



Size resolved metal distribution in the PM matter of the city of Turin (Italy)



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HIGHLIGHTS

- Evaluation of urban air quality by determination of various atmospheric pollutants.
- Different behaviours for elements depending on PM size distribution were identified.
- A scarce correlation between O₃, NO_x and VOCs and metals in PM was observed.
- PM metals, NO_x and VOC respond differently to particular weather phenomena.
- Airborne pollutants are subject to different transformation and deposition processes.

ARTICLE INFO

Article history:

Received 27 August 2015

Received in revised form

10 December 2015

Accepted 23 December 2015

Available online 21 January 2016

Handling Editor: Martine Leermakers

Keywords:

PM₁₀

BTEX

Ozone

Urban air quality

Size distribution of elements

Principal component analysis

ABSTRACT

A work on the characterization of the air quality in the city of Turin was carried out in different sampling periods, reflecting early autumn and winter conditions, including a snow episode during the early 2012 European cold wave.

The concentrations of 13 elements in eight size fractions of the aerosol were determined using inductively coupled plasma-mass spectrometry. The collection was carried out with a Andersen MkII cascade impactor.

The size distribution of elements allowed the identification of three main behavioural types: (a) elements associated with coarse particles (Cd, Cr, Cu, Fe, Mn, Mo and Sn); (b) elements found within fine particles (As, Co, Pb and V) and (c) elements spread throughout the entire size range (Ni and Zn).

Principal Component Analysis allowed to examine the relationships between the inorganic elements and to infer about their origin. Chemometric investigation and assessment of similarity in the distribution led to similar conclusions on the sources.

The concentration of gaseous trace pollutants (O₃, NO_x and VOCs) was determined. The concentrations of these pollutants are scarcely correlated with the metal contents of all the size classes of the PM. The differences found in the O₃, NO₂ and VOCs levels of the two winter campaigns due to the high photochemical reactivity in the period after the snow episode, do not reflect in differences in the metals distribution in the PM. Since PM metals, NO_x and VOC have common sources, this behaviour is due to relevant differences in the transformation and deposition processes.

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1. Introduction

Air pollution represents one of the greatest concerns of urban environments. In the last decades great attention was paid to

Particulate Matter (PM), due to the correlation between fine PM exposure and adverse health effects. The health burden associated to PM air pollution is one of the main environmental health concerns raised by World Health Organization (WHO, 2006). The European Environmental Agency indicates that, with respect to the PM₁₀ and PM_{2.5} thresholds and daily limit values, the Po Valley is one of the most critical areas in Europe (EEA, 2013).

Epidemiological and experimental studies have shown that particulate air pollution may induce and aggravate respiratory and

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cardiovascular diseases. Significant correlations between exposure to ambient air particles and increased morbidity and mortality have been demonstrated (Donaldson et al., 2002; Englert, 2004; Katsouyanni et al., 2001; Krewski and Rainham, 2007). Most of the available studies do not attribute the observed health effects to a particular characteristic of PM. Moreover, the exact physicochemical mechanism by which PM produces adverse effects is still unknown; one hypothesis implicates the oxidative potential of the particles or specific components. In particular, epidemiological studies have found this correlation for fine particle metrics, such as PM_{2.5} (Lall et al., 2005). The pro-inflammatory effect of urban fine and ultrafine PM on human bronchial epithelial cells resulted to be concentration dependent (Ramgolan et al., 2008; Baulig et al., 2009). Short term effects of the fine PM have also been found on daily mortality due to diseases of the cardiocirculatory system (Maté et al., 2010).

PM is a complex and heterogeneous mixture, whose composition (particle size distribution and chemical characteristics) changes with time and space and depends on emissions from various sources, atmospheric chemistry and weather conditions. The coarse fraction (2.5–10 µm) comes predominantly from natural sources (geological material, such as fugitive and resuspended dust, and biological material, such as pollen and endotoxins), and its composition changes depending on the geology of the site. The fine fraction (0.1–2.5 µm) is dominated by anthropogenic emissions: a mixture of carbon particles from combustion processes and secondary particles produced by photochemical reactions in the atmosphere (sulphate, nitrate, ammonium). The carbonaceous fraction consists of aggregates of organic and elemental carbon on which transition metals, organic compounds and biological constituents are adsorbed. Penetration in the respiratory system is strictly correlated with the aerodynamic radius. Thus, so concerning the adverse health effects, the distribution of toxic elements and compounds among the various size fractions of the PM is also of primary relevance.

Elements are released to the atmosphere from both anthropogenic and natural sources. Anthropogenic sources include fossil fuel combustion, industrial metallurgical processes, vehicle emission and waste incinerations. Natural sources include a variety of processes acting on crustal minerals, such as volcanism, erosion and surface winds, as well as from forest fires and the oceans.

Other than the effects on public health, the knowledge of the size distribution and size dependent chemical speciation is also important in identifying the sources and transformation processes during atmospheric transport. Indeed, different sources emit airborne PM with different size distribution, which are deposited at different rates. Particles with diameter in the range of 0.1–1.0 µm (accumulation mode) deposit slowly and can be transported far from emission sources with effects on remote areas (Allen et al., 2001). Instead, coarse particles (aerodynamic diameter > 2.7 µm) are usually emitted from local sources.

In general, as reported above, coarse particles mainly come from road dust re-suspensions, abrasion processes, crustal erosion and sea salts, whereas fine mode particles have been found to mainly derive from anthropogenic sources (combustion processes, high-temperature industrial activities, automotive traffic, etc.) (Handler et al., 2008). Therefore, source apportionment and toxicological information are enhanced by detailed knowledge of the size distribution of aerosol and of its chemical composition.

The size distribution of elements within atmospheric particles has been studied over cities around the world and a bimodal distribution of atmospheric particulates has been usually reported, e.g. in Beijing, China (Duan et al., 2012), New York, USA (Song and Gao, 2011), Kanazawa, Japan (Wang et al., 2006), San Roque, Spain (Sánchez de la Campa et al., 2011), Venice, Italy (Toscano et al.,

2011; Masiol et al., 2015) and Thessaloniki, Greece (Samara and Voutsas, 2006).

Concerning the situation in Turin, Italy, time series of the size-fractionated PM₁₀ mass concentration were registered. An overview of these was recently published (Casazza et al., 2013), considering winter data of years 1980, 2000 and 2011. These three years, as reported in the paper, can be associated to a known variation of sources characteristics, due to the introduction of new regulations and interventions aimed at air pollution mitigation. In particular, the first data (year 1980) are representative of the situation before the introduction of a stricter regulatory approach. The fuel desulphurization and the use of unleaded petrol were introduced just before year 2000, while, since year 2011, it is possible to record the effects of the introduction of regional regulations on domestic heating emission control. The data were considered as representative in the previous analyzed works, since the data values were quite stable along the years, following the same measurement conditions. A reduction of the absolute concentrations of PM is observable. Mean values were recorded of 63 µg/m³ (PM₁₀), 38 µg/m³ (PM_{2.5}) and 22 µg/m³ (PM₁) in year 1980; 59 µg/m³ (PM₁₀), 41 µg/m³ (PM_{2.5}) and 32 µg/m³ (PM₁) in year 2000. Finally we measured a mean value of 45 µg/m³ (PM₁₀), 34 µg/m³ (PM_{2.5}) and 29 µg/m³ (PM₁) in year 2011. The relative errors with respect to the gravimetric data, reported from previous works, fall within a lower 1% and an upper 10% limit. On the other side, the PM₁₀, PM_{2.5} and PM₁ distribution in PM₁₀ changed. In particular, the percentages of PM_{2.5} and PM₁ with respect to PM₁₀ mass are: 61% (PM_{2.5}) and 35% (PM₁) in year 1980; 70% (PM_{2.5}) and 54% (PM₁) in year 2000; 75% (PM_{2.5}) and 65% (PM₁) in year 2011. This means that, while the aerosol emissions have changed as absolute amount, the relative emission of smaller size aerosol particles has increased over the years. It should not be disregarded that these data are referred to stable good weather conditions, when the effects of scavenging are minimal and the emissions are not removed either by transport (wind) or by wet scavenging (precipitations). Thus, the aerosol deposition is limited to gravity effect in proportion to each single particle mass. While we can observe a general relative reduction of the coarse fraction, the PM₁ have increased from 35% to 65% of the total PM₁₀ mass concentrations over the 30-years period considered.

In order to provide further insights into the source identification and metal-health relationships, we carried out an ambient aerosol sampling focussing on the characteristics of trace metals in different size fractions. Three sets of size-segregated aerosol samples were collected at the centre of the city of Turin, in very different weather conditions. At the same time, VOC, NO_x and O₃ measurements were carried out in order to better assess the relationship between sources, the pollutant chemistry and the deposition processes.

The objectives of this study are: 1) to investigate the enrichment levels of selected trace metals and their size distributions, 2) to identify the major categories of sources contributing to the enrichments of trace metals, 3) to assess the influence of weather conditions on the concentrations of trace metals and their size distributions and 4) to verify their correlation with photochemical and gaseous pollutants.

2. Material and methods

2.1. Sampling location and size segregated PM collection

Turin is a metropolitan area characterized by a high density of residential and commercial premises and a very high volume of vehicular traffic; several industries, including power plants, chemical plants, plastic and metallurgical factories are located on

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