



# Aerosol and snow transfer processes: An investigation on the behavior of water-soluble organic compounds and ionic species



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## HIGHLIGHTS

- The air-snow transfer processes were evaluated using water soluble compounds.
- This is the first study about amino acids and sugar concentration in surface snow.
- Water soluble organic fractions of Antarctic aerosol and snow were investigated.

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## ABSTRACT

The concentrations of water-soluble compounds (ions, carboxylic acids, amino acids, sugars, phenolic compounds) in aerosol and snow have been determined at the coastal Italian base "Mario Zucchelli" (Antarctica) during the 2014–2015 austral summer. The main aim of this research was to investigate the air-snow transfer processes of a number of classes of chemical compounds and investigate their potential as tracers for specific sources.

The composition and particle size distribution of Antarctic aerosol was measured, and water-soluble compounds accounted for 66% of the PM<sub>10</sub> total mass concentration. The major ions Na<sup>+</sup>, Mg<sup>2+</sup>, Cl<sup>-</sup> and SO<sub>4</sub><sup>2-</sup> made up 99% of the total water soluble compound concentration indicating that sea spray input was the main source of aerosol. These ionic species were found mainly in the coarse fraction of the aerosol resulting in enhanced deposition, as reflected by the snow composition.

Biogenic sources were identified using chemical markers such as carboxylic acids, amino acids, sugars and phenolic compounds. This study describes the first characterization of amino acids and sugar concentrations in surface snow. High concentrations of amino acids were found after a snowfall event, their presence is probably due to the degradation of biological material scavenged during the snow event. Alcohol sugars increased in concentration after the snow event, suggesting a deposition of primary biological particles, such as airborne fungal spores.

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## 1. Introduction

Chemical markers provide unique information on the sources of aerosol and the chemical processes that took place during atmospheric transport. Their applicability to paleoclimatic studies of ice cores from polar regions, offers exceptional potential to evaluate the role and influence of aerosol in anthropogenic and natural forcing of climate change.

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Several studies have been conducted to evaluate the relationships between the concentrations of some markers in air and snow. So far, only the relationship between ionic compounds and carboxylic acids has been investigated since they are the most easily detectable in most matrices. Wolff et al. (1998) determined the most abundant ionic compounds in the aerosol, fresh snow and firn cores from three coastal Antarctic stations and highlighted the difficulties in studying processes in an area where environmental conditions change frequently. Investigations on the depositional fluxes of non-sea salt sulfate and methanesulfonic acid allowed an assessment of the spatial variation of marine biogenic sulfur (Minikin et al., 1998) to be made.

At Dome C on the Antarctic plateau, it was demonstrated that nitrate and chloride can be caught by the surface snow through dry deposition and adsorption processes (Udisti et al., 2004). The interactions of carboxylic acid at the air-snow interface has been studied in the Arctic (Narukawa et al., 2002), and underlined the compositional difference between aerosol and snow samples.

Ionic species such as  $\text{Na}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  are the most abundant compounds in Antarctic aerosol (Udisti et al., 2012) because the main source of aerosol in polar regions is fine sea spray emitted by bubble bursting because they are distant from any anthropogenic or continental emission sources.

Carboxylic acids (CA) may have different emission sources to the atmosphere (Kawamura et al., 1996a). Some anthropogenic organic pollutants that act as CA precursors, once emitted into the atmosphere they can undergo photochemical reactions or react with strong oxidants, such as ozone (Kawamura et al., 1996b). It is thought that the main natural source for CA is photochemical reactions after the release of alkenes by phytoplankton (Xu et al., 2013).

Several organic compound classes, such as sugars and amino acids, are emitted from biogenic sources into the atmosphere. Bioaerosol (bacteria, fungi, viruses, pollen, cell debris) contains primary saccharides such as glucose and sucrose (Medeiros et al., 2006). Arabinol and mannitol can be used as tracers for airborne fungal spores (Bauer et al., 2008), and are also the main carbohydrates in lichens and bacteria (Medeiros et al., 2006).

Levoglucosan, the most abundant anhydrosugar, is a specific atmospheric tracer of biomass burning (Simoneit and Elias, 2001). Some studies have demonstrated that the composition of phenolic compounds (PC) in atmospheric aerosol is linked to the type of vegetation that was burned (Simoneit, 2002), although Zangrando et al. (2016) have suggested that these compounds may have an additional marine source. A possible correlation between primary marine production and amino acids has also been identified (Barbaro et al., 2015b).

The main aim of this study was to investigate the behavior of water-soluble compounds during aerosol-snow transfer processes. Aerosol and snow samples were collected at Faraglione Camp, about 3 km from the Italian station "Mario Zucchelli", called MZS (Terra Nova Bay, Antarctica) during the 2014–2015 austral summer. Our research focused on several classes of compounds, such as ionic species ( $\text{Cl}^-$ ,  $\text{Br}^-$ ,  $\text{I}^-$ ,  $\text{NO}_2^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{PO}_4^{3-}$ , methanesulfonate (MSA),  $\text{NH}_4^+$ ,  $\text{Na}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ), CA ( $\text{C}_2$ -oxalic,  $\text{C}_2$ -glycolic,  $\text{C}_3$ -malonic,  $\text{C}_4$ -succinic,  $h\text{C}_4$ -malic,  $cis$ - $us\text{C}_4$ -maleic,  $trans$ - $us\text{C}_4$ -fumaric,  $\text{C}_5$ -glutaric,  $\text{C}_6$ -adipic,  $\text{C}_7$ -pimelic,  $a\text{C}_7$ -benzoic,  $\text{C}_8$ -suberic), sugars (monosaccharides, disaccharides, alcohol sugars and anhydrosugars), amino acids (AA) and PC (vanillin, syringyl and coumaryl compounds). This is the first study of potential relationships between sugars, AA and PC in aerosol and snow samples from the same site.

## 2. Material and methods

### 2.1. Collection and processing of aerosol samples

Aerosol samples ( $n = 7$ ) were collected using a multi-stage Andersen impactor (TE-6070, Tisch Environmental Inc., Cleves, OH, USA) placed at Campo Faraglione ( $74^\circ 41' \text{ S}$ ,  $164^\circ 06' \text{ E}$ ), 3 km south of the Italian MZS (Victoria Land, Antarctica) from the 6th of November 2014 to the 13th of January 2015.

Aerosol samples were collected on six pre-combusted (4 h at  $400^\circ \text{C}$  in a muffle furnace) quartz fiber filters with different cut-offs: the sampler accumulated particles with the following size fractions: 10.0–7.2  $\mu\text{m}$ , 7.2–3.0  $\mu\text{m}$ , 3.0–1.5  $\mu\text{m}$ , 1.5–0.95  $\mu\text{m}$ , 0.95–0.49  $\mu\text{m}$  on slotted filters and the  $<0.49 \mu\text{m}$  particle size on a

backup filter at an average sampling airflow of  $1.1 \text{ m}^3 \text{ min}^{-1}$ . Samples were collected for 10 days, as previous tests showed that this exposure time was necessary to quantify species at very low concentrations (Barbaro et al., 2015a, 2015b, 2016, 2017; Zangrando et al., 2016). Further sampling details are available in Table S2. Three field blank samples were collected by loading, carrying and installing the filter on the instrument with the air pump switched off, removing it after 10 min, following the same procedure used for installation and removal of the sample filters. Samples and blanks were wrapped in a double layer of aluminum foil and stored at  $-20^\circ \text{C}$  until analysis.

The pre-analytical protocol to determine the ionic species ( $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{NO}_2^-$ ,  $\text{SO}_4^{2-}$ , MSA,  $\text{I}^-$ ,  $\text{Br}^-$ ,  $\text{PO}_4^{3-}$ ,  $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ) and 12 CA ( $\text{C}_2$ -oxalic,  $\text{C}_2$ -glycolic,  $\text{C}_3$ -malonic,  $\text{C}_4$ -succinic,  $h\text{C}_4$ -malic,  $cis$ - $us\text{C}_4$ -maleic,  $trans$ - $us\text{C}_4$ -fumaric,  $\text{C}_5$ -glutaric,  $\text{C}_6$ -adipic,  $\text{C}_7$ -pimelic,  $a\text{C}_7$ -benzoic,  $\text{C}_8$ -suberic acids, Table S1) is described elsewhere (Barbaro et al., 2016). A quarter of each sample filter was broken into small pieces and placed into polyethylene tubes, using steel tweezers. All materials were previously cleaned by sonication with ultrapure water for 30 min at room temperature, and all sample handling was carried out under a class 100 laminar flow hood. Slotted filters were ultrasonicated for 30 min in 7 mL of ultrapure water, while backup filters were extracted with 15 mL of ultrapure water. Extracts were filtered through a 0.45  $\mu\text{m}$  polytetrafluoroethylene (PTFE) filter (ThermoFisher) to remove solid residues before analysis.

Half of each sample filter was used to determine AA, sugars and PC using a previously described method (Barbaro et al., 2015a, 2015b; Zangrando et al., 2016), where filter subsamples were spiked with a  $^{13}\text{C}$ -labelled internal standard mix (AA, PC and levoglucosan), ultrasonicated twice for 15 min in ultrapure water, and filtered through a 0.45- $\mu\text{m}$  PTFE filter before analysis.

### 2.2. Collection and processing of snow samples

Surface snow samples ( $n = 8$ ) were collected about 30 m from the aerosol sampling site. Snow samples were collected at 10-days intervals, when aerosol filter changing occurred (6 November, 17 November, 27 November, 7 December, 27 December 2014). Three consecutive snow samplings (10, 12 and 13 January 2015) were also performed during a snowfall event that lasted from the 8th to 11th of January 2015. Surface snow samples (ca. 1 cm in depth) were collected in 2-L polyethylene bottles, previously cleaned with ultrapure water and methanol. The samples were melted in an ISO 4 clean room and aliquoted into pre-cleaned vials prior to determination of ionic species and organic compounds. Before analysis for sugars, AA and PC the melted aliquots were spiked with a  $^{13}\text{C}$ -labelled internal standard mix.

### 2.3. Instrumental analysis

The quantification of all anions and CA was performed using an ion chromatograph (Thermo Scientific Dionex TM ICS-5000, Waltham, MA, USA), with an anion exchange column (Dionex Ion Pac AS11  $2 \times 250 \text{ mm}$ ) and guard column (Dionex Ion Pac AG11  $2 \times 50 \text{ mm}$ ), coupled with a single quadrupole mass spectrometer (MSQ PlusTM, ThermoScientific, Bremen, Germany). The ion chromatograph was equipped with a capillary system to simultaneously determine cations using a conductivity detector. Cations were separated using an Ion Pac CS19 – 4 mm capillary cation-exchange column ( $0.4 \times 250 \text{ mm}$ ) equipped with an Ion Pac CG19 – 4 mm guard column ( $0.4 \times 50 \text{ mm}$ ). Further details about the methods for anion and cation determinations are reported by Barbaro et al. (2016).

Quantifications of the AA were obtained using high-performance liquid chromatography with a chiral column

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