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Simulated air quality and pollutant budgets over Europe in 2008

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HIGHLIGHTS

- Major pollutant levels are simulated over Europe for the year 2008.
- Ozone levels are overestimated while aerosol levels are underestimated.
- Updated emissions over East Mediterranean result in better agreement with observations.
- Emission distributions and photochemistry lead to a north–south gradient.
- Surface ozone is affected by transport from upper troposphere over Europe

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ABSTRACT

Major gaseous and particulate pollutant levels over Europe in 2008 have been simulated using the offline-coupled WRFCMAQ chemistry and transport modeling system. The simulations are compared with surface observations from the EMEP stations, ozone (O_3) soundings, ship-borne O_3 and nitrogen dioxide (NO_2) observations in the western Mediterranean, tropospheric NO_2 vertical column densities from the SCIAMACHY instrument, and aerosol optical depths (AOD) from the AERONET. The results show that on average, surface O_3 levels are underestimated by 4 to 7% over the northern European EMEP stations while they are overestimated by 7–10% over the southern European EMEP stations and underestimated in the tropospheric column (by 10–20%). Particulate matter (PM) mass concentrations are underestimated by up to 60%, particularly in southern and eastern Europe, suggesting underestimated PM sources. Larger differences are calculated for individual aerosol components, particularly for organic and elemental carbon than for the total PM mass, indicating uncertainty in the combustion sources. Better agreement has been obtained for aerosol species over urban areas of the eastern Mediterranean, particularly for nss-SO_4^{2-}, attributed to the implementation of higher quality emission inventories for that area. Simulated AOD levels are lower than the AERONET observations by 10% on average, with average underestimations of 3% north of 40°N, attributed to the low anthropogenic emissions in the model and 22% south of 40°N, suggesting underestimated natural and resuspended dust emissions. Overall, the results reveal differences in the model performance between northern and southern Europe, suggesting significant differences in the representation of both anthropogenic and natural emissions in these regions. Budget analyses indicate that O_3 and peroxyacetyl nitrate (PAN) are transported from the free troposphere (FT) to the planetary boundary layer over Europe, while other species follow the reverse path and are then advected away from the source region.

1. Introduction

Air pollution can have significant impacts on ecosystems, human health, visibility and climate. Ozone (O_3) threatens human health (WMO, 2003) and vegetation (Fowler et al., 2009) and is an important greenhouse gas (IPCC, 2007). Fine and coarse particulate matter

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(PM) have adverse impacts on human health (Becker et al., 2003; Nel, 2005). They also scatter and absorb the radiation in the atmosphere and alter cloud properties (Ramanathan et al., 2001) influencing the climate at local to regional scales. The European Commission has taken measures in order to decrease the concentrations of air pollutants such as O_3 (EC, 2002), nitrogen dioxide (NO_2) and sulfur dioxide (SO_2; European Commission, 2001), PM_{10} and $PsM_{2.5}$ (PM with aerodynamic diameter less than 10 μm and 2.5 μm, respectively; European Commission, 2008).

Over Europe, long term observations show different trends for different pollutants. Colette et al. (2011) investigated the air quality changes over Europe between 1998 and 2007, mainly focusing on O_3, NO_2 and PM_{10}, using data from the European Monitoring and Evaluation Programme (EMEP; www.emep.int) and the European Environmental Agency AIRBASE (http://acm.eionet.europa.eu/databases/AIRBASE/) networks. They reported a slight increase in O_3 levels, particularly in urban areas, that was attributed to the robust decrease of NO_2 throughout Europe. The decrease in NO_2 levels was not sufficient to depress the O_3 levels, mainly because volatile organic compound (VOC) emissions did not change much. Significant decreases of PM_{10} levels were found over western Europe and increases in southern Europe (Colette et al., 2011). In agreement with these results, Wilson et al. (2012) found positive annual trends of rural O_3 in central and north-western Europe and significant negative trends in eastern and south-western parts of Europe between 1996 and 2005. Through the use of satellite products, Vrekoussis et al. (2013) showed the decrease in tropospheric NO_2 levels over Greece following the reduction in emissions, particularly from 2008 and onward, due to the economic crisis.

Chemistry and transport models enable analysis of atmospheric composition changes and understanding of relations and feedbacks between meteorology, emissions and chemistry. For Europe, several studies focused on investigating the present and future levels of air quality indicators (Kukkonen et al., 2012 and references therein). The evaluation of the performances of these models through comparisons with observations is essential to increase confidence in the projected changes (Russell and Dennis, 2000). In this respect, Matthias (2008) found 30–60% underestimation of surface PM_{10} levels, comparably better agreement for the chemical composition (15–20% underestimation) and satisfactory simulations of the aerosol optical depth (AOD) levels over Europe for the years 2000 and 2001. Depending on season and location, Zyrichidou et al. (2009) found model over- and underestimations of the tropospheric NO_2 columns retrieved from Global Ozone Monitoring Experiment (GOME)/European Remote Sensing (ERS2), SCanning Imaging Absorption spectroMeter for Atmospheric CHartograpHY (SCIAMACHY)/Environmental Satellite (Envisat), Ozone Monitoring Instrument (OMI)/AURA and GOME-2/Meteorological Operational (MetOp) satellite observations over south-east Europe for the period 1996–2001, which were attributed to the uncertainties in emissions. Pay et al. (2010) have shown that the high resolution CALIOPE–EU model system was able to successfully simulate surface O_3 and NO_2 levels with a mean normalized bias (*MNB*) of 6% and −17%, respectively, and reproduced most of the PM pollutant events ($r = 0.5$–0.6) with underestimations ($MNB = -50\%$ and −45% for PM_{10} and $PM_{2.5}$, respectively) for the year 2004. Appel et al. (2012) evaluated the U.S. EPA Community Multiscale Air Quality (CMAQ) model performance over North America and Europe for the year 2006. They found daytime O_3 levels overestimated (by ~8%) in winter and by ~2% in summer and year-long underestimations of PM_{10} and $PM_{2.5}$ levels over Europe by ~25 to 65%. Basart et al. (2012) used the high resolution (12 km × 12 km) CALIOPE air quality modeling system to evaluate the daily-to-seasonal aerosol variability over Europe for the year 2004. They found that PM_{10}, $PM_{2.5}$ and AOD levels were underestimated due to underestimations in the fine fraction of carbonaceous matter (organic and elemental carbon) and secondary inorganic aerosols (SIA). They showed that PM_{10} north of 40°N was dominated by SIA while south of 40°N high PM levels were associated with desert dust. In general, differences between observations and models were mainly attributed to uncertainties in emissions, and to a lesser extent, to boundary conditions and spatial resolutions of the models.

The air pollution levels over Europe are likely to increase in a changing climate (Katragkou et al., 2011; Im et al., 2011, 2012; Megaritis et al., 2013). Among these studies, Katragkou et al. (2011) performed simulations using the Regional Climate Model (RegCM) v.3 and Comprehensive Air Quality Model with extensions (CAMx), which showed increases in surface O_3 particularly over southern Europe and the Mediterranean basin by up to 6 ppbv from 2041–2050 to 2091–2100. Im et al. (2011) calculated a 1 ppb yr^{-1} increase in surface O_3 levels over the eastern Mediterranean using the Weather Research and Forecasting mesoscale meteorological model (WRF)–CMAQ modeling system when increasing air temperatures homogeneously from +1 to +5 K. Megaritis et al. (2013) increased the temperatures uniformly by 2.5 and 5 K using PMCAMx-2008 chemistry and transport model and found increases in sulfate and organic aerosols (OA) in southern Europe and decreases in ammonium nitrate levels, particularly in central Europe. Similar results have been reported by Im et al. (2012) with increases in OA levels in south-eastern Europe due to increases in temperatures but decreases in sulfate levels due to significant reductions in-cloud-production of sulfate.

In this manuscript, air quality levels over the European domain (Fig. 1) for the entire year of 2008 have been investigated using the WRF–CMAQ model system in the frame of the EU-funded projects: Megacity-Zoom for the Environment (CityZen; https://wiki.met.no/cityzen/start) and Evaluating the Climate and Air Quality Impacts of Short-lived Pollutants (ECLIPSE; http://eclipse.nilu.no). The model performance in simulating air quality levels has been evaluated via comparisons with surface and satellite observations, as well as O_3 soundings. The budgets of major gaseous and particulate pollutants over Europe are calculated for the first time using a mesoscale chemistry and transport model (WRF–CMAQ) on a relatively high spatial and temporal resolution compared to the previous studies that employed global chemistry models (Aan de Brugh et al., 2011; Pozzer et al., 2012).

2. Materials and methods

2.1. Model system

The WRF–ARW (v3.1.1; Skamarock and Klemp, 2008) has been offline-coupled with the CMAQ model, v4.7 (Byun and Schere, 2006; Foley et al., 2010). The simulation period covers the year 2008 (366 days), which was one of the focus years of the CityZen project, with a spin-up period of 20 days from December 2007. The model domain (Fig. 1) covers most of Europe, North Africa and the Middle East (from 18.98°N, 3.58°W to 49.82°N, 57.64°E) on a 30 km horizontal resolution, extending up to 16 km height on 23 vertical levels. The physical and chemical model configurations are provided in detail by Im and Kanakidou (2012). Monthly initial and boundary conditions for the CMAQ model have been extracted from the global chemistry-transport model TM4-ECPL (Myriokefalitakis et al., 2011) on a 3° × 2° spatial resolution and 34 vertical levels up to 0.1 hPa (~60 km).

2.2. Emissions

European anthropogenic emissions are provided from the emission inventory of the French National Institute of Industrial Environment and Risks (INERIS: https://wiki.met.no/cityzen/page2/emissions) on a 10 km × 10 km spatial resolution, which is a re-gridded product of the 50 km × 50 km EMEP inventory (http://www.ceip.at/) covering Europe. Details on the European anthropogenic emissions and chemical and temporal disaggregation of the emissions can be found in Im and Kanakidou (2012). The emissions from the remaining areas (i.e. North Africa and the Middle East) have been provided by the CIRCE global emission inventory on a 0.1° × 0.1° spatial resolution (Pozzer et al., 2012). The non-methane volatile organic compound (NMVOC) emissions are speciated