

## Trace Gas/Aerosol Boundary Concentrations and their Impacts on Continental-Scale AQMEII Modeling Domains

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## Abstract

Over twenty modeling groups are participating in the Air Quality Model Evaluation International Initiative (AQMEII) in which a variety of mesoscale photochemical and aerosol air quality modeling systems are being applied to continental-scale domains in North America and Europe for 2006 full-year simulations for model inter-comparisons and evaluations. To better understand the reasons for differences in model results among these participating groups, each group was asked to use the same source of emissions and boundary concentration data for their simulations. This paper describes the development and application of the boundary concentration data for this AQMEII modeling exercise. The European project known as GEMS (Global and regional Earth-system Monitoring using Satellite and in-situ data) has produced global-scale re-analyses of air quality for several years, including 2006 (<http://gems.ecmwf.int>). The GEMS trace gas and aerosol data were made available at 3-hourly intervals on a regular latitude/longitude grid of approximately 1.9° resolution within 2 “cut-outs” from the global model domain. One cut-out was centered over North America and the other over Europe, covering sufficient spatial domain for each modeling group to extract the necessary time- and space-varying (horizontal and vertical) concentrations for their mesoscale model boundaries. Examples of the impact of these boundary concentrations on the AQMEII continental simulations are presented to quantify the sensitivity of the simulations to boundary concentrations. In addition, some participating groups were not able to use the GEMS data and instead relied upon other sources for their boundary concentration specifications. These are noted, and the contrasting impacts of other data sources for boundary data are presented. How one specifies four-dimensional boundary concentrations for mesoscale air quality simulations can have a profound impact on the model results, and hence, this aspect of data preparation must be performed with considerable care.

Keywords: air quality modeling, boundary concentrations, model evaluation, AQMEII, GEMS

## 1. Introduction

The evaluation of regional- through continental-scale photochemical and aerosol air quality simulation modeling systems (PAQMs; also known as chemical-transport models or CTMs) has been a subject of considerable interest in recent years (Dennis et al., 2010; Vautard et al., 2007; McKeen et al., 2005). Such systems have been adopted by the air quality management and forecasting communities to provide estimates of future (10 years and longer) air quality based on planned emissions mitigation actions, as well as forecasts of short-term (1-3 days) air quality for public notice and alerts (Honoré et al., 2009; Eder et al., 2010). The results from these model applications have become increasingly visible and significant from the standpoint of public perceptions as well as having considerable economic, political, and social implications. Therefore, it is necessary that users of the models and consumers of the model results have sufficient confidence in these tools and their predictions to use for the intended applications. Such confidence can be obtained, in part, from evaluations of the models against real-world measurements for their particular applications.

To better foster a structured and coordinated approach to the PAQM evaluations at the international level, an Air Quality Model Evaluation International Initiative (AQMEII) was launched in 2008 as a collaboration between North American and European modeling groups (Rao et al., 2011; Galmarini and Rao, 2011; <http://aqmeii.jrc.ec.europa.eu>). AQMEII is aimed at providing a permanent forum to constantly monitor the state of advancement of regional-scale PAQMs and model evaluation methodologies through the organization of periodic workshops and modeling activities in which the different aspects of model performance evaluation are considered. In the first phase of AQMEII, an initial exercise has been launched in which more than twenty modeling groups in North America and Europe applied their regional-scale PAQMs to simulate air quality for the full-year of 2006. Each participating group has been requested to model both continents using common reference model input data sets, namely, gridded source emissions and lateral boundary concentrations for each continent. The focus of the study is on the application of the structured model evaluation framework discussed in Dennis et al. (2010) to these model simulation results, using a comprehensive observational database consisting of surface, aircraft, and satellite data for model evaluation and model inter-comparison.

This paper describes the development of a set of prescribed boundary concentrations for each continental model domain for use by all AQMEII modelers for this exercise, as well as some of the major impacts of these boundary concentrations on results in the interior of the model domain. Regional- or even continental-scale geographical extents of modeling domains require careful specification of the vertical and horizontal profiles of boundary concentrations since typical airflows over North America and Europe can traverse each continent in a few days to a week. The trace gas and aerosol concentrations as specified at the model's boundaries will affect the model simulation results since they are transported into the simulation domains, interacting with the model's internal processing of emissions, chemical transformations, deposition, etc. For this AQMEII exercise, the minimum spatial extent of each continent to be modeled by all participants (see Figures 1 and 2 in Rao et al., 2011) is defined by latitude/longitude boundaries:

North America:      Latitude: 25.5°N to 58.5°N      Longitude: 130°W to 59.5°W

Europe:              Latitude: 35.0°N to 70.0°N      Longitude: 15.0°W to 35°E

## **2. GEMS Re-analysis**

A variety of sources is used to specify boundary concentrations for retrospective regional-scale PAQM simulations. Ideally, observational data should be of sufficient density and quality to provide these specifications. However, model domain boundaries are often over the ocean or sparsely-monitored land areas. Satellite-based platforms provide data for only a few chemical species and only intermittently in space and time. Global-scale PAQMs of coarser resolution are another source for providing boundary concentrations to regional-scale models. For the AQMEII project, a combination of global-scale models with assimilation of satellite-based observational data was used to derive boundary concentrations. This hybrid analysis using model and observational data for a retrospective assessment is commonly referred to as “re-analysis”. In this case, data were derived from a multi-year re-analysis that included the target year of 2006 from the European Union-funded project of Global and regional Earth-system Monitoring using Satellite and in-situ data (GEMS; <http://gems.ecmwf.int>; Hollingsworth et al., 2008; GEMS, 2010).

The GEMS project was set up by thirty-one participating institutions coordinated by the European Centre for Medium-Range Weather Forecasts (ECMWF) to build and demonstrate the

core capability for providing a comprehensive range of services related to the chemical and particulate matter composition of the atmosphere. Among the demonstrated capabilities were data analyses and modeling systems for monitoring global distributions of atmospheric constituents, with focus areas of climate, air quality, and ultraviolet radiation, especially as they affect European communities. Global re-analysis products from GEMS are available for the period 2003-2008. These re-analyses make use of satellite observations allowing the retrieval of O<sub>3</sub>, CO, CH<sub>4</sub>, CO<sub>2</sub>, and aerosol optical depth during the AQMEII period of interest. Although GEMS is now concluded, the work conducted therein is being extended and improved through the new Monitoring Atmospheric Composition and Climate (MACC; <http://www.gmes-atmosphere.eu>) project.

Figure 1 illustrates the principal components of the GEMS global modeling system. The GEMS system was built within and around the ECMWF Integrated Forecasting System (IFS; <http://www.ecmwf.int/research/ifsdocs/>), a global operational weather forecasting model system, including the capability for four-dimensional variational data assimilation (Rabier et al., 2000). The IFS system is coupled with one of three global CTMs: MOZART3 (Kinnison et al., 2007), MOCAGE (Bousserez et al., 2007), or TM5 (Huijnen et al., 2010) through a special-purpose OASIS4 software coupler (Flemming et al., 2009). The main idea behind the coupled system is that the IFS computes only the transport of the assimilated reactive gases while the tendencies due to chemical conversion, deposition and emission injection are provided by one of the CTMs. The simulation of global aerosol and greenhouse gases is directly included within the IFS model (Morcrette et al., 2009). In this way, the IFS needs to handle only five additional chemical tracers, while the comprehensive schemes of the CTMs contain between 55 and 118 gaseous species. The coupled CTM is driven by meteorological data from the IFS with a coupling interval of one hour. For the AQMEII application, it is principally the IFS-MOZART3 configuration whose data has been processed for regional-scale model boundary concentrations.

The IFS uses a T159 spectrally-resolved global grid with a horizontal grid box size of about 125 km. MOZART3 uses a regular latitude/longitude grid of 1.875°x1.875°. The coupler performs horizontal bi-linear interpolations between the meteorological and CTM horizontal grids. The vertical coordinate is given by 60 hybrid-sigma pressure levels, with a model top at 0.1hPa. The same vertical coordinate is used by the IFS and all CTMs in the GEMS system to avoid the need for vertical interpolations. The coupling interval is 1-h which is the largest

acceptable time step for the IFS at a T159 resolution. Output is saved at 3-h intervals from the model simulations. Source emissions for the MOZART3 global simulations are specified as monthly averages for a base year of 2000 for anthropogenic trace gases (RETRO database; Schultz et al., 2009) and aerosols (EDGAR, <http://www.pbl.nl/en/themesites/edgar/index.html>; SPEW, Bond et al., 2004). Eight-day average wildfire emissions for the 2006 model application year are derived from the Global Fire Emissions Database (GFEDv2; van der Werf et al., 2006). Biogenic, sea salt, and dust emissions are parameterized within the model based on meteorological inputs (GEMS, 2010).

The advantage of using the GEMS re-analysis data to provide boundary concentrations for AQMEII simulations compared to other global model outputs is that the GEMS results include the assimilation of observations from satellite platforms. Figure 2 indicates the satellite data usage during the GEMS project, with the AQMEII target year of 2006 highlighted. There were multiple instruments available for measuring portions of the ozone (O<sub>3</sub>) column during 2006, including from SCIAMACHY, SBUV-2, and MLS instruments. Taken together, these data provide some vertical resolution to the O<sub>3</sub> column, with greatest fidelity in the stratosphere and upper troposphere (Flemming et al., 2011). In addition, CO columns are available from the MOPITT instrument, and aerosol optical depths (AODs) are derived from the MODIS instruments onboard the Terra and Aqua satellites. Information on atmospheric aerosols can be derived from the AOD retrievals. Complete details on the data assimilation system and implementation for GEMS can be found in Benedetti et al. (2009) and Inness et al. (2009).

The GEMS re-analysis outputs for 2006 were further processed by ECMWF for AQMEII use by interpolating all requested data for selected variables at 3-h intervals on an evenly spaced latitude/longitude grid (1.875°X1.875°) within specific geographical “cut-outs” from the global model domain. These cut-outs for Europe and for North America are illustrated in Figure 3. The spatial extent of each cut-out extends well beyond the minimum model domain sizes specified for AQMEII regional/continental domains such that AQMEII participants could use the data within the global cut-outs to derive the boundary concentrations for their own model exercises. Data from the lowest 47 IFS model layers (surface through 10hPa) were extracted within each cut-out over a full time period of 1 December 2005 through 31 December 2006, allowing for sufficient model spin-up time for the 2006 simulation. Table 1 lists the chemical and aerosol

species extracted for AQMEII use. These data were archived and made available to AQMEII participants by AQMEII collaborators in Météo-France.

Air quality modelers participating in AQMEII are then able to access these GEMS data and use them for specification of boundary concentrations. There are, however, additional assumptions and processing steps involved before the data can be used by the regional models. The GEMS data must be spatially interpolated for the boundaries of each regional model's native grid and temporally interpolated from the 3-h output interval to the 1-h boundary updates typically employed by the regional models. Also, the GEMS data contain fairly coarse chemical speciation of the gaseous organic compounds. Additional disaggregation of these organic compounds into the specific organic classes used by the tropospheric atmospheric chemistry mechanisms is usually necessary. In addition, the GEMS aerosol data for sea salt and dust may need to be redistributed based on the size distribution information carried within the regional model. Finally, the GEMS data provide information for most of the chemical and aerosol species needed at the models' boundaries that have significant transport influence. However, all pollutant species are not included in the GEMS data. Modelers may need to provide another source of boundary concentration data for aerosol sulfate and nitrate, for example, and additional gas-phase species that may be in their model, unless the boundaries are assumed to be zero, zero-gradient, or some fixed concentration.

There are a few caveats to note with regards to the use of the GEMS data for AQMEII. First, the SO<sub>2</sub> concentrations were calculated within IFS as a tracer using simple assumptions of emissions and prescribed loss. No chemical transformations were considered. Hence, recommendations were made to AQMEII modelers to use the SO<sub>2</sub> data with caution. Sea salt estimates were made as a function of wind speed and other environmental parameters in the IFS model. Based on the evaluation of the GEMS sea salt data for 2003, large overestimations of sea salt aerosol (over 400%) were observed over North America (GEMS, 2010). Preliminary analysis showed this to be true for 2006 as well, so AQMEII modelers were cautioned about the GEMS sea salt values. Estimates for sea salt over Europe, however, did not show these same tendencies of overestimation. In addition, organic carbon emissions from wildfires in the GEMS data set may have been overestimated in the lower model layers due to the lack of a plume-rise mechanism in the model and an overestimated persistence of the wildfires from the 8-day

resolution in the data being assimilated into the model. Cautions were therefore extended to AQMEII modelers regarding the use of organic carbon data from large wildfires.

### **3. Other Sources of Boundary Concentrations**

The modeling protocol for the AQMEII 2006 model simulations requested that participants derive their boundary concentrations from the GEMS re-analysis data described above. However, not all participants adhered strictly to this request. In fact, there was a variety of sources that were accessed for continental-scale model boundary concentrations. Table 2 presents examples of the various modeling systems used and the source of boundary concentration data in each instance. These data sources include other hemispheric and global modeling systems, as well as climatological or “background” tropospheric concentrations. Ideally, if all participants had used the same source for the boundary concentration data, as well as a common source for the emissions data, the data analysis and interpretation for the project would be assisted by minimizing confounding effects of different sources of data on model results. Some groups used the requested GEMS data source, as well as alternate sources, providing the data for sensitivity studies on the effect of alternate sources of boundary concentrations on their model simulations.

### **4. Impact of Boundary Concentrations on Continental Simulations**

With prevailing wind speeds across North America and Europe, air masses can traverse each continent within 3-5 days. Thus, the specification of inflow air quality boundary concentrations has the potential for significant impacts on continental model simulations, especially in areas of weaker internal model forcing from source emissions and atmospheric chemistry, and for chemical compounds having lifetimes of 3-5 days or longer. Pfister et al. (2011) used aircraft and satellite data during the ARCTAS-CARB field campaign during the summer of 2008 to evaluate the MOZART4 global chemical transport model’s simulation results for its chemical representativeness of chemical inflow to the U.S. west coast. The global model was shown to capture only about half of the observed free tropospheric air pollution variability. Sensitivity simulations with the regional WRF-Chem model, performed as part of the cited work, also showed that the temporal variability in the pollution inflow does clearly impact modeled surface

concentrations over California. It was suggested that time- and space-varying chemical boundary concentrations from global models provide useful input to regional models, but likely still lead to an underestimate of peak surface concentrations and the variability associated with the long-range transport of air pollution.

Hogrefe et al. (2011) performed long-term simulations with the Community Multiscale Air Quality (CMAQ) model using two sets of chemical boundary concentrations, one derived from time-invariant climatological vertical profiles and the other one from a global chemistry model. The comparison of both simulations revealed that lateral boundary concentrations have a significant impact on a regional air quality model's ability to simulate long-term O<sub>3</sub> variability and trends, especially for the middle and lower percentiles of the O<sub>3</sub> distribution. As an illustration, Figure 4 shows time series of May – September average daily maximum 1-h O<sub>3</sub> concentrations derived from observations and these two sets of CMAQ simulations for the time period from 1988 to 2005. It can be seen that the choice of boundary concentrations affects the magnitude of the mean concentrations as well as their inter-annual variability and trends. In this particular example, the CMAQ simulation using the time-invariant boundary concentrations shows better agreement with the observations in terms of absolute concentrations and trends while the CMAQ simulation using boundary concentrations derived from the global model shows better agreement in terms of inter-annual variability.

Li et al. (2002) used a five-year (1993-1997) simulation with the GEOS-Chem CTM and showed that North American pollution enhances surface O<sub>3</sub> in continental Europe by 2–4 ppbv on average in summer and by 5–10 ppbv during transatlantic transport episodes. Specifying the model continental-scale O<sub>3</sub> boundary concentrations correctly is significant in that the North American influence on surface O<sub>3</sub> in Europe is particularly strong at the thresholds used for the European air quality standards (55–65 ppbv). Simulating the daily variability of O<sub>3</sub> boundary concentrations was also shown to significantly improve both variability and biases of simulated daily O<sub>3</sub> maxima in Europe, in particular for the most frequent non-extreme values (Szopa et al., 2009).

Ratigejev et al. (2010) demonstrate that global CTMs have difficulty reproducing synoptic-scale pollution plumes during long-range transport. Numerical diffusion interacting with non-uniform atmospheric flows dissipates the plumes faster than ambient observations suggest. The authors state that novel numerical methods, such as adaptive grids or embedded Lagrangian

plumes, may circumvent the problem of accurately sustaining the plume integrity. Makar et al. (2010) evaluated ten different approaches for applying lateral and top climatological boundary concentrations for O<sub>3</sub> using the AURAMS regional CTM. They found that dynamic adjustments to the O<sub>3</sub> profile in response to the model-estimated tropopause height were needed to better match mass consistency between chemical and meteorological models. Their results highlight the importance of evaluating the boundary concentrations and mass consistency/correction algorithms with three-dimensional measurements.

#### *4.1 CMAQ Model – North America application*

The U.S. EPA contributed results to AQMEII from a 2006 North American simulation from the CMAQ model. The model domain included all of the continental U.S. (except Alaska), southern Canada, and northern Mexico. Meteorological data for the CMAQ simulation were derived from a continental model run of the Weather Research and Forecasting (WRF) model using four-dimensional data assimilation. The CMAQ model also made use of the standard protocol data provided by AQMEII for emissions and boundary concentrations (GEMS dataset). To assess the adequacy of the GEMS data for providing inflow O<sub>3</sub> boundary concentrations, we have examined the performance of the CMAQ model using observed data from the INTEX-B Ozone Network Study (IONS) of 2006 (Tarasick et al., 2010) for sounding locations near the west (inflow) coast of North America. The IONS 2006 study provided a total of 740 ozonesonde profiles from 23 sites across North America. Figure 5 displays the locations of these sites within the CMAQ modeling domain, with the shaded area indicating the region of interest for examining the boundary concentration impacts.

Figures 6a and 6b present the results of the CMAQ simulation and the observed O<sub>3</sub> vertical profile from the Trinidad Head site on the northern California coast averaged over all March (n=6) and August (n=30) profiles, respectively. For both months, the model and observed profiles agree fairly well at altitudes corresponding to the upper troposphere and stratosphere. Here, the model is greatly influenced by the boundary concentrations as there are no local emissions and little atmospheric chemistry to influence the estimated concentrations. However, in the lower and mid troposphere, the model significantly underestimates the O<sub>3</sub> concentrations compared to the observed profile. Also indicated on the figures are the concentrations from CMAQ attributable to a boundary concentration tracer for O<sub>3</sub>. In these calculations, the impact

of the spatially and temporally varying O<sub>3</sub> boundary concentrations was tracked using a tracer species that underwent advective, turbulent, and cloud transport, and wet and dry deposition similar to O<sub>3</sub>. Since the tracer was not subject to any chemical loss, the inferred impact on the net simulated O<sub>3</sub> may be slightly overestimated. Nevertheless, the tracer provides a direct estimate of the impact of the GEMS boundary concentrations on simulated O<sub>3</sub> patterns. With little local photochemistry occurring in March, it is evident that the lower portion of the profile is completely dictated by the boundary concentrations, while in August with more active photochemistry the lowest portion of the profile shows enhanced concentrations as compared to the boundary tracer. This is further illustrated in Figure 7 which presents the average fractional contribution of the boundary concentrations to the simulated vertical profile at Trinidad Head for the months of March and August 2006. It is evident that above 3-4 km the simulated O<sub>3</sub> variability is largely dictated by the boundary concentrations used in the simulation.

Similar analyses for the Kelowna site in British Columbia, Canada are presented in Figure 8. (There were 2 profiles available in March and 26 in August at Kelowna.) Results of these comparisons are comparable to those of the Trinidad Head site, although this site shows larger deviations in the upper portion of the profiles between CMAQ and the observed data. Results of comparisons at other west coast sites (not shown) demonstrate similar behavior, with CMAQ generally underestimating O<sub>3</sub> near the surface and in the lower and mid troposphere, with the greatest discrepancies in winter and the least in summer. From these profiles it appears that the O<sub>3</sub> boundary concentration tracer, as a surrogate for the GEMS data, has considerably underestimated the inflow of O<sub>3</sub> to the west coast of North America in the lower troposphere, especially during the winter and spring. It is not surprising that the GEMS data should better reflect the observed O<sub>3</sub> profile in the upper levels as compared to levels closer to the surface. The GEMS re-analyses have made extensive use of data assimilation for O<sub>3</sub> based on satellite retrievals. These derived measurements are most accurate for the stratospheric O<sub>3</sub> burden and become more uncertain in the lower portions of the profile. In the lower levels, the GEMS data are more reflective of the results of the MOZART3 model simulations which appear to have systematically underestimated tropospheric O<sub>3</sub> in the northeast Pacific region, and generated a low bias in the specified inflow concentrations for North America, possibly due to emissions uncertainties and an overestimation of dry deposition.

A distinct advantage of the data assimilation aspect of the GEMS re-analysis is that real-time events, such as large forest fires or dust storms, are seen by satellite sensors and can be incorporated in the model simulation. Figure 9 presents the vertical profiles of primary organic particulate matter (OM) along each boundary of the CMAQ North American domain averaged over the period of 21-30 June 2006 from the GEMS database. One can readily see the strong impact of a large wildfire occurring at the time in the Canadian boreal forest. The impact on the surface layer CMAQ estimates of primary organic particulate matter is seen in Figure 10 in which the 01 UTC concentrations are presented for 30 June 2006. The effects of the wildfires north of the CMAQ domain are evident as the boundary concentrations have been advected into the northern portion of CMAQ's computational domain. Satellite measurements confirm the elevated aerosol loadings in this area from the wildfires.

#### *4.2 CHIMERE Model – North America application*

The sensitivity of concentrations simulated by a regional model to chemical boundary concentrations was tested by IPSL-France using the CHIMERE model (Bessagnet et al., 2004), by using in separate simulations the GEMS boundary concentrations provided to AQMEII (simulation A) and the boundary concentrations typically used in CHIMERE studies, as provided by monthly climatologies of the LMDzINCA global model (Hauglustaine et al., 2004) for gas-phase species and the GOCART model for aerosol species (Ginoux et al., 2001). For this case (simulation B), model runs were carried out with the same settings as for simulation A for other model parameters, except that boundary concentrations were kept constant within each month but varied along the model boundaries. It must be noted that CHIMERE only simulates concentrations within the lower atmosphere since it has a top boundary at 500 hPa. Concentrations within the modeling domain are thus sensitive to both lateral and top boundary concentrations.

The mean O<sub>3</sub> surface concentration differences between simulations B and A have been calculated for each season (Winter=DJF, Spring=MAM, Summer=JJA, Fall=SON) and are represented in Figure 11. The sensitivity to O<sub>3</sub> boundary concentrations differs from one season to another. In winter and spring, strong winds and vertical mixing induce a larger sensitivity to boundary concentrations than in summer and fall. For instance, in winter, seasonal mean concentration differences between the two simulations in the center of the domain and those near

the boundaries vary by a factor of 2 or so. In contrast, in summer and fall the concentration differences vary by more than a factor of 5 between the center of the domain and the regions near the boundaries, indicating that boundary concentrations have a relatively smaller impact on the inner portions of the domain compared to winter and spring. However, in all seasons studied, the impact of boundary concentrations extends inland far from the boundaries. The Central-East U.S. shows the smallest influence from the boundary concentrations.

Note also that the difference between mean LMDzINCA-driven and MOZART-driven simulations remains positive across the domain, because the LMDzINCA O<sub>3</sub> boundary concentrations are higher than the MOZART ones. This difference is largest in the winter season, reaching about 15 ppb, while in summer it reaches 10 ppb. The magnitude of these differences in the seasonal mean concentrations caused by different boundary concentrations is comparable to those shown in Hogrefe et al. (2011; see Figure 11 therein).

#### *4.3 CHIMERE Model – Europe application*

The CHIMERE model has also been applied over Europe using the TNO emissions inventory at a 0.25° horizontal resolution by INERIS-France. As in the North American case, the model domain extends vertically to 500hPa. To assess the impact of the temporal resolution of the boundary concentrations (BCs) on air quality modeled in the regional domain, the whole year 2006 has been run twice with 3-hourly GEMS BCs for both gaseous species and aerosols (3HR), and with a monthly climatology derived from the same dataset (gas and aerosols as well; CST). In the CST run, for a given time in a month, the model is driven with constant BCs; no interpolation is performed between two consecutive months.

Figure 12 displays the average difference between the simulations with 3-hourly BCs and the monthly climatology for O<sub>3</sub>. With both sets of BCs derived from the same global simulations, these plots would exhibit a difference of zero throughout the domain if the regional model behaved as a linear operator. Since that is obviously not the case, these plots reflect the combined impact of (1) the non-linearity of the regional model together with (2) the skewness of the O<sub>3</sub> distribution at the boundaries. For instance, the difference is consistently negative at the southern boundary showing that the mean BCs (used in the CST run) lead to an overestimation of O<sub>3</sub> compared to the 3HR BCs. On the western and northern parts of the domain, the situation is less straightforward. In summer (JJA), climatological BCs lead to an overestimation of O<sub>3</sub>, but

in spring and fall they yield an underestimation compared to time-varying fields. It is likely that stratospheric intrusions into the troposphere captured in the GEMS re-analysis (which may have an impact in the CHIMERE model down to the surface by means of vertical mixing) play an important role on these patterns. As isolated, yet very concentrated, layers of O<sub>3</sub>, these events have a larger impact on the average than on the median concentrations. Depending on their geographical and seasonal variability they could thus be responsible for the patterns observed in Figure 12.

Figure 13 shows the difference of standard deviation between the 3HR and CST simulation results for both O<sub>3</sub> and PM<sub>10</sub> concentrations. Using the 3HR fields at the boundary has a noticeable impact on the outskirts of the model domain. Since Europe is mainly affected by sporadic and large dust outbreaks from the Sahara, the southern boundary displays higher variability with 3HR BCs. The eastern part of the domain includes a fire emission zone in Russia, which leads to higher standard deviations in this region. Since sea salt BCs have not been used from the GEMS dataset there are no specific patterns observed in the western and northern parts of the domain. Table 3 lists the global standard deviation of daily mean concentrations of O<sub>3</sub>, NO<sub>2</sub>, and PM<sub>10</sub> for all European air quality monitoring stations taken from the AIRBASE dataset (all available stations). For short-lived species like NO<sub>2</sub>, the time-varying BCs have a negligible impact. However, for both O<sub>3</sub> and PM<sub>10</sub>, using the 3HR fields at the boundaries contributes to obtaining a slightly larger variability that is more in agreement with the observations for O<sub>3</sub> and NO<sub>2</sub>. The time variability is impaired for PM<sub>10</sub> showing that the predictability of dust events (intensity and occurrence) remains difficult as shown by Menut et al. (2009). If dust models can provide a better measure of variability on seasonal or monthly bases, these models could better predict dust concentrations over Europe on a daily basis. It should be noted that the estimates provided by this comparison at the station locations overly weight the center of the domain, where stations are by far more numerous and the impact of BCs is less noticeable.

Recently Pfister et al. (2011) conducted similar sensitivity experiments with the WRF-Chem model in which boundary concentrations for North American inflow were derived from the MOZART4 global chemical transport model using 3-hourly varying data as well as an experiment using boundary data averaged over their simulation period (14-30 June 2008; during ARCTAS-CARB field experiments). Their results focused on inflow to California during that

period, and much like the results presented here, the variability in boundary concentrations was better captured with the higher temporal resolution. One difference that was noted was in the mean O<sub>3</sub> concentrations at the boundaries, where Pfister et al. (2011) reported the same mean O<sub>3</sub> values irrespective of the temporal averaging at the boundaries, while the current study noted differences in the means based on the temporal averaging. This discrepancy in the findings is likely due to the longer simulation period used here (one year) compared to the 17-day simulation period in the Pfister study. The longer simulation allowed for additional anomalous events, such as stratospheric intrusions of O<sub>3</sub> into the lower troposphere, to affect the average in non-linear fashion.

#### *4.4 CMAQ Model – Europe application*

The CMAQ model was applied over the European domain for the year 2006 using the input datasets prescribed for AQMEII (including the GEMS boundary concentrations) by the University of Hertfordshire-UK. An evaluation of the CMAQ calculations, for the continental-scale domains in North America and Europe, is given in Appel et al. (2011; this issue). To examine the impacts of the boundary concentrations on the model results, an additional simulation was performed for 2006 using boundary concentrations provided by the global chemical transport model GEOS-Chem, version 8-03-01 (see [http://wiki.seas.harvard.edu/geos-chem/index.php/Main\\_Page](http://wiki.seas.harvard.edu/geos-chem/index.php/Main_Page)). The GEOS-Chem model was run with 2° x 2.5° horizontal cell size with 47 hybrid pressure-sigma vertical levels. The model was driven by assimilated meteorological data from the Goddard Earth Observing System (GEOS-5) at the NASA Global Modeling and Assimilation Office (GMAO). We used the chemistry mechanism NO<sub>x</sub>-O<sub>x</sub>-hydrocarbon-aerosol to simulate O<sub>3</sub> and aerosols (Jacob, 2000; Bey et al., 2001). The aerosol components included sulfate, nitrate, ammonium, black carbon, organics, mineral dust, and sea salt (see Park et al., 2004; Hu et al., 2007). The emission inventories were separated into four source categories: anthropogenic, biomass, biofuel, and biogenic. Sources of mineral dust and sea salt are dealt with separately. The anthropogenic emissions were obtained from the Global Emissions Inventory Activity (GEIA) dataset. Biomass burning and biofuel-use emissions were derived from Duncan et al. (2007). Biogenic emissions included isoprene, methyl butenol, acetone, and alkene.

As in Section 4.1, we focus on the adequacy of the boundary concentration data for setting the inflow of O<sub>3</sub> into the modeling domain. Figure 14 presents the observed and modeled vertical distributions of O<sub>3</sub> at Lerwick, Shetland Mainland, UK, for the year 2006. The Lerwick Observatory is situated in a remote location representative of background (inflow) atmospheric concentrations. The time-height evolution of O<sub>3</sub> in Figure 14a was compiled from measurements by a UK Met Office ozonesonde of the Electrochemical Concentration Cell (ECC) type (Komhyr et al., 1995). The two model calculations agree fairly well with the observations at altitudes above 6-8 km (i.e., in the upper troposphere/lower stratosphere). While the GEMS re-analysis included the assimilation of O<sub>3</sub> data from satellite observations, the GEOS-Chem simulation included stratospheric O<sub>3</sub> chemistry based on a climatological representation of species sources and sinks. Both techniques appeared to work well in reproducing the O<sub>3</sub> profile at the higher altitudes. Interestingly, the agreement degrades in the lower troposphere when using the GEMS boundary concentrations, while it remains fair when using those provided by GEOS-Chem. The difference between the observed O<sub>3</sub> concentrations and those of the simulation using the GEMS boundary concentrations is most dramatic for the first four months of 2006, with low biases as large as 20 ppb.

Similar findings can be observed in Figure 15, where the observed and modeled ground-level O<sub>3</sub> concentrations at Mace Head, Republic of Ireland, are shown. The location of the monitoring station on the Atlantic Coast makes it a representative site for background concentrations of substances in the atmosphere. The O<sub>3</sub> concentration at the site, simulated using the GEMS boundary concentrations, appears to be biased low for the first four months of 2006. Afterwards, the two model calculations give comparable results. This highlights the importance of boundary concentrations in setting the baseline concentrations in the modeling domain.

## **5.0 Summary and Conclusions**

The AQMEII project on regional-scale air quality model system evaluation and inter-comparison has proposed the use of a common set of boundary concentrations to be specified to the regional modelers for all to use in an effort to minimize differences across the models from this particular aspect of the modeling protocol. The GEMS re-analysis air quality dataset has been provided for this purpose from the ECMWF for the AQMEII focus year of 2006. The re-

analysis is produced by assimilating satellite observations of select chemical and aerosol species into a coupled model system, which consists of ECMWF's IFS and the MOZART3 CTM. The resulting dataset provides spatially- and temporally-resolved information on concentrations of most of the key transported species of interest to the AQMEII regional modelers for use in their modeling systems. Each regional modeling group then must cast these data into forms directly usable by their particular model. This processing may include further spatial and temporal interpolations as well as chemical speciation of the trace gases and aerosols for the particular chemical and aerosol mechanisms used by the model.

This study reveals that specification of O<sub>3</sub> profiles from the GEMS dataset at the boundaries of the North American and European modeling domains for the 2006 simulations offered good agreement in the upper troposphere and lower stratosphere with an independent set of observations from ozonesondes. For the North American domain, the O<sub>3</sub> boundary concentrations throughout 2006 were underestimated in the GEMS dataset in the lower to mid troposphere, with greater biases in winter and spring and lower biases in summer. In the European simulations the GEMS dataset yielded O<sub>3</sub> boundary concentrations that were consistent with ozonesonde observations except for the first quarter of 2006 when the lower tropospheric O<sub>3</sub> values were biased low by as much as 20 ppb. SO<sub>2</sub> concentrations in the GEMS dataset were derived from simple assumptions and not based on a complete chemical description in the global modeling. Hence, AQMEII modelers were cautioned regarding their use for continental boundary concentrations. For particulate matter we found that organic carbon from large wildfires was well detected by the GEMS data assimilation system, but that concentrations could be overestimated near the surface due to lack of a plume rise mechanism and eight-day temporal averaging of fire emissions. Sea salt was greatly overestimated near the boundaries of the North American domain, although the bias was less over the North Atlantic near the borders of the European domain. The GEMS dataset did not provide estimates of particulate sulfate or nitrate.

Specification of boundary concentrations is a required element in modeling with limited-area air quality models, such as regional- to continental-scale PAQMs. The limited area models are typically quite sensitive to these specified concentrations, especially in areas of weaker internal forcing by emissions and chemistry within the model's computational domain. Sensitivity simulations performed with the CHIMERE model emphasize that the impact of O<sub>3</sub> boundary

concentrations can extend far into the model domain beyond the boundaries. These results also show that boundary concentrations derived from monthly O<sub>3</sub> climatologies can deviate substantially from more temporally-resolved concentrations. The tropospheric impacts of stratospheric O<sub>3</sub> intrusion events, for example, can be greatly damped or eliminated by monthly averages.

Long-duration simulations, such as the full-year simulations performed within the AQMEII project, require boundary concentrations that reflect not only day-to-day variations but also seasonal and inter-annual changes in the global environment. Use of global CTMs to provide these boundary concentrations is a logical and convenient mechanism for their specification. It should be noted, however, as seen in these AQMEII model results, as well as the results presented in Hogrefe et al. (2011), that the global models may contain errors or biases in their simulated results that can then propagate into the regional models through the boundaries and affect the results within the model domain. The use of data assimilation in the global models can help minimize these errors, but cannot eliminate them. The assimilated satellite observations provide mainly vertically integrated column values, which makes it more difficult for the assimilating model to obtain realistic concentration profiles close to the surface. Examining several sources of boundary concentrations, such as alternate global CTMs, may provide useful information to modelers on ranges of boundary concentrations to consider and enable quantification of uncertainty associated with this model input variable. With the tightening of air quality standards and the imposition of emissions control programs over the past few decades, air pollution levels have generally been declining in many nations, leading to the need to properly quantify background pollution as an “irreducible” portion of the local pollutant burden. Therefore, the process of specifying the boundary concentrations for limited area models is an important issue and must be performed with careful scrutiny to assure the best possible outcome from regional-scale model simulations.

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Table 1. Gas-phase reactive chemical and aerosol species extracted from GEMS data

<b>Gas-phase Reactive Chemical Species<sup>1</sup></b>		
O <sub>3</sub> (ozone)	HNO <sub>3</sub> (nitric acid)	C <sub>2</sub> H <sub>6</sub> (ethane)
CO (carbon monoxide)	HO <sub>2</sub> NO <sub>2</sub> (peroxynitric acid)	ISOP (isoprene)
CH <sub>2</sub> O (formaldehyde)	PAN (peroxy acetyl nitrate)	TOLUENE (sum of C <sub>7</sub> ,C <sub>8</sub> ,C <sub>9</sub> aromatics)
NO (nitrogen oxide)	CH <sub>4</sub> (methane)	BIGENE (>C <sub>3</sub> alkenes)
NO <sub>2</sub> (nitrogen dioxide)	CH <sub>3</sub> CHO (acetaldehyde)	BIGALK (>C <sub>3</sub> alkanes)
<b>Aerosol Species<sup>2</sup></b>		
Sea Salt (0.03-0.5 micrometer)	Desert Dust (0.03-0.55 micrometer)	OM (organic matter)
Sea Salt (0.5-5 micrometer)	Desert Dust (0.55-0.9 micrometer)	BC (black carbon)
Sea Salt (5-20 micrometer)	Desert Dust (0.9-20 micrometer)	SO <sub>2</sub> (sulfur dioxide-gas) <sup>3</sup>
<b>Other Variables</b>		
T (temperature) <sup>4</sup>	PS (surface pressure) <sup>5</sup>	

<sup>1</sup>volume mixing ratios; units: mole mole<sup>-1</sup>

<sup>2</sup>units: μg m<sup>-3</sup>; size distribution bins are chosen so that roughly 10, 20 and 70 percent of the total mass of each aerosol type are in the three successive bins

<sup>3</sup>SO<sub>2</sub> here is based on simple assumptions of emissions and prescribed loss; no active chemistry

<sup>4</sup>units: °K; used in conversion between molar and mass mixing ratios for gas-phase species

<sup>5</sup>units: Pa; used in conversion of model layer number to atmospheric pressure

Table 2. Boundary Concentration Data Sources Used by AQMEII Participants

<b>AQMEII Participant</b>	<b>Model System</b>	<b>Source of Boundary Concentration Data</b>
ZAMG - Austrian Weather Service - AT	ALADIN/CAMx	CECILIA model
Environment Canada - CA	GEM/AURAMS	Climatological chemical boundary concentrations with dynamic O <sub>3</sub> adjustments (Makar et al., 2010)
Paul Scherrer Institute - CH	WRF/CAMx	GEMS re-analysis
Leibniz Institute for Tropospheric Research - DE	COSMO	GEMS re-analysis
HZG Research Centre - DE	CCLM/CMAQ	GEMS re-analysis
University of Aarhus - DK	MM5v3/DEHM	DEHM hemispheric simulation
Barcelona Supercomputing Centre - ES	WRF/CMAQ/DREAM8b	GEMS re-analysis
Finnish Meteorological Institute - FI	ECMWF/SILAM	GEMS re-analysis
CEREA - FR	POLYPHEMUS	GEMS re-analysis
INERIS/IPSL - FR	CHIMERE	GEMS re-analysis; LMDzINCA model
Meteorological Service of Croatia - HR	EMEP/HIRLAM-PS	EMEP model
TNO - NL	LOTOS-EUROS	GEMS re-analysis
Kings College London - UK	WRF/CMAQ	STOCHEM model
University of Hertfordshire - UK	WRF/CMAQ	GEMS re-analysis; GEOS-Chem
Environmental Protection Agency - US	WRF/CMAQ	GEMS re-analysis
Environ Corporation - US	WRF/CAMx	GEMS re-analysis

Table 3. Standard deviation of daily means at the location of surface air quality monitoring stations for O<sub>3</sub>, NO<sub>2</sub> and PM10 in the data and in the two CHIMERE model simulations (3HR and CST).

	Obs. data	3HR	CST
O <sub>3</sub> (µg/m <sup>3</sup> )	26.17	21.67	21.26
NO <sub>2</sub> (µg/m <sup>3</sup> )	16.48	13.11	13.12
PM10 (µg/m <sup>3</sup> )	22.75	24.11	23.93

## Figure Captions

**Fig 1.** Schematic of the components of the GEMS modeling system (figure provided courtesy of M.G. Schultz, FZ-Jülich).

**Fig. 2.** Timeline of satellite data usage for variables used in the GEMS re-analysis. AQMEII modeling is focused on 2006.

**Fig. 3.** Domain cut-outs from global GEMS re-analysis grid used for providing boundary concentrations for (a) Europe and (b) North America.

**Fig. 4.** Time series of May – September average daily maximum 1-h  $O_3$  concentrations for observations and two sets of CMAQ simulations. CMAQ/Profile refers to the CMAQ simulations utilizing time-invariant climatological vertical profiles for the specification of boundary concentrations while CMAQ/Global refers to the CMAQ simulations utilizing boundary concentrations derived from a global chemistry model. The time series represent spatial averages over the location of all  $O_3$  monitors in the modeling domain. Further details on these simulations are provided in Hogrefe et al. (2011).

**Fig. 5.** Locations of IONS-2006 North American ozonesonde launch sites within the CMAQ modeling domain. Shaded area represents the analysis region for inflow air from the western boundary of the domain.

**Fig. 6.** Mean  $O_3$  concentrations for (a) March 2006 and (b) August 2006 for vertical profiles at Trinidad Head, California (US). Observed mean concentrations (with standard deviations) are indicated by gray circles; CMAQ mean concentrations are indicated by open circles; boundary-tracer concentrations are indicated by triangles.

**Fig. 7.** Fractional contribution of the boundary concentrations to the simulated mean vertical  $O_3$  distributions during March and August 2006 at Trinidad Head.

**Fig. 8.** Same as Fig. 6, except for Kelowna, British Columbia (Canada).

**Fig. 9.** Vertical profiles from the GEMS re-analysis database of primary organic particulate matter (OM) along each boundary of the CMAQ model North American domain averaged over the period of 21-30 June 2006.

**Fig. 10.** CMAQ model-predicted average primary organic aerosol on 30 June 2006 at 01 UTC using GEMS boundary concentrations.

**Fig. 11.** Mean seasonal distribution for 2006 of the difference of surface  $O_3$  concentration between the CHIMERE simulation using the LMDzINCA (gas phase) and GOCART (aerosols) boundary concentrations and that using the GEMS boundary concentrations. Concentration differences are in ppb. The horizontal resolution of the simulations is 36 km. Each panel corresponds to a seasonal mean.

**Fig 12.** Average difference of modeled O<sub>3</sub> (ppb) at the surface in the CHIMERE model between the simulation driven by 3-hourly boundary concentrations and the monthly climatology; DJF (December, January, February), MAM (March, April, May), JJA (June, July, August), SON (September, October, November).

**Fig. 13.** Difference in standard deviation of modeled O<sub>3</sub> (left, ppb) and PM10 (right,  $\mu\text{g}/\text{m}^3$ ) at the surface in the CHIMERE model between the simulation driven by 3-hourly BCs and when a monthly climatology is used at the boundaries.

**Fig 14.** Vertical distribution of O<sub>3</sub> at Lerwick, Shetland Mainland, UK, for the year 2006 as (a) observed, (b) simulated with CMAQ model using the GEMS boundary concentrations, and (c) simulated with CMAQ model using the GEOS-Chem boundary concentrations.

**Fig. 15.** Time series of observed (black) and CMAQ-simulated (colors) 1-h O<sub>3</sub> concentrations at Mace Head, Republic of Ireland, for the year 2006 for (a) the simulation using the GEMS boundary concentrations and (b) that using the GEOS-Chem boundary concentrations. Color variations in simulated time series represents different seasons.

Figure 1

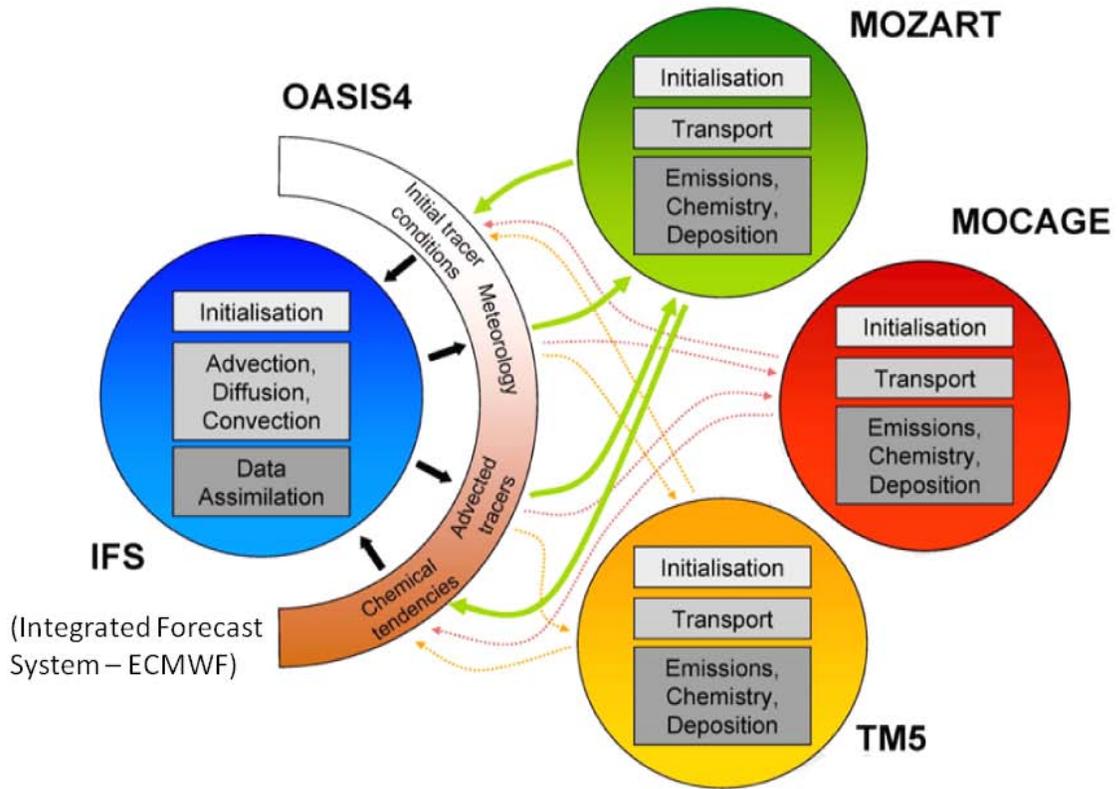


Figure 2

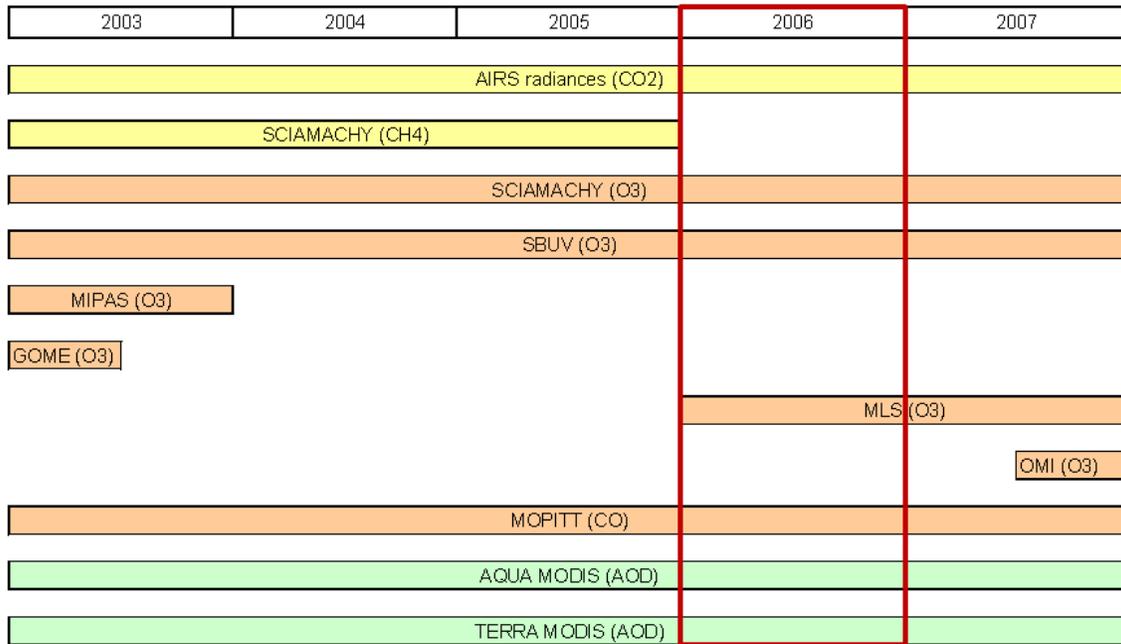


Figure 3 (a)

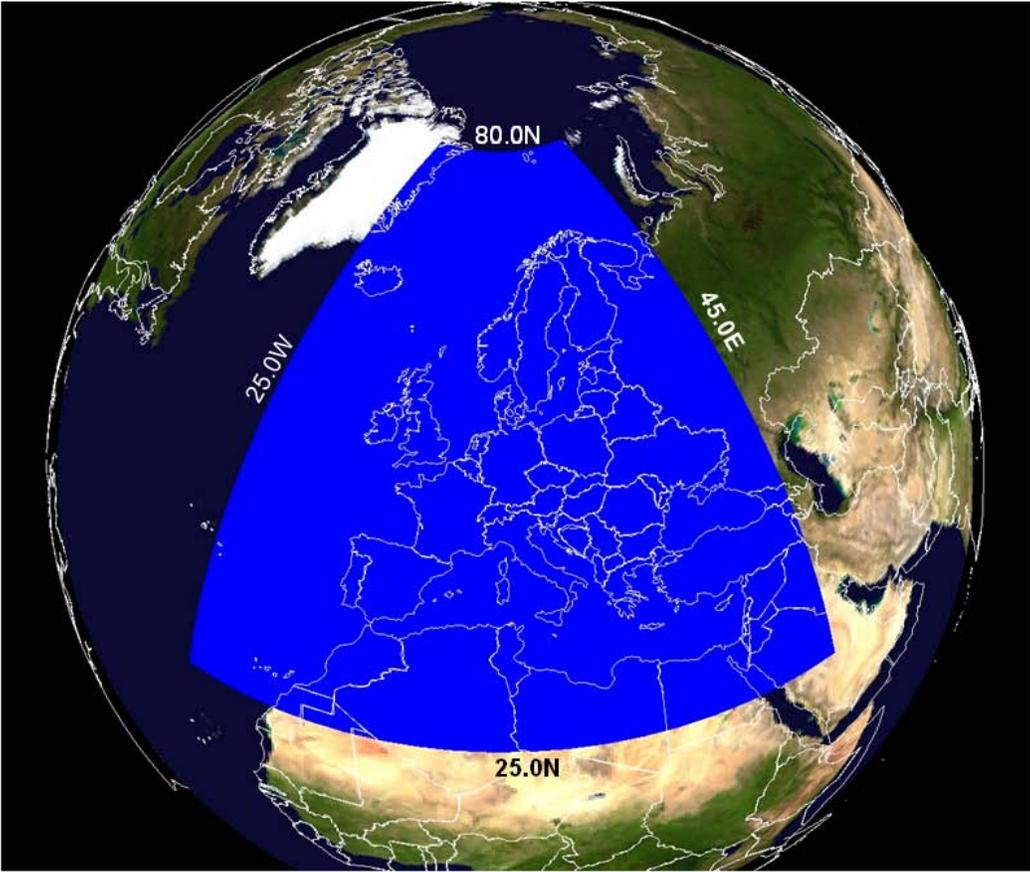


Figure 3b



Figure 4.

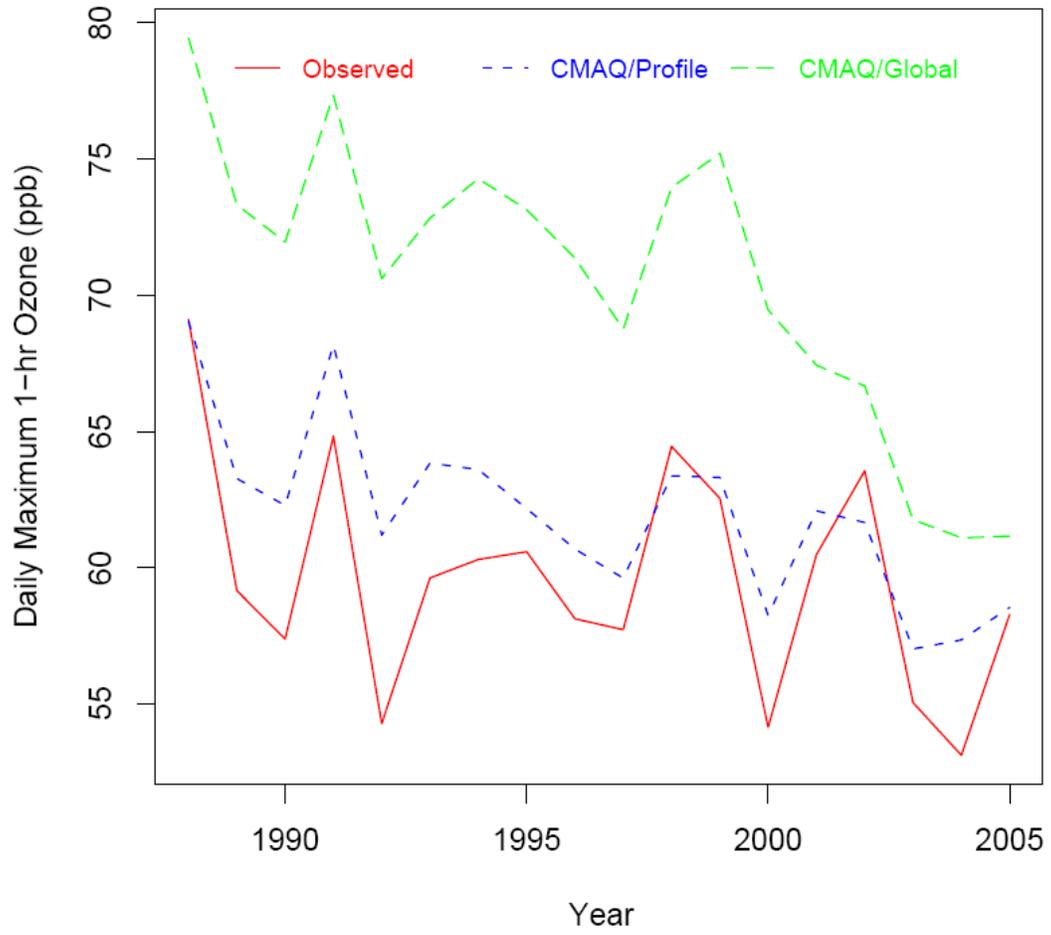


Figure 5

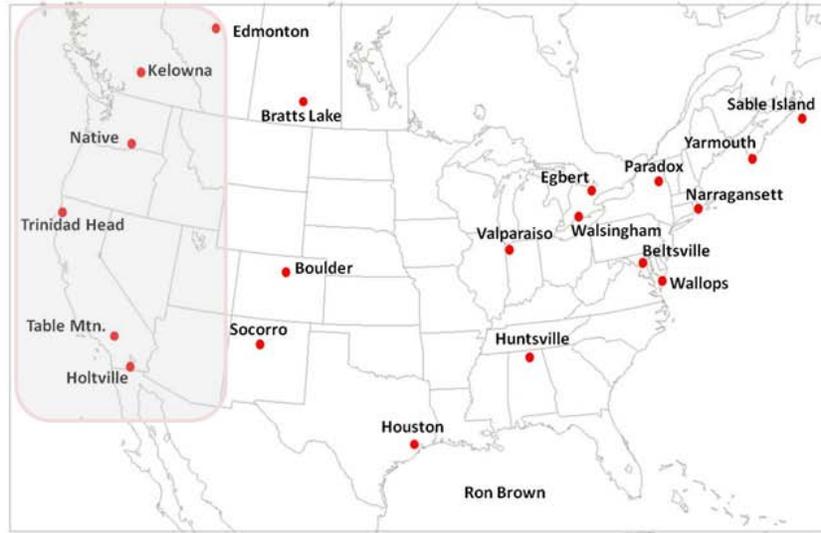


Figure 6a

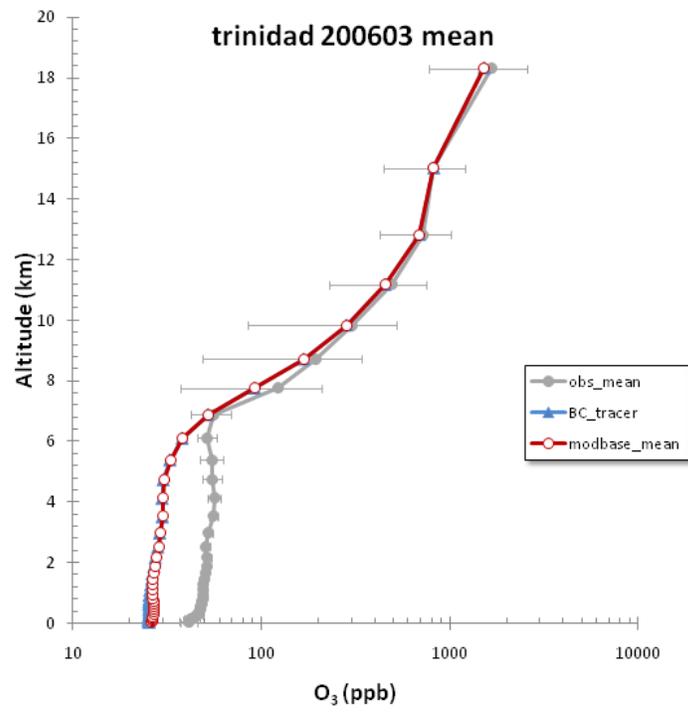


Figure 6b

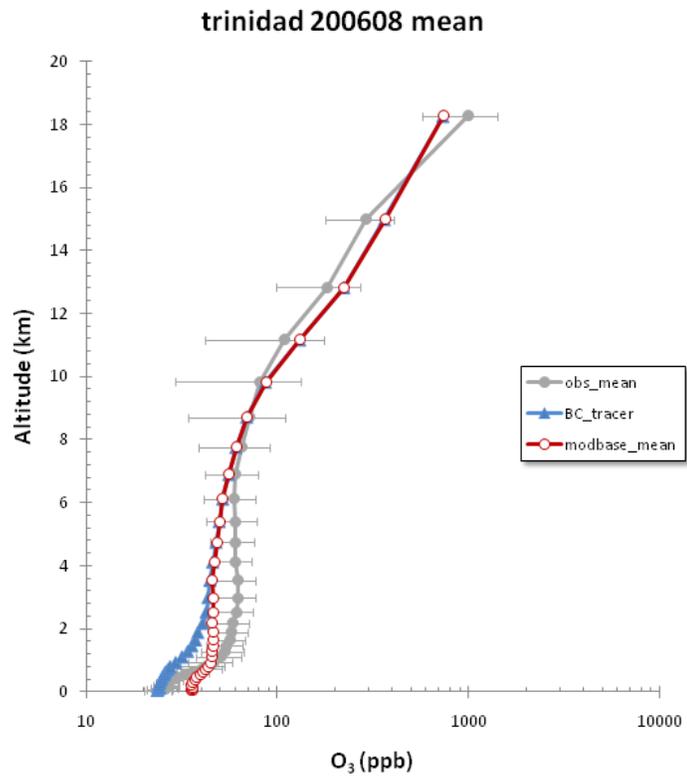


Figure 7

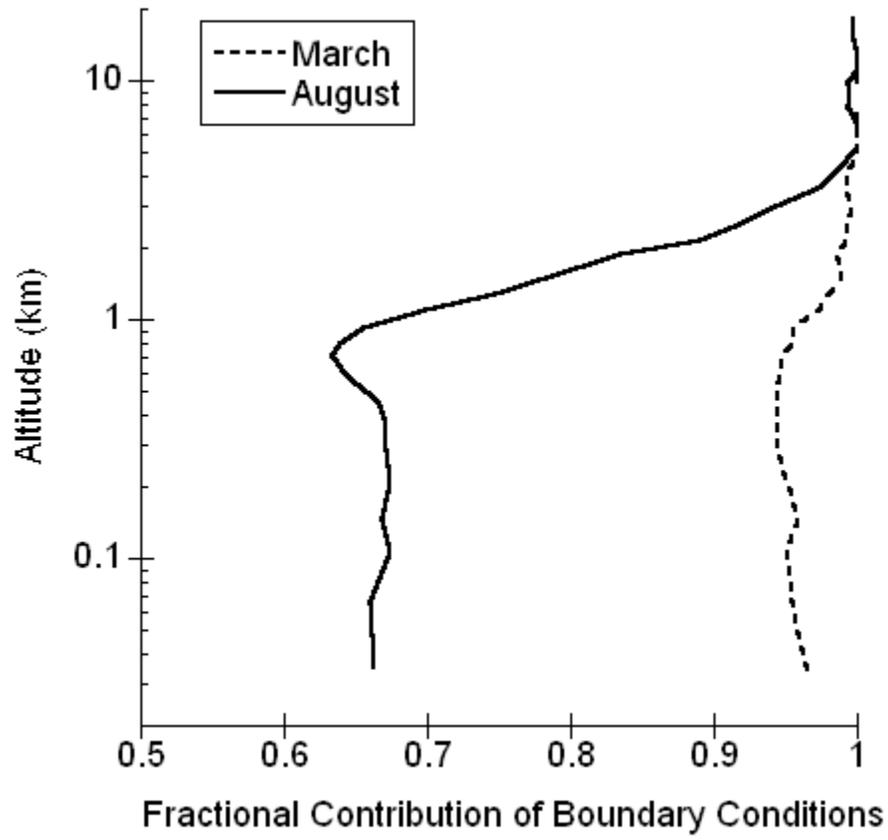


Figure 8a

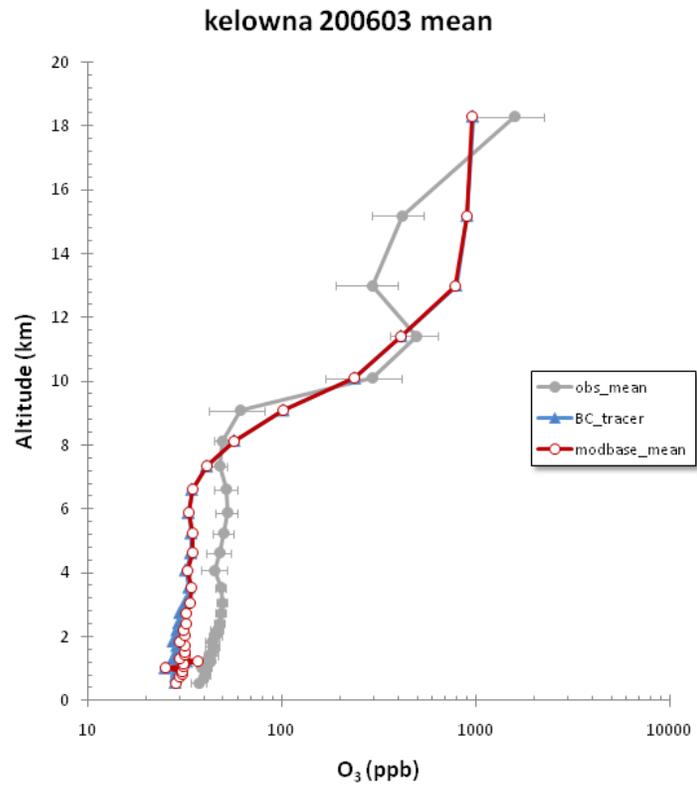


Figure 8b

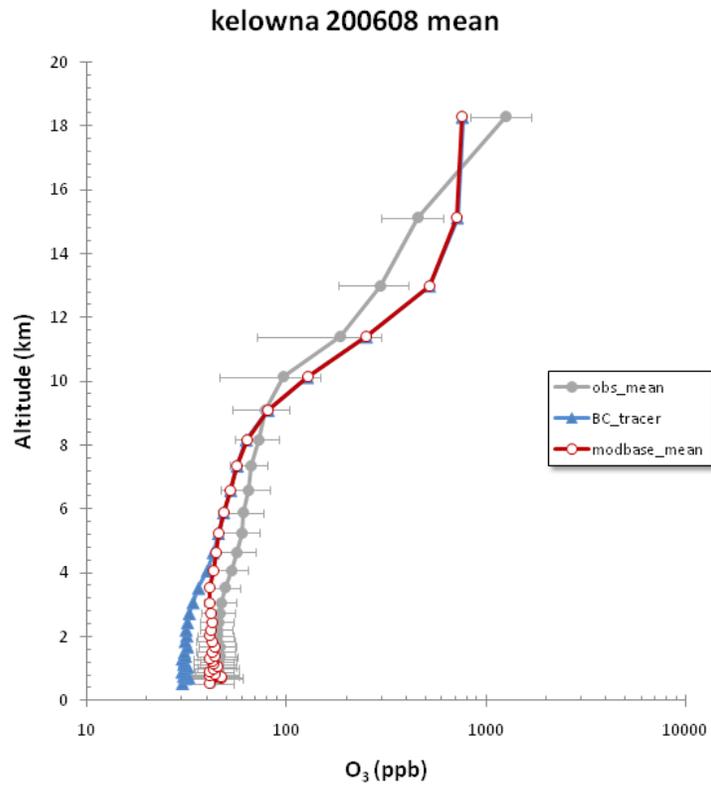


Figure 9

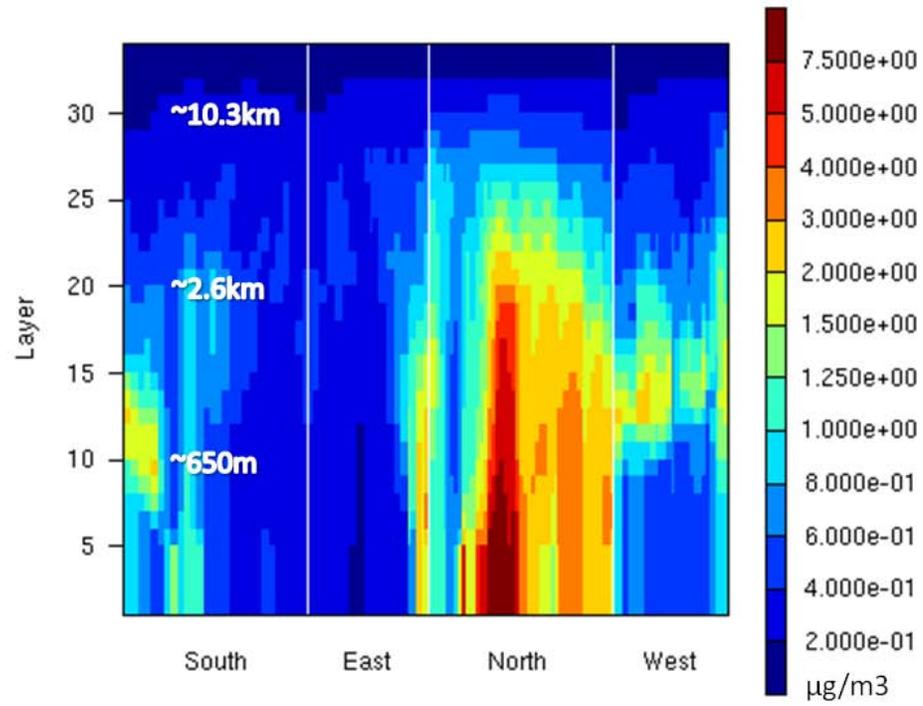


Figure 10

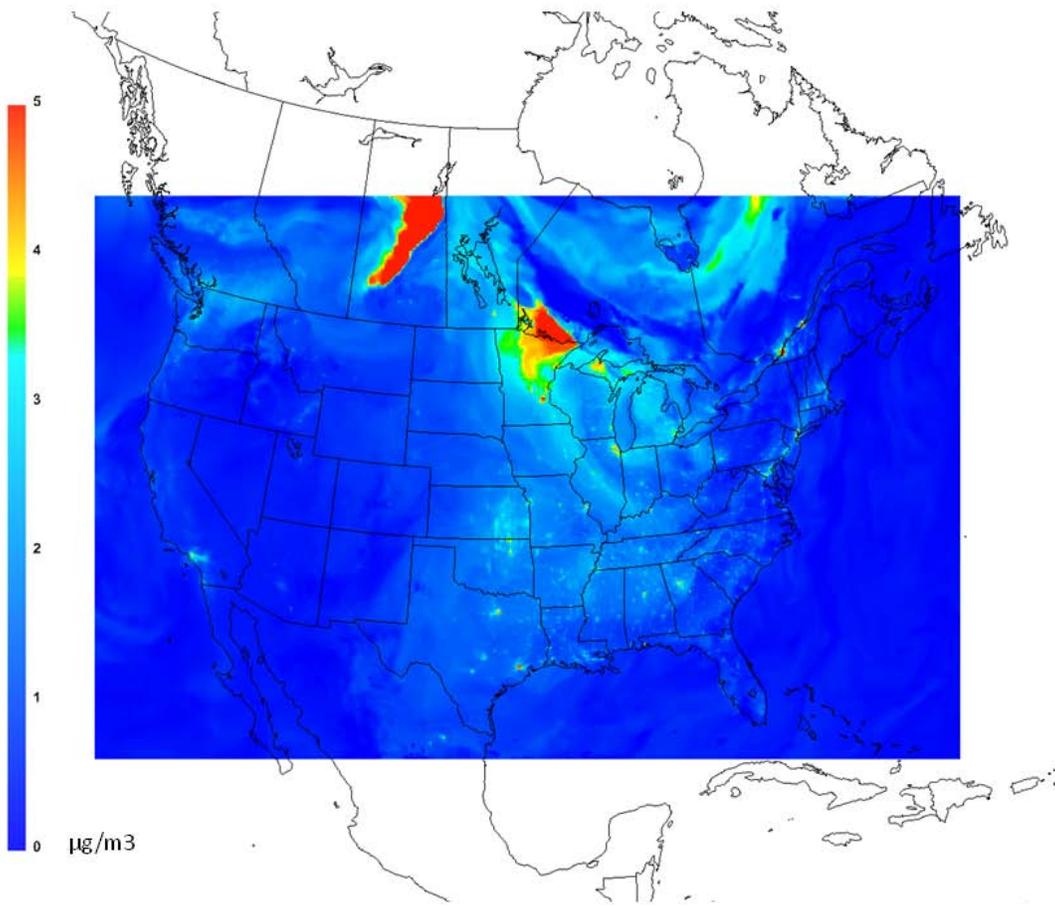


Figure 11

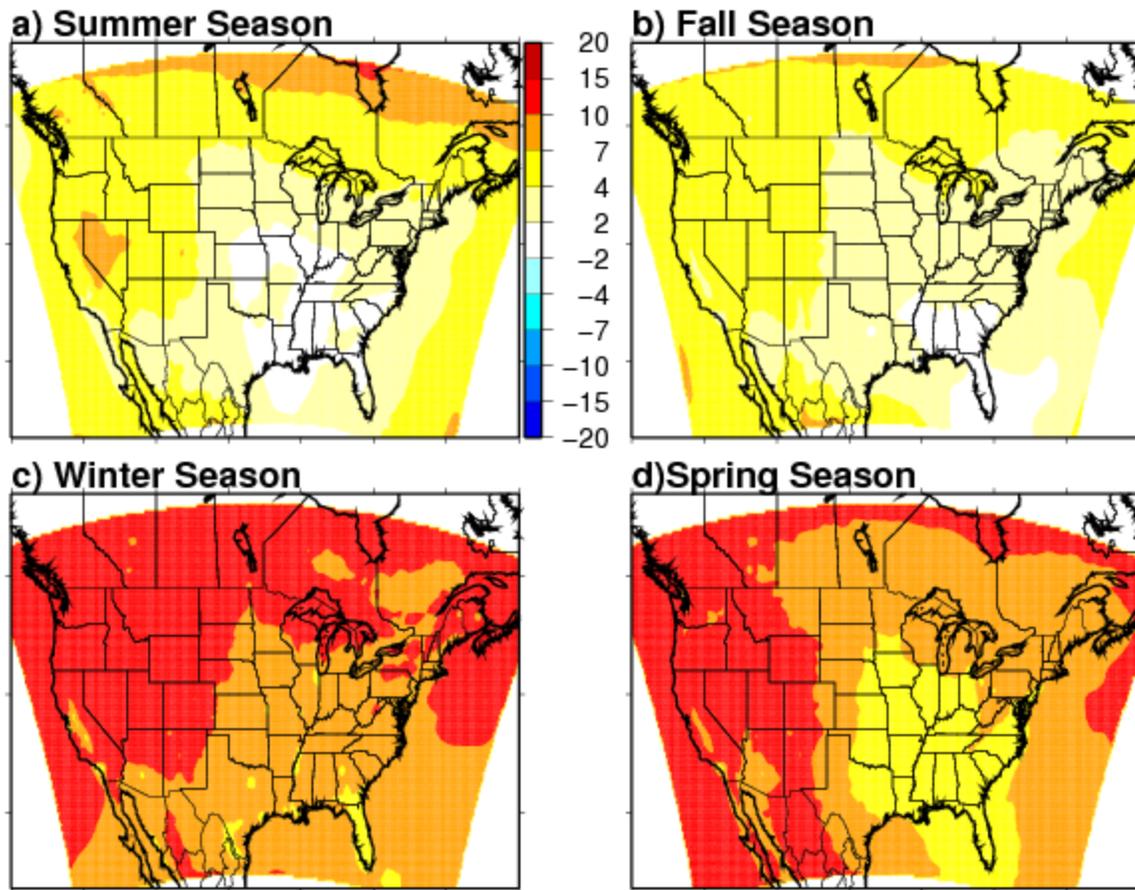


Figure 12.

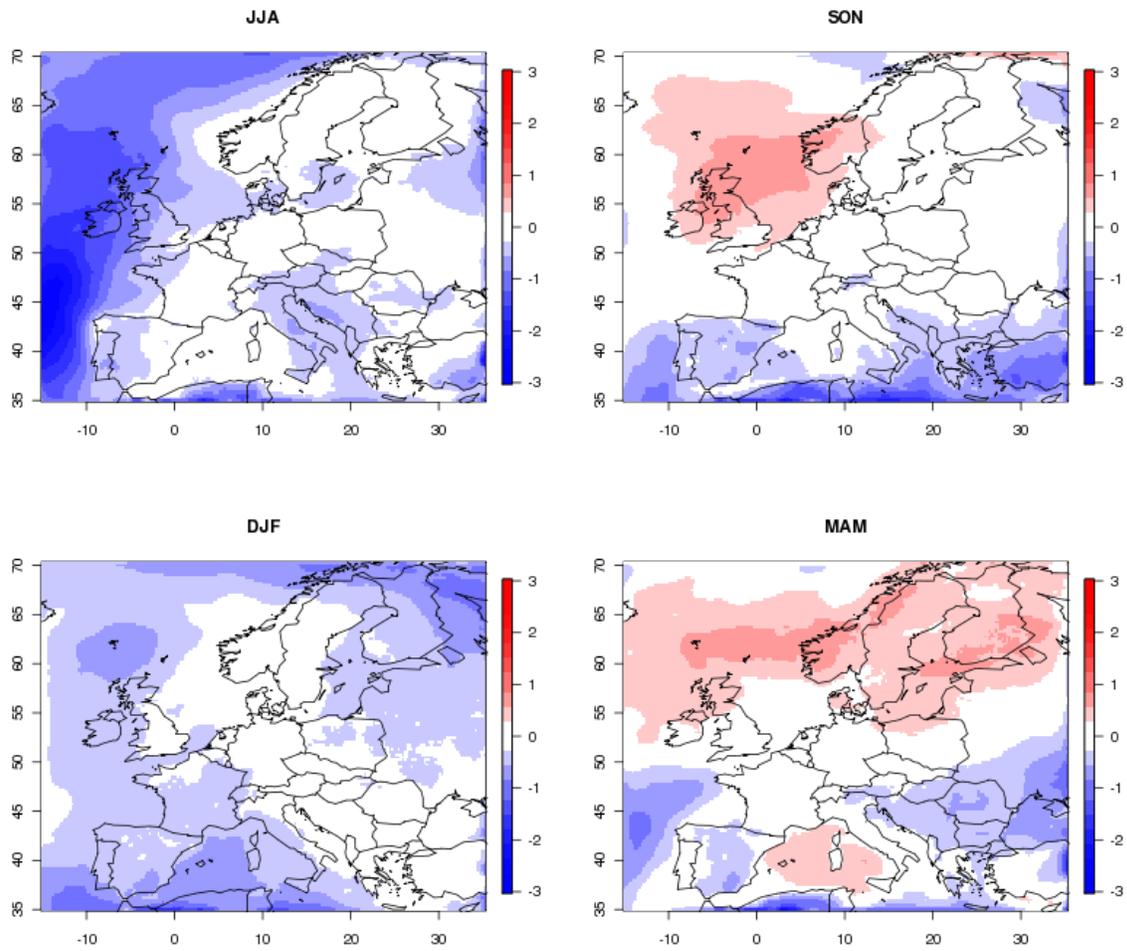


Figure 13.

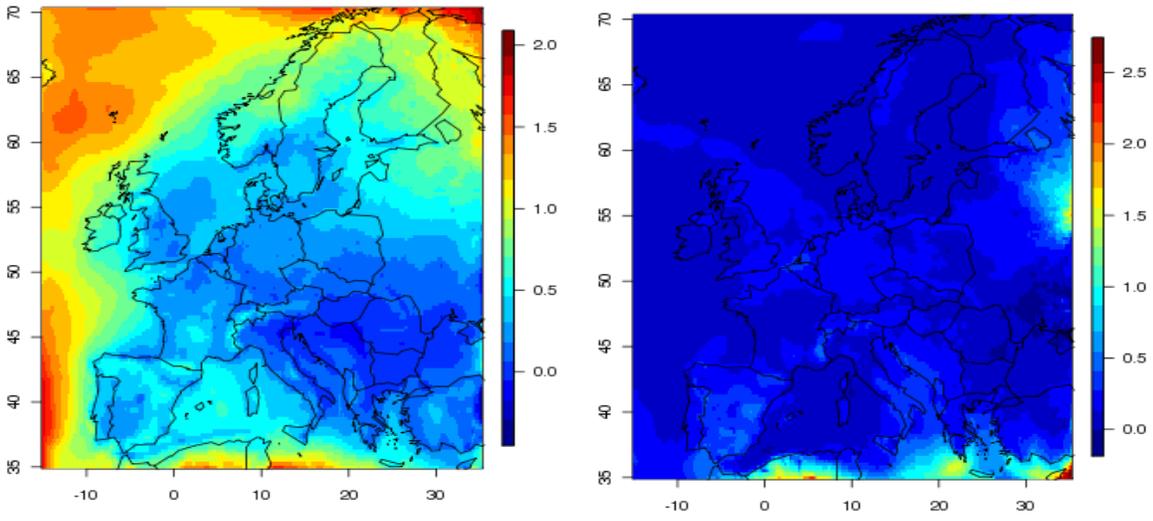


Figure 14.

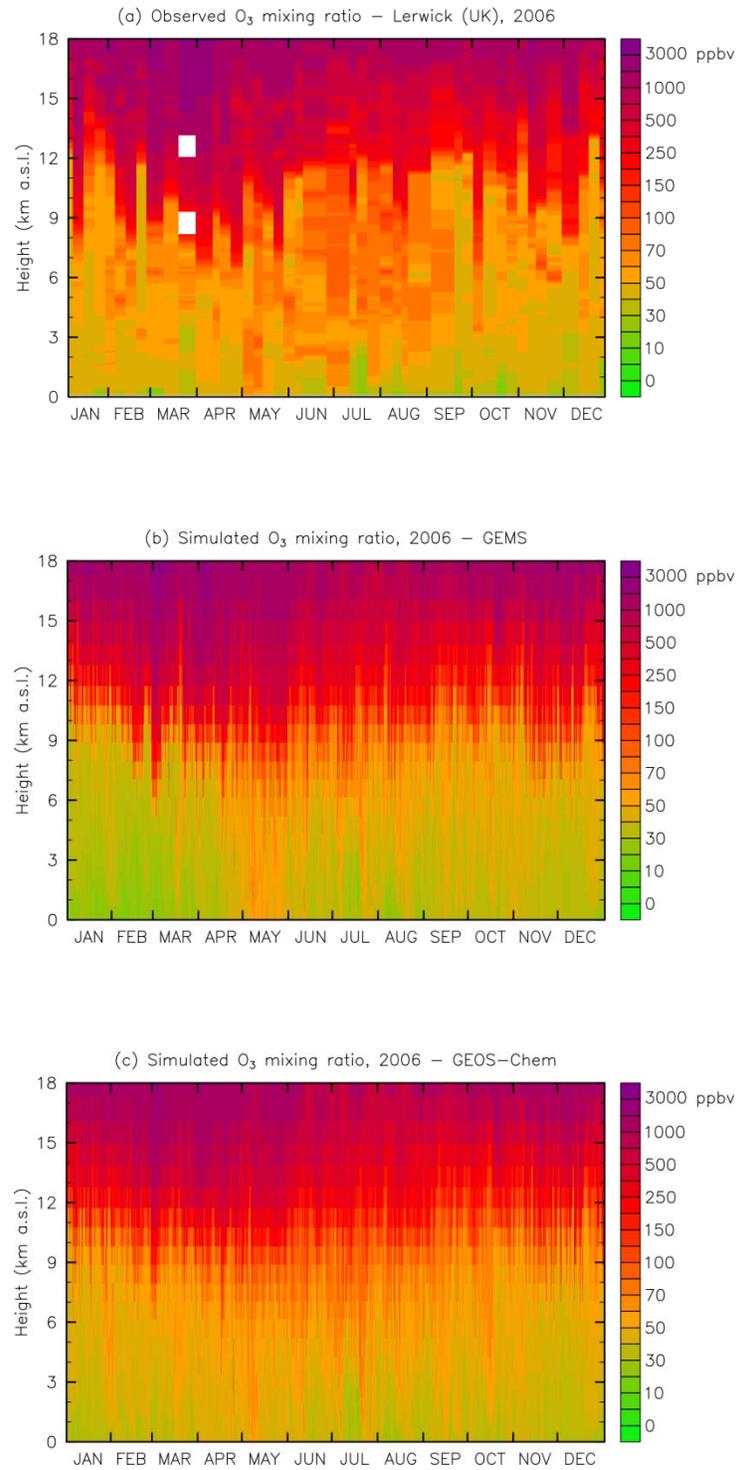


Figure 15.

