



## Rainwater chemistry at a Mediterranean inland station (Avignon, France): Local contribution versus long-range supply

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### ABSTRACT

In order to investigate the role of local contribution versus long-range transport in the rainwater chemistry at a site in the North-western Mediterranean area, we collected 90 daily wet samples, 75 dry samples (corresponding to bulk sampling during dry periods) and 102 sequential wet sub-samples from October 1997 to March 1999. Major ions were analyzed ( $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ,  $\text{HCO}_3^-$ ) along with the determination of pH and electrical conductivity. To discriminate the fraction of components supplied by long- and medium-range transport from that derived from local scavenging, a classification of precipitation chemistry according to air masses back trajectories and sources has been established. Three different origins were identified: (1) a Western one associated with high amount of rainfall and an equivalent contribution of terrestrial, marine and anthropogenic components, (2) a Northern and North-eastern origin characterized by low rainfall heights and a high load in anthropogenic and terrestrial components, (3) a Southern origin which presented the highest concentration in sea-salt components and a high participation of anthropogenic components. Determination of the local contribution has been achieved through the characterization of dry deposition rates and the investigation of the evolution of rain chemistry during events. As only 21 to 32% of the total concentration is removed in the first fraction, it appeared that the below-cloud scavenging process does not dominate the removal of atmospheric components in the region. The evolution of the rain chemistry during an event is perturbed by increases of concentrations that could be due either to long-range inputs (of humid air parcels coming from the near Mediterranean Sea or occurrence of Saharan dust) or to the scavenging of local sources of pollutants and terrestrial material.

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### 1. Introduction

The chemical composition of rainfall depends strongly on the atmosphere. The concentrations of chemical species in

precipitation vary widely in relation to a multiplicity of factors: type and distribution of aerosols sources, transport, chemical species and scavenging processes of species. Meteorological factors are often considered to be the predominant factor (Plaisance et al., 1997; Seto and Hara, 2006; Strayer et al., 2007) in relation with the two main processes (Mouli et al., 2005; Pelicho et al., 2006; André et al., 2007) that are usually described. The first process refers to the washout of the below-cloud atmosphere during precipitation events by raindrops which scavenge and dissolve particles and gases along their fall; the washout components are mainly of local/regional origin. The second process, called “rainout”, corresponds to

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the condensation of water vapor on aerosol particles during the formation of cloud droplets and incorporation of gases surrounding the droplets by aqueous-phase reactions. This process corresponds mainly to the incorporation of long-range transported components which may be traced back by air masses trajectories.

The atmospheric circulation over Western Europe is directly linked to oceanic circulation in the North Atlantic and to the associated position of the atmospheric and oceanic polar fronts. When the Azores high pressure cell is attenuated and latitudinally lowered, mainly in autumn–winter, rainfall fronts from the Atlantic Ocean cross the Iberian Peninsula from West to East before reaching the Western Mediterranean (Fons, 1979). Sporadic but intense winds from the Sahara desert, loaded with fine dust particles, blow towards the Western Mediterranean mainly from spring to autumn (Loÿe-Pilot and Martin, 1996). The Southern France is also affected by southerly airflow due to cyclonic circulation over the Mediterranean Sea. Precipitation related to these moist southerly winds becomes particularly important during autumn and spring. In winter and spring, an advection can occur from Central and Eastern Europe, produced by a depression crossing over Europe to the Italian Peninsula. These scarce situations give very little precipitation. There is then a broad diversity in the origin of the air masses associated with rainfall in the Western Mediterranean region. This topic has been widely investigated (Loÿe-Pilot et al., 1986; Loÿe-Pilot and Morelli, 1988; Rodà et al., 1990; Durand et al., 1992; Camarero and Catalan, 1993; Quereda Sala et al., 1996; Cénac et al., 1998; Ulrich et al., 1998; Balestrini et al., 2000; Panettiere et al., 2000). Three main sources of constituents strongly affect the composition of atmospheric aerosols and precipitation in the Mediterranean area: marine, terrigenous supplied by regional soil erosion or eolian dusts transported from North Africa and anthropogenic, delivering polluted aerosols from continental Europe (Loÿe-Pilot and Morelli, 1988; Bergametti et al., 1989; Gullu et al., 1998). The natural (marine, terrigenous, biological) and anthropogenic (industry, traffic, heating, agriculture) sources are difficult to discriminate, but their relative contribution can be estimated by a coupling of chemical composition and meteorological data (Chansin, 1988; Loÿe-Pilot and Morelli, 1988; Cénac et al., 1998; Avila and Alarcon, 1999).

The study of the chemical composition of the atmospheric deposition at the Mediterranean station of Avignon (South-eastern France) began in 1997 and was part of a more global project that aims at defining the aquifers input signal through chemical (major ions) and isotopic (oxygen-18 and deuterium) tools (Celle, 2000; Celle-Jeanton et al., 2001). The objective of the present paper is to determine the relative proportions of washout (dissolution of gases and impaction of particles contained in the air column below the cloud during the fall of raindrops) and rainout (incorporation of gases and aerosols into

the cloud during its formation and movement) to the rainfall chemistry. Thus the nature of the site is fundamental to the relative proportions of the two contributions: local incorporation could be of the utmost importance at a site with basic soils (Camarero and Catalan, 1993) providing a large neutralizing contribution. Contribution of long-range transport and local input has been evaluated by investigating the chemical content of rainwater and its variations during the rain events in relation with the air masses back trajectories and by determining the deposition rates of wet and dry periods.

## 2. Sampling and analyses

Wet and dry deposition samples were collected at the University of Avignon (43°57'N, 4°49'E, 30 m) from October 1997 to March 1999. The sampler was a plastic rain gauge with a 400 cm<sup>2</sup> aperture, placed 1 m above the roof of one of the university buildings. The bucket was kept clean and covered in the laboratory until installation, which occurred when weather forecasts predicted rainfall. This procedure was chosen to minimize dry deposition in wet samples although it could be not completely avoided. 90 wet samples were collected on a daily basis. During dry periods, the collector was continuously open to the atmosphere. At the end of the dry periods, the sampler was washed with 200 ml of deionized water to collect the dry deposition. The 75 dry samples corresponded to dry periods lasting from one to eleven days. After each sampling, the device was washed and thoroughly rinsed with deionized water and dried.

In order to investigate the local scavenging components and follow the evolution of the rain chemical composition along rain events, we implemented a sequential sampling of heavy rainfalls. In this specific purpose, a sequential collector was developed, based on a modified version of the Bourrié's device (Bourrié, 1978; Celle, 2000) and placed near the total rain collector. This collector of 962 cm<sup>2</sup> area is feeding a set of vials which are successively filled by the way of a three-way pipe. Mixing between successive vials does not exceed 5% (Lastennet, 1994). The first three vials of 150 ml each were followed by three vials of 500 ml, and two of 1000 ml, for a total volume of 3.95 l, enabling the collection of rains up to 41 mm in 8 fractions. This sequence of increasing sample volumes is best suited to detecting initial chemical variations.

Upon the sampling, the collector was taken to the laboratory where pH, electrical conductivity and HCO<sub>3</sub><sup>-</sup> were measured in unfiltered samples (rainwater for wet samples or distilled water for dry sample). Concentrations of bicarbonate were determined by titration with N/2000 sulfuric acid. After these measurements, samples were filtered through 0.4 µm pore size Nuclepore® filters and stored in dark at 4 °C for later analysis of the major ions (Mg<sup>2+</sup>, Ca<sup>2+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>,

**Table 1**

Major ionic constituents (µeq l<sup>-1</sup>) of the 90 sampled rains during the study period (Oct 97–March 99)

	Cl <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	HCO <sub>3</sub> <sup>-</sup>	Na <sup>+</sup>	K <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	NH <sub>4</sub> <sup>+</sup>	H <sup>+</sup>
Arithmetic mean	60.2	77.0	45.7	41.0	49.8	13.3	117.8	16.7	29.2	11.9
Weighted mean	37.2	47.0	24.0	13.0	30.7	5.7	51.7	10.0	14.1	22.6
Minimum	1.1	7.9	1.6	<LoD	0.4	<LoD	5.0	<LoD	<LoD	<LoD
Maximum	380.0	360.8	228.2	524.6	300.4	104.6	1047.0	85.6	273.1	100.0
Standard deviation	83.7	63.2	44.7	101.9	68.9	20.8	158.9	19.6	53.4	18.8

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