



# Influence of marine, terrestrial and anthropogenic sources on ionic and metallic composition of rainwater at a suburban site (northwest coast of Spain)



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## HIGHLIGHTS

- Chemical composition (major ions and trace metals) of rainwater was assessed.
- Major ions and trace metal wet deposition fluxes were obtained.
- Trace metals solubilities were assessed.
- Sources contributions of ions and metals from SF and NSF of rainwater were identified.
- Lineal regression between Al and Co concentrations in both rainwater fractions was found.

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## ABSTRACT

In the present research, the rainwater chemistry of soluble (SF) and non-soluble (NSF) fractions is studied over a one a half year period (from March 2011 to August 2012) at a suburban site (Oleiros, A Coruña, Spain). The monthly rainfall in this region during the studied period ranged from 10 to 137 mm, while the NSF ranged from 0.9 to 54 mg L<sup>-1</sup>. More rainfall occurs within October–January. Eighteen samples, which provide information pertaining to the monthly variation in chemistry, were analyzed. Trace metals (Al, As, Ba, Co, Cu, Cr, Fe, Mn, Ni, Pb, Sr, V, Zn) were enclosed in the study of both fractions of the rainwater. Major inorganic ions (Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup> and NH<sub>4</sub><sup>+</sup>) were also enclosed in the study of the SF of the rainwater. After partition coefficients analysis, univariate and principal components analysis (PCA) and air mass back trajectories analysis, three sources were found for the ionic and metal composition of the SF of rainwater; terrestrial (Ca<sup>2+</sup>, non sea salt SO<sub>4</sub><sup>2-</sup>, Al and Fe), marine (Mg<sup>2+</sup>, Na<sup>+</sup>, Cl<sup>-</sup>) and anthropogenic (K<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup>, Fe, Mn, Pb, Sr, V and Zn). Results also suggest ubiquitous sources for Ba, Co, Cu, Cr and Ni. One source (terrestrial) was found for NSF of rainwater.

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## 1. Introduction

Rainwater chemistry is being the subject of researches in all over the world because of increased concern for environmental problems caused by air pollution and acid rain. Those environmental problems are mainly consequence of the increase in energy consumption, derived from rapid economic development. Pollutants in the atmosphere stay there until they are removed by cleansing processes such as dry deposition, scavenging and washout by rain

(Church et al., 1990). Thus, rainwater acquires dissolved pollutants from the dissolution of gaseous and particulate material present in the atmosphere. Rainwater chemistry studies also allowing the knowledge of biogeochemical cycles of pollutants in the atmosphere; helps us to understand the relative contribution of different sources and fluxes of atmospheric pollutants, and provide information on the prevailing state of the atmospheric environment of that area.

In the last decade, the study of inorganic chemistry of rainwater collected at urban, rural and suburban sites in Iberian Peninsula has been reported (Alastuey et al., 2001; Ávila, and Roda, 2002; Hontoria et al., 2003; Vázquez et al., 2003; Ávila and Alarcón,

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2003; Encinas et al., 2004; Ávila and Rodrigo, 2004; Santos et al., 2008; Bacardit and Camarero, 2009; Calvo et al., 2010; Montoya-Mayor et al., 2011; Santos et al., 2011; Dueñas et al., 2012; Izquierdo and Ávila, 2012; Calvo et al., 2012).

Although the increasing concern about inorganic pollutants determination in rainwater in all over the world (including Spain), there are few studies on major ions (Vázquez et al., 2003) concentrations in rainwater deposited at suburban regions of the northwest of Iberian Peninsula. In addition, trace metals concentrations are not available. The aim of this paper is to understanding of the rainwater chemistry (including trace metals), to identify possible sources of major ions and trace metals that contribute to its chemical composition and finally to establish data of those species at a coastal suburban site of the northwest of peninsular Iberian, where data are not available.

## 2. Sampling and experimental methods

### 2.1. Details and description of the study area

The sampling site (Oleiros), a suburban area near to A Coruña, is situated on the northwest coast of Spain (latitude 43.2°N, longitude 8.17°W, at 120 m above the sea level). Sampling site is located in a residential area immediate vicinity is agricultural lands, forests and the sea (Fig. 1). The population of this town (33,765 (2008 census)), with low traffic which is increased at summer due to tourism (there are several beaches in the vicinity). The economic activity is very small (stores and shopping centers catering to the local population). Most locals work, study and do much of their daily activities in the neighboring city of La Coruña, located 8 km. Oleiros has experienced remarkable growth over the past 20 years, moving from a rural to a residential area. Climate of zone is humid oceanic, characterized by low thermal oscillation and abundant rainfall, where the prevailing winds are from the north in summer and south in winter.

### 2.2. Rainwater: collection and treatment

The rainwater samples were collected weekly with a wet collector (Sedimentary particles sampler MCV PS) supplied with a

bottle of 10 L (MCV SA, Barcelona, Spain) in University Institute of Environment (Pazo de Lóngora) located almost in the central part of the study area. Sample was filtered into pre-cleaned polypropylene bottles using pre-weighed Munktell quartz filters (Falun, Sweden) and solid and liquid phases were stored. After month ended, aqueous phases were combined and analyzed. By the same way, filters were combined and acid digested. The site is surrounded uniformly by almost all activities that exist in the study area. Samples were collected ~2.5 m above the ground level on the roof of an immission monitoring station using a 305 mm diameter PVC funnel, which was cleaned before collection. Particles adhering to the funnel were washed with a portion of sampled rainwater. The washing sample was collected in the same 10 L bottle used for sample collection. Then the bottle is removed and replaced by another clean bottle. 18 samples were collected from March 2011 to August 2012, which provide information pertaining to the monthly variation in chemistry. After total rainwater sample volume, pH and conductivity was measured. All the filtered samples were preserved at 4 °C in a refrigerator until subjected for major inorganic ion and trace metals analysis by CE and ICP-MS, respectively.

To avoid metal contamination during collection and analysis of precipitation samples, all plastic ware and glassware was washed with ultrapure water of 18 MΩ cm resistance (Milli-Q water-purification system, Millipore, Bedford, MA, USA) and kept for 48 h in 10% (v/v) nitric acid (ultraclean nitric acid 69–70% (Baker, Phillipsburg, PA, USA)), then rinsed several times with ultra-pure water before use. After collection, sample manipulation and analysis were carried out into a class – 100 clean room.

### 2.3. Solid fraction of rainwater: collection and treatment

After quartz filters were weighted (to achieve the total non-soluble masses), filter portions were transferred to a PFA bombs with the addition of 2.5 ml of nitric acid (Baker, Phillipsburg, PA, USA) and 5 ml of hydrofluoric acid (Baker), and heated at 90 °C for 24 h. The mixture was driven to dryness after the addition of 2.5 ml of perchloric acid (Baker) and 1 ml of nitric acid. Once total dryness was reached, 1.25 ml of nitric acid was added and the solution made up to 25 ml with Milli-Q water and stored at 4 °C until subjected for trace metals analysis by ICP-MS (Piñeiro-Iglesias et al., 2003).

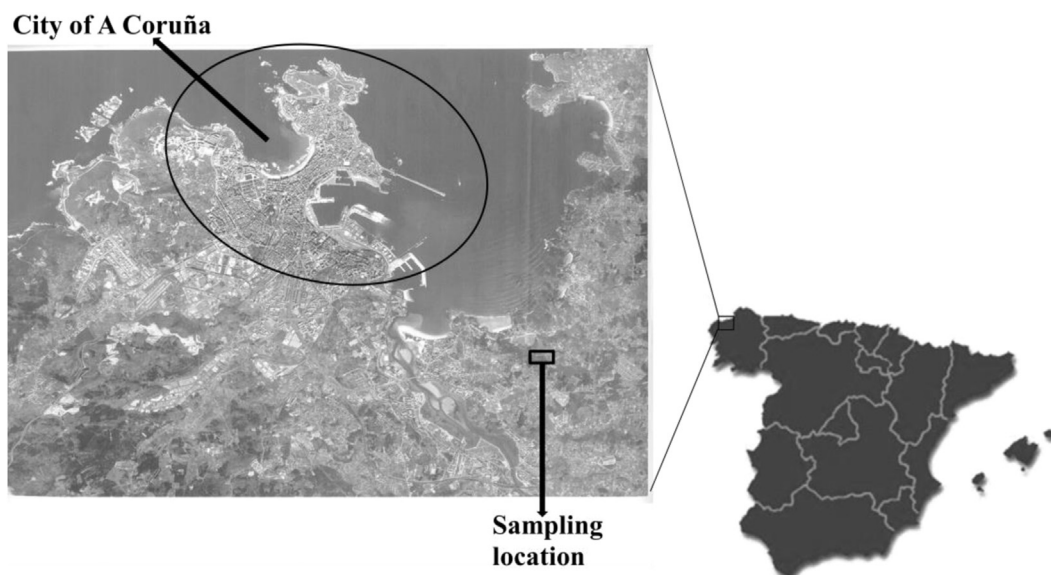


Fig. 1. Sampling location.

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