Science of the Total Environment 639 (2018) 417–427

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Seasonal and spatial variations of magnetic susceptibility and potentially toxic elements (PTEs) in road dusts of Thessaloniki city, Greece: A one-year monitoring period

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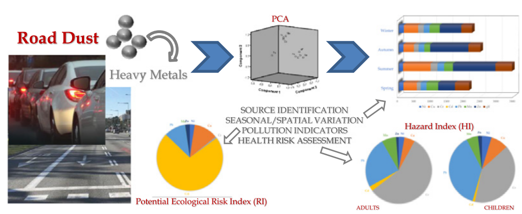
HIGHLIGHTS

- Elemental concentrations and magnetic susceptibility indicate a significant anthropogenic load.
- Three factors influenced heavy metal content, with traffic-related non-exhaust emissions contributed most.
- Seasonality for both geochemical and magnetic records were revealed.
- Road dusts indicate a high potential ecological risk, especially related to Cd.
- Cr and Pb were appeared as high-risk contaminants for children's health.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 31 January 2018
Received in revised form 19 April 2018
Accepted 11 May 2018
Available online xxxx

Editor: P. Kassomenos

Keywords:
Road dust
Heavy metals
Seasonality
Ecological risk
Health risk

ABSTRACT

A one-year sampling campaign of road dusts was carried out at 10 distinct sites in the broader area of the city of Thessaloniki, Greece and concentrations of heavy metals (HMs) along with magnetic susceptibility were evaluated. The concentrations of HMs in road dusts were higher than their local background values, while magnetic parameters indicated a significant anthropogenic load. Principal component analysis (PCA) identified non-exhaust vehicular emissions, oil/fuel combustion and industrial activities as major sources of heavy metals accounted for approximately 73% of the total variance. A significant seasonal variability for Cr, Cu, Mn, and χ_{lf} was observed with constantly higher values during summer. Moreover, variations among urban and industrial sites were more pronounced for Cr, Cu, Zn, and χ_{lf}, while they displayed insignificant variations across all urban sites. On the contrary, concentration peaks in the urban cluster were observed for Cd, Mn, and Ni coinciding with the port area. Based on multiple pollution indices, a severe polluted area was revealed, while potential ecological risk index (RI) indicated a high potential ecological risk with Cd being regarded as the pollutant of high concern. The health risk assessment model indicated ingestion as the major exposure pathway. For both adults and children, Cr and Pb had the highest risk values, mainly recorded in the urban cluster underscoring the need of potential measures to reduce road dust in urban environments.

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https://doi.org/10.1016/j.scitotenv.2018.05.170

1. Introduction

Road dust (RD) is a composite environmental medium consisting mainly of solid matter composed of both natural and anthropogenic constituents. Deposition of suspended matter, displaced soil, vehicular emissions (exhaust and non-exhaust), building deterioration, etc. contribute directly to road dust load (Liu et al., 2014).

RD emissions captured the interest of researchers as they have been widely documented to be the main components of atmospheric particulate matter (PM). RD dominates the total suspended particulate (TSP) mass, while it highly contributes to finer fractions (PM_{10} and $PM_{2.5}$) influencing their annual mean levels (Manoli et al., 2002; Pant and Harrison, 2013; Querol et al., 2008; Samara et al., 2003). Manoli et al. (2002) found that road dust dominated the coarse particle fraction (3.0–10 μm) in the center of Thessaloniki, accounting for 57% to its ambient levels, while it was also an important contributor (28%) to the fine size fraction (<3.0 μm). Moreover, a source apportionment study carried out in Thessaloniki (Samara et al., 2003), estimated that contribution of road dust to ambient PM_{10} at three sites within the city ranged between 19 and 22%. Moreover, lack of legislation concerning non-exhaust emissions and the prospect of non-exhaust particles to dominate both emissions and contributions to air quality over the coming years, raise concern over RD contributions (Amato et al., 2014).

RD is a source and a sink of multiple inorganic contaminants in urban environments (Christoforidis and Stamatis, 2009; Sysalova and Szakova, 2006). Heavy metals (HMs) as being potentially harmful elements (PHEs), and non-biodegradable, have attracted research attention. Over 400 articles in the SCOPUS database, the last 10 years, focus on heavy metals introduced into urban environment through RD particles yielding insight into the contamination level, the sources and the risks associated with heavy metals in road dusts. Different contamination and risk indicators (e.g. EF, I_{geo}, potential ecological risk) have been combined in order to provide a more reliable assessment (Aminiyan et al., 2017; Men et al., 2018; Trujillo-González et al., 2016). Moreover, the spatial variability of metal concentrations has been extensively documented (Aminiyan et al., 2017; Men et al., 2018; Trujillo-González et al., 2016), while in contrast the data concerning seasonal variability are rather limited (Fergusson and Kim, 1991; Massadeh and Snook, 2002; Pathak et al., 2013). The non-carcinogenic and carcinogenic risks to both adults and children through the exposure to road dust have also been evaluated (Men et al., 2018; Najmeddin et al., 2017). On the other hand, the advantages and mechanisms of the observed relationship between anthropogenic pollution and magnetic signature of road dusts and soils have been extensively reported in the international literature (Aidona et al., 2016; Bourliva et al., 2017b; Goddu et al., 2004; Jordanova et al., 2014; Tan et al., 2018; Yang et al., 2012; Zhang et al., 2011) confirming the effectiveness of magnetic mapping to delineate hot spots with increased anthropogenic load.

Previous sporadic studies conducted in Thessaloniki area reported elevated concentrations of heavy metals (Ewen et al., 2009; Misaelides et al., 1989; Samara et al., 2003), while a more recent one based on an intensive sampling in the city center focused on the mineralogical and morphological characteristics of the dust particles, on the chemical forms of heavy metals, and on a health risk assessment of the potentially toxic metals in road dusts (Bourliva et al., 2017a). In the present study, a temporal evaluation over a 12-month period has been performed at both urban and industrial sites allowing the assessment of the possible effects that arise from the choice of sampling location and the sampling season on heavy metal contents of road dusts. The main objectives of the present study are a) to estimate heavy metal contents and analyze the sources and their contributions to the heavy metals in RD, b) to describe spatial and temporal variations, and c) to evaluate contamination levels and associated health risks.

2. Materials and methods

2.1. Study area

The city of Thessaloniki (40°62′E, 22°95′N) constitutes a modern European commercial and cultural metropolis, and one of the most important trade and communication centers, situated in the heart of the Balkan Peninsula. With a population of over 1,000,000 inhabitants, it is the second largest city of Greece and one of the largest urban agglomerations in the Balkans. It is a coastal city surrounded by numerous residential suburbs while an extended industrial area is located northwesterly of the city. Moreover, at the west of the city and very close to the city center, operates the Thessaloniki's port, one of the largest ports in the East Mediterranean basin, with transport facilities for passengers (approximately 200,000 passengers/year) and goods (a total annual traffic capacity of 16 million tons). Despite the important size of the city, the absence of a developed public transport system (metro or tram) lead to increased road traffic into the city center of over 400,000 cars on a daily basis (Tolis et al., 2015b).

The climate in the broader Thessaloniki area is typically Mediterranean characterized by low precipitation levels (rain during 33% of the year) and low wind velocity values. The meteorological parameters during the campaign were available from the Meteorological Station of the Aristotle University of Thessaloniki. Temperature values ranged between 7.3 °C and 28.3 °C, while relative humidity exhibited a range of 32.4–67.8%. The prevailing wind directions during the sampling period were SE and SSE. The overall wind speed was at low levels during the whole sampling period with an average value of 2 m/s. The meteorological parameters per month for the one-year sampling period are given in Table S1 (Supplementary material).

2.2. Sampling locations and sampling methods

Road dust samples for a one-year period were collected, from March 2010 to March 2011 (a total of 120 samples) from the city of Thessaloniki, Northern Greece. The scheduled plan included a one-day sampling procedure every month (so as to avoid time variability among sampling sites) after a previous one-week (at least) dry period. Ten sampling locations, involving sites influenced by both urban and industrial activities, were selected as illustrated in Fig. 1. Specifically, eight sites were in the historic center of the city (U1-8), while two sampling sites (I1-2) were in the vicinity of specific industrial units (cement industry and electroplating plant). Additionally, road dust samples from two background sites (B1, B2) were collected (Fig. 1), at a distance from the city center but still in the urban area. In particular, site B1 was located in upper part of the city (around 174 m above sea level), in a less populated residential area with limited vehicular traffic, while site B2 was located in a peri-urban forest area. The dust samples were collected by sweeping an area of about 1 m^2 from road edges. Emphasis was given during sampling to suppress fine particles' loss by sweeping directly into a plastic bag. The amount of material from each sampling site varied between 50 and 150 g. After sampling, each sample was air-dried, mechanically sieved through a 0.5 mm sieve, divided into two sub-samples and stored at −4 °C. The first sub-sample was used to measure mass specific magnetic susceptibility (χ), while the other sub-sample was sieved through 0.063 mm sieve and the finer size fraction (<63 μm) was used for subsequent chemical analyses.

2.3. Analytical procedures

The mass specific magnetic susceptibility (χ) of dust samples was measured at low (0.46 kHz) and high (4.6 kHz) frequency using a Bartington MS2 laboratory magnetic susceptibility meter (Bartington Ltd., UK), equipped with a dual frequency MS2B sensor. Magnetic susceptibility value provides an indication of the concentration within the sample of strongly ferromagnetic minerals, such as magnetite.