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Scavenging of black carbon in Chilean coastal fogs

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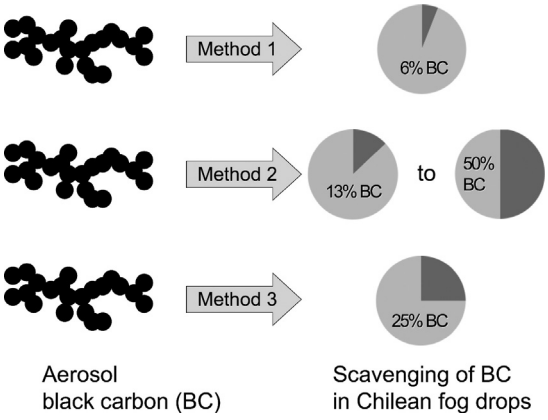
HIGHLIGHTS

- Black carbon and other particle components were measured in Chilean fogs
- Fog-scavenging coefficients for black carbon ranged from six to 50%
- Higher fog scavenging coefficients were determined for other aerosol components

GRAPHICAL ABSTRACT



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ABSTRACT

In November/December 2013 a pilot experiment on aerosol/fog interaction was conducted on a coastal hill in the suburbs of Valparaíso, Chile. Passages of *garúa* fog were monitored with continuous recordings of a soot photometer and an optical aerosol spectrometer. An optical fog sensor and an automatic weather station provided meteorological data with which the aerosol could be classified. High-resolution back trajectories added meteorological information. From filter samples, optical and chemical aerosol information was derived. Scavenging coefficients of black carbon (BC) and measured particulate mass below 1 μm diameter (PM1) were estimated with three approaches. Averaging over all fog periods of the campaign yielded a scavenging coefficient of only 6% for BC and 40% for PM1. Dividing the data into four 90°-wind sectors gave scavenging factors for BC ranging from 13% over the Valparaíso, Viña del Mar conurbation to 50% in the marine sector (180°–270°). The third, and independent approach was achieved with two pairs of chemical aerosol samples taken inside and outside fogs, which yielded a scavenging coefficient of 25% for BC and 70% for nonseasalt sulfate. Whereas fogs occurred rather infrequently in the beginning of the campaign highly regular daily fog cycles appeared towards the end of the experiment, which allowed the calculation of typical diurnal cycles of the aerosol in relation to a fog passage.

1. Introduction

Clouds strongly control the life cycle of the atmospheric aerosol (e.g., Heintzenberg, 2012). To some extent, the inverse also holds with aerosols controlling the life cycle of clouds (e.g., Siebesma et al., 2009).

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The formation of clouds leads to their scavenging of particles and gases. Through physical and chemical in-cloud processes, the size distribution of aerosol particles is altered and particulate mass may be added to the particles. In precipitating clouds, the opposite happens eliminating particles and gases from the atmosphere. Irrespective of these complex processes, cloud dynamics leads to some redistribution of the aerosol in the cloudy atmosphere.

The complicated interdependency of atmospheric clouds and aerosols has strong consequences for two major current atmospheric issues: climate change (e.g., Rosenfeld et al., 2014) and air pollution (e.g., Ramanathan & Feng, 2009). Anthropogenic aerosol changes affect climate both, directly, and through their interaction with clouds. Besides, in populated regions of the Earth anthropogenic air pollution with gases and particles is of major concern. For two reasons black carbon (BC) or soot is one of the most important anthropogenic aerosol components. One, the absorption of solar radiation due to BC in several compartments of the climate system is recognized as a major component in Earth's energy balance (Bond et al., 2013). Two, concerns in public health are related to toxic substances connected with BC (Davidson et al., 2005).

Fogs are low clouds in contact with Earth's surface where most of the anthropogenic sources of atmospheric pollution are located. Thus, the effects of fogs on atmospheric aerosols should be of major interest in fog prone areas. Nevertheless, the number of studies relating to the interaction of aerosol and fogs is rather limited. The chemical content of fog water has been studied for a long time (cf. the summary in Heintzenberg, 1992). Mainly two issues have caused that the uptake of BC in fogs has received less attention. Quantitative sampling of black carbon containing fog water is difficult due to the tendency of BC to stick to sampling surfaces (Ogren et al., 1985), which can only be avoided by sophisticated sampling equipment that prevents the fog water from contact with the sampler (Hallberg et al., 1992; Ogren et al., 1985). The second issue concerns problems related to the analysis of BC in general (Dlugi, 1989; Engström & Leck, 2011; Heintzenberg & Winkler, 1991; Ogren & Charlson, 1983, 1984; Ogren et al., 1984).

These technical problems combined with a high variability in fog processes and atmospheric pollution lead to highly variable results on BC scavenging in fogs. Smallest values of 5–6% have been reported by Hallberg et al. (1992) in polluted radiation fogs in Northern Italy and by Collett et al. (2008) in Californian radiation fogs. Later experiments with different instrumentation at the same site in Northern Italy yielded values around 39% (Gilardoni et al., 2014). In radiation fogs over Southern Germany Dlugi (1989) determined 18%, which is rather near the value of 26% given by Gundel et al. (1994) in Californian advection fogs. However, for similar Californian fogs and with another experimental approach Hansen and Novakov (1989) reported BC-scavenging fractions up to 80% and Benner et al. (1989) even 90% in laboratory fogs.

Because of the specific geographic and oceanographic settings fogs are very frequent in otherwise dry coastal regions of Chile (Weischet, 1966) and thus have been widely studied as a water resource for human consumption (Cereceda & Schemenauer, 1998; Schemenauer & Cereceda, 1994). There are several operational fog water sites in the country. With increasing water scarcity and/or population this method of providing water from fog is likely to be utilized more widely in Chile. However, only two chemical fog investigations directed at the quality of drinking water from collected Chilean fogs are known to the authors (Schemenauer & Cereceda, 1992; Sträter et al., 2010). In relation to air pollution fogs have not been studied in Chile while there are urban and rural areas where both phenomena coexist frequently. Below we report results of a pilot study for future systematic fog investigations concerning the scavenging of air pollution in Chilean fogs. This study comprised a simple field experiment on a coastal Chilean hill in the suburbs of Valparaíso. This area is frequently plagued by heavy fogs. Knoche (1931) reported more than 50 fog days per year and 25 to 50 days of the regional advected sea fog *garúa*. While providing the first data on aerosol composition near the Chilean coast the study was mainly aimed at adding new data on fog scavenging of BC.

2. Field experiment

On a hill in the suburbs of Valparaíso close to the Pacific coast a pilot experiment for the investigation of polluted fogs was set up (cf. Fig. 1). The instrumentation was mounted on the roof of the administration building of the Valparaíso Extension Zone Logistics Support, ZEAL (71.636° W, 33.0693° S) some 10 m above the ground level, which was about 450 m above sea level. The nearest distance to the Pacific is some 3 km towards the west. In the whole sector from about 300° through north to 90° in wind direction fogs having developed over the cold coastal waters of the Humboldt current pass over the cities of Valparaíso, Viña del Mar, Quilpué and their suburbs with about 700,000 inhabitants.

3. Instrumental and ancillary data

For the physical and chemical characterization of the aerosol in and about fogs, several sensors, samplers and analyses were combined.

3.1. Continuous recording of BC

A homemade continuous soot photometer (CSP) of the type aethalometer (Bond et al., 1999; Hansen et al., 1982) provided a continuous record of BC. A mass-flow controller maintained the sample flow to about 1 L min^{-1}. The CSP has been characterized and compared to other techniques of BC analysis by E. Engström (Engström & Leck, 2011; Engström, 2009). A homemade cyclone excluded particles (and fog drops) larger than 1 μm in diameter from the sample air. The cut-off characteristic of this cyclone was verified with two Grimm optical particle spectrometers of the type detailed below.

3.2. Combustion analysis of CSP filters

For the filters in the CSP quartz-fiber filters of the type PALLFLEX Tissuequartz® were chosen in order to be able to analyze them with conventional combustion techniques after the field experiment. With this post-experiment analysis elemental carbon (EC) and organic carbon (OC) were determined on each of the CSP filters thus establishing a calibration of the optical attenuation results of the CSP in terms of EC. The carbon analyses were made with a Sunsetlab OCEC Dual Optics Lab Instrument according to the EUSAAR-2 temperature protocol (Cavalli et al., 2010). According to the manufacturer the precision of the instrument measured as a relative standard deviation, typically falls into the 4–6% range. For the accuracy Sunsetlabs give relative standard deviations of less than 5% and for the OC/EC speciation 5–10%.

3.3. Collection of ambient aerosol particles with filter cassettes

For post-experiment optical and wet-chemical analysis 37 mm diameter filter-cassettes were preloaded with 0.4 μm pore size polycarbonate Nuclepore® filters. The exposed filter surface was masked to 8 mm diameter (0.5 cm^2). Engström and Leck (2011) describe the details of the setup. The geometric configuration of the sample spot on the filter surface was used to optimize the analytical conditions (i.e., the signal to noise ratio) for the post-sampling analyses (BC and particulate ionic mass) on the filters (Hansson et al., 1986). Two of these cassettes were mounted at the same time, each downstream of a cyclone of the same type as upstream of the CSP in order to exclude super-micrometer particles from sampling. A mass-flow controller maintained the sample flow of about 2 L min^{-1}. A fog sensor (see below) switched the sample pump between the two cassettes.