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Ozone attributed to Madrid and Barcelona on-road transport emissions: Characterization of plume dynamics over the Iberian Peninsula

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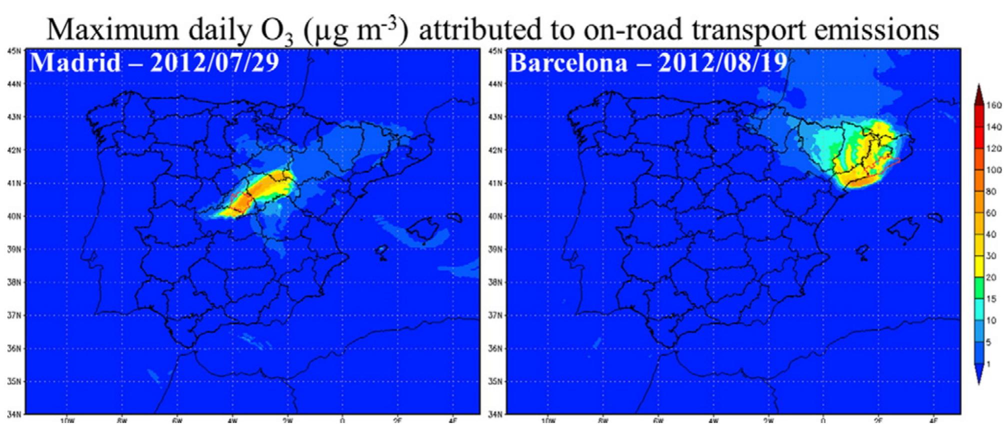
HIGHLIGHTS

- On-road transport is the main anthropogenic source of NO_x/NMVOCs in Spanish cities.
- Synoptic and mesoscale dynamics drive Madrid and Barcelona O_3 plumes, respectively.
- Less than 25% of daily O_3 concentration is attributed to urban traffic emissions.
- Long-range transport towards Spain is the main source of O_3, especially in winter.
- The O_3 attributed to traffic emissions explains the concentration peaks in summer.

GRAPHICAL ABSTRACT



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ABSTRACT

Despite the ~30% emission decrease of the main tropospheric ozone (O_3) precursors in Spain in the 2001–2012 period, the O_3 concentration in summer still exceeds the target value for the protection of the human health of the Air Quality Directive (2008/50/EC). On-road transport is the main anthropogenic contributor to O_3 precursor's emissions in Madrid and Barcelona metropolitan areas (65%/59% of NO_x, 40%/33% of NMVOC, and 67%/85% of CO emissions) but this contribution to O_3 formation is not well understood. The present work aims at increasing the understanding on the role of on-road transport emissions from main Spanish urban areas in O_3 dynamics over Spain under typical circulation types. For that purpose, the Integrated Source Apportionment Method is used within the CALIOPE modelling system (WRF/CMAQ/HERMES/BSC-DREAM8b). The results indicate that the daily maximum O_3 concentration attributed to the on-road transport emissions from Madrid ($O_{3T\text{-}MAD}$) and Barcelona metropolitan areas ($O_{3T\text{-}BCN}$) contribute up to 24% and 8% to total O_3 concentration, respectively, within an area of influence of 200 km. The contribution of $O_{3T\text{-}MAD}$ and $O_{3T\text{-}BCN}$ is particularly significant (up to 80–100 μg m^{-3} in an hour) to the O_3 concentration peak during the central hours of the day in the high O_3 concentration season (April–September). The maximum $O_{3T\text{-}MAD}$ concentration is calculated within the metropolitan area of Madrid but the plume, channelled by the Tajo and the Henares valleys, affects large areas of the Iberian Peninsula. The $O_{3T\text{-}BCN}$ plume is more driven by sea-land and mountain-valley breezes than by the synoptic advection and its maximum concentration is usually registered over the Mediterranean Sea.

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The O_3 concentration transported long-range to the Iberian Peninsula is significant in the area of influence of Madrid and Barcelona, being maxima under cold (70–96%) and minima in warm circulation types (35–70%).

1. Introduction

Tropospheric ozone (O_3) is a matter of concern because it is responsible of ~17,400 premature deaths each year in the European Union (European Environmental Agency, 2014a), it is a greenhouse gas with positive radiative forcing (0.40 ± 0.2 W m^{-2}, Myhre et al., 2013) that favours the global warming, and it is a powerful oxidant that can damage vegetation and reduce crop productivity (Chuwah et al., 2015).

O_3 is photochemically produced close to the ground from a series of precursors emitted by anthropogenic and natural sources, namely nitrogen oxides (NO_x), non-methane volatile organic compounds (NMVOC), and carbon monoxide (CO). Moreover, O_3 is transported over intercontinental scales favoured by its lifetime (c.a. 25 days) in the atmosphere (European Environmental Agency, 2013; Monks et al., 2014). Wild and Akimoto (2001) have suggested that Europe is the continent of the Northern Hemisphere most prone to be affected by intercontinental effects due to rapid transport of O_3 and its precursors from North America.

In Europe, the O_3 rural background concentration doubled between the 1950s and 2000 (Cooper et al., 2014) and it has had a negative trend (−1.44%) in the 2003–2012 decade (European Environmental Agency, 2014a). However, the target value for the protection of the human health, as defined by the Air Quality Directive 2008/50/EC, is still exceeded in large areas of Southern and Central Europe where high temperature and solar radiation occur in summer (European Environmental Agency, 2013). In Spain, there is an upward trend in O_3 concentration in the last decade (2001–2012), which is in line with a decrease of NO_x emissions and with an increase in surface solar radiation (Querol et al., 2014; Santurtún et al., 2015). On average for the 2010–2012 period, the O_3 target value was exceeded in 48 of the 135 Spanish air quality zones (close to urban and industrial areas, mainly in the center and south of the country and along the Mediterranean coast, Ministerio de Agricultura, Alimentación y Medio Ambiente MAGRAMA, 2013) leading to an exposition to O_3 concentration above the target value of 14.5% of the urban population (European Environmental Agency, 2014b).

In Spain, the emission of NO_x, NMVOC and CO has decreased by 32%, 38%, 27%, respectively, in the 2001–2012 period due to the effect of emission reduction policies and from 2008 also as a consequence of the economic crisis (European Environmental Agency, 2014b). The on-road transport is the anthropogenic activity with the largest contribution to the O_3 precursor's emissions in Madrid and Barcelona cities, the two biggest Spanish urban areas. However, there are not studies quantifying the contribution of on-road transport emissions from main Spanish urban areas to O_3 concentration in Spain.

Identifying the activity sources and the areas contributing to pollutant concentration is a fundamental task to design and implement effective abatement strategies and air quality plans as stated in the Air Quality Directive 2008/50/EC. Source apportionment modelling techniques such as the Integrated Source Apportionment Method (ISAM) within the Community Multi-scale Air Quality Model (CMAQ) can be used to analyse the contributions of different areas and sources to O_3 pollution and support the diagnosis of the origin of the problem (Cohan et al., 2005; Tran et al., 2014; Kwok et al., 2015). Furthermore, in order to establish efficient mitigation measures that minimize the impact of O_3 on population and ecosystems, it is vital to understand the O_3 transport dynamics which depend on meteorological conditions (Kassomenos et al., 1998; Jacob and Winner, 2009; Santurtún et al., 2015).

The objective of the present work is twofold. First, to determine the contribution of on-road transport emissions to the surface O_3 concentration within the area of influence of Madrid and Barcelona O_3 plumes. Second, to characterize the O_3 plume dynamics attributed to urban on-road transport emissions in Madrid and Barcelona areas under the typical synoptic conditions that affect the Iberian Peninsula (IP).

The paper is organized as follows. Section 2 describes the days of study, the target areas as well as the CALIOPE air quality forecast system (CALIOPE-AQFS) and its setup for O_3 source apportionment studies. Section 3 quantifies the relative contribution of Madrid and Barcelona on-road transport emissions to the total O_3 concentration under typical circulation types (CTs), and it analyses the O_3 plume dynamics attributed to those sources over the IP. Finally, conclusions are given in Section 4.

2. Methods

2.1. Days of study

The present work is performed for one representative day of each of the six synoptic circulation types that typically affect the IP (Table 1). These typical CTs and their representative day were identified by Valverde et al. (2014) using an objective synoptic classification derived for air quality purposes over the present climate (1983–2012). Despite the statistical limitation of using a single day to analyse pollution dynamics valid for a complete CT, the synoptic conditions on the representative day have proven to consistently represent those of the CT (Valverde et al., 2014) and are therefore useful for our purposes. In the resulting six CTs, the advection towards the IP depends on the location and strength of the actions centres that control the circulation at synoptic scale over South-Western Europe, mainly the Azores high and the Scandinavian low.

2.2. Target areas

Madrid and Barcelona metropolitan areas are taken as source areas to be tagged because they are the two biggest cities in the country in terms of population (6 and 5 million inhabitants, respectively) and number of vehicles (13.7% and 11.4% of total vehicles in Spain in 2012 in Madrid and Barcelona provinces, respectively; Dirección General de Tráfico, DGT, 2015). Furthermore, those areas have dissimilar topographic constraints and weather conditions that can lead to different O_3 plume dynamics. On the one hand, in the Madrid area the climate is Mediterranean continental and the main topographic features are the Tajo valley in the S/SE and the Guadarrama mountains (2200–2400 masl) with a NE–SW orientation, north of the metropolitan area of Madrid (Fig. 1c). On the other hand, the Mediterranean climate in the Barcelona area is strongly influenced by its complex topography (Fig. 1d) with two sets of mountain ranges parallel to the coast, the Pre-coastal (1000–1500 masl) and the Coastal chains (~500 masl).

Madrid and Barcelona metropolitan areas have been defined according to the Spanish Ministry of Public Works and Transport (Ministerio de Fomento MFom, 2013) which define the boundaries taking into account the number of inhabitants per municipality, population density, demographic and commuting dynamics, and transport networks (Madrid ~1000 km^2; Barcelona ~3200 km^2; Fig. 1b). On-road transport in Madrid and Barcelona is the most important contributor to the overall emissions of O_3 precursors. In the Madrid metropolitan area, the total on-road transport emissions fluxes range 22–36 kg_{NO2} day^{-1} km^{-2}; 5.8–9.7 $kmol_{NMVOCs}$ day^{-1} km^{-2}; and 1.7–2.8 kg_{CO} day^{-1} km^{-2}; the same order of magnitude as in the Barcelona metropolitan area where they range 22–31 kg_{NO2} day^{-1} km^{-2}; 4.8–6.2 $kmol_{NMVOCs}$ day^{-1} km^{-2}; and 1.9–2.8 kg_{CO} day^{-1} km^{-2} (Guevara et al., 2014). In Madrid/