sb, Pb and Bi. Analysis was realized after acid digestion, considering about 5 mg of particles placed into a PTFE flask with $HNO_3/HF/HClO_4$ (4:1:0.5 v/v/v Suprapur®, Merck®) at 130 C for 4 h. After evaporation at 170 C and then cooling, the solution was diluted with MilliQ water up to 10 mL and acidified up to 0.2% HNO_3, prior to analysis. The quantification method was validated from the analysis of a certified aerosol sample from American NIST (Standard reference Materials, no.1648). Uncertainties in determining water soluble ions and elements concentrations were less than 3% (relative standard deviation from three analytical replicates).

Major source identification

Constrained weighted-non-negative factorization matrix The model used in this study comes at this stage in the form of a weighted non-negative matrix factorization model and respects to a certain extent the maximum number of recommendations on the use of modeling in source apportionment studies. Briefly, similar to the principles of positive matrix factorization, the non-negative matrix factorization use the factorization approach of X = G × F under positivity constraints ($g_{ik} \geq 0$) and ($f_{kj} \geq 0$), where the X matrix is the measured dataset, and G and F are the contribution at the source profile matrices respectively, g_{ik} and f_{kj} are elements from the G and F matrices. By default, the NMF algorithm does not consider any differed variance data in the calculations. Building a new version of the NMF that considers individual variances on the data input was essential and formed an important extension to the model. This upgrade is included in the "weighted NMF" based algorithm which enables the consideration of individual measurement uncertainties in the calculations. The uncertainties are inserted in the model in a matrix form, and the calculation equation turns to the following form:

$$\{G,F\} = \arg\min_{G,F} \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{x_{ij} - (GF)_{ij}}{\sigma_{ij}} \right)^2$$

Where, x_{ij} represents the current element drawn from the observations matrix, G is the contributions matrix, F is the source profiles matrix, and $\sigma_{ij}$is the general element of the uncertainties matrix. Furthermore, the weighted NMF algorithm has been modified in order to take into account the a-priori knowledge on the source chemical composition by applying constraints. In the so-called constrained weighted non negative factorization model, constraints are applied to the source profile composition in order to guide the calculation within iterations. Constraints can be inserted in two forms: the first one is the "equality" defines the presence or absence of an element the source profile, and eventually forces its concentration to a specific value, as PMF/ME-2 (positive matrix factorization–multilinear engine 2) from USEPA (United States Environmental Protection Agency) can do [9], [10]. The second type of constraint is "inequality" and imposes an order of abundance for a given species between the different source profiles (for example [X]source-1 > [X]source-2 and [X]source-3 > [X]source-4).

These "inequality" constraints are preferred when compared to the equality ones because of their flexibility and lightness on the calculation. In fact, these soft constraints do not force the NMF model to calculate under stringent conditions, but only guide it to respect an order of abundance in species between the different source profiles. The accessibility to the NMF source code adds more advantages to this algorithm over the PMF, and allows additional developments and implementation compared to the PMF software which appears like a "black box". Calculations using PMF are also known to be time consuming (Paatero and Tapper, 1994), whereas calculation times are lighter using the NMF algorithm (Gobinet *et al.*, 2006). Finally, the use of constraints allows to refine source profiles and at the same time to be less subject to local minima during the calculation process. The introduction of constraints constitutes a major advance in the source apportionment calculation field. Additional information about the constrained weighted NMF model is given in (Delmaire *et al.*, 2010 and Hleis *et al.*, 2010).

Source identification

A preliminary selection of samples according to the wind direction was undertaken on all samples in the sector 250° - 60° (thus covering the industrial and the marine sector).

The application of the weighted NMF model on these samples was made in two stages

- In a first step, the model was applied to samples under sector 250° -60° but not having an industrial pronounced influence (in terms of element contents emitted by the installations in the steel industry). This step is used to identify the sources of particulate profiles that explain the background pollution.
- In a second step, all of the samples in the 250° -60° sector were considered.

This process is gradual because the characteristics of particulate background profiles (first stage) may be kept in the 2nd stage. The algorithm performs a first set of iterations using a projected gradient derived from NMF toolbox Lab (http: //www.bsp.brain.riken.jp/ICALAB/nmflab.html) with standardization between each step until a stopping criterion. Then, a second phase will start following a least squares alternating (ALS), with a constraint of normalization and positivity. Standard deviation of the relative values (RSD) of the analytical measurement by ICP-AES and CI were used to express the uncertainty about the levels of elements and ions. For values below the limit of detection (LOD), the concentration value is set to LDD / 2. For missing values, the concentration is replaced by the average. For values below the limit of detection and missing values, the relative uncertainty is set at 100% and 400% respectively (Polissar *et al.*, 1998; Liu *et al.*, 2005). In addition, starting uncertainties are sometimes adjusted by most authors in the analysis (Prendes, 1999; Song *et al.*, 2001). Constraints for the considered factors were deduced from the literature (Amato *et al.*, 2009; Mooibroek *et al.*, 2011). It includes the composition of sea-water and continental crust (Hans Wedepohl, 1995), for natural and urban background sources, the analysis of raw materials, the characterization of dust collected inside or directly under the influence of the facilities (Hleis *et al.*, 2013 and Hleis, 2010). This significant load of information was intentionally used in the model because it will efficiently guide the calculation process to result in refined results, mostly when modeling complicated cases such as our study.

The result shows 4 profiles sources: sea salt, aged sea salt, secondary aerosols and crustal (Figure 3)

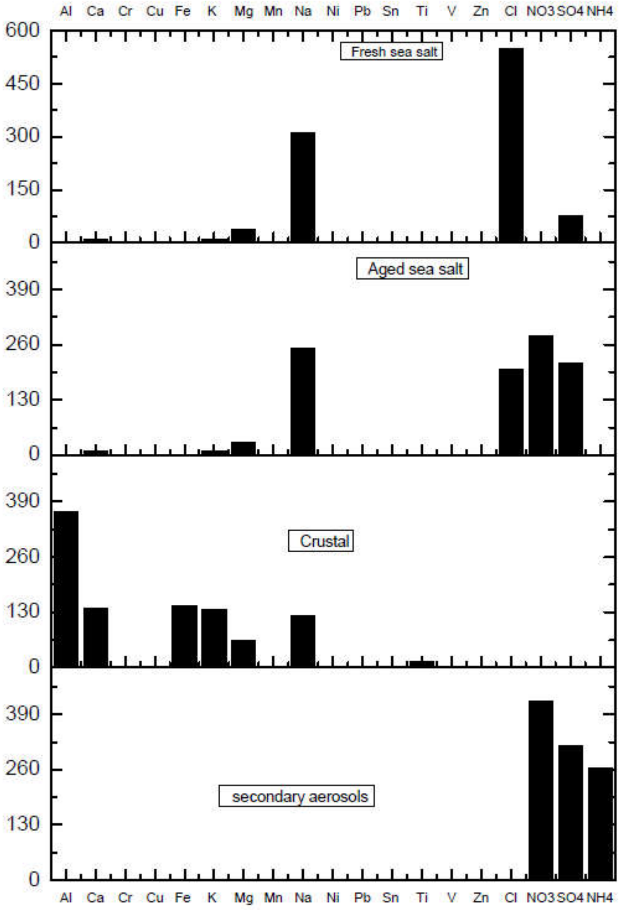

Figure 3. Profiles of non-industrial sources found by the model under constraints (ng/m^3)

Four particles types were evidenced: sea salts, aged sea-salts, secondary inorganic aerosols and crustal particles. Their respective profiles were in good agreement with results from literature (Evans *et al.*, 2004 and Harkel *et al.*, 1997 and Zhao and Gao, 2008). We can assume that the composition of such emissions does not vary with time, and they represent the main sources contributing to atmospheric particulate background.

We have therefore chosen to freeze these profiles retaining the values obtained by applying the weighted NMF model under constraints. As we aim to identify the industrial emission sources which additionally influence the level of atmospheric particulate at our sampling site. In the second step, all samples collected on the sector 250° -60° (industrial and not industrial) were considered for modeling. We looked for industrial sources profiles. We must not forget that the integrated steelwork complex gathers processes devoted to coke making, iron ores sintering, steel production and slag treatments. A ferromanganese alloys production plant concerned by atmospheric emissions is also located next to the above industrial site. The particulate emissions have different chemical compositions from one process to another and do not always occur simultaneously. For these reasons, it is not adequate to use a single profile to track the impact of this industrial site on the atmospheric particulate level but rather 5.

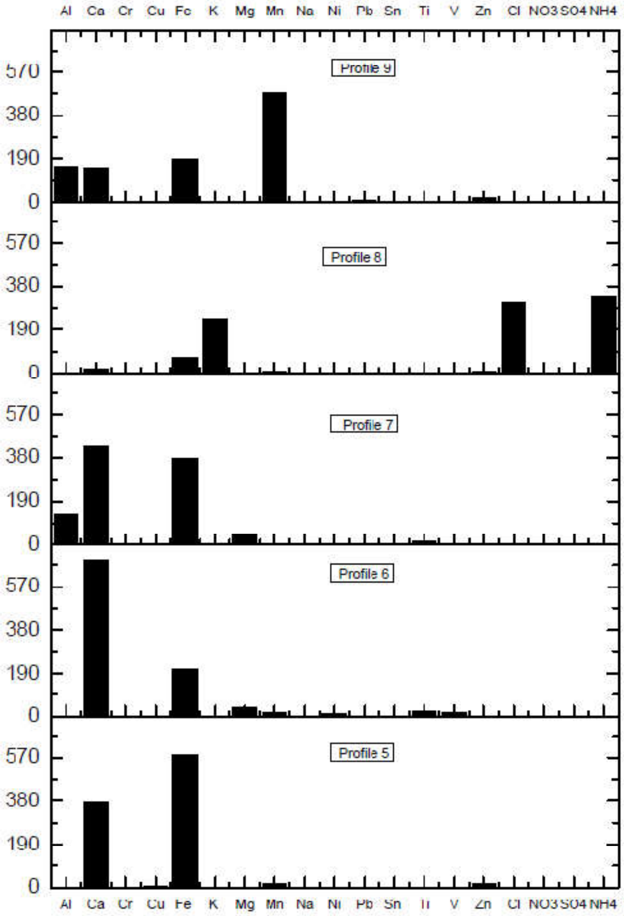

Figure 4: Profiles of industrial sources (ng/m^3)

The case 9 sources presented relevant results (Figure4). In fact, the integrated steelwork complex gathers processes devoted to coke making, iron ores sintering, steel production and slag treatments. A ferromanganese alloys production plant concerned by atmospheric emissions is also located next to the above industrial site. The particulate emissions have different chemical compositions from one process to another and do not always occur simultaneously. For these reasons, it is proposed that 5 industrial sources are expected to contribute significantly to airborne particulate matter at a local scale (Laversin *et al.*, 2006 and Laversin *et al*). By comparing these profiles with experimental data defined for each source [28], the profile issued from profile 5 is referred to as iron rich particles emitted by blast furnaces or steel plant. These two sources have got very similar chemical identities. These features have also been reported for similar processes located on other sites all over the world (Machemer *et al*., 2004 and Oravisjärvi, 2003).

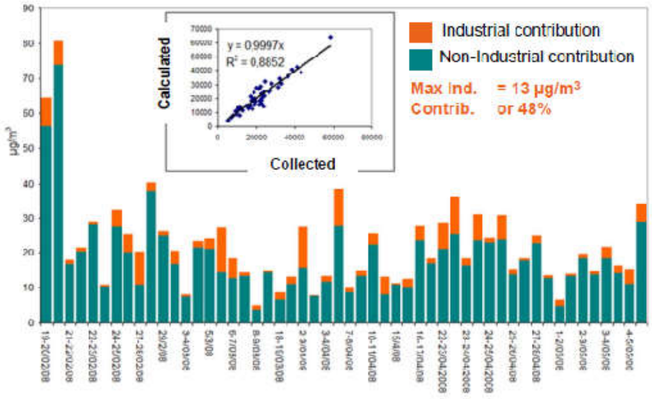

Figure 5: Contribution of the different sources of pollution

The fifth profile is characterized by a high proportion of calcium and iron and then various metallic impurities. Unambiguously, this profile can be attributed to fine particles which gather a set of undesirable elements in steel composition and which are separated in the form of slag. Such emissions are classified as fugitive emissions. The seventh profile highlights species in the following order: Fe>Ca>Al>Mg>Mn>K>Na. It is ascribed to ores sintering plant source and such particles correspond to fugitive emissions depending on all handling performed on sintered ores. It comes from the heating of raw materials. However, a lack of potassium and sodium may be noted. The eighth source profile is expected to be mainly composed with Cl , K, and Fe. It stands for the sintering chimney point source. But it can be noticed that $NH_4^+$is over estimated. Finally, a ferromanganese plant source may be recognized from the ninth and it is to be noted that the expected profile is very well recovered.

Source contribution

Figure5 represents the evolution of the contributions of the different sources over the sampling period. These contributions are expressed as % of total inorganic content. It appears that the contribution of non-industrial sources of contributions (so-called "background") are dominant. Their contributions are high in the case of secondary inorganic aerosols (~ 41% on average) and marine aerosols (freshly issued aged ~ + 39%). However, the contribution of crustal source is low (2%). The cumulative contribution of industrial sources is an average of about 17%; it will be discussed more extensively below. Figure5 shows that the contribution of the industrial emissions varies: the cumulated industrial contributions is sometimes very low, but conversely, it reaches, during certain days, nearly 45% of the total inorganic content, when, with certain weather conditions, our receptor site is directly under the influence of the industrial sector. In order to get an idea of the rest, we also showed the result of correlation between total inorganic "collected" in the atmosphere and the inorganic total "calculated" by the model. We noted that the total inorganic content "calculated" explains 99.9% of the total inorganic "collected" with a correlation coefficient (r^2=0.8852). This may be compared to results of other studies on other sites worldwide. In a study on the contributions of sources to the particulate content in an industrial area in the north-east Spain, Viana *et al*. (2007) explained the PM_{10} content with the contributions from five sources. They found a good correlation (r^2=0.83) between PMF calculated data and sampling data. In a similar study carried out in an industrial area in Korea J.-M, Lim *et al*. (2010) attributed the load of PM10 to the contributions from 9 sources , the calculated PM10 contents then explained 98% of the content determined by chemical analysis and the correlation between the data was r^2=0.77. Overall, the non-industrial contribution outweighs the industrial contribution, the industry averaging 17% of the total collected. These results are comparable with those of several other work on nearby industrial plant sites. Oravisjärvi *et al* (2003) studied the composition and origin of fine particles in a site close to a steel plant in Finland. They attribute 44% of PM2.5 contributions to "long distance (secondary aerosols), 14% metallurgical emissions. Mugica *et al* (2009 & 2002), in a study on a site nearindustrial plants in Mexico, attributed 11% of the particulate content to secondaryaerosols, 10% to industrialemissions, 26% to resuspendedparticles M. Koçak *et al* (Koçak *et al.,* 2009) in their study of the composition and origin of the particles collected on a rural site, allocate a large share (60%) of PM10 to crustal and marine aerosols. They also attribute 10 and 20% respectively to the particles resulting from combustion processes and secondary aerosols.

Acknowledgment

This work was supported by ArcelorMittal.

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