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Atmospheric composition in the Eastern Mediterranean: Influence of biomass burning during summertime using the WRF-Chem model

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HIGHLIGHTS

- The Etesian flow pattern determines the distribution of aerosols over the AS.
- The Etesian flow pattern determines the paths of biomass burning (BB) emissions.
- The BB activity increases $PM_{2.5}$ concentrations up to 5.8 μg m^{-3}.
- The effect of the biogenic emissions is evident inside/above the PBL.
- Simulations with MOZART boundaries increase O_3 by 50%.

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ABSTRACT

The composition of the atmosphere over the Aegean Sea (AS) during an 'Etesian' outbreak under the influence of biomass burning (BB) activity is investigated. Simulations with the fully coupled WRF-Chem model during the Aegean-GAME campaign (29/8-9/9/2011) are used to examine the BB effect over the region. Two distinct Etesian flow patterns characterized by different transport conditions are analysed. The influence of the off-line calculated BB emissions on the atmospheric chemical composition over the AS under these conditions is estimated. In addition, sensitivity runs are used to examine the influence of the biogenic emissions calculated on-line and the realistic representation of the stratosphere-troposphere exchange processes are investigated through the time-varying chemical boundary conditions from the MOZART global chemical transport model. The horizontal and vertical distributions of gaseous and aerosol species are simulated under long-range transport conditions and interpreted in relation to the evolution of the Planetary Boundary Layer (PBL). In the case of a weaker synoptic system (medium-range transport conditions), even a small variability of meteorological parameters in limited areas become critical for the spatial distribution of gases and aerosols. The BB activity increases O_3, $PM_{2.5}$ and organic matter concentrations up to 5.5 ppb, 5.8 μg m^{-3} and 3.3 μg m^{-3}, respectively. The spatial extent of the simulated BB plumes is further examined by comparison with airborne measurements of hydrogen cyanide (HCN). The estimated effect of biogenic emissions on O_3 and $PM_{2.5}$ concentrations is either positive or negative (±6 ppb for O_3 and up to ± 1 μg m^{-3} for $PM_{2.5}$) depending on the emission algorithm employed. The realistic representation of the chemical boundary conditions reproduces an observed layer rich in O_3 above 4 km, but also increases O_3 concentrations inside the PBL by up to 40%.

1. Introduction

The Mediterranean is at a crossroad of air masses coming from Europe, Asia and Africa (Lelieveld et al., 2002), where

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anthropogenic emissions blend with gaseous and aerosol species from natural and biomass burning (BB) sources (Monks et al., 2009). In particular, during the warm period, a persistent northerly flow over the Eastern Mediterranean known as "Etesian winds" (Tyrlis et al., 2014) favours the transport of polluted air masses originating from northeastern Europe, Russia and the Black Sea, yielding high ozone (O_3) and particulate matter (PM) concentrations that often exceed air quality limits (Gerasopoulos et al., 2006a, c).

Many studies have focused on understanding the physical and chemical mechanisms leading to O_3 enhancement under the Etesian regime using airborne measurements (e.g., MINOS and PAUR; respectively Lelieveld et al., 2002; Zerefos et al., 2002), and atmospheric models (Roelofs et al., 2003). The high summertime O_3 concentrations observed in the boundary layer of the Eastern Mediterranean has been attributed to long-range transport (Gerasopoulos et al., 2006b; Kourtidis et al., 2002; Kouvarakis et al., 2002) and to anticyclonic influence in combination with the persistent northerly flow over the Aegean Sea (AS) (Kalabokas et al., 2007), which is a subregion of the Mediterranean basin north of Crete (Schicker et al., 2010). The significant role of the entrainment of air masses rich-in-O_3 from the upper troposphere has also been pointed out from the analysis of ground measurements (Gerasopoulos et al., 2006b, c), modelling results (Roelofs et al., 2003; Safieddine et al., 2014; Zanis et al., 2014; Zerefos et al., 2002), observed vertical profiles in the framework of the Measurement of Ozone and Water Vapour by Airbus in Service Aircraft project (MOZAIC) (Kalabokas et al., 2013) and satellite observations (Doche et al., 2014). Although modelling studies have investigated the contribution of different emission sources on summertime O_3 (Curci et al., 2009; Im and Kanakidou, 2012; Richards et al., 2013; Safieddine et al., 2014), the efficiency of photochemical processes inside the boundary layer is still not adequately quantified (Zanis et al., 2014).

A number of long- and short-term ground measurements of the properties of PM have also been conducted in the area (e.g., Gerasopoulos et al., 2006a, 2007; Hildebrandt et al., 2010; Kalivitis et al., 2008; Koçak et al., 2011; Kopanakis et al., 2013; Lazaridis et al., 2006; Querol et al., 2009). The interpretation of these measurements has focused on their daily and seasonal variability, PM chemical composition and on the investigation of natural events (i.e., mainly mineral dust events observed during spring and autumn). The vertical profile of aerosols, which has mainly been based on satellite and ground-based remote sensing observations (e.g., Gerasopoulos et al., 2007; Kalivitis et al., 2007) has been investigated to a lesser extent. Pollution events of fine aerosols during summer have been associated with transport from Central, Northern and Eastern Europe and Istanbul (Astitha and Kallos, 2009; Formenti et al., 2002; Kallos et al., 2007; Koçak et al., 2011; Lelieveld et al., 2002; Mihalopoulos et al., 1997; Querol et al., 2009), and less with local emissions and photochemistry (Gerasopoulos et al., 2006a, 2007). In addition, under northern winds, BB emissions attributed to fires in countries bordering the Black Sea (Salisbury et al., 2003; Sciare et al., 2008) or on various Greek islands (Bougiatioti et al., 2014, 2015) affect the air masses over the AS. While various emission sources and long-range transport of air masses over the Eastern Mediterranean play an important role in the PM composition over the AS, modelling studies to date have mainly investigated their effects near the surface (Athanasopoulou et al., 2014; Fountoukis et al., 2011; Im and Kanakidou, 2012; Lazaridis et al., 2006; Sotiropoulou et al., 2004).

In the present modelling study, we investigate the chemical composition of the lower troposphere over the AS during Etesian winds under the influence of BB activity. We use the regional on-line WRF-Chem atmospheric model (excluding the effect of aerosol-cloud radiation feedbacks) to simulate the horizontal and vertical distribution of various atmospheric variables and to quantify the effect of BB emissions. The simulations cover an Etesian outbreak during the period between 29/8/2011 and 9/9/2011, but the analysis focuses on two distinct Etesian patterns characterized by different transport conditions (medium- and long-range; Tombrou et al., 2015). The performance of the WRF-Chem is evaluated using airborne and ground-based observations performed during the Aegean-GAME (Aegean Pollution: Gaseous and Aerosol airborne Measurements) campaign (Bezantakos et al., 2013; Tombrou et al., 2015). The observations from the UK Facility for Airborne Atmospheric Measurements (FAAM) BAe-146 research aircraft cover the whole Aegean, inside and above the PBL (up to 4.5 km). In addition, airborne hydrogen cyanide (HCN) observations are used as a tracer to identify the location and spatial extent of BB plumes.

The paper is organized as follows: section 2 describes the set-up of the regional chemical/dynamical model, the simulations performed and the measurements used for the evaluation of the model. This section also presents the airborne HCN measurements. The interpretation of the modelling results and the comparison against the airborne observations is presented in section 3, while section 4 discusses the contribution of BB emissions on the chemical composition of the atmosphere over the AS. Section 5 presents numerical sensitivity experiments on the contribution of the on-line calculated biogenic emissions and the time-varying chemical boundary conditions from the MOZART global model.

2. Methodology and data

2.1. Model set-up

Version 3.3 of the Weather Research and Forecasting (WRF) model coupled with Chemistry (Fast et al., 2006) is implemented in this study. The numerical simulations have been performed applying triple nesting and LAT-LON projection. The first domain (D01) covers the extended area of Europe (Fig. 1a; from 14.5° W to 44.5° E longitude and from 23° to 77° N latitude, with a resolution of 0.5° × 0.5°), the second nested domain (D02) covers the extended area of Greece and Italy (Fig. 1c, from 4.83° to 32.17° E longitude and from 29.33° to 50.17° N latitude, with a resolution of 0.167° × 0.167°), while the third (D03) is centred on the extended area of Greece (from 18.44° to 29.05° E longitude and from 34.11° to 42.55° N latitude, with a resolution of 0.056° × 0.056°). The atmosphere is vertically resolved in 35 full sigma levels (model top at 50 hPa), with the first being at ~ 10 m above ground level (agl).

Input data for the initial, lateral and boundary conditions have been obtained from the National Centers for Environmental Prediction (NCEP) operational Global Final (FNL) Analyses. Land use and soil category data sets are used from the US Geological Survey (USGS). The parameterizations used are summarized in Table S1.

The RADM2 mechanism (Stockwell et al., 1990) is applied to simulate the gas phase chemistry. The aerosol modules used are the Modal Aerosol Dynamics Model for Europe (MADE) (Ackermann et al., 1998) for inorganic species, and the Secondary Organic Aerosol Model (SORGAM) (Schell et al., 2001) for secondary organic aerosols (SOA). In both MADE and SORGAM, the size distribution is described with three log-normal modes (Aitken, accumulation, and coarse). The species treated by the modules are the main inorganic ions (SO_4^{2-}, NO_3^-, NH_4^+, Na^+, Cl^-), elemental carbon (EC), primary organic aerosols (POA), SOA, a primary unspeciated $PM_{2.5}$ fraction covering all the unspeciated/unknown fine particles ($PM_{2.5\text{-unsp}}$), condensed water on the aerosol particles, and three species for the coarse mode (i.e., anthropogenic, marine, and soil derived