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Atmospheric Chemistry and Physics Discussions

Source-sector contributions to European ozone and fine PM in 2010 using AQMEII modeling data

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Abstract. Source apportionment modeling provides valuable information on the contributions of different source sectors and/or source regions to ozone (O_3) or fine particulate matter ($PM_{2.5}$) concentrations. This information can be useful in designing air quality management strategies and in understanding the potential benefits of reducing emissions from a particular source category. The Comprehensive Air quality Model with Extensions (CAMx) offers unique source attribution tools, called the Ozone and Particulate Source Apportionment Technology (OSAT/PSAT), which track source contributions. We present results from a CAMx source attribution modeling study for a summer month and a winter month using a recently evaluated European CAMx modeling database developed for Phase 3 of the Air Quality Model Evaluation International Initiative (AQMEII). The contributions of several source sectors (including boundary conditions representing transport of emissions from outside the modeling domain) to O_3 or $PM_{2.5}$ concentrations in Europe were calculated using OSAT and PSAT, respectively. Evaluation focused on 16 major cities and on identifying source sectors that contributed above 5%. Boundary conditions have a large impact on summer and winter ozone in Europe and on summer $PM_{2.5}$, but are only a minor contributor to winter $PM_{2.5}$. Biogenic emissions are important for summer ozone and $PM_{2.5}$. The important anthropogenic sectors for summer ozone are transportation (both on-road and non-road), energy production and conversion, and the industry sector. In two of the 16 cities, solvent and product also contributed above 5% to summertime ozone. For summertime $PM_{2.5}$, the important anthropogenic source sectors are the energy sector, transportation, industry, and agriculture. Residential wood combustion is an important anthropogenic sector in winter for $PM_{2.5}$ over most of Europe, with larger contributions in central and eastern Europe and the Nordic cities. Other anthropogenic sectors with large contributions to wintertime $PM_{2.5}$ include energy, transportation, and agriculture.

1 Introduction

Photochemical grid models (PGMs) such as the Comprehensive Air quality Model with Extensions (CAMx; Ramboll Environ, 2014) and the Community Multiscale Air Quality (CMAQ) model (Byun et al., 2006), are widely used in air quality management to assess the effectiveness of potential control strategies for secondary pollutants such as O_3 and $PM_{2.5}$. Source

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Published: 11 November 2016

Atmospheric Chemistry and Physics Discussions

apportionment analysis is an important component of this process to identify source sectors and/or regions that are contributing to O_3 and $PM_{2.5}$. The traditional approach to source attribution analysis is "brute-force" or "zero-out" sensitivity analysis in which the emissions from a given source sector are removed to quantify the contribution of that sector. This approach is expensive and impractical for assessing the contributions of a large number of source categories and suffers the limitation that the sum of zero-out impacts over all sources will not equal the total concentration (Koo et al., 2009). Tagged species methods, such as the Ozone and Particulate Source Apportionment Technology (OSAT/PSAT) in CAMx (Dunker et al., 2002; Yarwood et al., 2007), can efficiently track contributions from many source sectors and/or regions and provide source contributions that sum to the total concentration. These methods are increasingly being used to help understand complex air quality issues (e.g., Wagstrom et al., 2008; Burr and Zhang, 2011;Baker and Kelly, 2014; Collet et al., 2014; Wang et al., 2009; Li et al., 2012; Skyllakou et al., 2014).

Many source attribution studies in Europe have used receptor models and back trajectory analysis for inert pollutants, i.e., primary particulate matter (e.g., Querol et al., 2001; 2004; 2009; Belis et al., 2011; 2013; Viana et al., 2014). Source attribution studies in Europe for secondary pollutants, such as ozone and secondary $PM_{2.5}$, have used PGMs with the zero-out approach and Skyllakou et al. (2014) used the CAMx PSAT approach to distinguish the contributions of local and regional sources to fine PM in Paris. Reis et al. (2000) studied the impact of road traffic emissions on regional ozone levels in Europe by zeroing-out traffic emissions. Derwent et al. (2005) used a similar approach to determine the contribution of shipping emissions to ozone and acid deposition in Europe. Sartelet et al. (2012) estimated the contributions of biogenic and anthropogenic emissions to O_3 and PM concentrations in Europe and North America by zeroing-out one source category at a time. Under the TRANSPHORM (Transport related Air Pollution and Health Impacts-Integrated Methodologies for Assessing Particulate Matter) program, the source contributions of transport emissions (road transport, shipping, aviation) to O_3 and PM contributions in Europe were assessed using WRF-CMAQ and the zero-out approach (TRANSPHORM, 2014). Derwent et al. (2008) conducted sensitivity studies with a global chemical transport model to understand the effects of long-range transport from North America and Asia to surface ozone in Europe.

All of the above PGM source attribution studies for Europe investigated the contributions of a limited number of source categories. More recently, Tagaris et al. (2015) used the zero-out approach with CMAQ to calculate the contributions of emissions from 10 Standard Nomenclature for Air Pollution (SNAP) source sectors to air quality over Europe for a summer month (July 2006). Here, we use CAMx with OSAT and PSAT to calculate the contributions of SNAP sectors, biogenic emissions, and sources outside the modeling domain (boundary conditions) to cities in Europe. We make use of the CAMx modeling database for 2010 developed as part of the Air Quality Model Evaluation International Initiative (AQMEII).

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AQMEII is a joint regional air quality model evaluation effort between the North American and European modeling communities to improve the understanding of uncertainties in model predictions of ozone and $PM_{2.5}$, and to use this knowledge to guide model improvements (Rao et al., 2011). In Phase 1 of the AQMEII, a large number of offline photochemical air quality models were applied and evaluated in Europe and North America for the year 2006 using consistent inputs to the extent possible (Rao et al., 2011; Galmarini et al., 2012). The second phase of AQMEII was dedicated to the evaluation of online coupled chemistry-meteorology models over both continents with a primary focus on the year 2010 (Galmarini et al., 2015). In Phase 3, the focus is on diagnostic evaluation through sensitivity studies on model inputs and spectral decomposition of model errors (Galmarini et al., 2016; Solazzo and Galmarini, 2016; Solazzo et al., 2016). As part of Phase 3, a CAMx modeling database has been developed and evaluated for Europe (Solazzo et al., 2016) and this database was used in the study described in this paper.

2 Model Setup

2.1 Model configuration, domain, and inputs

Solazzo et al. (2016) developed a 2010 CAMx database for Europe and applied CAMx version 6.1 (Ramboll Environ, 2014) over the European Union (EU). The simulations used the Carbon Bond 2005 (CB05) gas phase chemistry (Yarwood et al., 2005) and the Coarse-Fine (CF) aerosol scheme. CAMx was applied for the whole year for a domain covering Europe and a portion of Africa. The domain (see Fig. 1) is defined in a Lambert Conic Conformal projection that includes 270 by 225 grid cells with a 23 km horizontal grid resolution.

Input meteorological data were generated using WRF-Chem 3.4.1, the coupled chemistry version of the Weather Research and Forecasting (WRF) Model (Skamarock et al., 2008), driven by the European Centre for Medium-Range Weather Forecasts (ECMWF) analysis fields. WRF-Chem was used rather than WRF to obtain emission estimates for wind-blown dust. Analysis nudging for wind speed, temperature and water vapor mixing ratio was employed within the boundary layer, with a nudging coefficient of 0.0003 sec-1. The WRF vertical grid with 33 layers extends from the surface to a fixed pressure of 50 hPa (about 20 km), with a surface layer depth of 24 m. The WRFCAMx pre-processor (version 4.2; Ramboll Environ, 2014) was used to create CAMx input files collapsing the 33 layers used by WRF to 14 layers in CAMx but keeping layers up to 230 m above ground level identical to the WRF layers.

Anthropogenic emissions for calendar year 2009 were derived from the TNO-MACC_II emission inventory (Kuenen et al., 2014; Pouliot et al., 2015) resolved by SNAP sector (see Table 1). The primary data sources were national emission inventories developed by European countries in accordance with guidance provided by the European Environment Agency (EEA, 2013). SNAP sector 34 combines “industrial combustion” (sector 3) with “industrial processes” (sector 4) to mitigate

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
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Atmospheric Chemistry and Physics Discussions

inconsistent classification of sources to sector 3 or 4, as discussed by Kuenen et al. (2014). Supplemental Section A provides a summary of SO_2, NOx and $PM_{2.5}$ emissions from the 9 SNAP sectors for the summer and winter months and presents spatial maps of total surface emissions and surface emissions for some sectors.

Biogenic VOC emissions were estimated by applying the Model of Emissions of Gases and Aerosols from Nature (MEGAN; Guenther et al., 2012) v2.04. Sea salt emissions were estimated using published algorithms (de Leeuw et al., 2000; Gong, 2003). Dust emissions were based on the GOCART (Ginoux et al., 2001; 2004) model implemented in WRF-Chem (Zhao et al., 2010). Chemical boundary conditions were derived from the Monitoring Atmospheric Composition and Climate (MACC) project using the Composition–Integrated Forecast System (C-IFS) model (Flemming et al., 2015).

2.2 Model performance evaluation summary

Solazzo et al. (2016) conducted a detailed model performance evaluation of CAMx for 2010 in the framework of the AQMEII Phase 3 project. Here we present a brief summary of model performance using a set of ground-based observations belonging to the Airbase network (http://www.eea.europa.eu/data-and-maps/data/airbase-the-european-air-quality-database-8) and covering most of the computational domain. Only background stations are considered in the analysis. Furthermore, analysis was carried out for the whole set, including Urban, Suburban and Rural sites (All Background, AB), as well as for Rural Background sites only (RB), since they are the most adequate for comparing model results over a 23 km-grid resolution. The comparative analysis among AB and RB sets provides an indication of the possible degradation in model performance when focusing on urban areas.

The model performance was evaluated over the whole year and based on daily mean concentrations. Only stations that had more than 75% of data availability on a yearly basis have been included in the comparisons. The number of available stations ranges according to the chemical species. The highest availability was noted for NO_2, with more than 2500 stations. Ozone and SO2 were available at more than 1500 sites, while for PM_{10} more than 2300 sites were available. $PM_{2.5}$ observations were available in about 700 sites over all Europe, with about 300 sites corresponding to RB stations.

Table 2 provides a domain wide summary of model performance for the AB and RB sets of stations. The statistical performance measures used in the evaluation are defined in Supplemental Section B. The yearly mean of the observed SO_2 concentration is 2.3 ppb, while the modeled value is 1.2, corresponding to a 48 % low bias. Similar results are noted at RB sites (NMB = -41%). NO_2 yearly mean concentrations are clearly underestimated when all background sites are considered. However, when the analysis is limited only to RB sites, which are more suitable for comparisons with a model run using a 23 km horizontal grid resolution, there is a noticeable improvement in model performance. The NMB improves from -56.3 % to -29.4% and the NME decreases from 60% to 47.8%. As a consequence, the RMSE drops from 11.7 to 5.4 ppb, and the daily

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
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Atmospheric Chemistry and Physics Discussions

correlation grows from 0.52 to 0.60. The under-estimation of NO_2 concentrations may be because the grid resolution is too coarse to resolve many of the monitoring locations. or alternatively indicates that NOx emissions are under-estimated in this inventory.

Annual mean ozone concentrations at AB sites are overestimated (NMB = 21.1%), while the standard deviation (SD) of the yearly series is correctly reproduced (standard deviation of 11.3 ppb observed versus 12 ppb modeled). Similar performance for SD is observed at RB sites, together with a clear improvement in terms of the yearly mean, as pointed out by the decrease of NMB and NME. These results suggest that the ozone bias at AB sites is partially due to overestimation at urban and suburban sites, where the horizontal grid resolution is insufficient to resolve ozone suppression at monitors by nearby sources of NOx. This hypothesis is confirmed by analysis of the Ox concentration ($Ox = O_3+NO_2$) that removes the local effect of NOx titration. Ox concentrations at both AB and RB sites are very well reproduced in terms of both mean and SD. Also, the temporal variation of Ox concentrations is well reproduced as shown by the correlation value (0.64 and 0.67 at AB and RB sites, respectively).

PM_{10} concentrations are underestimated at AB sites (NMB = -19%), but the bias for RB sites is small (NMB = 0.4% and FB = 3.5%). Conversely, the NME (and FE) remains high for both sets of stations. In particular, the NME increases from 51.7% at AB sites to 55% at RB sites. The temporal correlations are also low (< 0.3) in both cases. The overall performance suggests that CAMx correctly captured the yearly mean burden of aerosol but not its temporal evolution. This could be due to compensating errors among different sources that could be either underestimated or overestimated. The correlations for $PM_{2.5}$ are better (correlation = 0.48 and 0.52 at AB and RB sites, respectively), although there is more underestimation bias for $PM_{2.5}$ than for PM_{10}. These results suggest that coarse PM mass is likely overestimated and its temporal evolution is not correctly reproduced by CAMx. Note that a large fraction of the coarse PM can be attributed to dust and/or sea salt sources and there are large uncertainties in estimating emissions from these sources.

Supplemental Section B provides additional details and discussion on the spatial and temporal performance of the model.

2.3 Source attribution modeling

The source attribution modeling with CAMx used the OSAT and PSAT tagged species methods in CAMx version 6.1 (Ramboll Environ, 2014). In addition to the SNAP sector emission categories, the contributions of biogenic emissions, dust and sea salt emissions (for PM), and boundary conditions were explicitly tracked. Secondary organic aerosol (SOA) was not apportioned by PSAT because of the high computer memory requirement to track SOA categories on the large CAMx modeling grid. The total biogenic and anthropogenic SOA were both available from the CAMx CF aerosol scheme.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
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Published: 11 November 2016

Atmospheric Chemistry and Physics Discussions

The source attribution simulations were conducted for a summer month (August 2010) and a winter month (February 2010). A model spin-up period of one week (last week of January 2010 for the winter simulation and the last week of July 2010 for the summer simulation) was used to minimize the influence of initial conditions.

3 Results

We selected 16 cities, representing the Nordic countries, countries in western, central, and eastern Europe, and countries along the Mediterranean coastline, for the source attribution analysis. The contributions of the various source sectors to ozone and $PM_{2.5}$ concentrations were calculated for these cities and are discussed in this section.

3.1 Ozone source apportionment-summer

The European standard for ozone is based on the maximum daily 8 hour mean (not to exceed a threshold of 120 µg/m3, about 60 ppb, for 25 days averaged over 3 years). Accordingly, the source apportionment results for ozone are presented for the maximum daily 8 hour average (referred to as H1MDA8) for the summer month. Ozone results for the winter month are not discussed here because H1MDA8 levels at all the selected cities are less than the threshold and because boundary conditions dominate the ozone levels in winter, with contributions at the 16 cities ranging from a low of 61% to a high of 96%. Figure 2 shows the spatial pattern of calculated H1MDA8 ozone concentrations across the modeling domain. Over most of western and northern Europe, ozone levels are below 60 ppb. The highest ozone values (about 120 ppb) are predicted near Moscow, Russia. The 60 ppb level is exceeded in some of the Mediterranean countries and in parts of central and eastern Europe.

The source attribution results for summertime H1MDA8 ozone in each city are reported in Table 3 for contributors of 5% or more. In the 4 cities near the Mediterranean from Lisbon, Portugal in the west to Istanbul, Turkey in the east, H1MDA8 ozone in August 2010 is estimated to be above or very close to the standard of 60 ppb. Boundary conditions are the largest contributor to H1MDA8 ozone in all 4 Mediterranean cities, with contributions ranging from 26% to 34% from east (Istanbul, Athens) to west (Barcelona, Lisbon). Contributions from on-road transport (SNAP 7) are the next largest (20% to 24%) at 3 of the 4 cities (Lisbon, Barcelona, Athens). At Istanbul, the second highest contribution (24%) comes from biogenic emissions, while on-road transport is the third largest contributor at 15%. Non-road transport (SNAP 8; 12% to 18% contribution) and biogenic emissions (15% to 24% contribution) are also significant contributors at all 4 locations. The other anthropogenic sectors contributing 5% or more to summertime ozone in the Mediterranean cities are industry (SNAP 34; 6 to 8% contribution) and the energy sector (SNAP 1; 5 to 8% contribution).

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
Manuscript under review for journal Atmos. Chem. Phys.
Published: 11 November 2016

Boundary conditions are again an important contributor in the 4 cities in central and eastern Europe making the largest contribution in Minsk (25%) and Warsaw (28%) and the second largest contribution in Budapest (29%) and Kiev (21%). Road transport is the largest contributor (35%) in Budapest, while biogenic emissions are the largest contributor (33%) in Kiev. Road transport is the second largest contributor in Warsaw (25%) and biogenic emissions are the second largest contributor (23%) in Minsk. Biogenic emissions contribute less in Budapest (10%) and Warsaw (14%) than in the other two cities. Other important contributing source sectors in the central and eastern European cities are the energy sector (9% in Kiev to 17% in Warsaw), the non-road sector (7% to 10% contribution) and the industry sector (5% to 7% contribution).

In all 4 cities in western Europe (Paris, London, Amsterdam, and Berlin), H1MDA8 ozone concentrations are below the 60 ppb threshold. Boundary conditions are the largest contributor (29% to 59%) to HD1MA8 ozone at all 4 cities, and contribute more than half the HD1MA8 ozone in London and Paris and nearly 50% in Berlin. Road transport is the next largest contributor in Paris (13%) and Berlin (17%), while non-road transport (12%) and biogenic emissions (21%) are the second largest contributors in London and Amsterdam, respectively. Road transport contributions in London and Amsterdam rank third at 11% and 19%, respectively. Non-road transport contributes less than 10% to HD1MA8 ozone in Paris, Amsterdam and Berlin. The energy sector is an important contributor (13%) in Berlin, and contributes 5% to 6% in London and Amsterdam. The energy sector contribution in Paris is small (less than 3%), since France derives over 75% of its electricity from nuclear energy. The solvent and product use sector (SNAP 6) contributes 6% and 10% to summertime ozone in Paris and Amsterdam, respectively, but its contributions in London and Berlin are less than 3%.

As for the cities in western Europe, H1MDA8 ozone levels in the Nordic cities (Oslo, Copenhagen, Stockholm, Helsinki) are below the European threshold of 60 ppb. Boundary conditions again play an important role for ozone in these cities, and are the largest contributors in 3 of the 4 cities. Road transport is the largest contributor (24%) in Stockholm followed by boundary conditions (21%). Road transport contributions to the other 3 cities range from 12% in Oslo to 23% in Copenhagen. Non-road transport is an important contributing sector (14 to 21%) and its contributions in Oslo and Helsinki are higher than on-road transport. Biogenic emissions are also important contributors in all 4 cities, with contributions ranging from 12 to 20%. The energy sector contributes 12 to 13% in Helsinki, Stockholm and Copenhagen, but its contribution to H1MDA8 ozone in Oslo is slightly less than 5%.

3.2 $PM_{2.5}$ source apportionment-summer

The European standard for fine PM is an annual average concentration of 25 μg/m^3. Since we obtained source attribution for only two months, our discussion of the $PM_{2.5}$ source attribution focuses on the summer and winter monthly average concentrations. Figure 3 shows the spatial pattern of monthly mean $PM_{2.5}$ concentrations for August 2010 across the modeling domain. The highest $PM_{2.5}$ concentrations are along the southern and southeastern boundaries of the domain and in

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
Manuscript under review for journal Atmos. Chem. Phys.
Published: 11 November 2016

Atmospheric Chemistry and Physics Discussions

the Mediterranean countries. Transport of Saharan dust from North Africa explains the high values along the southern domain boundary. Removing the dust component from the calculated total $PM_{2.5}$ concentrations reduces the highest concentrations along the southern boundary by a factor of 2.

Table 4 shows the source attribution results for monthly mean $PM_{2.5}$ concentrations in August 2010. In the Mediterranean cities of Lisbon, Barcelona, Athens and Istanbul, boundary conditions are the largest contributors to mean August $PM_{2.5}$ concentrations, with contributions ranging from 38% to 49%. Non-road transport and SOA are the second and third largest contributors in Lisbon and Barcelona. In Athens, the energy sector and non-road transport are the second and third largest contributors, while in Istanbul the industry sector is the second largest contributor and SOA and the energy sector are the third largest contributors. Road transport contributions are less than 5% in Istanbul and less than 10% in Lisbon and Athens. The highest on-road transport contribution to the selected Mediterranean cities is 10% in Barcelona. The industry sector contributions in all 4 Mediterranean cities are 5% or more, while the SOA contributions in the 4 cities are 8% or more. The agriculture sector (SNAP 10) contribution to August 2010 mean $PM_{2.5}$ concentrations is 7 to 8% in Athens and Istanbul and less than 5% in the other 2 cities.

Boundary conditions are important contributors to monthly average $PM_{2.5}$ concentrations at cities in central and eastern Europe as well, as shown in Table 3, but the relative BC contributions in these regions are lower than those in southern Europe. BCs are the largest contributors in Minsk and Kiev, while the energy sector is the largest contributor in Budapest and Warsaw. The energy sector contributions range from 9% in Kiev to 24% in Warsaw. SOA are also important contributors in all 4 cities and are the second largest contributors in Minsk (18%) and Kiev (17%). The agriculture sector also has a large contribution in all 4 cities (12% to 14%), suggesting that ammonia emissions from agricultural activity leads to formation of particulate nitrate. The industry sector contributes from 6% to 9% of $PM_{2.5}$ concentrations in the 4 cities. Road transport contributions are 8% in Budapest and Minsk and 10% in Warsaw, but less than 5% in Kiev. Non-road transport contributions are more than 5% in the 4 cities, but less than 10%.

Boundary conditions are not large contributors to the August monthly average $PM_{2.5}$ concentrations in any of the 4 western European cities. Boundary condition contributions range from 9% in Amsterdam to 14% in Paris and Berlin. SOA are the largest contributors in London, Paris, and Berlin, while non-road transport is the largest contributor (28%) in Amsterdam. Non-road transport is an important contributor in the other 3 cities as well, with contributions ranging from 14% in Berlin to 23% in London. The energy sector has a 15% contribution in Berlin, but less than 10% in the other 3 cities. Agriculture has a large contribution (14%) in Paris, but lower contributions in Berlin (8%) and Amsterdam (6%). Agriculture contributions to the mean August 2010 $PM_{2.5}$ concentrations in London are less than 5%. Road transport is an important but not a major contributor (12 to 13%) in any of the 4 western European cities.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
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Published: 11 November 2016

Atmospheric Chemistry and Physics Discussions

The source attribution results for the 4 cities in the Nordic countries show the decreasing influence of boundary conditions in the northern portion of the modeling domain. Boundary condition contributions are not as large as for some of the cities to the south and range from 10 to 15%. SOA and non-road transport are the highest contributors in Oslo and contribute about 25% each. SOA is the largest contributor (about 31%) in Helsinki and Stockholm, while non-road transport is the largest contributor in Copenhagen. Non-road transport is the second highest contributor in Stockholm. Energy sector emissions contribute from 7 to 12% to monthly mean $PM_{2.5}$ concentrations, while the on-road transport sector contributes 9 to 13%. Residential combustion (SNAP 2) contributes 11% in Oslo but less than 5% in the other 3 Nordic cities.

3.3 $PM_{2.5}$ source apportionment-winter

Figure 4 shows the spatial distribution of monthly mean $PM_{2.5}$ concentrations for February 2010 across the modeling domain. The highest $PM_{2.5}$ concentrations are again along the southern boundary of the modeling domain, but the influence of boundary conditions further inside the domain is not to the same extent as for the summertime $PM_{2.5}$ concentrations, as shown in Table 5. High $PM_{2.5}$ concentrations are also predicted over Poland and we see from Table 5 that, from the 16 cities selected for the analysis, the highest $PM_{2.5}$ concentration (38 $\mu g/m^3$) is in Warsaw.

As mentioned above, Table 5 shows that boundary condition contributions to wintertime $PM_{2.5}$ concentrations in cities along the Mediterranean coastline are much lower than summertime contributions, particularly at cities in the west, such as Lisbon and Barcelona, where BC contributions are less than 5%. BC contributions are slightly higher than 10% in the eastern Mediterranean cities (Athens and Istanbul). There are some variabilities in source contributions among the 4 Mediterranean cities. In Lisbon, SOA is the single largest contributor, explaining nearly 50% of the winter month average $PM_{2.5}$. Residential combustion is the next largest contributor at 15%, followed by non-road transport at 13%. Non-road transport is the largest contributor (21%) in Barcelona, followed by SOA, on-road transport and residential combustion with comparable contributions (17% to 18%). Residential combustion is the largest contributor in both Athens (20%) and Istanbul (25%). Non-road transport is the next highest contributor in Athens while on-road transport, industry and boundary conditions are the second highest contributors (11%) in Istanbul. Energy sector contributions are more important in the eastern Mediterranean cities (9 to 10%) than in the western cities (less than 5% in Lisbon and 7% in Barcelona). Road transport contributions in Lisbon and Athens are 10% or less. Dust emissions within the modeling domain contribute 10% of the $PM_{2.5}$ in Athens.

At the 4 selected cities in central and Eastern Europe, residential combustion is the single largest contributor to wintertime $PM_{2.5}$, with contributions ranging from 29 to 38%. Boundary condition contributions are less than 5% in all 4 cities. Road transport and the energy sector are the second highest contributors in Budapest (17 to 18%) followed by agriculture at 15%.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
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Published: 11 November 2016

Atmospheric Chemistry and Physics Discussions

In Kiev, agriculture and the energy sector are the second highest contributors (11% to 12%) followed by on-road and non-road transport at 10% and industry at 9%. Agriculture is the second highest contributor in Minsk (16%) followed by the energy sector and on-road transport (12% to 13%). In Warsaw, the second highest contributions to wintertime $PM_{2.5}$ are from on-road transport and agriculture (16 to 17%) while the energy sector contributes 12%. Non-road and industry contributions in Warsaw are comparable and less than 10%.

There is significant variability in the source sector $PM_{2.5}$ contributions among the cities in western Europe. In London, non-road transport and SOA are the largest contributors (23%) followed by on-road transport (19%) and residential combustion (11%). Contributions from agriculture and the energy sector to wintertime $PM_{2.5}$ in London are about 7%. In Amsterdam, on-road and non-transport are the largest contributors (18 to 19%), residential combustion ranks second (16%) and agriculture and SOA contribute 12 to 13%. The energy sector contributes 10% of wintertime $PM_{2.5}$ in Amsterdam, while the industry sector contributes 7%. In both Paris and Berlin, residential combustion is the largest contributor (30% and 24%, respectively). However, there are differences in the contributions of the other source sectors in these 2 cities. SOA and on-road transport contributions rank second in Paris at about 16% followed by non-road transport at 13%, and 6 to 8% contributions from agriculture and the energy and industry sectors. In Berlin, on-road transport also ranks second but the contribution of SOA is only about 6%. Agriculture (15%), the energy sector (12%), non-road transport (11%) and industry (7%) are also significant contributors to wintertime $PM_{2.5}$ in Berlin.

Table 5 shows that, for all 4 cities in the Nordic countries, the contribution of boundary conditions is less than 5%. The largest contributors in Oslo and Helsinki are residential combustion sources (47% and 33%, respectively). The non-road and on-road transport sectors have significant contributions as well in these two cities (16% and 11% in Oslo, respectively and 14% and 18% in Helsinki, respectively). SOA, the energy sector and agriculture contribute 5 to 7% and 7 to 9% of the wintertime $PM_{2.5}$ in Oslo and Helsinki, respectively. Residential combustion is also the largest contributor in Copenhagen (20%) but it is followed closely by non-road transport (19%). Road transport contributes 14% of the wintertime $PM_{2.5}$ in Copenhagen and agriculture, the energy sector and SOA contribute about 11 to 12%. Industry contributions in Copenhagen are about 6%. Road transport is the largest contributor (22%) in Stockholm but residential combustion and non-road transport are significant contributors as well with contributions of about 19% and 16%, respectively. SOA contributes 14% to wintertime $PM_{2.5}$ in Stockholm while the energy sector contributes about 10% and agriculture and industry contribute 6 to 7%.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
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Published: 11 November 2016

Atmospheric Chemistry and Physics Discussions

4 Discussion

The source attribution analysis results show that long-range transport of ozone from beyond Europe has a strong influence on summertime ozone in August 2010 over most of Europe. The background summertime ozone contribution, simulated by the boundary condition tracer in the OSAT simulation, is about 26 to 34% in southern Europe and 20 to 30% in central and eastern Europe. The boundary condition contributions in western Europe are larger, ranging from about 30 to 60%. In the Nordic cities, BC contributions range from about 20% in Stockholm to 40% in Oslo. Wintertime ozone levels are below the EU standard and dominated by boundary conditions (60% to over 90%). The contribution of intercontinental transport (from North America and, to a smaller extent, from Asia) to ozone levels in Europe has been studied extensively through data analysis and modeling (e.g., Parrish et al., 1993; Wild and Akimoto, 2001; Lelieveld et al., 2002; Li et al., 2002; Naja et al., 2003; Trickl et al., 2003; Derwent et al., 2004; 2008; Auvray and Bey, 2005; Fehsenfeld et al., 2006; Guerova et al., 2006; Richards et al., 2013).

Summertime ozone contributions from biogenic emissions range from about 10% to 30%. At the cities selected for the analysis, the largest biogenic contribution of 33% is in Kiev, while the lowest contribution of 8% is in London. For anthropogenic emission sectors, the combined transportation sector (on-road and non-road transport) contributions range from 30 to 40% in cities along the Mediterranean coastline, cities in central and eastern Europe, and cities in northern Europe. In western Europe, the combined transport sector has a contribution of 20 to 30%. Contributions from the on-road transport sector are generally higher than those from the non-road transport sector, except for a few cities. The two transport sector contributions are comparable (within 3%) in Barcelona, Istanbul, London, and Oslo. Non-road transport contributions are slightly higher than on-road contributions in Oslo and Helsinki. These results are qualitatively consistent with those of Tagaris et al. (2015) who found that the on-road transport sector was the largest overall anthropogenic source sector contributing to July 2006 ozone concentrations in Europe with non-road transport contributions ranking second. Pouliot et al. (2015) noted that emissions from on-road transport in Europe decreased from 2006 to 2009 while emissions from shipping increased. This explains some of the higher contributions of non-road transport to ozone concentrations in some cities that were calculated in our study. The largest contributions of the energy sector were in central and Eastern Europe (9% to 17%) and in the Nordic cities (5% to 13%). Industry contributions to summertime ozone were important for the Mediterranean cities and cities in central and eastern Europe, with contributions ranging from 5% to 9%.

For summertime ozone, the total contribution from sources that cannot be controlled within Europe (i.e., the boundary conditions and biogenic emissions) ranges from 39% to 69%. The largest non-controllable contributions are 69% in Paris and 64% in London where the H1MDA8 city center ozone concentrations are 44 ppb and 41 ppb, respectively, well below the 60 ppb threshold. However, lower ozone levels are not necessarily associated with higher non-controllable contributions, or vice-versa. For example, the H1MDA8 ozone concentration in Copenhagen is 44 ppb with anthropogenic sources

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
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Published: 11 November 2016

contributing nearly 60%. The highest H1MDA8 ozone concentrations among the selected cities are predicted in Istanbul (73 ppb) and Kiev (70 ppb), and the non-controllable contributions are 50% and 54%, respectively.

Boundary conditions constitute a large fraction (40 to 50%) of the August 2010 average $PM_{2.5}$ concentrations in the Mediterranean cities. The influence of boundary conditions decreases from southern to northern Europe. This decreasing south-to-north gradient suggests that the Mediterranean cities were influenced by long-range transport of dust emissions from North Africa. These results are qualitatively consistent with numerous studies on the transport of Saharan dust and its contributions to PM levels in the Mediterranean Basin and other parts of Europe (e.g., Querol et al., 2001; 2004; 2009; Lyamani et al., 2005; Escudero et al., 2005; 2007a; 2007b; Vanderstraeten et al., 2008; Marconi et al., 2014). In contrast, there is an increasing south-to-north gradient in contributions of SOA (organic $PM_{2.5}$ formed in the atmosphere from precursor VOC species) to summertime $PM_{2.5}$ levels. Modeled SOA in Europe and North America is primarily associated with biogenic emissions (e.g., Sartelet et al., 2012). The contributions of SOA to summer PM range from 8 to 15% in the Mediterranean cities to 23 to 31% in the Nordic cities.

The anthropogenic source sector contributions to summertime average $PM_{2.5}$ vary with region. The important anthropogenic sectors in summer are the transport sector (both on-road and non-road), the energy sector, the industry sector, and agriculture. The contributions of other anthropogenic source sectors to the mean monthly $PM_{2.5}$ are generally less than 10%, with the exception of the solvent and product use sector, which has a contribution of over 10% in Amsterdam.

The source attribution results for wintertime $PM_{2.5}$ are significantly different from the summertime results. The contributions of boundary conditions are generally less than 5% with the exception of the eastern Mediterranean cities of Athens and Istanbul, where the BC contributions are 12 and 11%, respectively. SOA contributions are small (less than 10%) to moderate (about 20%) at most locations, except in Lisbon, where the SOA contribution is nearly 50%.

The important anthropogenic sectors for wintertime $PM_{2.5}$ are residential combustion, the combined transport sector (on-road and non-road), the energy sector, and agriculture. Residential combustion contributions in winter are much higher than in summer and range from over 10% in London to nearly 50% in Oslo. Residential combustion is the largest contributor in 11 of the 16 cities studied in this work. Higher winter contributions from this sector are consistent with residential wood burning for heating in winter (e.g., Denier van der Gon et al., 2015), particularly in northern Europe (e.g., Krecl et al., 2008). As shown in Supplemental Section A, primary $PM_{2.5}$ emissions from residential combustion are a factor of 10 higher in winter than in summer.

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Our model results are subject to limitations in model formulation and input data. Model performance evaluations presented here and by others, such as AQMEII Phase 3 contributors (see Solazzo et al., 2016), can suggest where modeling uncertainties exist and how they can influence source contributions. For example, when differences between modeled and observed concentrations are mostly driven by meteorology we may expect, as a first approximation, that the relative source contributions are reasonable even though the absolute contributions are not well captured. In contrast, discrepancies related to emission inventories or model processes can be expected to bias both the absolute and relative contributions of specific sources. For example, model underestimation for OA could result from missing emission categories (e.g., intermediate VOC) and/or biased inventories (e.g., uncertain biogenic emissions) and/or biased model SOA schemes and these errors would have different influences on source contributions. Quantifying source contributions can help assess when uncertainties are influential, keeping in mind that errors that underestimate impacts from a specific source may be less obvious than overestimation. Like others, we conclude that for the AQMEII European modeling domain, uncertainties in NOx emissions are important to both O_3 and $PM_{2.5}$, uncertainties in SOA formation algorithms and precursor emissions are important for $PM_{2.5}$, and that dust emission estimates are uncertain.

Competing interests. The authors declare that they have no conflict of interest.

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Atmospheric Chemistry and Physics Discussions

Table 1. SNAP sector classification of anthropogenic emissions.

Sector Number	Description
1	Energy industries (e.g., power generation and refineries
2	Non-industrial (residential) combustion
34	Industry*
5	Extraction and distribution of fossil fuels
6	Solvent and other product use
7	Road transport (includes exhaust, evaporative, tire/brake/road wear)
8	Non-road transport (includes rail, aircraft, shipping, construction equipment)
9	Waste treatment
10	Agriculture

*Sector 34 combines "industrial combustion" (SNAP 3) with "industrial processes" (SNAP 4) to mitigate inconsistent classification of sources to sector 3 or 4 (see Kuenen et al., 2014)

Table 2. CAMx model performance metrics at All (AB) and Rural (RB) background Airbase sites. Metrics are computed for daily mean concentrations for calendar year 2010.

	SO_2 [ppb]		NO_2 [ppb]		O_3 [ppb]		O_x [ppb]		PM_{10} [μg/m^3]		$PM_{2.5}$ [μg/m^3]	
Parameter	AB	RB	AB	RB	AB	RB	AB	RB	AB	RB	AB	RB
# Observations	550113	90446	954709	141241	646965	144139	561059	108438	842896	115022	267121	36378
Observed Mean	2.3	1.6	14.0	6.9	26.3	29.2	36.7	34.6	27.8	21.7	17.5	14.5
Modeled Mean	1.2	0.9	6.1	4.9	31.8	31.6	36.8	35.8	22.6	21.8	14.0	13.9
Observed S.D.	4.6	2.2	10.1	6.1	11.3	11.1	10.3	10.1	22.0	17.0	15.9	13.5
Modeled S.D.	1.4	1.0	4.8	4.2	12.0	12.2	10.6	10.7	13.8	13.5	9.2	9.2
Mean BIAS	-1.1	-0.6	-7.9	-2.0	5.6	2.4	0.0	1.1	-5.3	0.1	-3.4	-0.6
NMB (%)	-47.9	-40.9	-56.3	-29.4	21.1	8.2	0.0	3.3	-18.9	0.4	-19.7	-4.2
Mean Error	1.7	1.1	8.4	3.3	8.6	7.4	6.8	6.6	14.4	11.9	8.3	7.1
NME (%)	72.1	69.2	60.0	47.8	32.7	25.2	18.5	19.1	51.7	55.0	47.3	49.3
FB (%)	-45.4	-37.6	-73.1	-29.3	19.7	6.4	-0.3	2.9	-15.4	3.5	-13.7	4.2
FE (%)	81.9	76.5	81.6	55.6	33.5	28.2	19.5	20.0	53.1	52.6	49.8	50.1
Correlation	0.24	0.32	0.52	0.59	0.66	0.69	0.64	0.67	0.28	0.30	0.48	0.52
RMSE	4.6	2.2	11.7	5.4	11.1	9.5	8.9	8.5	23.1	18.3	14.5	11.8
IoA	0.3	0.5	0.6	0.7	0.8	0.8	0.8	0.8	0.5	0.5	0.6	0.7

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
Manuscript under review for journal Atmos. Chem. Phys.
Published: 11 November 2016

Atmospheric Chemistry and Physics Discussions

Table 3. Sectors contributing 5% or more to summertime H1MDA8 ozone concentrations.

City (ppb)	Sector* Contributions (%)					
Barcelona (58)	BC (28)	SNAP 7 (21)	SNAP 8 (18)	Biogenic (15)	SNAP 34 (7)	SNAP 1 (5)
Lisbon (61)	BC (34)	SNAP 7 (20)	Biogenic (19)	SNAP 8 (11)	SNAP 34 (6)	SNAP 1 (6)
Athens (69)	BC (26)	SNAP 7 (24)	SNAP 8 (16)	Biogenic (15)	SNAP 1 (8)	SNAP 34 (6)
Istanbul (73)	BC (26)	Biogenic (24)	SNAP 7 (15)	SNAP 8 (13)	SNAP 34 (9)	SNAP 1 (8)
Minsk (58)	BC (25)	Biogenic (23)	SNAP 7 (19)	SNAP 1 (15)	SNAP 8 (10)	--
Budapest (63)	SNAP 7 (35)	BC (29)	SNAP 1 (11)	Biogenic (10)	SNAP 8 (7)	SNAP 34 (5)
Warsaw (66)	BC (28)	SNAP 7 (24)	SNAP 1 (17)	Biogenic (14)	SNAP 8 (7)	SNAP 34 (7)
Kiev (70)	Biogenic (33)	BC (21)	SNAP 7 (18)	SNAP 8 (10)	SNAP 1 (9)	SNAP 34 (6)
London (41)	BC (56)	SNAP 8 (12)	SNAP 7 (11)	Biogenic (8)	--	--
Paris (44)	BC (59)	SNAP 7 (13)	Biogenic (10)	SNAP 8 (6)	SNAP 6 (6)	--
Amsterdam (51)	BC (29)	Biogenic (21)	SNAP 7 (19)	SNAP 6 (10)	SNAP 8 (8)	SNAP 1 (6)
Berlin (56)	BC (46)	SNAP 7 (17)	SNAP 1 (13)	Biogenic (11)	SNAP 8 (6)	--
Copenhagen (44)	BC (29)	SNAP 7 (23)	SNAP 8 (14)	SNAP 1 (13)	Biogenic (12)	SNAP 34 (5)
Oslo (50)	BC (41)	Biogenic (20)	SNAP 8 (14)	SNAP 7 (12)	--	--
Helsinki (50)	BC (31)	SNAP 8 (21)	SNAP 7 (17)	SNAP 1 (13)	Biogenic (13)	--
Stockholm (57)	SNAP 7 (24)	BC (21)	SNAP 8 (18)	Biogenic (18)	SNAP 1 (12)	--

*See Table 1 for anthropogenic (SNAP) sector descriptions

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
Manuscript under review for journal Atmos. Chem. Phys.
Published: 11 November 2016

Atmospheric Chemistry and Physics Discussions

Table 4. Sectors contributing 5% or more to summertime monthly mean $PM_{2.5}$ concentrations.

City (μg/m^3)	Sector* Contributions (%)						
Lisbon (11)	BC (45)	SNAP 8 (18)	SOA (15)	SNAP 34 (6)	SNAP 7 (5)	--	--
Barcelona (12)	BC (40)	SNAP 8 (19)	SOA (11)	SNAP 7 (10)	SNAP 34 (5)	--	--
Athens (16)	BC (38)	SNAP 1 (15)	SNAP 8 (10)	SOA (9)	SNAP 10 (8)	SNAP 7 (7)	SNAP 34 (6)
Istanbul (17)	BC (49)	SNAP 34 (11)	SOA (8)	SNAP 1 (8)	SNAP 10 (7)	SNAP 8 (6)	--
Budapest (10)	SNAP 1 (23)	BC (23)	SNAP 10 (13)	SOA (13)	SNAP 34 (9)	SNAP 7 (8)	SNAP 8 (5)
Warsaw (13)	SNAP 1 (24)	BC (21)	SOA (13)	SNAP 10 (12)	SNAP 7 (10)	SNAP 8 (8)	SNAP 34 (8)
Minsk (13)	BC (27)	SOA (18)	SNAP 10 (14)	SNAP 1 (14)	SNAP 7 (8)	SNAP 8 (7)	SNAP 34 (7)
Kiev (13)	BC (37)	SOA (17)	SNAP 10 (12)	SNAP 1 (9)	SNAP 8 (9)	SNAP 34 (6)	--
Berlin (8)	SOA (19)	SNAP 1 (15)	SNAP 8 (14)	BC (14)	SNAP 7 (12)	SNAP 34 (10)	SNAP 10 (8)
London (10)	SOA (32)	SNAP 8 (23)	SNAP 7 (13)	BC (12)	SNAP 1 (7)	SNAP 34 (5)	--
Paris (11)	SOA (18)	SNAP 8 (16)	SNAP 10 (14)	BC (14)	SNAP 7 (13)	SNAP 34 (8)	SNAP 1 (8)
Amsterdam (13)	SNAP 8 (28)	SOA (23)	SNAP 7 (13)	SNAP 1 (9)	BC (9)	SNAP 34 (6)	SNAP 10 (6)
Oslo (8)	SNAP 8 (25)	SOA (25)	SNAP 2 (11)	BC (10)	SNAP 7 (9)	SNAP 1 (7)	SNAP 34 (6)
Stockholm (8)	SOA (31)	SNAP 8 (15)	BC (15)	SNAP 1 (12)	SNAP 7 (10)	SNAP 34 (7)	--
Helsinki (8)	SOA (31)	BC (15)	SNAP 8 (15)	SNAP 7 (13)	SNAP 1 (10)	SNAP 34 (5)	--
Copenhagen (11)	SNAP 8 (26)	SOA (23)	BC (11)	SNAP 7 (10)	SNAP 1 (10)	SNAP 10 (7)	SNAP 34 (6)

*See Table 1 for anthropogenic (SNAP) sector descriptions

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
Manuscript under review for journal Atmos. Chem. Phys.
Published: 11 November 2016

Atmospheric Chemistry and Physics Discussions

Table 5. Sectors contributing 5% or more to wintertime monthly mean $PM_{2.5}$ concentrations.

City (μg/m^3)	Sector* Contributions (%)							
Lisbon (13)	SOA (47)	SNAP 2 (15)	SNAP 8 (13)	SNAP 7 (7)	SNAP 34 (6)	--	--	--
Barcelona (13)	SNAP 8 (21)	SOA (18)	SNAP 7 (18)	SNAP 2 (17)	SNAP 10 (7)	SNAP 1 (7)	SNAP 34 (7)	--
Athens (15)	SNAP 2 (20)	SNAP 8 (17)	SOA (13)	BC (12)	Dust (10)	SNAP 7 (10)	SNAP 1 (9)	--
Istanbul (26)	SNAP 2 (25)	SNAP 7 (11)	BC (11)	SNAP 34 (11)	SNAP 1 (10)	SNAP 8 (10)	SNAP 10 (9)	SOA (6)
Budapest (30)	SNAP 2 (29)	SNAP 7 (18)	SNAP 1 (17)	SNAP 10 (15)	SNAP 8 (7)	SNAP 34 (7)	--	--
Minsk (30)	SNAP 2 (33)	SNAP 10 (16)	SNAP 1 (13)	SNAP 7 (12)	SNAP 8 (10)	SNAP 34 (7)	--	--
Kiev (31)	SNAP 2 (37)	SNAP 10 (12)	SNAP 1 (11)	SNAP 8 (10)	SNAP 7 (10)	SNAP 34 (9)	--	--
Warsaw (38)	SNAP 2 (34)	SNAP 7 (17)	SNAP 10 (16)	SNAP 1 (12)	SNAP 8 (7)	SNAP 34 (6)	--	--
London (21)	SNAP 8 (23)	SOA (23)	SNAP 7 (19)	SNAP 2 (11)	SNAP 10 (7)	SNAP 1 (6)	--	--
Paris (25)	SNAP 2 (30)	SOA (16)	SNAP 7 (16)	SNAP 8 (13)	SNAP 10 (8)	SNAP 1 (6)	SNAP 34 (6)	--
Amsterdam (26)	SNAP 7 (19)	SNAP 8 (18)	SNAP 2 (16)	SNAP 10 (13)	SOA (12)	SNAP 1 (10)	SNAP 34 (7)	--
Berlin (32)	SNAP 2 (24)	SNAP 7 (18)	SNAP 10 (15)	SNAP 1 (12)	SNAP 8 (11)	SNAP 34 (7)	SOA (6)	--
Stockholm (17)	SNAP 7 (22)	SNAP 2 (19)	SNAP 8 (16)	SOA (14)	SNAP 1 (10)	SNAP 10 (7)	SNAP 34 (6)	--
Oslo (19)	SNAP 2 (47)	SNAP 8 (16)	SNAP 7 (11)	SOA (7)	SNAP 1 (6)	SNAP 10 (5)	--	--
Helsinki (21)	SNAP 2 (33)	SNAP 7 (18)	SNAP 8 (14)	SNAP 1 (9)	SOA (9)	SNAP 10 (7)	SNAP 34 (5)	--
Copenhagen (24)	SNAP 2 (20)	SNAP 8 (19)	SNAP 7 (14)	SNAP 10 (12)	SNAP 1 (12)	SOA (11)	SNAP 34 (6)	--

*See Table 1 for anthropogenic (SNAP) sector descriptions

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
Manuscript under review for journal Atmos. Chem. Phys.
Published: 11 November 2016

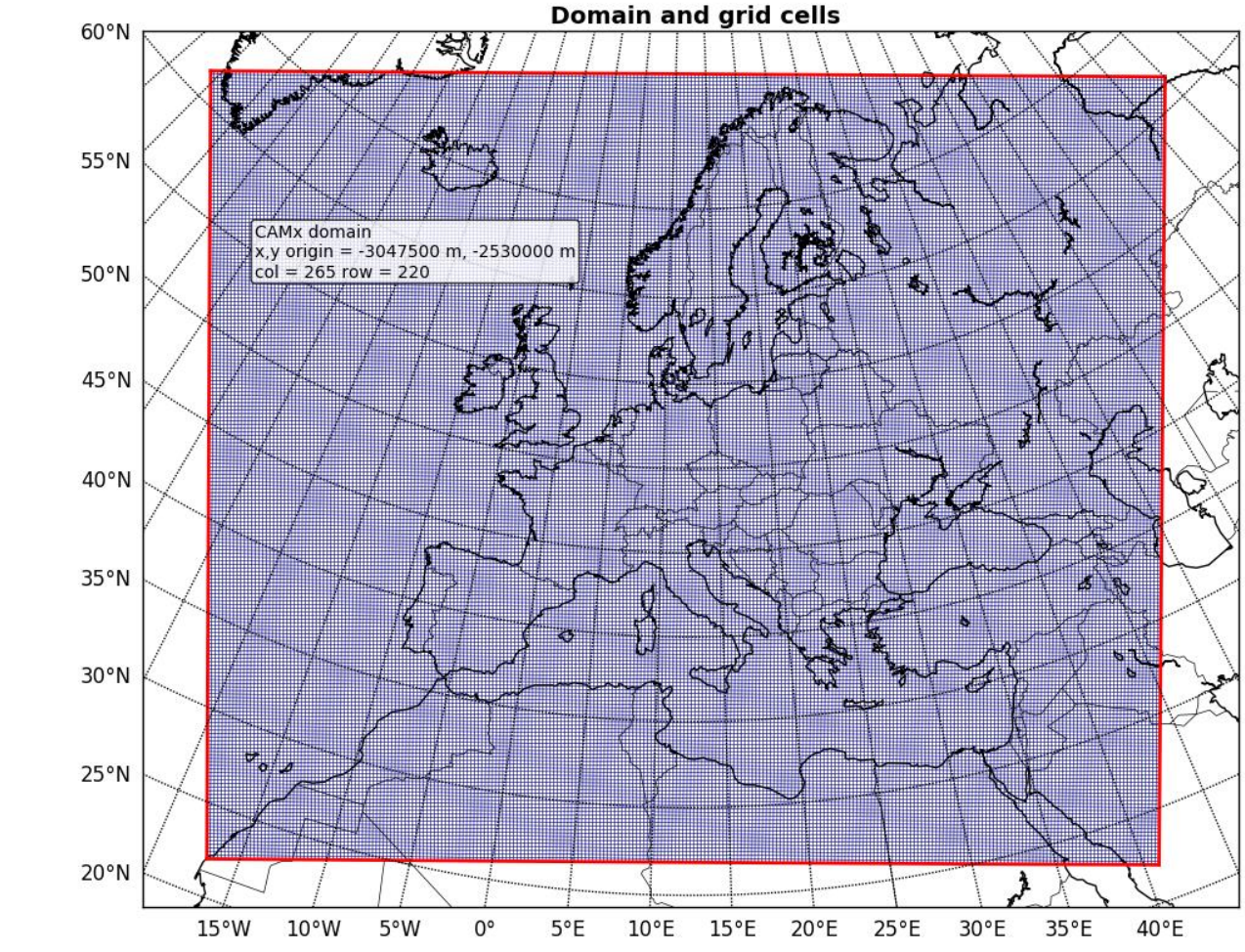

Figure 1. CAMx modeling domain with 270 by 225 grid cells at 23 km horizontal grid resolution.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
Manuscript under review for journal Atmos. Chem. Phys.
Published: 11 November 2016

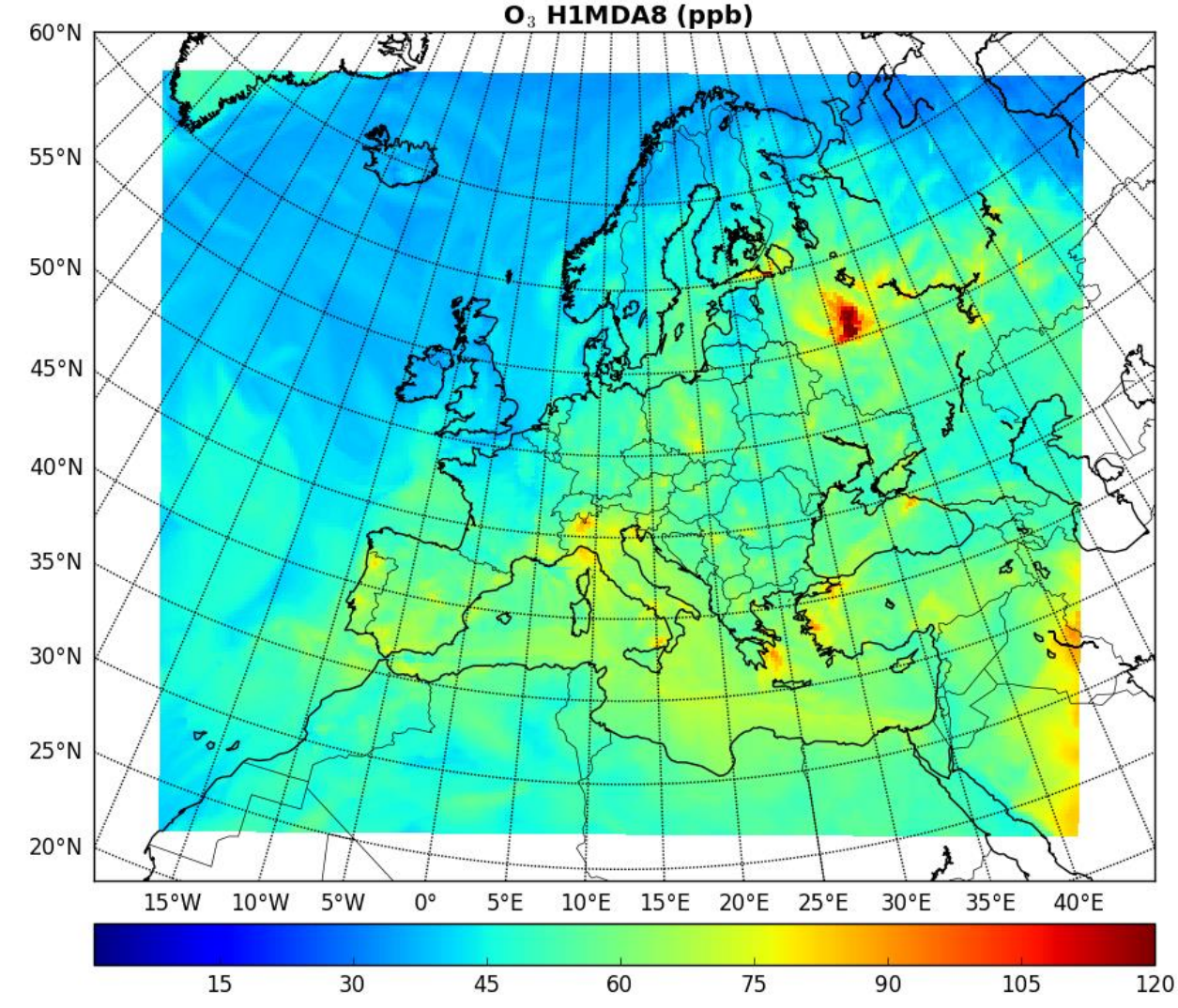

Figure 2. Predicted maximum daily 8-hour average ozone concentrations during August 2010.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
Manuscript under review for journal Atmos. Chem. Phys.
Published: 11 November 2016

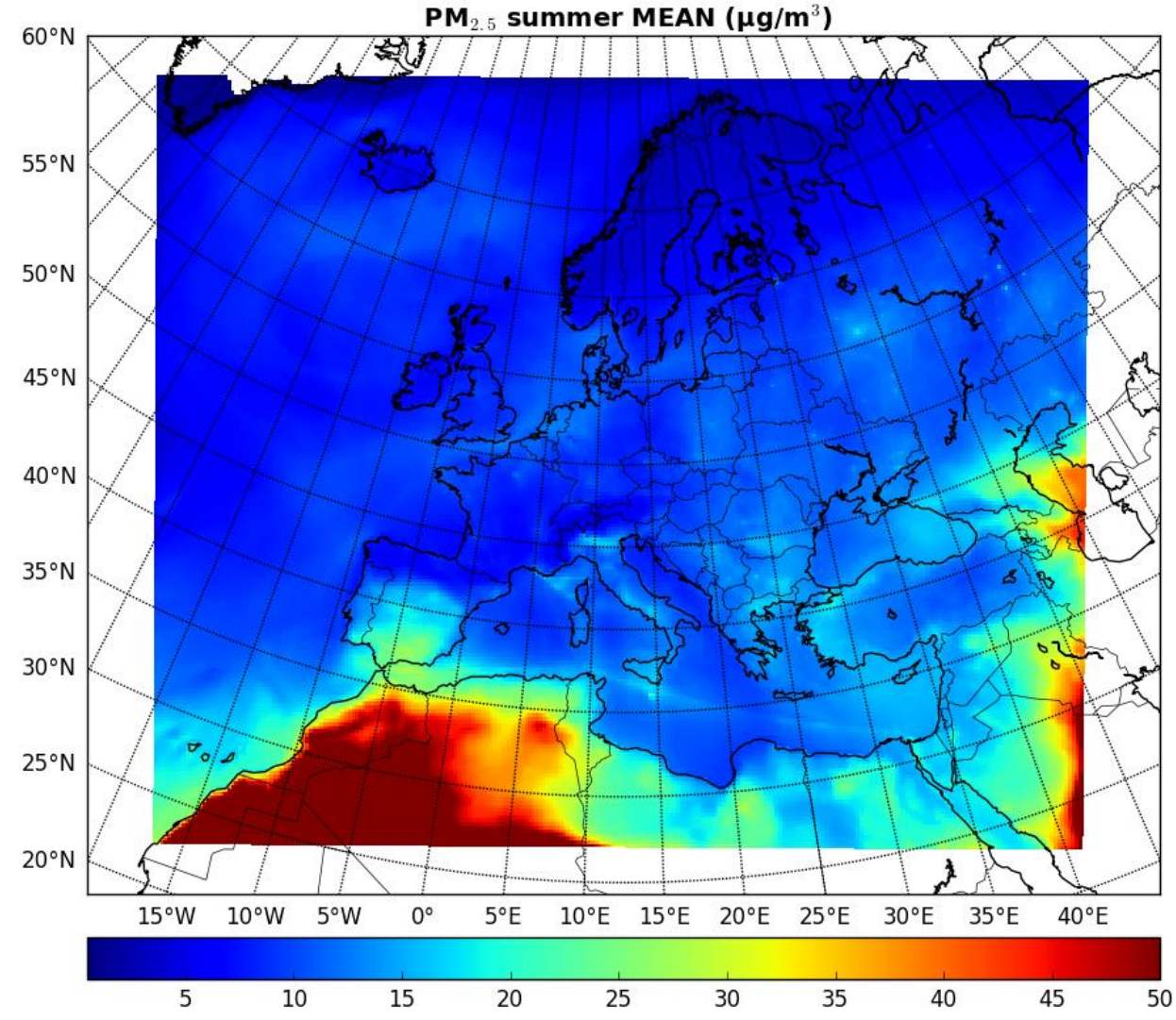

Figure 3. Predicted monthly average $PM_{2.5}$ concentrations during August 2010.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-973, 2016
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Published: 11 November 2016

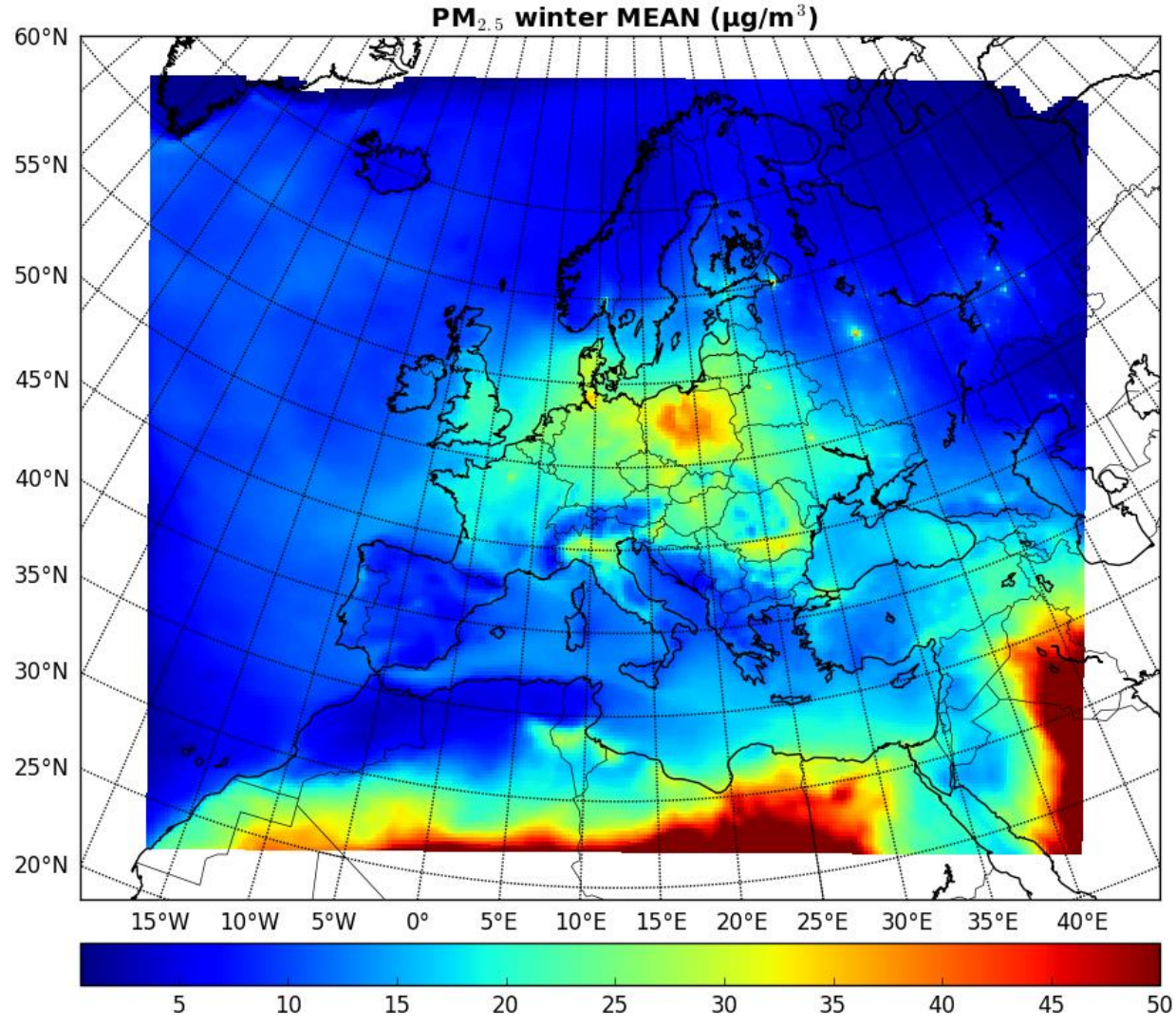

Figure 4. Predicted monthly average $PM_{2.5}$ concentrations during February 2010.