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Use of beryllium-7 as a surrogate to determine the deposition of metal and polycyclic aromatic hydrocarbon through urban aerosols in Nantes, France



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HIGHLIGHTS

- We measured ⁷Be, metals and polycyclic aromatic hydrocarbons (PAHs) in urban aerosols.
- ⁷Be has a unimodal distribution and is associated with submicron aerosols.
- The activity median aerodynamic diameter (AMAD) of ⁷Be varies from 0.50 to 0.59 μm.
- ⁷Be can be used as a surrogate to determine trace metal and PAHs deposition.
- Micropollutants have the same dry deposition velocity as ⁷Be.

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ABSTRACT

The aerodynamic size distribution of the natural radionuclide beryllium-7 (7Be) and associated trace metals, pesticides and polycyclic aromatic hydrocarbons (PAHs) in Nantes (France) was determined during 6 campaigns (conducted from June 2010 to March 2012) using a low-pressure impactor. The activity distribution of 7Be , as measured by a gamma ray spectrometer, was associated with the accumulation mode (with around 85% of the mass $< 1~\mu m$) and explained by post condensation processes on the airborne particle. The mass size distribution of heavy metals and PAHs was mainly focused on the accumulation mode as well. Trace metals generally show a unimodal distribution, although bimodal distributions for Zn, Cu, As and Cr could also be observed; these were attributed to re-suspension processes. For PAHs, the size distribution remained unimodal with a mass median diameter (MMD) ranging from 0.31 to 0.68 μm , thus confirming the urban origin. Pesticide concentrations in the atmosphere were low and did not allow determining the size distribution. Given their identical aerodynamic distribution as 7Be , trace metals and PAHs are expected to display a similar dry deposition velocity, which will then allow us to calculate the dry deposition fluxes over this urban area.

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1. Introduction

Good air quality is considered essential to human health. Nonetheless, air pollution remains an important problem throughout

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the world [...] and over 2 million people die prematurely each year due to poor indoor and outdoor air quality (WHO, 2000). The atmosphere is regularly contaminated by gaseous, liquid or solid pollutants. Atmospheric particles originate from either natural or anthropogenic sources (Slezakova et al., 2008; Thorpe and Harrison, 2008). Aerosols are included as atmospheric pollutants and exert multiple effects on human health and the environment (Hochstetler et al., 2011; Ravindra et al., 2006; Dall'Osto and Harrison, 2006; Shi et al., 2003; Bérubé et al., 1999). Moreover, atmospheric particles interact either directly or indirectly with the earth energy balance

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and can therefore alter the global climate (IPCC, 2007). The impact of dust aerosols on atmospheric chemistry can also be significant by virtue of providing a large surface area for heterogeneous reactions (Hochstetler et al., 2011).

In urban environments, atmospheric pollutants are either natural or anthropogenic in origin. Natural sources consist mainly of volcanoes, forest fires, soil erosion (especially in dry regions) and oceans (marine aerosols, especially in costal cities). Among the anthropogenic sources are road traffic, rail and air traffic, heating, industrial activity, construction sites, agriculture and incineration plants (Thorpe and Harrison, 2008). Along with these primary pollutants are secondary pollutants (e.g. ozone, NO₂) resulting from the chemical transformation of primary pollutants or from reactions between the gases themselves.

Urban aerosols are typically polydispersed, with particle sizes ranging from a few nanometers to several microns. Fine particles (i.e. with a mean diameter $<1~\mu m$, hereafter referred to as $PM_1)$ may be of natural or anthropogenic (combustion) origin (Widory et al., 2004); in an urban environment, the anthropogenic origin would be the main source of this particle category. Moreover, fine particles can be formed in the atmosphere, in which case they result from secondary chemical processes. Coarse particles (>1 μm , hereafter denoted PM > 1) are often of mechanical origin (erosion, abrasion, construction, etc.).

Beryllium-7 (7 Be) is a radioactive nuclide ($t_{1/2} = 53.3$ d) formed in the upper troposphere and lower atmosphere by means of spallation processes (Papastefanou and Ioannidou, 1995). Once formed, this nuclide rapidly associates with submicron aerosols, which constitute a major reservoir of pollutants in the atmosphere (Ioannidou, 2011, 2012). Its residence time in the troposphere is on the order of one week (Papastefanou and Ioannidou, 1995, 1996), and its concentration in the atmosphere depends, among other things, on precipitation, relative humidity, temperature and wind speed (Pinera-Garcia et al., 2012).

Numerous studies have already been conducted on atmospheric particles and urban aerosols, with an emphasis on large cities (Papastefanou and Ioannidou, 1995; Allen et al., 2001; Samara and Voutsa, 2005; Xie et al., 2005; Perez et al., 2008; Amato et al., 2009; Pey et al., 2010; Ioannidou, 2011). Few studies however have been devoted to small urban catchments.

A long-term study is currently being carried out on the "*Pin Sec*" catchment (Nantes, France) under the supervision of the Research Institute of Urban Sciences and Techniques (IRSTV), with the aim of assessing water and both pollutant and energy balances for a small urban catchment. In this context, special attention is paid to measuring dry deposition by means of ⁷Be.

The objective of this paper is to draw a comparison between trace metal, PAH, pesticide and ⁷Be aerosol size distributions and then prove that this radionuclide offers a good surrogate for quantifying dry pollutant deposition. The pollutants studied during this step are: trace metals, polycyclic aromatic hydrocarbons (PAHs), and pesticides.

2. Materials and methods

2.1. Study area

The sampling site was the small $Pin\ Sec$ catchment located east of the city of Nantes (see Fig. 1), which was urbanized mostly during the 20th century; this site is limited on the northeast by a tram line embankment. The land use is mainly composed of residential dwellings and 5-story multi-family buildings. Impervious surfaces, as estimated from Nantes' Metropolitan urban cadastral survey, cover approx. 45% of the catchment while the mean slope equals about 1.1%. The highest altitude of the catchment lies roughly 28 m above sea level in the northern part and the lowest is at $+17\ m$ to the southeast.

The climate in Nantes (47°13′N, 01°33′W, 80 km from the Atlantic coast) is oceanic. Prevailing wind directions come from the south-southwest and north-northwest sectors with maximum wind speeds typically from the southwest, while moderate and weak winds are most often from the west. Average annual

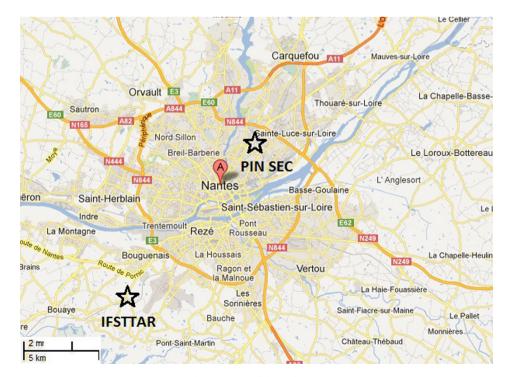


Fig. 1. Location of the study sites.

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