



# The geochemical fingerprinting of geogenic particles in road deposited dust from Tehran metropolis, Iran: Implications for provenance tracking

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## ABSTRACT

Earlier studies focused on tracking anthropogenic sources of toxic trace metals associated with urban deposited dusts, while research on the provenance of natural-sourced dust particles deposited in the urban environment is still rare. We studied the geochemical signature of geogenic particles in street deposited dust (SDs) of Tehran metropolis, Iran, in order to infer their provenance and intensity of post-deposition weathering. The chemical index of alteration (CIA) and Ce anomalies suggested that SDs are chemically immature and partially weathered. The normalized REE pattern and positive Eu anomalies in conjunction with  $\text{SiO}_2/\text{Al}_2\text{O}_3$  values implied the derivation of SDs from intermediate to fully mafic igneous rocks. The REE composition and  $\text{La}_N/\text{Sm}_N$ ,  $\text{Gd}_N/\text{Yb}_N$  and  $\text{La}_N/\text{Yb}_N$  values of Tehran SDs were similar to those found in a Post-Achaean Australian Shale (PAAS) reference, indicating that dust sources from local rocks of Archean greenstone shales in the north of Tehran were the dominant contributor to Tehran SDs. However, CaO enrichment of the SDs in comparison with PAAS implied dilution and mixing with windborne carbonates originating from Alborz formations, as well as from intrinsic anthropogenic sources of the urban environment. Abundance patterns of the major elements and REEs as well as scatter plots of light rare earth elements (LREE) to heavy rare earth elements (HREE) vs.  $\text{Eu}/\text{Eu}^*$  and  $\Sigma\text{REE}$  vs. CaO (%) confirmed that conclusion. These results improved our knowledge about the nature of SDs and have implications for further investigations of urban deposited dust.

## 1. Introduction

In recent decades, considerable attention has been paid to deposited street dust (SD) and its potential to affect the quality of air, runoff and soil, as well as endanger human health in urban environments (Calvillo et al., 2015; Dean et al., 2017; Padoan et al., 2017; Franco et al., 2017; Zafra et al., 2017). Although, SDs have a wide range of intrinsic (vehicular traffic, tyre tread, brake lining, road salt, building deterioration, construction and renovation, and adjacent industries) and extrinsic sources (displaced surface soil, plant and leaf litter, and atmospheric deposition) (Magiera et al., 2011; Ram et al., 2012; Norouzi et al., 2017; Modaihsh et al., 2017), most studies have attempted to identify intrinsic or anthropogenic sources of SD particles (Wang and Lu, 2011; Yuan et al., 2014; Zhang et al., 2012) and there have been a very few studies on geogenic sources of SD particles. Hopke et al. (1980) found that soil material formed approximately 75% of SDs, emphasizing the need to understand the geogenic sources of urban deposited dust as well as anthropogenic sources.

Due to the complex and highly heterogeneous nature of SDs,

tracking the geogenic provenance of SDs is complicated. Moreover, extrinsically-sourced particles may experience long or short-range aeolian transport that results in geochemical and morphological changes. Since REEs are not easily fractionated during sedimentation and weathering, REE patterns provide an index to average provenance composition (Moreno et al., 2006). As a consequence, geochemistry of REEs in combination with major and trace elements has been evaluated by many researchers in provenance tracking studies, particularly to understand the source of terrigenous sediments (Bhuiyan et al., 2011; Ali et al., 2014; Négrel et al., 2015; He et al., 2015; Um et al., 2017).

Source identification approaches employ elemental ratios, which are more representative than individual concentrations, as they are able to eliminate the dilution effects resulting from inputs of other components (Wei et al., 2004). For instance, Ferrat et al. (2011) studied different REE ratios in desert particles of the eastern Tibetan Plateau and found that a combination of  $\text{Y}/\Sigma\text{REE}$ ,  $\text{La}/\text{Yb}$ ,  $\text{Y}/\text{Tb}$ ,  $\text{Y}/\text{La}$  and the Eu anomaly could be used to distinguish possible sources. Armstrong-Altrin et al. (2015) applied different ratios of major and trace elements in combination with REEs and determined that continental slope

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sediments in the southwestern Gulf of Mexico were classified as shale and originated mainly from intermediate rocks. In contrast, the concentrations of trace elements in urban areas are highly influenced by anthropogenic inputs. For instance, antimony and copper may derive from brake-wear and mechanical abrasion of vehicle parts (Jiries, 2001; Daher et al., 2014) and lead is the most well-known gasoline additive (Sezgin et al., 2004; Al-Khashman, 2007). Therefore, identifying natural sources of urban SD particles using trace element ratios is questionable and thus should be eliminated from such studies.

The chemical composition of deposited loess and loess-like sediments is directly dependent on the mineral composition of the dust sources, post-depositional weathering process and transportation of sediments (Ahmad and Chandra, 2013; Babeesh et al., 2017). Hence, elemental ratios in combination with mineralogical studies have been used to reveal the nature of weathering and depositional environment at the source regions, which, in turn, is controlled by climatic and tectonic factors (Verma and Armstrong-Altrin, 2013; Schatz et al., 2015; Xie et al., 2018). Deposited urban dust particles are believed to be predominantly accumulated by sub-aerial deposition (Sutherland, 2003) and act approximately the same as loess deposits, so the same approach can be employed to detect how intensely SDs have been weathered.

In general, no single methodology yields unequivocal information regarding dust provenance (Marx et al., 2005). Hence, in the current study, a number of approaches were used to identify the provenance of geogenic particles in SDs, including the mineralogical and morphological study of particles in contribution with geochemical studies of REE and major elements. The objective of the present study is to determine the optimum combination of approaches which reliably establishes the dominant provenance of SDs. We need to especially consider and distinguish the effects of anthropogenic pollution on the geochemistry of SDs from those of climate and weathering. The focus of the present study is on the Tehran metropolis. The ambient deposited dust of such industrialized and populated cities can include high concentrations of elements and determination of background values provided by extrinsic sources assists environmentalists to set geogenic concentration thresholds for elements in the urban environment.

## 2. Study area

The study area is in the southern vicinity of Alborz folded highlands (Fig. 1). The facies younger than upper Precambrian observed in the Alborz include shallow evaporite deposition, dolomite, and limestone along with sedimentary facies of shallow trenches including clastic sediments, alkaline and sub-alkaline volcanic deposits, flysch and ophiolite (GSI, 2011). The predominant Precambrian rocks of Alborz area include continental carbonates such as dolomites, while upper Cambrian consists of clastic sandstone formations (GSI, 2011). These formations originated as continental sediments and granitic and metamorphic rocks. Dolomitic limestones of Elika formation (Mesozoic) and interactions of tuffaceous shale (Karaj formation), limestone, sandstone, siltstone, and conglomerate (Cenozoic) occurred in the north of Tehran (GSI, 2011).

Damavand stratovolcano, the highest peak of the Alborz Mountain range (hereafter referred to as “Alborz”), acts as a barrier against movement of air pollution and dust particles and lead to accumulation of dust particles on the south-western slopes of the mountain, where Tehran city is located. Loess deposits occur on the northeastern slope of the Alborz, 380 km northeast of Tehran (Khadjeh et al., 2004). Tehran, with > 8.2 million people (Statistical Center of Iran, 2011) is located on early quaternary and Pliocene alluvial and moraine deposits, resulting from river activities and seasonal inundations, extending toward the south from the foothills of the Alborz. The bedrock of Tehran is a Tertiary formation, mostly Eocene lava, outcropped in the mountainous areas in the north and Triassic and Cretaceous dolomite limestone in the eastern heights of the city. The study area was 100 km<sup>2</sup> of Tehran

plain with an average elevation of 1250 m above sea level (Fig. 1).

A significant portion of the southern and central areas of province includes highly erodible rocks. Much of the northern areas of the province include moderately erodible rocks, whereas resistant formations cover a few scattered areas in north and south of the province. Unplanned agricultural activities and expansion of impermeable surfaces resulting from construction activities in the area, intensify erosion in the southern and south-western area of Tehran province.

According to the Köppen climate classification, this region is semi-arid with continental climate characteristics. The annual temperature varies from 0 to 37 °C. The mean annual rainfall is 245.3 mm, mostly occurring in February and March (Iran Meteorological Organization, 2015). The prevailing westerly and south-westerly winds predominate in the upper atmosphere throughout the year, while local winds are mostly impacted by the topography. There are over 7000 industrial units in Tehran, with 30% and 54% of them located in west and south of the city, respectively.

## 3. Materials and methods

Thirty composite SDs were collected adjacent to the curb of the two sides of the street, by sweeping an area of about 30 m<sup>2</sup> using a plastic brush and shovel, based on EPA method AP-42 (Fig. 1). All samples were kept in pre-cleaned polyethylene bags and were transported to the laboratory. There were no dust grains coarser than 250 μm in SDs. Thus, extraneous matter such as paving stone and asphalt, small pieces of brick and concrete, leaves and other debris were removed using a 250 μm sieve. The texture of SDs was determined by the hydrometry method (Hakanson, 1980) and classified according to United States Department of Agriculture (USDA) classification system using Rockwork software (version 14). Afterward, samples were dried in an oven at 90 °C for 20 h. A 50 g aliquot of each of the 5 dried SDs selected for their spatial distribution in the study area were sieved into a series of sieves using shaker dry sieving (nylon material) for 20 min. The weight percentages of SDs composed of different size fractions were then determined.

Next, all dried and 250 μm sieved SDs were mixed with LiBO<sub>2</sub>/Li<sub>2</sub>B<sub>4</sub>O<sub>7</sub> flux. Crucibles were fused in a furnace. The cooled bead was dissolved in ACS grade nitric acid and analyzed by ICP-MS for REEs and major elements. Loss on ignition (LOI) was determined by igniting a sample split (250 μm) then measuring the weight loss. Ten surface soils were collected from the central district of Tehran. Each sample included at least two sub-samples collected from an area of 3 m<sup>2</sup> from top 0–5 cm soil using a small plastic shovel. Soil samples were prepared as were the SDs. A 0.5 g aliquot of each sample was digested in aqua regia at 90 °C in a microprocessor controlled digestion block for 2 h to obtain the pseudo-total concentration of major elements. The digested sample was diluted and analyzed using inductively coupled plasma mass spectrometry (ICP-MS). According to Shapiro-Wilk and Kolmogorov-Smirnov tests, the data for all major oxides in soil samples except for K<sub>2</sub>O, Na<sub>2</sub>O and MnO were normally distributed. Thus, the median concentrations of major elements in soil samples were used as local surface soil (LSS) reference in this study (Table 1).

The validity of determinations of concentrations of major elements and REEs was evaluated by co-analysis of a standard reference material (STD SO-19, Bureau Veritas Inc.). Two replicates were used to isolate sources of variation in the digestion process. The coefficient of variation (VC) was < 1%. Recovery (%) of major elements was generally > 98%. All REEs were recovered within 10% of SRM, except for Tb, Ho, and Lu, for which recovery was 89.4%, 89.2% and 88.7%, respectively. Blank samples were analyzed in the same manner as regular samples and concentrations of all REE and major elements were below detection. Multi-element standards including GXR1 and GXR4 from USGS from Geochemical Reference Materials and Certificates, were used as certified reference material for major element analysis in soil samples. The recovery of all major elements was > 80%, which is within the

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