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Characterization of aerosols above the Northern Adriatic Sea: Case studies of offshore and onshore wind conditions

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HIGHLIGHTS

- This paper is a contribution of the study of coastal aerosol properties over the Mediterranean.
- We study the sea-spray and the anthropogenic contributions in the aerosol concentrations measured in the Mediterranean coast.
- One of the objectives is to implement an accurate source term for the sea-spray aerosols in the aerosol transport model.

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ABSTRACT

Aerosol particles in coastal areas result from a complex mixing between sea spray aerosols locally generated at the sea surface by the wind-waves interaction processes and a continental component resulting from natural and/or anthropogenic sources. This paper presents a physical and chemical analysis of the aerosol data acquired from May to September 2014 in the Adriatic Sea. Aerosol distributions were measured on the Acqua Alta platform located 15 km off the coast of Venice using two Particle Measuring System probes and a chemical characterization was made using an Ion Chromatography analysis (IC). Our aim is to study both the sea-spray contribution and the anthropogenic influence in the coastal aerosol of this Mediterranean region. To this end, we focus on a comparison between the present data and the aerosol size distributions measured south of the French Mediterranean coast. For air masses of marine origin transported by southern winds on the French coast and by the Sirocco in the Adriatic, we note a good agreement between the concentrations of super-micrometer aerosols measured in the two locations. This indicates a similar sea surface production of sea-spray aerosols formed by bubble bursting processes in the two locations. In contrast, the results show larger concentrations of submicron particles in the North-Western Mediterranean compared to the Adriatic, which result probably from a larger anthropogenic background for marine conditions. In contrast, for a coastal influence, the chemical analysis presented in the present paper seems to indicate a larger importance of the anthropogenic impact in the Northern Adriatic compared to the North-Western Mediterranean.

1. Introduction

The estimation of the atmospheric aerosol impact on climate remains an important scientific challenge (Intergovernmental Panel on Climate Change (IPCC)). In particular, in contrast with the radiative forcing attributed to greenhouse gases, the uncertainties related to aerosol radiative forcing remain very large (IPCC, 2013). This is due to their heterogeneous spatial and temporal distribution, their different origins (natural and anthropogenic) and their physical and chemical behaviour in the free troposphere. In coastal areas, aerosols may be of either natural or anthropogenic origin. The sea-spray aerosols represent a major contribution to the coastal aerosol mass (Yoon et al., 2007; Piazzola et al., 2009). Sea-spray

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particles result from the primary production through breaking waves (e.g., Monahan et al., 1986), which provide aerosols of diameters $\geq$0.5 μm. In addition, a generation of smaller particles results from secondary production due to gas-particle conversion (e.g., Fitzgerald, 1991). Sea-spray is made primarily of sodium chloride (NaCl) and small amounts of other salts such as sulfate, calcium and potassium, but it can also contain significant amounts of organic carbon. The organic component was mainly attributed to bubble bursting processes, due to the predominantly insoluble and surface active character of organic carbon in the marine aerosol particles (Ceburnis et al., 2008). Although it is well-recognized that marine aerosols have a significant influence on the coastal urban air quality through their ability to have chemical and physical interactions with gases and other aerosol species (Knipping and Dabdub, 2003), their interactions with anthropogenic pollutants in coastal areas are still largely unknown. In this context, we need first to obtain a good knowledge of the sea-spray generation processes and its atmospheric transport. To this end, Tedeschi and Piazzola (2011) developed the Marine Aerosol Concentration Model (MACMod) dedicated to the atmospheric transport of marine aerosols in the Marine Atmospheric Boundary Layer (MABL). Such a transport model needs to have accurate input conditions, as the source terms characteristics of the sea-spray production. However, uncertainties on the sea-spray source function (hereinafter S3F) are still large (De Leeuw et al., 2011). Demoisson et al. (2013, hereinafter, D13) recently proposed a new formulation of the S3F characteristics of the North-Western Mediterranean using the aerosol size distributions measured at different locations south of the French coast on board the ship Atalante in May 2008. The implementation of this latter sea-spray formulation in MACMod contributed to increase its performance for the Mediterranean (D13). One of the questions we need to reply is to what extent this formulation can be used in other Mediterranean areas. In addition, a better knowledge is required on atmospheric interactions between the sea-spray and other atmospheric compounds. Physico-chemical analysis of the aerosol in different Mediterranean areas is then needed.

The aim of this paper is to study the sea-spray contribution and the anthropogenic influence in the coastal aerosol measured in the Mediterranean. To this end, we used the aerosol size distributions acquired in the Northern Adriatic between May and September 2014 on board the Acqua Alta platform located near the coast of Venice. The results are compared to the aerosol size distributions recorded on board the ship Atalante south of the French coast in May 2008 that were used for the implementation of the S3F proposed by D13 for the Mediterranean. In addition, to evaluate the anthropogenic influence in the aerosols measured in the Northern Adriatic, we present a chemical characterization of aerosols concentrations. The composition of aerosols in the study area is also then compared to measurements made in May 2007 on the island of Porquerolles located south of the French coast (Piazzola et al., 2012; hereinafter P12). In Section 2, we describe the field sites of the present study, while Section 3 deals with the variation of the aerosol size distributions. Section 4 presents the chemical analysis, which allows for a good appreciation of the degree of anthropogenicity of the Western Mediterranean.

2. Field sites and experiments and general characterization of the areas

Our aim is to study the aerosol properties in the Northern Adriatic and compare the results to the data obtained south of the French Mediterranean coast. Alternatively, these two coastal regions are under influence of continental and marine air masses depending on the wind direction. The variation in wind direction can also be accompanied by changes of the fetch, which represents the distance over water for which the wind has blown constantly. Continental winds may then blow over the sea, giving the transported air masses a mixed character (defined as “coastal”). For our analysis, we have selected episodes characteristics of coastal and marine influence, i.e., short and large fetches, as found in coastal Mediterranean areas. Large fetches deal with sea conditions for which the waves tend to be fully developed. For the wind speed conditions analyzed in Section 3, this corresponds to fetches larger than 150 km, following the criterion proposed by (Hsu, 1986).

2.1. The Acqua Alta field site

Measurements in the Northern Adriatic took place on the Acqua Alta platform, which is an oceanographic tower (Fig. 1a) located in the Northern Adriatic Sea, 15 km off the coast of the Venice lagoon (Fig. 1b). The tower, which has been operational since the early 70s, is managed by the Institute of Marine Sciences - National Research Council (ISMAR-CNR) and commonly hosts a large variety of instruments. The main structure is a quadratic template jacket steel structure with four 0.60-m diameter main legs fixed to inserted steel pipes driven 22 m into the sea bed (Cavaleri, 2000). The aerosol probes (see below) were fixed on the second floor, at 7 m above marine surface level, on which there is a 5.5 $\times$ 4.0 m platform. These were connected to power generators and to an indoor room for data acquisition.

2.2. Sampling and analytical procedures

During the experimental campaigns conducted in the Northern Adriatic and in the North-Western Mediterranean, the aerosol data were acquired in the 0.1–45 μm size ranges using two particle measuring systems (PMS): one active scattering spectrometer probe (ASASP) and one classical scattering spectrometer probe (CSASP). The data were stored as the average over a 4-min interval. Prior to the experiments, the probes had been calibrated with latex particles of known sizes. It should be noted that a minimum of a factor of 3 is expected between the aerosol concentration simultaneously measured using two probes of the same type at the same location (Reid et al., 2006). To ensure a rather accurate comparison between the two sites (see Section 3), we used the same probes (not only the same model but the same devices) for the two experimental campaigns in order to have the same measurement error. However, the absolute concentration registered by a PMS probe may change over time and after servicing due to decreasing laser power or contamination of optics (Reid et al., 2006). The probes were then checked in the laboratory at the end of each series of experiments. The differences noted for each probe before and after the experiment were less than 15% for aerosol diameters smaller than 10 μm. This can be assumed as accurate since our aim was to study more specifically the sea-spray aerosols issued from bubble-bursting processes which deal, for a larger part, with particles of sizes smaller than 10 μm. Prior to the plots, all the aerosol size distributions were normalized to a relative humidity of 80%.

The experimental campaign in the Northern Adriatic consisted of three intensive measurement periods of 7–10 days between May and September. Among them, we have selected a number of ensembles of measurements performed during windy period of nearly constant direction (throughout the paper we use incoming direction). Each aerosol spectrum reported in Section 3 is an average of four to six aerosol size distributions.

For chemical characterization reported in Section 4, aerosols were sampled with a low pressure cascade impactor (Dekati) from 14.45 of 18th of June to 24.00 of 19th of June 2014 (about 33 h) and from 14.30 of 23rd of September to 12.00 25th of September 2014