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Research article

Impact of secondary inorganic aerosol and road traffic at a suburban air quality monitoring station

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ABSTRACT

PM10 from a suburban site in the northwest of Spain was assessed using data from chemical determinations, meteorological parameters, aerosol maps and five-day back trajectories of air masses. Temporal variations in the chemical composition of PM10 were subsequently related to stationary/mobile local sources and long-range transport stemming from Europe and North Africa.

The presence of secondary inorganic species (sulphates, nitrates and ammonium) in airborne particulate matter constituted one of the main focuses of this study. These chemical species formed 16.5% of PM10 on average, in line with other suburban background sites in Europe. However, a maximum of 47.8% of PM10 were recorded after several days under the influence of European air masses. Furthermore, the highest values of these three chemical species coincided with episodes of poor air circulation and influxes of air masses from Europe. The relationship between SO_4^{2-} and NH_4^+ ($R^2 = 0.57$, p-value<0.01) was found to improve considerably in summer and spring ($R^2 = 0.88$ and $R^2 = 0.87$, respectively, p-value<0.01), whereas NO_3^- and NH_4^+ ($R^2 = 0.55$, p-value<0.01) reproduced this pattern in winter ($R^2 = 0.91$, p-value<0.01). The application of a receptor model to the dataset led to the identification of notable apportionments due to road traffic and other types of combustion processes. In fact, large amounts of particulate matter were released to the atmosphere during episodes of biomass burning in forest fires. On isolated days, combustion was estimated to contribute up to 21.0 μg PM/m^3 (50.8% of PM10). The contribution from industrial processes to this source is also worth highlighting given the presence of Ni and Co in its profile. Furthermore, African dust outbreaks at the sampling site, characterised by an arc through the Atlantic Ocean, were usually associated with a higher concentration of Al_2O_3 in PM10.

Results evidenced the relevance of stationary (i.e., steelworks and thermal power station) and mobile sources in the air quality at the suburban site under study, with important apportionments of particulate matter coming from road traffic and as consequence of releasing precursor gases of secondary particles to the atmosphere.

1. Introduction

Human activities influence environmental sustainability. The release of pollutants to the environment must be managed in a way that reduces their impact on ecosystems (Schaubroeck et al., 2016). In particular, air quality plans have to be designed to abate gaseous precursors of secondary aerosol (inorganic and organic), which represents between 40 and 71% of PM10 in Europe depending on the location (Amato et al., 2016).

In the case of secondary inorganic aerosol (SIA), which includes sulphates, ammonium and nitrates, the gas-to-particle transformation is the most important contributor (Sun et al., 2015). The origin of its precursor gases (NO_X, SO_X and NH_3) may be either natural or anthropogenic. These gases may be released by local sources or originate from distant locations as a result of long-range transport, the latter being highly dependent on weather conditions

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(Di Gilio et al., 2015). Moreover, secondary inorganic compounds could be more concentrated during African dust outbreaks, together with mineral dust and other trace elements (Fernández-Camacho et al., 2016). The contribution of different sources to airborne particulate matter (PM) has been widely estimated in air quality studies via the application of what are known as receptor models (Jorquera and Barraza, 2013; Taiwo et al., 2014). These statistical approaches are based on pollutant concentrations determined at a sampling location (receptor site) and do not require direct measurements or emission inventories as in other types of source apportionment methodologies (Alleman et al., 2010). Examples of broadly implemented receptor models are Principal Component Analysis (PCA) and Positive Matrix Factorization (PMF) (Fernández-Olmo et al., 2016).

Although it is not necessary to characterise each possible source of PM to apply a receptor model (Padoan et al., 2016), it is essential to infer the emission sources correctly from the principal components provided by the models. The commonly named marker elements or tracers provide clues regarding the sources that may be associated with each factor given by the model. The perfect tracer would be unique to one source, constant (regardless of operational and environmental conditions), inert and very precisely measured (Khalil and Rasmussen, 2003). Nevertheless, it is not unusual to find the same element reported in the literature as a tracer of several sources, thus giving rise to dissimilar conclusions (Taiwo et al., 2014). Given PM is highly influenced by meteorology (Li et al., 2016), the aforementioned difficulty can be overcome via the study of meteorological factors that affect air quality. Dispersion of pollutants in the atmosphere, wind, solar radiation and topography are amongst the most important (Di Gilio et al., 2015). Some of these factors are essential to assess the seasonal behaviour of secondary species.

The aim of this study was to identify relevant sources of PM10 at a suburban location where road traffic and nearby industrial activities were thought to influence air quality. Special attention was paid to secondary inorganic species and days with higher levels of pollution, when these species represented the largest fraction of PM10. To this end, results from a receptor multivariate model, several meteorological parameters, five-day back trajectories of air masses and aerosol maps were simultaneously analysed. Variations in the chemical composition of PM10 were subsequently related to stationary/mobile local sources and long-range transport stemming from Europe and North Africa. Results were compared with other studies carried out in the area that used a different approach and with other European suburban background sites in order to provide useful information for managing air quality.

2. Materials and methods

2.1. Receptor site

The sampling area was situated at a suburban site (43°31′23.1″N 5°37′16.2″W) in the east of Gijón, a city in north-western Spain. Fig. 1 shows the location of the sampling station, as well as the main industrial activities in the area: port facilities, a cement plant, coalfield, coal-fired power plant, steelworks and industrial complexes. Activities at the complexes encompass aluminium production, galvanizing, the manufacture of refractory products, iron casting, the melting of aluminium scrap and the production of metallic structures and wires (Gobierno del Principado de Asturias, 2014).

2.2. Gravimetrical and chemical determinations

Three hundred and seventy-five daily samples of PM10 were collected over microfibre filters during the period July 2013–July 2014, using a high-volume sampler MCV CAV-A/MSb (30 m^3/h). The filter material was composed of either glass or quartz microfibres. PM10 levels were gravimetrically determined from all the samples. Fifty-two PM10 samples were collected over quartz matrixes once a week and, subsequently, chemically processed to analyse their composition. The sampling day was alternated to obtain samples of all days of the week, thirteen samples being collected at weekends and twenty-three during raining days (above 1.0 mm).

2.2.1. Analytical and indirect determinations

The PM10 samples were acid digested; Al, As, Ba, Be, Bi, Ca, Cd, Ce, Co, Cr, Cu, Fe, K, La, Li, Mg, Mn, Mo, Na, Ni, Pb, Rb, Sb, Se, Sn, Sr, Ta, Ti, Tl, V, Zn and Zr in PM10 were determined by inductively coupled plasma mass spectrometry. Organic and elemental carbon (OC and EC) were quantified by means of a thermo-optical method. Ion chromatography was used to analyse four soluble chemical species (Cl^-, NH_4^+, NO_3^- and SO_4^{2-}). Al_2O_3 was stoichiometrically calculated from Al. Organic matter (OM) was estimated from organic carbon (OC), applying a factor of 1.2, which has been used in sampling locations close to traffic (Amato et al., 2016). Further details about the equipment and the methodology are provided in the supplementary material.

2.3. Receptor model: PCA-MLRA

The receptor model used to study source apportionment was PCA, applying Statistica software. The chemical compositions of the fifty-two samples were used in the PCA, twenty-eight chemical species being introduced in the analysis as variables (see supplementary material). Varimax rotation, a procedure to maximise the explained variance, was used to explore the relationship between variables. Principal components with eigenvalues higher than 1 were retained, as in Negral et al. (2008). Multi-Linear Regression Analysis (MLRA) was used to model PM10 as the dependent variable, as well as the concentration of the chemical species used in the PCA. The percentage contribution of a specific factor to the predicted PM10 was calculated as the sum of the predicted PM10 for the fifty-two samples for this factor divided by the total concentration of predicted PM10 considering all factors.

2.4. Meteorological parameters

Fig. 1 shows the wind rose obtained from data recorded at the sampling location by the Spanish Meteorology Agency (AEMET) since October 2013. The predominant wind directions between October 2013 and July 2014 were East, Southeast and West. Information from AEMET on maximum wind speed and wind direction for each sampling day was used to build polar plots of the concentration of the analysed chemical species in PM10. These graphs were useful in identifying their most likely sources, given that they would be situated in the wind directions that have higher values (Yi and Hwang, 2014). Other tools used to support data interpretation were: dust concentrations predicted by the SKIRON forecasting model, images from the BSC-DREAMS8b model, the NAAPS Global Aerosol Model and the analysis of five-day isentropic back trajectories (ending at 12:00 UTC) calculated at three heights (750, 1500, 2500 m above sea level) with HYSPLIT model from NOAA (Stein et al., 2015). The study of five-day back trajectories led to classifying the air mass origin as Northern Atlantic (AN), North-western Atlantic (ANW), Western Atlantic (AW), South-western Atlantic (ASW), Northern African (NAF), Mediterranean (ME), European (EU), and Regional (RE).