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Assessment of atmospheric trace metal deposition in urban environments using direct and indirect measurement methodology and contributions from wet and dry depositions

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HIGHLIGHTS

- Dry, Wet and Bulk Atmospheric Depositions were measured in an urban residential area.
- Dry Depositions were measured using a new method based on ^{7}Be deposition velocities.
- ^{7}Be deposition method appears better than the subtraction Bulk minus Wet Depositions.
- The mean contribution of Dry to Bulk Deposition was significant for Zn, Cu, V and Pb.
- A detailed evaluation of uncertainties validates the method.

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ABSTRACT

Bulk Atmospheric Deposition (BAD), Wet Atmospheric Deposition (WAD) and Dry Atmospheric Deposition (DAD) were all measured within an urban residential area in Nantes (France) over a 9-month period (27 February - 10 December 2014). The objectives of this study were to compare 2 methods for measuring dry and wet atmospheric depositions in the urban environment (DAD and WAD: direct method; BAD and WAD: indirect one), and to characterize as well the variations and relative contributions of these depositions. Trace metals (As, Cd, Cr, Cu, Ni, Pt and V) were used to carry out these comparison and quantification. BAD was collected with two open polyethylene containers (72 × 54 × 21 cm), while WAD was collected by means of an automated rainwater collector and DAD was determined from both air measurements (recorded by an air sampler) and ^{7}Be deposition velocities. The comparison based on a detailed evaluation of uncertainties showed a significant difference between the direct and indirect methods. Dry and wet depositions varied widely from one month to the next. Zn and Cu were the most abundant elements in both dry and wet depositions. The mean contribution of DAD to the bulk atmospheric deposition during this 9-month study was significant for Zn, Cu and V (about 25%) as well as for Pb (approx. 60%). For this relatively unpolluted urban residential catchment, the contribution of atmospheric deposition to global load at the catchment outlet was low, between 10% and 20% for Zn, Cu, V and Pb, 25% for Cr and about 30% for Ni. For other urban sites exhibiting high atmospheric pollution however, the atmospheric contribution to the global pollution load could be much greater. An accurate and representative estimation of DAD thus proves critical.

1. Introduction

Air pollution has been on the rise over the past several decades and remains a major health hazard throughout the world (CITEPA, 2013; Dall'Osto et al., 2013; Hochsteller et al., 2011; Ravindra et al.,

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2006; Dall'Osto and Harrison, 2006; Shi et al., 2003). Atmospheric particles play a predominant role in pollutant transport mechanisms. Chemical elements attached to particles can be transported across long distances and transferred to ecosystems depending on meteorological conditions, the physical and chemical characteristics of particles, and the type of substrate (Connan et al., 2013). Aerosols have an impact on acid rain, plant photosynthesis and the hydrological cycle via their interactions with clouds (Tombette, 2007; Percot, 2012). The toxicity of aerosols depends on their composition and any trace metal loads; the presence of polycyclic aromatic hydrocarbons, endotoxins and other organic compounds may also influence their toxicity (Massamba et al., 2013). Among these sources, trace metal pollutants are still of major concern in urban environments (Gaspéri et al., 2014). The atmospheric pollutants in urban environments are either of natural or anthropogenic origin. Natural sources consist mainly of volcanoes, forest fires, soil erosion (especially in dry regions) and oceans (marine aerosols, coastal cities in particular). Among the anthropogenic sources are: road traffic, rail and air traffic, heating, industrial activities, building construction, agriculture, and incineration plants (Thorpe and Harrison, 2008; Gouzy and Ducos, 2008). These primary pollutants are supplemented by secondary pollutants (e.g. ozone, NO_2) resulting from either the chemical transformation of primary pollutants or reactions between the gases themselves (Azimi, 2004; Agglomération d'Avignon, 2014). The residence time of particles in the air mainly depends on their formation height, their composition and the micrometeorological conditions (primarily atmospheric turbulence). It also depends on size distribution of trace elements (Pan and Wang, 2015). This time is thus highly variable and difficult to calculate (Azimi, 2004; CFHA, 2007; Tombette, 2007).

Fine particles (i.e. with a mean diameter < 1 μm, PM_1) may be of natural or anthropogenic origin (Widory et al., 2004); in an urban environment, the anthropogenic origin would constitute the main source of this particle category. Moreover, fine particles can form in the atmosphere, in which case they would result from secondary chemical processes. Coarse particles ($PM_{2.5}$<x < PM_{10}) are often of mechanical origin (erosion, abrasion, construction, etc.). In the low troposphere, $PM_{0.1}$ (nanoparticles) and coarse aerosols (>PM_{10}) display the shortest residence time in air (a few minutes to a few hours), while aerosols with an aerodynamic diameter between 0.1 and 1 μm have the longest residence time in air (roughly 10 days) (Azimi, 2004; CFHA, 2007; Tombette, 2007). In the stratosphere, the residence time of atmospheric particles can last several years. Particles are removed from the atmosphere by means of either precipitation scavenging (wet deposition) or dry deposition processes (Connan et al., 2013).

The urban environment is very complex due to the juxtaposition of various surfaces such as grass, bitumen, glass, facade coating, slate, tile and zinc. Until now however, very few studies have taken this diversity into account when measuring dry deposition on urban surfaces (Roupsard, 2013; Percot, 2012; Roupsard et al., 2013; Percot et al., 2013). Dry deposition sampling remains a complicated procedure, although it has been significantly improved over the past thirty years. In the 1980's and 1990's, solid dry surfaces, e.g.Teflon trays, different types of filters, polyethylene or glass buckets/funnels and Petri dishes, were used for the direct measurement of dry deposition (Davidson et al., 1985; Noll et al., 1988; Davidson and Wu, 1990). Nonetheless, the surface characteristics of these types of collectors still exerted an impact on the deposition quantity. Greased surfaces were subsequently used to measure dry depositions (Shahin et al., 2002; Sakata and Asakura, 2011; Okubo et al., 2013; Guo et al., 2014). The results of this latter study plus the works using nearly the same collection methods (i.e. Yi et al., 1997a; Yi et al., 1997b; Odabasi et al., 2001; Shahin et al., 2002) indicated that greased solid surfaces provided more satisfactory results than dry solid surfaces, yet interaction problems remained between the surfaces used and the depositions, regardless of the fact that a single surface is not representative of the variety of urban surfaces.

The objective of this paper is to assess wet and dry atmospheric depositions on a small urban watershed using two methods to measure dry deposition: 1) the so-called “indirect method”, i.e.the subtraction of wet deposition (collected with an automated rain collector) from bulk deposition determined from a polyethylene container; and 2) the “direct method” (Percot et al., 2016) that takes into account the nature of urban surfaces (glass, tile, bitumen, zinc sheets, grass, facade coating, slate.) as well as long-term turbulence and local micrometeorology. To our knowledge, this study is the first one showing a comparison between 2 methods aimed at assessing atmospheric deposition and providing a detailed evaluation of uncertainties.

Trace metals (As, Cd, Cr, Cu, Ni, Pt and V) were analyzed. This study was carried out within the framework of the “Nantes Observatory of Urban Environments” (ONEVU), a long-term observatory dedicated to evaluating water, energy and pollutant fluxes in both urban and suburban systems.

2. Materials and methods

2.1. Experimental site

The sampling site was located on the flat roof of the 2-storey *Aquasim* Building at the CSTB (Scientific and Technical Center for Building Research) facility east of the city of Nantes, about 1 km from the ring road and 3 km from the Pin Sec catchment (31 ha, see Fig. 1). The land use in this zone is mainly composed of residential and tertiary buildings.

Meteorological data, including wind speed and direction, temperature, relative humidity and barometric pressure, were also measured during each sampling period using a meteorological station on the CSTB site. The climate in Nantes (47°13′N, 01°33′W, 50 km from the Atlantic coast) is oceanic. Prevailing wind directions were from the south-southwest and north-northwest sectors with maximum wind speeds typically from the southwest, while moderate and weaker winds most often blew from the west. Average annual precipitation over the past 30 years equals 802 mm (data provided by Météo France Weather Agency). The total rainfall recorded during the 9-month study period was close to this average, however August and November were very rainy while September was exceptionally dry (only 3 mm vs. an average of 70 mm).

2.2. Sampling procedure

Bulk Atmospheric Deposition (BAD), Wet Atmospheric Deposition (WAD) and Dry Atmospheric Deposition (DAD) were all collected over the course of 9 monthly campaigns conducted from 27 February to 10 December 2014. More precisely, the mean campaign duration lasted 29 days; for the sake of simplicity, the sampling period was assumed to extend from March to December. Furthermore, no sampling was performed in April due to technical problems.

2.2.1. Bulk deposition

Bulk deposition samples were collected by means of two open polyethylene containers (72 × 54 × 21 cm) set 1.1 m above ground level (Gaspéri et al., 2014). The containers were connected to 10-L polyethylene bottles (pre-cleaned with HNO_3 and MilliQ water). The samples were collected after each rainfall event, stored in 5-L