



Seasonal and spatial variations of atmospheric trace elemental deposition in the Aliaga industrial region, Turkey

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ABSTRACT

Atmospheric bulk deposition (wet + dry deposition) samples ($n = 40$) were collected concurrently at ten sites in four seasons between June 2009 and April 2010 in the Aliaga heavily industrialized region, Turkey, containing a number of significant air pollutant sources. Analyses of trace elements were carried out using inductively coupled plasma–mass spectrometry (ICP–MS). While there were significant differences in the particulate matter (PM) deposition fluxes among the sampling sites, seasonal variations were not statistically significant (Kruskal–Wallis test, $p < 0.05$). Both PM deposition and elemental fluxes were increased at the sampling sites in the vicinity of industrial activities. The crustal elements (i.e., Ca, Mg) and some anthropogenic elements (such as Fe, Zn, Mn, Pb, Cu, and Cr) were high, and the highest fluxes were mostly measured in summer and winter seasons. The enrichment factor (EF) and principal component analysis (PCA) was applied to the data to determine the possible sources in the study area. High EF values were obtained for the anthropogenic elements such as Ag, Cd, Zn, Pb, Cu and Sb. The possible sources were identified as anthropogenic sources (i.e., iron–steel production) (45.4%), crustal and re-suspended dust (27.1%), marine aerosol (7.9%), and coal and wood combustion (8.2%). Thus, the iron–steel production and its related activities were found to be the main pollutant sources for this region.

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1. Introduction

Atmospheric particulate matter (PM) may be emitted into the atmosphere by a wide variety of sources: natural (i.e., soil erosion, sea spray, volcanic activities) and anthropogenic (i.e., industrial activities, traffic emissions, residential heating, fossil fuel combustion including coal and biomass burning). PM could also be formed by the chemical transformation of organic compounds or inorganic gaseous in the atmosphere as secondary organic aerosol (SOA) (Kroll and Seinfeld, 2008) and secondary inorganic aerosol (SIA) (Belis et al., 2013). Atmospheric PM is generally defined as a mixture of solid and/or liquid particles that remain individually dispersed in air and it is identified as one of the most significant of air pollutants in terms of environmental

and health impacts. Many studies indicated that PM can affect the climate (Tainio et al., 2013), reduce the visibility (Polissar et al., 2001; Chang et al., 2009), and affect other ecosystems (aquatic, soil, or vegetation) (Odabasi et al., 2010; Hofman et al., 2013; Im et al., 2013). High PM concentrations may also play a role in the severity and incidence of respiratory diseases such as aggravated asthma, coughing and painful breathing, chronic bronchitis, and decreased lung functions (Ostro et al., 1999; Zheng, 2011; Chen et al., 2013).

PM is removed from the atmosphere by depositional processes (wet and dry) (Odabasi et al., 2002; Motelay-Massei et al., 2005; Okubo et al., 2013). Atmospheric PM may vary in size from a few nanometers to several tenths of micrometers. The residence time of particles (and associated trace elements) in the atmosphere depends on their size. While the rate of deposition is slower for fine particles, the coarse particles settle out quickly near their sources by sedimentation or impaction

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processes (Marx et al., 2005; Perrino, 2010). Thus, deposition is a significant pathway for transferring trace elements from the atmosphere to the terrestrial surfaces and aquatic ecosystems.

It is important for PM and its elemental concentrations and compositions to be measured, monitored and determined in urban and industrial regions in order to assess whether it has negative effects on biotic or abiotic environments. Bulk deposition sampling (i.e., sampling the total deposition fluxes without separating as wet or dry) provides some advantages compared to ambient air monitoring, such as having low cost, being simple, no need for electrical power, and low maintenance (Clarke et al., 2010). The deposition fluxes of atmospheric PM and the ratio of wet to dry deposition are controlled by emission sources, atmospheric concentrations, distance to receptor sites, and meteorological conditions (i.e., wind speed, prevailing wind directions, frequency, amount and intensity of precipitation) (Mijic et al., 2010; Okubo et al., 2013). The characterization of bulk deposition samples is important for identifying the variability and sources of the atmospheric pollutants (Odabasi et al., 2002; Avila and Rodrigo, 2004; Azimi et al., 2005; Motelay-Massei et al., 2005). During the past decades, many studies on the deposition fluxes of trace elements in rural, urban and/or industrial areas have been widely used to estimate the influence of atmospheric inputs of trace elements to the environmental compartments (Golomb et al., 1997; Wong et al., 2003; Dundar, 2006; Papaefthymiou and Anousis, 2006; Sharma et al., 2008; Balakrishna and Pervez, 2009; Wang et al., 2009; Mijic et al., 2010; Cereceda-Balic et al., 2012). As a result of these studies, it is pointed out that the atmospheric PM mass and trace elemental deposition originate from two main source categories: wind entrained soil from arid/semi-arid regions and anthropogenic sources, i.e., fossil fuel combustion (V, Mo, Ni, and Cr), traffic and road emissions (Cr, Mn, Fe, Br, Co, Cu, Zn, Ba, Sb, and Pb), ferrous metal production (Ag, As, Cr, Cd, Cu, Pb, Fe, Mn, Sn, Zn), non-ferrous metal production (Cu, Zn, Cd) and residential heating (coal and wood combustion) (As, Hg, K, Sb, Pb, Se, Rb, and Tl) (Schauer et al., 1996; Bilos et al., 2001; Pacyna and Pacyna, 2001; Molnar et al., 2005; Aksoy et al., 2009; Kara et al., 2011; Nalbandian, 2012; Song et al., 2012; Zhang et al., 2013). Recent studies have indicated that trace elements particularly emitted from anthropogenic sources can be persistent, toxic, widely dispersed in the environment (air, water and soil compartments), and interact with different natural components, cause threat to human health and they have harmful effects on the biosphere (Kabata-Pendias and Mukherjee, 2007; Alloway, 2012; Srinivas and Sarin, 2013). Therefore, it is important to monitor the trace elemental levels in urban and industrial areas.

The objectives of this study were (1) to determine and characterize the spatial and seasonal variations of atmospheric PM and trace elemental bulk deposition fluxes, and (2) to identify the possible sources of trace elements in bulk deposition in a heavily industrialized region (Aliaga, Turkey).

2. Materials and methods

2.1. Study area and sampling sites

The Aliaga region (38°40'–38°54'N and 26°50'–27°03'E) is located in the western part of Turkey on the coast of the Aegean Sea and contains a number of significant air pollutant

sources including five scrap iron–steel processing plants with electric arc furnaces (EAFs), several steel rolling mills, a large petroleum refinery, a petrochemical complex, a natural gas-fired power plant, a fertilizer plant, ship breaking yards, coal storage and packing, scrap storage and classification sites, large slag and scrap piles, heavy road traffic, very intense transportation activities including ferrous scrap trucks and busy ports used for transportation. Aliaga town with a population of ~60,000, several villages and agricultural areas and some resorts are also located within the region. The region is mainly classified within the local climate of the Mediterranean Sea that is characterized by hot, dry summers and cool, wet winters and springs. While the annual rainfall reaches to 688 mm, the average rainfall amounts were 10, 40, 117 and 54 mm for summer, autumn, winter, and spring sampling periods, respectively (TSMS, 2013). Monthly average temperatures during the sampling periods were measured as 28.2, 21.0, 10.6 and 16.1 °C for July, October, January and April, respectively. The prevailing winds are northwest and southeast in the study area. The wind roses generated using WRPLOT View (Lakes Environmental, Canada) for four sampling periods are shown in Fig. S1. The mean wind speed was 3.1 m/s during the sampling period while the annual prevailing wind directions were WNW (26.5%), NW (13.3%), N (13.0%) and S (8.8%).

In the present study, the bulk deposition samples were collected at ten sites (Aliaga town, ship-breaking yards, steel production area, coal plants area, organized industrial zone, and five villages) in the study area. Locations of the sampling sites, industrial activities and settlements for the region are illustrated in Fig. 1 and detailed information on the sampling sites is listed in Table S1.

2.2. Sample collection and analysis

A total of 40 atmospheric bulk deposition samples were collected using open polyethylene funnel samplers (26 cm diameter and 30 cm height) with a collecting chamber fitted on a stand at about 2 m above the ground (Aas et al., 2009). Both dry and wet particulate depositions were collected during 30 days. Sampling campaigns were conducted at ten sampling sites between June 2009 and April 2010 for four sampling periods representing summer, autumn, winter and spring. Before sampling, all samplers were rinsed with 10% (v/v) solution of nitric acid (HNO₃) and then with deionized water (18.2 MΩ/cm). At the end of the sampling, the bulk deposition samplers were rinsed with 500 mL of 10% (v/v) HNO₃ (65%, Merck, Suprapur) solution in order to transfer the particulate matter completely from the collecting chamber to the collecting bottle. The collecting bottles were sealed in the field with plastic covers and transferred immediately to the laboratory. The collected solutions were evaporated at 100 °C in a beaker (borosilicate glass). The residuals (PM mass) were weighted with a microbalance (GEC Avery, capable of weighing 0.1 mg). Then, the samples were prepared for analysis by ICP-MS by dissolving them in 30 mL HNO₃ (65%, Merck, Suprapur) and 10 mL HCl (30%, Merck, Suprapur) and digesting for 2 h by heating at 90 °C on a hot plate and waiting for 24 h at 20 °C). The samples were diluted to 100 mL with deionized water and filtered through 0.45 μm porosity PTFE filters. All procedures were also applied for the blank samples. Analysis of trace elements (Ag, Al, As, B, Ba, Be, Bi, Ca, Cd, Ce, Co, Cr, Cu, Dy, Er, Eu,

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