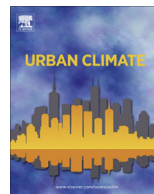




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Discussion on graphical methods to identify point sources from wind and particulate matter-bound metal data

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ABSTRACT

The aim of the present work is to use graphical methods based on the evaluation of selected trace metals (Mn, Cu, Cr, V and Ni) and wind direction monitoring data to identify sources of trace metal in the main urban areas of the Cantabria Region (Northern Spain). These graphical methods take into account the frequency of wind in each sector and the measured concentration of trace elements in PM₁₀. The comparison between the contribution of wind and selected trace metals to each sector is presented in polar diagrams. The main conclusions derived from these diagrams are compared to those obtained from radial diagrams based on pollutant concentration roses computed from daily metal levels and hourly wind direction data. The procedure, based on plotting the ratio between the contribution of trace metals and wind data to each sector on polar diagrams, may result in an easier interpretation. Finally, both procedures are applied to data from three sampling sites located in Santander Bay, to study the influence of point sources on the levels of Mn. The analysis of the results shows that similar conclusions were obtained from both methods. These methods are primarily recommended when large emissions are produced by local point sources.

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1. Introduction

The levels and composition of particulate matter (PM) are directly linked to the proven adverse effects on human health (Pope and Dockery, 2006). The levels of suspended particulate matter with an aerodynamic diameter less than 10 μm (PM10) in some European areas usually exceed the yearly and daily limit values of 40 and 50 $\mu\text{g}/\text{m}^3$ respectively, given by the European Air Quality Directive 2008/50/EC (Querol et al., 2008; Putaud et al., 2010). In addition to natural emissions, the large anthropogenic contributions of particles from local sources increase the level of PM10 and micropollutants such as heavy metals. In this context, some metals are good tracers of local emissions (Moreno et al., 2006); therefore, the analysis of their presence in particulate matter at receptor sites may help in identifying point sources that affect the local air quality.

Three main groups of source apportionment techniques are usually reported in the literature (Viana et al., 2008). The first group consists of source-receptor modelling by means of deterministic models. This approach is the most mathematically complex and requires high quality emission datasets from inventories or direct measurements of pollutants (Maes et al., 2009), to model the dispersion, transformation, transport and deposition of such contaminants (Bessagnet et al., 2004). The second group of models is based on the statistical evaluation of the pollutants measured at receptor sites. These methods have been widely applied in source apportionment studies of different environmental matrices such as rainwater (Juntto and Paatero, 1994), bulk deposition (Mijic et al., 2010) and airborne particles (Pandolfi et al., 2008). Major components, trace metals and organic compounds are usually considered in this type of analysis where chemical mass balance (CMB), principal component analysis (PCA) and positive matrix factorization (PMF) are the main techniques (Viana et al., 2008). The third group consists of methods based on the evaluation of monitoring data, for example, the correlation of meteorological variables, such as wind direction, with the levels of air pollutants (Henry et al., 2002; Somerville et al., 1996). Graphical methods based on the evaluation of trace metals and wind direction monitoring data may be used to identify their sources.

The relationship between the levels of single pollutants and the wind direction is usually reported by means of pollutant concentration roses, which are polar diagrams that show how air pollution depends on wind direction (Eilers, 1991). If an ambient air quality monitoring station is markedly influenced by a source of the pollutant measured, the pollutant concentration rose shows a peak towards the local source (Cosemans and Kretzschmar, 2004; Eilers, 1991). Rose analysis is a commonly used tool in source apportionment on local scales (Lenschow et al., 2001; Rigby et al., 2006) and for identifying local point sources (Fernández-Camacho et al., 2010; Henry et al., 2002; Somerville et al., 1994). Pollutant concentration roses require highly time-resolved concentration data due to the variability of most meteorological parameters over a long sampling period. However, trace metal levels are usually determined in PM10 samples that are collected in sampling periods of 24/48 h. Different strategies have been used to solve this. Firstly, highly time-resolved particulate composition data can be obtained from direct measurement of the particles at the receptor sites by aerosol spectrometry (e.g., ATOFMS, Time-of-Flight Mass Spectrometer) and can be plotted against the average wind direction data (Snyder et al., 2009). A second option is to use low time-resolved metal concentration data (e.g., 24 h) and highly time-resolved wind direction data (e.g., 1 h). Cosemans et al. (2008) used modified pollutant concentration roses to demonstrate that SO_2 reference roses calculated from hourly concentration and wind direction data are similar to those obtained from modified pollutant concentration roses computed by mathematical methods from 24 h concentration data and 1 h wind direction data. Gladtke et al. (2009) used calculated PM10 and metal concentration roses in an industrial area from 24 h surplus concentration data and 0.5 h wind direction data to allocate the individual shares of emitting facilities in an integrated steel plant; surplus concentration data were first obtained by subtracting the background levels of these pollutants measured in sites not directly affected by the point sources. A third approach is to use daily metal concentration data and hourly wind direction data for the calculation of the contribution of each wind sector to the measured concentrations of elements. Yatin et al. (2000) calculated the fractional contribution of each wind sector to measured concentrations of the elements in an aerosol in Ankara (Turkey) to analyse possible source directions. In a later work, (Qin and Oduyemi, 2003) identified the source direction that affects the PM10 data in

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