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Original article

Detection and simulation of wildfire smoke impacting a Mediterranean urban atmosphere

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ABSTRACT

The combined use of chemical analysis of organic molecules in atmospheric aerosols (PM_1) collected in situ in Barcelona and optical measurements with a light detection and ranging (LIDAR) instrument allowed the characterization of the smoke plume from a wildfire that reached the city in July 2012. Analysis of the chemical composition of the aerosols collected on 23 July 2012 confirmed the large effect of biomass burning on urban air quality during a period of several hours. Typical biomass burning tracers, such as levoglucosan, dehydroabietic acid and polycyclic aromatic hydrocarbons (PAH) were enhanced at the same time as the aerosol concentrations in the boundary layer increased. According to air-mass trajectory modeling, the biomass burning particles originated from a severe wildfire burning 120 km northeast of the city. On the following days, no significant contribution of wildfire smoke was found in the urban air, although the lidar detected particles aloft. A Lagrangian particle dispersion model (FLEXPART) was used to simulate the transport of aerosols ($PM_{2.5}$) and carbon monoxide (CO), and the simulated concentrations in Barcelona were compared to in-situ measurements. FLEXPART simulated the onset of the wildfire smoke plume event in the urban center in the early morning of 23 July successfully; by contrast, the fast passage of the plume at the surface and the decoupling of the cleaner boundary layer from the persistent smoke plume aloft was not well captured. This was attributed to the fact that the model did not capture the local sea-breeze circulation well enough.

1. Introduction

Aerosols have adverse effects on human health (Pérez et al., 2009; Pope et al., 2002) and influence the Earth's radiative balance (Forster et al., 2007). In urban areas, and especially in the Mediterranean region, high background aerosol levels are often observed due to anthropogenic emissions (i.e. traffic, industry, etc.) and stable atmospheric conditions with high solar insolation, resulting in accumulation of pollutants within the planetary boundary layer (PBL) (Millán et al., 1997; Pérez et al., 2010; Pey et al., 2008). Moreover, the Mediterranean area is often exposed to Saharan dust outbreaks that occur about 15 times per year. During these events, the background aerosol levels are increased by between two and four times in the Western Mediterranean and more than ten times in the Eastern Mediterranean (Querol et al., 2009). Wildfires are also common in the Mediterranean area in summer and constitute an additional pollution source that further increases the aerosol levels (Phuleria et al., 2005; Rodríguez et al., 2002; Verma et al., 2009). Exposure to aerosols from wildfires has been associated to toxic effects such as lung cell damage and airway inflammation (Barrett et al., 2006; Thoring et al., 1982). Although these effects are not exclusively linked to biomass burning

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particles, there is evidence that they are more toxic than those from other emission sources on an equal-dose basis (Wegesser et al., 2009).

Recent studies for the urban area of Barcelona (Catalonia, Spain) showed a low-to-moderate impact of regional wildfire smoke and Saharan dust on the urban aerosol, which is normally dominated by local fossil fuel combustion sources (van Drooge et al., 2012). Simultaneous measurements of the vertical distribution of the aerosols in the atmospheric column showed that Saharan dust and wildfire smoke layers were mainly situated and transported in the free troposphere above the PBL (Sicard et al., 2011, 2012), with low impact on the air quality at ground level.

The present study focuses on the impact of a wildfire event that affected the urban area of Barcelona during a short period of time in July 2012 (see chronicle of wildfire in SI.1). On Sunday July 22nd (22J) a wildfire started along a highly transited highway near the border between France and Spain, in the Alt Empordà region, a forested area 120 km northeast of Barcelona. The fire burned for the next few days and was officially controlled on 25 July and completely extinguished on 30 July. As a result of the wildfire, four persons died, approximately 14,000 ha of mainly forested areas were burnt, as well as several farms and houses, and one camping site.

This paper describes the influence of the smoke plume generated by the fire on the urban aerosol concentrations in Barcelona by analyzing PM_1 samples and remote sensing lidar (Light Detection and Ranging) measurements, as well as satellite imagery and air mass trajectory modeling. The Lagrangian particle dispersion model FLEXPART was used (Stohl et al., 1998, 2005) to study the pathways of sampled air masses to the measurement station and to identify the contribution of the wildfire event.

2. Materials and methods

2.1. PM_1 filter sampling and organic tracer compound speciation

Atmospheric particulate matter with an aerodynamic diameter less than 1 μm (PM_1) was collected in the center of Barcelona (41°23′01.6″N; 2°10′45.8″E; 50 m above ground level (agl)) between 19 July and 24 July on pre-fired quartz filters (2500QAT-UP; Pallflex, Pall Corporation, USA) with 12 h time resolution (08:00 to 20:00 UT and 20:00 to 08:00 UT) using a Digitel-DH80 Hivol-sampler (Digitel Elektronic AG, Switzerland).

A detailed description of the applied analytical procedure is given in Fontal et al. (2015). Briefly, PM_1 filter samples were extracted using a mixture of dichloromethane and methanol (2:1 (v/v) (Merck, Germany)). The sample extracts were filtered over 0.45 μm teflon membrane filter (Whatman, USA) in order to remove particles and concentrated by vacuum rotary evaporation to 1 mL. Aliquots of 25 μL of the extract were evaporated under a gentle stream of N_2. Then, 25 μL of bis(trimethylsilyl)trifluoroacetamide (BSFTA) + 1% trimethylchlorosilane (TMCS) (Supelco, USA) and 10 μL of pyridine (Merck, Germany) were added for derivatization of the saccharides, acids and polyols to their trimethylsilyl esters at 70 °C during 1 h. Samples were injected in a gas-chromatograph (GC) coupled to a mass spectrometer (MS) (Thermo Trace GC Ultra – DSQ II, Thermo Scientific, USA) equipped with a 60 m fused capillary column (RXi® 5Si MS 0.25 mm × 0.25 μm film thickness; Restek, USA). A MS selective detector was operated in full scan (*m/z* 50–650) and electron impact (70 eV) modes. The applied method allows quantitative determination of carboxylic acids, dicarboxylic acids (and derivates), saccharides, aliphatic hydrocarbons (n-alkanes), polycyclic aromatic hydrocarbons (PAH) and hopanes. Method efficiency was assessed from recoveries of surrogate standards added to the filters prior to extraction. Mean recoveries were higher than 70% for all compounds. Field blank levels were low, always below 30% of the levels found in the samples. All reported concentrations were corrected by surrogate recovery and blank levels. Limit of quantification of the individual organic compounds was 0.06 ng/m^3 for sugars, 0.03 ng/m^3 for n-alkanes and 0.001 ng/m^3 for PAH and hopanes.

All statistical calculations were performed using the SPSS v.17 package (SPSS Inc., Chicago, III.). Unless otherwise noted, significance levels of the log–log correlations were set at $p < 0.01$.

2.2. Lidar measurements

Profiles of the aerosol stratification were obtained by the EARLINET (European Aerosol Research Lidar Network) lidar located in the center of Barcelona (41.39 N, 2.11 E, 115 m asl). The system employs a Nd:YAG laser emitting pulses at 355, 532 and 1064 nm at a repetition rate of 20 Hz. In reception, the backscattered elastic radiation and also the nitrogen (at 387 and 607 nm) and the water vapor (at 407 nm) Raman-shifted backscattered radiation is recorded. The vertical resolution is 3.75 m and the typical time resolution is 1 min.

A CIMEL sun-photometer, part of the Aerosol Robotic Network (AERONET), situated 600 m away from the lidar system was used to provide the aerosol optical depth (AOD) in the whole atmospheric column in the spectral bands of 440, 675, 870 and 1020 nm. On 23 July, the aerosol optical coefficient profiles (backscatter and extinction) were retrieved by averaging the two-components elastic lidar inversion algorithm (Fernald, 1984; Klett, 1985; Sasano and Nakane, 1984) using a constant lidar ratio of 50 sr. On 24 and 25 July, sun-photometer data were simultaneously available with the lidar measurements, so that the two-component elastic lidar inversion algorithm could be constrained with the sun-photometer-derived AOD (Reba et al., 2010). The retrieval of the PBL height was made using the gradient method (Sicard et al., 2006).

2.3. FLEXPART simulations

Forward simulations of the fire pollution plumes were made with the Lagrangian particle dispersion model FLEXPART (Stohl et al., 1998, 2005). The model was driven with ECMWF (European Centre for Medium-Range Weather Forecasts) operational analysis data with 0.18° × 0.18° latitude/longitude resolution, 91 vertical model levels and 3 h time resolution. Simulations were made both for a passive carbon monoxide (CO) tracer and a $PM_{2.5}$ aerosol tracer undergoing dry deposition and scavenging by precipitation.

The emissions used for the simulations were taken from information available on the fire. The coordinates of the starting point of the wildfire and the consecutive chronicle of its evolution through time and space were obtained from the Fire Brigade reports (see SI.1 for the translation of the report). Based on the geographical position of the fire and satellite images (see SI.2), the surface area of the wildfire was estimated for the consecutive days (SI.3). The time resolution in relation to burnt area was not conclusive, especially on 22 July, when the fire was most active (SI.2). Accordingly, two scenarios (plume 1 and 2) were adapted with different burning areas on 22 July between 14:00 and 15:00 h (plume 1 and 2: 33 km^2 and 24 km^2) and 15:00 and 17:00 h (plume 1 and 2: 9 km^2 and 19 km^2) (SI.3). The areas burnt consisted of Mediterranean forests (76%) and agricultural fields (19%), and a mixture of different surfaces, including urbanized areas (5%). For Mediterranean vegetation types, Alves et al. (2011) estimated emission factors of 231 g CO/kg dry biomass burnt, and 9 g/kg for $PM_{2.5}$. The biomass of