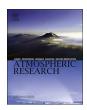
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Characterization of biomass burning from olive grove areas: A major source of organic aerosol in PM_{10} of Southwest Europe



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ABSTRACT

The inorganic and organic geochemistry of aerosol particulate matter (APM) was studied in a major olive grove area from Southwest Europe (Baena, Spain). The biomass consists of olive tree branches and the solid waste resulting of the olive oil production. Moreover, high PM_{10} levels were obtained (31.5 $\mu g m^{-3}$), with two days of exceedance of the daily limit of 50 $\mu g m^{-3}$ (2008/50/CE; EU, 2008) during the experimental period. A high mean levoglucosan concentration was obtained representing up 95% of the total mass of the isomers analysed (280 ng m⁻³), while galactosan and mannosan mean concentrations were lower (8.64 ng m⁻³ and 7.86 ng m⁻³, respectively). The contribution of wood smoke in Baena was estimated, representing 19% of OC and 17% of OM total mass. Positive matrix factor (PMF) was applied to the organic and inorganic aerosols data, which has permitted the identification of five source categories: biomass burning, traffic, mineral dust, marine aerosol and SIC (secondary inorganic compounds). The biomass burning category reached the highest mean contribution to the PM_{10} mass (41%, 17.6 μg m⁻³). In light of these results, the use of biomass resulting from the olive oil production for residential heating and industry must be considered the most important aerosol source during the winter months. The results of this paper can be extrapolated to other olive oil producing areas in the Mediterranean basin. Therefore, a fuller understanding of this type of biomass combustion is required in order to be able to establish appropriate polices and reduce the environmental impact on the population.

1. Introduction

The Mediterranean basin accounts for 5 Mha of the 10 Mha worldwide of cultivated area of olive groves comprising 850 million olive trees (IOOC, 2010). In the European Union, the olive grove plantations are the principal type of agricultural land use, which produces 91% (14 million tonnes) of the world's olive oil production (López-Piñeiro et al., 2011). The Mediterranean basin contributes up the 70% of worldwide olive oil production (3 \times 10 6 Mg per year, IOOC, 2010). The major producing countries are Spain (4.6 million tonnes), Greece (2.3 million tonnes) and Italy (2.0 million tonnes), which make up the 93% of the European crop (FAOSTAT, 2016). Olive groves are mostly located in rural areas, where air quality is characterized by low concentrations of gaseous pollutants and atmospheric particulate matter (APM) in terms of geochemical background compared to other zone categories (traffic, urban and industrial) (Lenschow et al., 2001). However, the common practice in rural areas of burning biomass for the

purposes of cooking and heating in the cold season has a negative impact on air quality (Smidt and Herman, 2004; Ancelet et al., 2013).

The general process of land clearing produces 95% of the global biomass burning anthropogenic emissions of atmospheric pollutants to the atmosphere. It comprises mainly forest transformation to agricultural and pastoral lands (Andreae and Merlet, 2001), disposal of agricultural wastes and home heating (Crutzen and Andreae, 1990; Schüle, 1990; Szidat et al., 2007; Sandradewi et al., 2008). However, the significance of APM emissions from these various types of biomass combustion is poorly quantified (Kuhns et al., 2004; Urbanski et al., 2011).

Emissions from biomass burning are the major source of primary carbonaceous aerosols (Bond et al., 2004), contributing up to 75% of the global combustion primary organic particulate matter. Combustion of olive tree branches is considered the most significant sources of fine aerosol during the winter months in many southern regions of Europe (Puxbaum et al., 2007; Gelencsér et al., 2007). The fresh particles from

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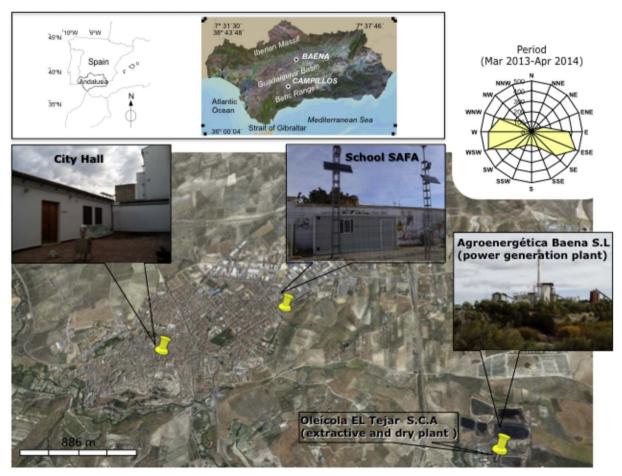


Fig. 1. Schematic map of the study area with location of monitoring sites and the power generation plant within the industrial complex. Instruments used in the study also appear in the pictures of the specific sites. City Hall: PM10 by high volume sampler. School: BC and ultrafine particles by CPC and MAAP instruments, respectively. The wind rose diagram for the monitoring period (from November 2013 to March 2014) is shown as an inset.

biomass burning are contained in the submicron size range and composed approximately of 80% organic compounds, 5–10% black carbon (BC) and 10–15% inorganic salts (potassium, sulphate, chloride and nitrate) (Reid et al., 2005). The main individual organic compound identified is levoglucosan, which results from thermal decomposition of cellulose (Shafizadeh, 1984; Shafizadeh et al., 1979). For instance, in Greece, 2300 \pm 600 tonnes of organic aerosol are emitted every year from the burning of branches. The composition of these fresh particles is 80% organic matter (OM), 8–10% black carbon (BC), 5% potassium, 3–4% sulphate, 2–3% nitrate and 0.8% chloride (Kostenidou et al., 2013). Papadakis et al. (2015) estimated the emissions from the burning of olive tree branches (a mean value of 5570 tonnes of fine organic aerosols and 230 tonnes of elemental carbon per year) using an emission inventory PMCAMx (three-dimensional chemical transport model) during the winter period.

The environmental and health effects of APM are related to their physical and chemical properties. Some studies have indicated a relation between particles generated from combustion processes and inflammatory, cytotoxic and genotoxic effects and also oxidative stress (Happo et al., 2013; Uski et al., 2014). Hata et al. (2014) identified that the size range of particles resulting from biomass combustion is < 100 nm (specifically 0.43 μ m), which contains high levels of toxic polycyclic aromatic hydrocarbons (PAHs) and water-soluble organic carbon (WSOC).

It is very important to highlight that in Spain, the world's leading producer of olive oil, few studies has been made concerning the chemical characterization of APM from this productive sector and still even less the influence of its emissions on air quality and human health.

Spanish olive groves dedicate a total of around 2.3 Mha to olive oil production and 170,840 ha to table olives, with Andalusia being the main producing region in the world with 1.5 Mha for olive oil and table olives production (AgroES, 2016). Olive oil manufacture generates large amounts of solid waste with high humidity (water content up to 80%), which is treated and transformed in olive oil refineries. The oil extraction process also produces large quantities of wastewater. In Spain, one million cubic meters of wastewater resulting from the process of washing and tanks are generated by oil mills every year. Current production techniques have adopted a two-phase system, which generates oil, waste water and a mixed solid-liquid waste called "alperujo" (a portmanteau from "orujo", essentially pulp, and "alpechin", the liquid waste). The alperujo is treated in secondary extraction plants to produce olive pomace oil. The dry residue resulting from this process ("orujillo") can be used in the production of biomass, agricultural fertilizer, dyes and antioxidants.

In recent years, several rural monitoring stations at olive groves in Southern Spain have registered poor air quality, specifically high number of days exceeding the PM_{10} (APM of aerodynamic diameter lower than $10\,\mu m$) daily limit value (50 μg m $^{-3}$, after the 2008/50/EC European Directive) (MAGRAMA, 2014). For this reason monitoring of air quality in olive groves in the Southern Europe (Baena, in the province of Cordoba, Spain) was performed between November 2013 and March 2014. Analysis of ultrafine particles (UFP) and black carbon (BC) was carried out using an Ultrafine Condensation Particle Counter (CPC), and a Multi-Angle Absorption Photometer (MAAP) to determine the chemical composition (organic and inorganic) of the PM_{10} . The chemical of organic aerosol is of critical interest to this study, due to the

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