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

Kenneth Schere[a*], Johannes Flemming[b], Robert Vautard[c], Charles Chemel[d,e], Augustin Colette[f], Christian Hogrefe[g], Bertrand Bessagnet[f], Frederik Meleux[f], Rohit Mathur[a], Shawn Roselle[a], Rong-Ming Hu[e], Ranjeet S. Sokhi[e], S.T. Rao[a], Stefano Galmarini[h]

[a]Atmospheric Modeling and Analysis Division, National Exposure Research Laboratory, Office of Research and Development, US Environmental Protection Agency, Research Triangle Park, NC 27711, USA
[b]European Centre for Medium-Range Weather Forecasts (ECMWF), Reading, UK
[c]Laboratoire des Sciences du Climat et de l'Environnement, IPSL, CEA/CNRS/UVSQ, Gif/Yvette, FRANCE
[d]National Centre for Atmospheric Science (NCAS), Centre for Atmospheric & Instrumentation Research (CAIR), University of Hertfordshire, Hatfield, UK
[e]Centre for Atmospheric & Instrumentation Research (CAIR), University of Hertfordshire, Hatfield, UK
[f]Institut National de l' Environnement Industriel et des Risques (INERIS), Parc Technologique Alata, Verneuil en Halatte, FRANCE
[g]Bureau of Air Quality Analysis and Research, New York State Department of Environmental Conservation, Albany, NY 12233, USA
[h]European Commission-Joint Research Center, Institute for Environment and Sustainability, I-21027 Ispra, ITALY

[*]Corresponding author: Tel: +01-919-5413795; fax: +01-919-5411379. E-mail: schere.kenneth@epa.gov. U.S. EPA-E243-04, Research Triangle Park, NC 27711 USA

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1   **Abstract**

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3   Over twenty modeling groups are participating in the Air Quality Model Evaluation International

4   Initiative (AQMEII) in which a variety of mesoscale photochemical and aerosol air quality

5   modeling systems are being applied to continental-scale domains in North America and Europe

6   for 2006 full-year simulations for model inter-comparisons and evaluations. To better

7   understand the reasons for differences in model results among these participating groups, each

8   group was asked to use the same source of emissions and boundary concentration data for their

9   simulations. This paper describes the development and application of the boundary

10   concentration data for this AQMEII modeling exercise. The European project known as GEMS

11   (Global and regional Earth-system Monitoring using Satellite and in-situ data) has produced

12   global-scale re-analyses of air quality for several years, including 2006 (http://gems.ecmwf.int).

13   The GEMS trace gas and aerosol data were made available at 3-hourly intervals on a regular

14   latitude/longitude grid of approximately 1.9-degree resolution within 2 "cut-outs" from the

15   global model domain. One cut-out was centered over North America and the other over Europe,

16   covering enough spatial domain for each modeling group to extract the necessary time- and

17   space-varying (horizontal and vertical) concentrations for their mesoscale model boundaries.

18   Examples of the impact of these boundary concentrations on the AQMEII continental

19   simulations are presented to quantify the sensitivity of the simulations to boundary conditions.

20   In addition, some participating groups were not able to use the GEMS data and instead relied

21   upon other sources for their boundary concentration specifications. These are noted, and the

22   contrasting impacts of other data sources for boundary data are presented. How one specifies

23   four-dimensional boundary concentrations for mesoscale air quality simulations can have a

24   profound impact on the model results, and hence, this aspect of data preparation must be

25   performed with considerable care.

26

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

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1. Introduction

The evaluation of regional- through continental-scale photochemical and aerosol air quality simulation modeling systems (PAQMs) 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; 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 are using their regional-scale PAQMs to simulate a full-year (2006) retrospective continental application. Each participating group has been requested to model both continents using common reference model input data sets, namely the gridded source emissions and the 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 model results. 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 as this material is transported into the simulation domains and interacts 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 is
defined by latitude/longitude boundaries:

North America: Latitude: 25.5^{o}N to 58.5^{o}N   Longitude: 130°W to 59.5^{o}W

Europe: Latitude: 35.0^{o}N to 70.0^{o}N   Longitude: 15.0^{o}W to 35°E

(See Figures 1 and 2 in Rao et al., 2011.)

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).

90   The GEMS project was set up by thirty-one participating institutions coordinated by the

91   European Centre for Medium-Range Weather Forecasts (ECMWF) to build and demonstrate the

92   core capability for providing a comprehensive range of services related to the chemical and

93   particulate matter composition of the atmosphere. Among the demonstrated capabilities were

94   data analyses and modeling systems for monitoring global distributions of atmospheric

95   constituents, with focus areas of climate, air quality, and ultraviolet radiation, especially as they

96   affect European communities. Global re-analysis products from GEMS are available for the

97   period 2003-2008. These re-analyses make use of satellite observations allowing the retrieval of

98   O_3, CO, CH_4, CO_2, and aerosol optical depth during the AQMEII period of interest. Although

99   GEMS is now concluded, the work conducted therein is being extended and improved through

100   the new Monitoring Atmospheric Composition and Climate project (MACC; http://www.gmes-

101   atmosphere.eu).

102   Figure 1 illustrates the principal components of the GEMS global modeling system. The

103   GEMS system was built within and around the ECMWF Integrated Forecasting System (IFS;

104   http://www.ecmwf.int/research/ifsdocs/), a global operational weather forecasting model system,

105   including the capability for four-dimensional variational data assimilation (Rabier et al., 2000).

106   The IFS system is coupled with one of three global chemical transport models (CTMs):

107   MOZART3 (Kinnison et al., 2007), MOCAGE (Bousserez et al., 2007), or TM5 (Huijnen et al.,

108   2010) through a special-purpose OASIS4 software coupler (Flemming et al., 2009). The main

109   idea behind the coupled system is that the IFS computes only the transport of the assimilated

110   reactive gases while the tendencies due to chemical conversion, deposition and emission

111   injection are provided by one of the CTMs. The simulation of global aerosol and greenhouse

112   gases is directly included within the IFS model (Morcrette et al., 2009). In this way, the IFS

113   needs to handle only five additional chemical tracers, while the comprehensive schemes of the

114   CTMs contain between 55 and 118 gaseous species. The coupled CTM is driven by

115   meteorological data from the IFS with a coupling frequency of one hour. For the AQMEII

116   application, it is principally the IFS-MOZART3 configuration whose data has been processed for

117   regional-scale model boundary concentrations.

118   The IFS uses a T159 spectrally-resolved global grid with a horizontal grid box size of about

119   125 km. MOZART-3 uses a regular latitude/longitude grid of 1.875^{o}x1.875^{o}. The coupler

120   performs horizontal bi-linear interpolations between the meteorological and CTM horizontal

121   grids. The vertical coordinate is given by 60 hybrid-sigma pressure levels, with a model top at

122   0.1hPa. The same vertical coordinate is used by the IFS and all CTMs in the GEMS system to

123   avoid the need for vertical interpolations. The coupling interval is 3600 s which is the largest

124   acceptable time step for the IFS at a T159 resolution. Output is saved at 3-h intervals from the

125   model simulations. Source emissions for the MOZART-3 global simulations are specified as

126   monthly averages for a base year of 2000 for anthropogenic trace gases (RETRO database;

127   Schultz et al., 2009) and aerosols (EDGAR, http://www.pbl.nl/en/themasites/edgar/ index.html;

128   SPEW, Bond et al., 2004). Eight-day average fire emissions for the 2006 model application year

129   are derived from the Global Fire Emissions Database (GFEDv2; van der Werf et al., 2006).

130   Biogenic, sea salt, and dust emissions are parameterized within the model based on

131   meteorological inputs (GEMS, 2010).

132   The advantage of using the GEMS re-analysis data to provide boundary concentrations for

133   AQMEII simulations compared to other global model outputs is that the GEMS results include

134   the assimilation of observed data derived from satellite platforms. Figure 2 indicates the satellite

135   data usage during the GEMS project, with the AQMEII target year of 2006 highlighted. There

136   were multiple instruments available for measuring portions of the ozone (O_3) column during

137   2006, including from SCIAMACHY, SBUV-2, and MLS instruments. Taken together these data

138   provide some vertical resolution to the O_3 column, with greatest fidelity in the stratosphere and

139   upper troposphere (Flemming et al., 2011). In addition, CO columns are available from the

140   MOPITT instrument, and aerosol optical depths (AODs) are derived from the MODIS

141   instruments onboard the Terra and Aqua satellites. Information on atmospheric aerosols can be

142   derived from the AOD retrievals. Complete details on the data assimilation system and

143   implementation for GEMS can be found in Benedetti et al., 2009 and Inness et al., 2009.

144   The GEMS re-analysis outputs for 2006 were further processed by ECMWF for AQMEII use

145   by interpolating all requested data for selected variables at 3-h intervals on a regular

146   latitude/longitude ($1.875^{o} X 1.875^{o}$) grid within specific geographical "cut-outs" from the global

147   model domain. These cut-outs for Europe and for North America are illustrated in Figure 3. The

148   spatial extent of each cut-out extends well beyond the minimum domain sizes specified for

149   AQMEII regional/continental domains such that AQMEII participants could use the data within

150   the global cut-outs to derive the boundary concentrations for their own model exercises. Data

151   from the lowest 47 IFS model layers (surface through 10hPa) were extracted within each cut-out

152 over a full time period of 1 December 2005 through 31 December 2006, allowing for sufficient

153 model spin-up time for the 2006 simulation. Table 1 lists the chemical and aerosol species

154 extracted for AQMEII use. These data were archived and made available to AQMEII

155 participants by collaborators in Météo-France.

156   Air quality modelers participating in AQMEII then are able to access these GEMS data and

157 use them for specification of boundary concentrations. There are, however, additional

158 assumptions and processing steps involved before the data can be used by the regional models.

159 The GEMS data must be spatially interpolated for the boundaries of each regional model's native

160 grid and temporally interpolated from the 3-h output interval to the 1-h boundary updates

161 typically employed by the regional models. Also, the GEMS data contain fairly coarse chemical

162 speciation of the gaseous organic compounds. Additional disaggregation of these organic

163 compounds into the specific organic classes used by the tropospheric atmospheric chemistry

164 mechanisms is usually necessary. In addition, the GEMS aerosol data for sea salt and dust may

165 need to be redistributed based on the size distribution information carried within the regional

166 model. Finally, the GEMS data provide information for most of the chemical and aerosol

167 species needed at the models' boundaries that have significant transport influence. However, all

168 species are not included in the GEMS data. Modelers may need to provide another source of

169 boundary concentration data for aerosol sulfate and nitrate, for example, and additional gas-

170 phase species that may be in their model, unless the boundaries are assumed to be zero, zero-

171 gradient, or some fixed concentration.

172   There are a few caveats to note with regards to the use of the GEMS data for AQMEII. First,

173 the SO_2 concentrations were calculated within IFS as a tracer using simple assumptions of

174 emissions and prescribed loss. No chemical transformations were considered.

175 Recommendations were made to AQMEII modelers to use the SO_2 data with caution. Sea salt

176 estimates were made as a function of wind speed and other environmental parameters in the IFS

177 model. Based on evaluation of the GEMS sea salt data for 2003, quite significant

178 overpredictions of sea salt aerosol (over 400%) were observed over North America (GEMS,

179 2010). Preliminary analysis showed this to be true for 2006 as well, so AQMEII modelers were

180 cautioned on the GEMS sea salt values. Estimates for sea salt over Europe, however, did not

181 show these same tendencies for overestimation. In addition, organic carbon emissions from fires

182 in the GEMS data set may have been overestimated in the lower model layers due to the lack of a

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183   plume-rise mechanism in the model and an overestimated persistence of the fires from the 8-day

184   resolution in the data being assimilated into the model. Cautions were therefore extended

185   regarding the organic carbon data from large fires for AQMEII use.

186

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

188   The modeling protocol for the AQMEII 2006 model simulations requested that participants

189   derive their boundary concentrations from the GEMS re-analysis data described above.

190   However, not all participants adhered strictly to this request. In fact, there were a variety of

191   sources that were accessed for the continental-scale model boundary concentrations. Table 2

192   presents examples of the various modeling systems used and the source of boundary

193   concentration data in each instance. These data sources include other hemispheric and global

194   modeling systems, as well as climatological or "background" tropospheric concentrations.

195   Ideally, if all participants had used the same source for the boundary concentration data, as well

196   as a common source for the emissions data, the data analysis and interpretation for the project

197   would be assisted by minimizing confounding effects of different sources of data on model

198   results. Some groups used the requested GEMS data source, as well as alternate sources,

199   providing the data for sensitivity studies on the effect of alternate sources of boundary

200   concentrations on their model simulations.

201

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

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204   With typical wind speeds across North America and Europe, inflow air masses can traverse

205   each continent within as few as 3-5 days. Thus the specification of inflow air quality boundary

206   concentrations has the potential for significant impacts on continental model simulations,

207   especially in areas of weaker internal model forcing from source emissions and atmospheric

208   chemistry, and for chemical compounds having lifetimes of this order of magnitude. Pfister et al.

209   (2011) used aircraft and satellite data during the ARCTAS-CARB field campaign during the

210   summer of 2008 to evaluate the MOZART-4 global chemical transport model's simulation

211   results for its chemical representativeness of chemical inflow to the U.S. west coast. The global

212   model was shown to capture only about half of the observed free tropospheric air pollution

213   variability. Sensitivity simulations with the regional WRF-Chem model, performed as part of

214   the cited work, also showed that the temporal variability in the pollution inflow does clearly

215   impact modeled surface concentrations over California. It was suggested that time- and space-

216   varying chemical boundary conditions from global models provide useful input to regional

217   models, but likely still lead to an underestimate of peak surface concentrations and the variability

218   associated with long-range transport of air pollution.

219   Hogrefe et al. (2011) performed long-term simulations with the Community Multiscale Air

220   Quality (CMAQ) model using two sets of chemical boundary conditions, one derived from time-

221   invariant climatological vertical profiles and the other one from a global chemistry model. The

222   comparison of both simulations revealed that lateral boundary conditions have a significant

223   impact on a regional air quality model's ability to simulate long-term O_3 variability and trends,

224   especially for the middle and lower percentiles of the O_3 distribution. As an illustration, Figure 4

225   shows time series of May – September average daily maximum 1-h O_3 concentrations derived

226   from observations and these two sets of CMAQ simulations for the time period from 1988 to

227   2005. It can be seen that the choice of boundary conditions affects the magnitude of the mean

228   concentrations as well as their inter-annual variability and trends. In this particular example, the

229   CMAQ simulation using the time-invariant boundary conditions shows better agreement with the

230   observations in terms of absolute concentrations and trends while the CMAQ simulation using

231   boundary conditions derived from the global model shows better agreement in terms of inter-

232   annual variability.

233   Li et al. (2002) used a five-year (1993-1997) simulation with the GEOS-Chem CTM and

234   showed that North American pollution enhances surface O_3 in continental Europe by 2–4 ppbv

235   on average in summer and by 5–10 ppbv during transatlantic transport events. Specifying the

236   model continental-scale O_3 boundary concentrations correctly is significant in that the North

237   American influence on surface O_3 in Europe is particularly strong at the thresholds used for the

238   European air quality standards (55–65 ppbv). Simulating the daily variability of O_3 boundary

239   concentrations was also shown to significantly improve both variability and biases of simulated

240   daily O_3 maxima in Europe, in particular for the most frequent non-extreme values (Szopa et al.,

241   2009).

242   Ratigejev et al. (2010) demonstrate that global CTMs have difficulty reproducing synoptic-

243   scale pollution plumes during long-range transport. Numerical diffusion interacting with non-

244   uniform atmospheric flows dissipates the plumes faster than ambient observations suggest. The

245   authors state that novel numerical methods, such as adaptive grids or embedded Lagrangian

246   plumes, may circumvent the problem of accurately sustaining the plume integrity. Makar et al.

247   (2010) evaluated ten different approaches for applying lateral and top climatological boundary

248   concentrations for O_3 using the AURAMS regional CTM. They found that dynamic adjustments

249   to the O_3 profile in response to the model-estimated tropopause height were needed to better

250   match mass consistency between chemical and meteorological models. Their results highlight

251   the importance of evaluating boundary concentrations and mass consistency/correction

252   algorithms with three-dimensional measurements.

253

254   *4.1 CMAQ Model – North America application*

255   The U.S. EPA contributed results to AQMEII from a 2006 North American simulation from

256   their CMAQ model. The model domain included all of the continental U.S. (except Alaska),

257   southern Canada, and northern Mexico. Meteorological data for the CMAQ simulation were

258   derived from a continental model run of the Weather Research and Forecasting (WRF) model

259   using four-dimensional data assimilation. The CMAQ model also made use of the standard

260   protocol data provided by AQMEII for emissions and boundary concentrations (GEMS dataset).

261   To assess the adequacy of the GEMS data for providing inflow O_3 boundary concentrations, we

262   have examined the performance of the CMAQ model using observed data from the INTEX-B

263   Ozonesonde Network Study (IONS) of 2006 (Tarasick et al., 2010) for sounding locations near

264   the west (inflow) coast of North America. The IONS 2006 study provided a total of 740

265   ozonesonde profiles from 23 sites across North America. Figure 5 illustrates the locations of

266   these sites within the CMAQ modeling domain, with the shaded area indicating the region of

267   interest for examining the boundary concentration impacts.

268   Figures 6a and 6b present the results of the CMAQ simulation and the observed O_3 vertical

269   profile from the Trinidad Head site on the northern California coast averaged over all March

270   (n=6) and August (n=30) profiles, respectively. For both months, the model and observed

271   profiles agree fairly well at altitudes corresponding to the upper troposphere and stratosphere.

272   Here, the model is greatly influenced by the boundary concentrations as there are no local

273   emissions and little atmospheric chemistry to influence the estimated concentrations. However,

274   in the lower and mid troposphere, the model significantly underestimates the O_3 concentrations

275   compared to the observed profile. Also indicated on the figures are the concentrations from

276   CMAQ attributable to a boundary concentration tracer for O_3. In these calculations, the impact

277   of the spatially and temporally varying O_3 boundary conditions was tracked using a tracer

278   species that underwent advective, turbulent, and cloud transport, and wet and dry deposition

279   similar to O_3. Since the tracer was not subject to any chemical loss, its inferred impact on the net

280   simulated O_3 may be slightly overestimated. Nevertheless, the tracer provides a direct estimate

281   of the impact of the GEMS boundary concentrations on simulated O_3 distributions and trends.

282   With little local photochemistry occurring in March, it is evident that the lower portion of the

283   profile is completely controlled by the boundary concentrations, while in August with more

284   active photochemistry the lowest portion of the profile shows enhanced concentrations as

285   compared to the boundary tracer. This is further illustrated in Figure 7 which presents the

286   average fractional contribution of the boundary conditions to the simulated vertical profile at

287   Trinidad Head for the months of March and August 2006. It is evident that above 3-4 km the

288   simulated O_3 variability is largely dictated by the boundary condition specification.

289   Similar analyses for the Kelowna site in British Columbia, Canada are presented in Figure 8.

290   (There were 2 profiles available in March and 26 in August at Kelowna.) Results of these

291   comparisons are comparable to those of the Trinidad Head site, although this site shows larger

292   deviations in the upper portion of the profiles between CMAQ and the observed data. Results of

293   comparisons at other west coast sites (not shown) demonstrate similar behavior, with CMAQ

294   generally underestimating O_3 near the surface and in the lower and mid troposphere, with the

295   greatest discrepancies in winter and the least in summer. From these profiles it appears that the

296   O_3 boundary concentration tracer, as a surrogate for the GEMS data, has considerably

297   underestimated the inflow O_3 to the west coast of North America in the lower troposphere,

298   especially during the winter and spring. It is not surprising that the GEMS data should better

299   reflect the observed O_3 profile in the upper levels as compared to levels closer to the surface.

300   The GEMS re-analyses have made extensive use of data assimilation for O_3 based on satellite

301   retrievals. These derived measurements are most accurate for the stratospheric O_3 burden and

302   become more uncertain in the lower portions of the profile. In the lower levels, the GEMS data

303   are more reflective of the results of the MOZART-3 model simulations which appear to have

304   systematically underestimated tropospheric O_3 in the northeast Pacific region, and generated a

305   low bias in the specified inflow concentrations for North America, possibly due to emissions

306   uncertainties and an overestimation of dry deposition.

307   A distinct advantage of the data assimilation aspect of the GEMS re-analysis is that real-time

308   events, such as large forest fires or dust storms, are seen by satellite sensors and can be

309   incorporated in the model simulation. Figure 9 shows the vertical profiles of primary organic

310   particulate matter (OM) along each boundary of the CMAQ North American domain averaged

311   over the period of 21-30 June 2006 from the GEMS database. One can readily see the strong

312   impact of a large wildfire occurring at the time in the Canadian boreal forest. The impact on

313   surface layer CMAQ estimates of primary organic particulate matter is seen in Figure 10 in

314   which the 01 UTC concentrations are presented for 30 June 2006. The effects of the wildfires

315   north of the CMAQ domain are evident as the boundary concentrations have been advected into

316   the northern portion of CMAQ's computational domain. Satellite measurements confirm the

317   elevated aerosol loadings in this area from the wildfires.

318

319   *4.2 CHIMERE Model – North America application*

320   The sensitivity of concentrations simulated by a regional model to chemical boundary

321   conditions was tested by IPSL-France using the CHIMERE model (Bessagnet et al., 2004), by

322   using in separate simulations the GEMS boundary conditions provided to AQMEII (Simulation

323   A) and the boundary conditions typically used in CHIMERE studies, as provided by monthly

324   climatologies of the LMDzINCA global model (Hauglustaine et al., 2004) for gas-phase species

325   and the GOCART model for aerosol species (Ginoux et al., 2001). For this simulation (B),

326   carried out in exactly the same setting as for Simulation A for other model parameters, boundary

327   conditions are constant within each month but vary along model boundaries. It must be noted

328   that CHIMERE only simulates concentrations within the lower atmosphere: it has a top boundary

329   at 500 hPa. Concentrations within the modeling domain are thus sensitive to both lateral and top

330   boundary concentrations.

331   The mean O_3 surface concentration differences between Simulations B and A have been

332   calculated for each season (Winter=DJF, Spring=MAM, Summer=JJA, Fall=SON) and are

333   represented in Figure 11. The sensitivity to O_3 boundary concentrations differs from one season

334   to another. In winter and spring, strong winds and vertical mixing induce a larger sensitivity to

335   boundary concentrations than in summer and fall. For instance, in winter, seasonal mean

336   concentration differences between the two simulations in the center of the domain and those near

337   the boundaries vary by a factor of two or so. In contrast, in summer and fall the concentration

338   differences vary by more than a factor of 5 between the center of the domain and the regions near

339   the boundaries, indicating that boundary conditions have a relatively smaller impact on the inner

340   portions of the domain compared to winter and spring. However, in all seasons studied, the

341   impact of boundary conditions extends inland far from the boundaries. The Central-East U.S.

342   shows the smallest influence from the boundary conditions.

343   Note also that the difference between mean LMDzINCA-driven and MOZART-driven

344   simulations remains positive across the domain, because the LMDzINCA O_3 boundary

345   conditions are higher than the MOZART ones. This difference is largest in the winter season,

346   and reaches about 15 ppb, while in summer it reaches 10 ppb. The magnitude of these

347   differences in seasonal mean concentrations caused by different boundary conditions is

348   comparable to those shown in Hogrefe et al. (2011; see figure 11 therein).

349

350   *4.3 CHIMERE Model – Europe application*

351   The CHIMERE model has also been applied over Europe using the TNO emissions inventory

352   at a 0.25° horizontal resolution by INERIS-France. As in the North American case, the model

353   domain extends vertically to 500hPa. To assess the impact of the temporal resolution of the

354   boundary concentrations (BCs) on air quality modeled in the regional domain, the whole year

355   2006 has been run twice with 3-hourly GEMS BCs for both gaseous species and aerosols (3HR),

356   and with a monthly climatology derived from the same dataset (gas and aerosols as well; CST).

357   In the CST run, for a given time in a month, the model is driven with constant BCs; no

358   interpolation is performed between two consecutive months.

359   In Figure 12 we display the average difference between the simulations with 3-hourly BCs and

360   the monthly climatology for O_3. With both sets of BCs derived from the same global

361   simulations, these plots would exhibit a null geographical variability if the regional model

362   behaved as a linear operator. Since that is obviously not the case, these plots reflect the

363   combined impact of (1) the non-linearity of the regional model together with (2) the skewness of

364   the O_3 distribution at the boundaries. For instance, the difference is consistently negative at the

365   southern boundary showing that the mean BCs (used in the CST run) lead to an overestimation

366   of O_3 compared to the 3HR BCs. On the western and northern parts of the domain, the situation

367   is less straightforward. In summer (JJA), climatological BCs lead to an overestimation of O_3, but

368   in spring and fall they yield an underestimation compared to time-varying fields. It is likely that

13

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_3, 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_3 and PM10 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 been removed in the GEMS dataset there are no specific patterns observed in the western and northern parts of the domain. Table 3 shows the global standard deviation of daily mean concentrations of O_3, NO_2, and PM10 for all European air quality monitoring stations taken from the AIRBASE dataset (all available stations). For short-lived species like NO_2, the time-varying BCs have a negligible impact. However, for both O_3 and PM10, using the 3HR fields at the boundaries contributes to obtaining a slightly larger variability that is more in agreement with the observations for O_3 and NO_2. The time variability is impaired for PM10 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 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 MOZART-4 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

14

400   better captured with the higher temporal resolution. One difference that was noted was in the

401   mean O_3 concentrations at the boundaries, where Pfister et al. (2011) reported the same mean O_3

402   values irrespective of the temporal averaging at the boundaries, while the current study noted

403   differences in the means based on the temporal averaging. This discrepancy in the findings is

404   likely due to the longer simulation period used here (one year) compared to the 17-day

405   simulation period in the Pfister study. The longer simulation allowed for additional anomalous

406   events, such as stratospheric intrusions of O_3 into the lower troposphere, to affect the average in

407   non-linear fashion.

408

409   *4.4 CMAQ Model – Europe application*

410   The CMAQ model was applied over the European domain for the year 2006 using the input

411   datasets prescribed for AQMEII (including the GEMS boundary concentrations) by the

412   University of Hertfordshire-UK. An evaluation of the CMAQ calculations, for the continental-

413   scale domains in North America and Europe, is given in Appel et al. (2011; this issue). To

414   examine the impacts of the boundary concentrations on the model results, an additional

415   simulation was performed for 2006 using boundary concentrations provided by the global

416   chemical transport model GEOS-Chem, version 8-03-01 (see http://wiki.seas.harvard.edu/geos-

417   chem/index.php/Main_Page). The GEOS-Chem model was run at a 2º x 2.5º horizontal

418   resolution with 47 hybrid pressure-sigma vertical levels. The model was driven by assimilated

419   meteorological data from the Goddard Earth Observing System (GEOS-5) at the NASA Global

420   Modeling and Assimilation Office (GMAO). We used the chemistry mechanism NO_x-O_x-

421   hydrocarbon-aerosol to simulate O_3 and aerosols (Jacob, 2000; Bey et al., 2001). The aerosol

422   components included sulfate, nitrate, ammonium, black carbon, organics, mineral dust, and sea

423   salt (see Park et al., 2004; Hu et al., 2007). The emission inventories were separated into four

424   source categories: anthropogenic, biomass, biofuel, and biogenic. Sources of mineral dust and

425   sea salt are dealt with separately. The anthropogenic emissions were obtained from the Global

426   Emissions Inventory Activity (GEIA) dataset. Biomass burning and biofuel-use emissions were

427   derived from Duncan et al. (2007). Biogenic emissions included isoprene, methyl butenol,

428   acetone, and alkene.

429   As in Section 4.1, we focus on the adequacy of the boundary concentrations data for setting

430   the inflow of O_3 into the modeling domain. Figure 14 presents the observed and modeled

15

431   vertical distributions of O_3 at Lerwick, Shetland Mainland, UK, for the year 2006. The Lerwick

432   Observatory is situated in a remote location representative of background (inflow) atmospheric

433   concentrations. The time-height evolution of O_3 in Figure 14a was compiled from measurements

434   by a UK Met Office ozonesonde of the Electrochemical Concentration Cell (ECC) type (Komhyr

435   et al., 1995). The two model calculations agree fairly well with the observations at altitudes

436   above 6-8 km (i.e., in the upper troposphere/lower stratosphere). While the GEMS re-analysis

437   included the assimilation of O_3 data from satellite observations, the GEOS-Chem simulation

438   included stratospheric O_3 chemistry based on a climatological representation of species sources

439   and sinks. Both techniques appeared to work well in reproducing the O_3 profile at the higher

440   altitudes. Interestingly, the agreement degrades in the lower troposphere when using the GEMS

441   boundary concentrations, while it remains fair when using those provided by GEOS-Chem. The

442   difference between the observed O_3 concentrations and those of the simulation using the GEMS

443   boundary concentrations is most dramatic for the first four months of 2006, with low biases as

444   large as 20 ppb.

445   Similar findings can be observed in Figure 15, where the observed and modeled ground-level

446   O_3 concentrations at Mace Head, Republic of Ireland, are shown. The location of the monitoring

447   station on the Atlantic Coast makes it a representative site for background concentrations of

448   substances in the atmosphere. The O_3 concentration at the site, simulated using the GEMS

449   boundary concentrations, appears to be biased low for the first four months of 2006. Afterwards,

450   the two model calculations give comparable results. This highlights the importance of boundary

451   concentrations in setting the baseline concentrations in the modeling domain.

452

453   **5.0 Summary and Conclusions**

454

455   The AQMEII project on regional-scale air quality model system evaluation and inter-

456   comparison has proposed the use of a common set of boundary concentrations to be specified to

457   the regional modelers for all to use in an effort to minimize differences across the models from

458   this particular aspect of the modeling protocol. The GEMS re-analysis air quality dataset has

459   been provided for this purpose from the ECMWF for the AQMEII focus year of 2006. The re-

460   analysis is produced by assimilating satellite observations of select chemical and aerosol species

461   into a coupled model system, which consists of ECMWF's IFS and the MOZART-3 CTM. The

462   resulting dataset provides spatially- and temporally-resolved information on concentrations of

463   most of the key transported species of interest to the AQMEII regional modelers for use in their

464   modeling systems. Each regional modeling group then must cast these data into forms directly

465   usable by their particular model. This processing may include further spatial and temporal

466   interpolations as well as chemical speciation of the trace gases and aerosols for the particular

467   chemical and aerosol mechanisms used by the model.

468   In this study we found that specification of O_3 profiles from the GEMS dataset at the

469   boundaries of the North American and European modeling domains for the 2006 simulations

470   offered good agreement in the upper troposphere and lower stratosphere with an independent set

471   of observations from ozonesondes. For the North American domain, the O_3 boundary

472   concentrations throughout 2006 were underestimated in the GEMS dataset in the lower to mid

473   troposphere, with greater biases in winter and spring and lower biases in summer. In the

474   European simulations the GEMS dataset yielded O_3 boundary concentrations that were

475   consistent with ozonesonde observations except for the first quarter of 2006 when the lower

476   tropospheric O_3 values were biased low by as much as 20 ppb. SO_2 concentrations in the GEMS

477   dataset were derived from simple assumptions and not based on a complete chemical description

478   in the global modeling. AQMEII modelers were cautioned regarding their use for continental

479   boundary concentrations. For particulate matter we found that organic carbon from large fires

480   was well detected by the GEMS data assimilation system, but that concentrations could be

481   overestimated near the surface due to lack of a plume rise mechanism and eight-day temporal

482   averaging of fire emissions. Sea salt was greatly overestimated near the boundaries of the North

483   American domain, although the bias was less over the North Atlantic near the borders of the

484   European domain. The GEMS dataset did not provide estimates of particulate sulfate or nitrate.

485   Specification of boundary concentrations is a required element in modeling with limited-area

486   air quality models, such as regional- to continental-scale CTMs. The limited area models are

487   typically quite sensitive to these specified concentrations, especially in areas of limited internal

488   forcing by emissions and chemistry within the model's computational domain. Sensitivity

489   simulations performed with the CHIMERE model emphasize that the impact of O_3 boundary

490   concentrations can extend far into the model domain beyond the boundaries. These studies also

491   show that boundary concentrations derived from monthly O_3 climatologies can deviate

492   substantially from more temporally-resolved concentrations. The tropospheric impacts of

17

493 stratospheric O_3 intrusion events, for example, can be greatly damped or eliminated by monthly

494 averages.

495   Long-duration simulations, such as the full-year simulations performed within the AQMEII

496 project, require boundary concentrations that reflect not only day-to-day variations but also

497 seasonal and inter-annual changes in the global environment. Use of global CTMs to provide

498 these boundary concentrations is a logical and convenient mechanism for their specification. It

499 should be noted, however, as seen in these AQMEII model results, as well as the results

500 presented in Hogrefe et al. (2011), that the global models may contain errors or biases in their

501 simulated results that can then propagate into the regional models through the boundaries and

502 affect the results within the model domain. The use of data assimilation in the global models can

503 help minimize these errors, but cannot eradicate them. The assimilated satellite observations

504 provide mainly vertically integrated column values, which makes it more difficult for the

505 assimilating model to obtain realistic concentration profiles close to the surface. Examining

506 several sources of boundary concentrations, such as alternate global CTMs, may provide useful

507 information to modelers on ranges of boundary concentrations to consider. With the tightening

508 of air quality standards and the imposition of emissions control programs, air pollution levels

509 have generally been declining in many nations, leading to the need to better quantify background

510 pollution as an "irreducible" portion of the local pollutant burden. Therefore the process of

511 specifying boundary concentrations for limited area models is an important issue and must be

512 performed with careful scrutiny to assure the best possible outcome from regional-scale model

513 simulations.

514

534 **References**

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673

674   Table 1. Gas-phase reactive chemical and aerosol species extracted from GEMS data
675

Gas-phase Reactive Chemical Species[1]		
O_3 (ozone)	HNO_3 (nitric acid)	C2H6 (ethane)
CO (carbon monoxide)	HO_2NO_2 (peroxynitric acid)	ISOP (isoprene)
CH_2O (formaldehyde)	PAN (peroxy acetyl nitrate)	TOLUENE (sum of C7,C8,C9 aromatics)
NO (nitrogen oxide)	CH_4 (methane)	BIGENE (>C3 alkenes)
NO_2 (nitrogen dioxide)	CH_3CHO (acetaldehyde)	BIGALK (>C3 alkanes)
Aerosol Species[2]		
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_2 (sulfur dioxide-gas)[3]
Other Variables		
T (temperature)[4]	PS (surface pressure)[5]	

676   [1]volume mixing ratios; units: mole mole^{-1}
677   [2]units: $\mu g\ m^{-3}$; size distribution bins are chosen so that roughly 10, 20 and 70 percent of the total mass of each
678   aerosol type are in the three successive bins
679   [3]SO_2 here is based on simple assumptions of emissions and prescribed loss; no active chemistry
680   [4]units: ^oK; used in conversion between molar and mass mixing ratios for gas-phase species
681   [5]units: Pa; used in conversion of model layer number to atmospheric pressure
682

683   Table 2. Boundary Concentration Data Sources Used by AQMEII Participants
684

AQMEII Participant	Model System	Source of Boundary Concentration Data
ZAMG - Austrian Weather Service - AT	ALADIN/CAMx	CECILIA model
Environment Canada - CA	GEM/AURAMS	Climatological chemical boundary concentrations with dynamic O_3 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

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686

687   Table 3. Standard deviation of daily means at the location of surface air quality monitoring
688   stations for O_3, NO_2 and PM10 in the data and in the two CHIMERE model simulations (3HR
689   and CST).
690
691

	Obs. data	3HR	CST
O_3 (µg/m3)	26.17	21.67	21.26
NO_2 (µg/m3)	16.48	13.11	13.12
PM10 (µg/m3)	22.75	24.11	23.93

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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 conditions while CMAQ/Global refers to the CMAQ simulations utilizing boundary conditions 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 conditions to the simulated mean vertical O_3 distributions during March and August 2006 at Trinidad Head.

Fig. 8. Same as Fig. 5, 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 conditions and that using the GEMS boundary conditions. Concentration differences are in ppb. The horizontal resolution of the simulations is 36 km. Each panel corresponds to a seasonal mean.

740   **Fig 12.** Seasonal mean of the bias in modeled O_3 (ppb) at the surface in the CHIMERE model:
741   difference between the simulation driven by 3-hourly boundary conditions and the monthly
742   climatology, JFD (January, February, December), MAM (March, April, May), JJA (June, July,
743   August), SON (September, October, November).
744
745   **Fig. 13.** Difference in standard deviation of modeled O_3 (left, ppb) and PM10 (right, $\mu g/m^3$) at
746   the surface in the CHIMERE model between the simulation driven by 3-hourly BCs and when a
747   monthly climatology is used at the boundaries.
748
749   **Fig 14.** Vertical distribution of O_3 at Lerwick, Shetland Mainland, UK, for the year 2006 as (a)
750   observed, (b) simulated with CMAQ model using the GEMS boundary concentrations, and (c)
751   simulated with CMAQ model using the GEOS-Chem boundary concentrations.
752
753   **Fig. 15.** Time series of observed (black) and CMAQ-simulated (colors) 1-h O_3 concentrations at
754   Mace Head, Republic of Ireland, for the year 2006 for (a) the simulation using the GEMS
755   boundary concentrations and (b) that using the GEOS-Chem boundary concentrations. Color
756   variations in simulated time series represents different seasons.
757
758

Figure 1

Figure 2

	2003	2004	2005	2006	2007

AIRS radiances (CO2)

SCIAMACHY (CH4)

SCIAMACHY (O3)

SBUV (O3)

MIPAS (O3)

GOME (O3)

MLS (O3)

OMI (O3)

MOPITT (CO)

AQUA MODIS (AOD)

TERRA MODIS (AOD)
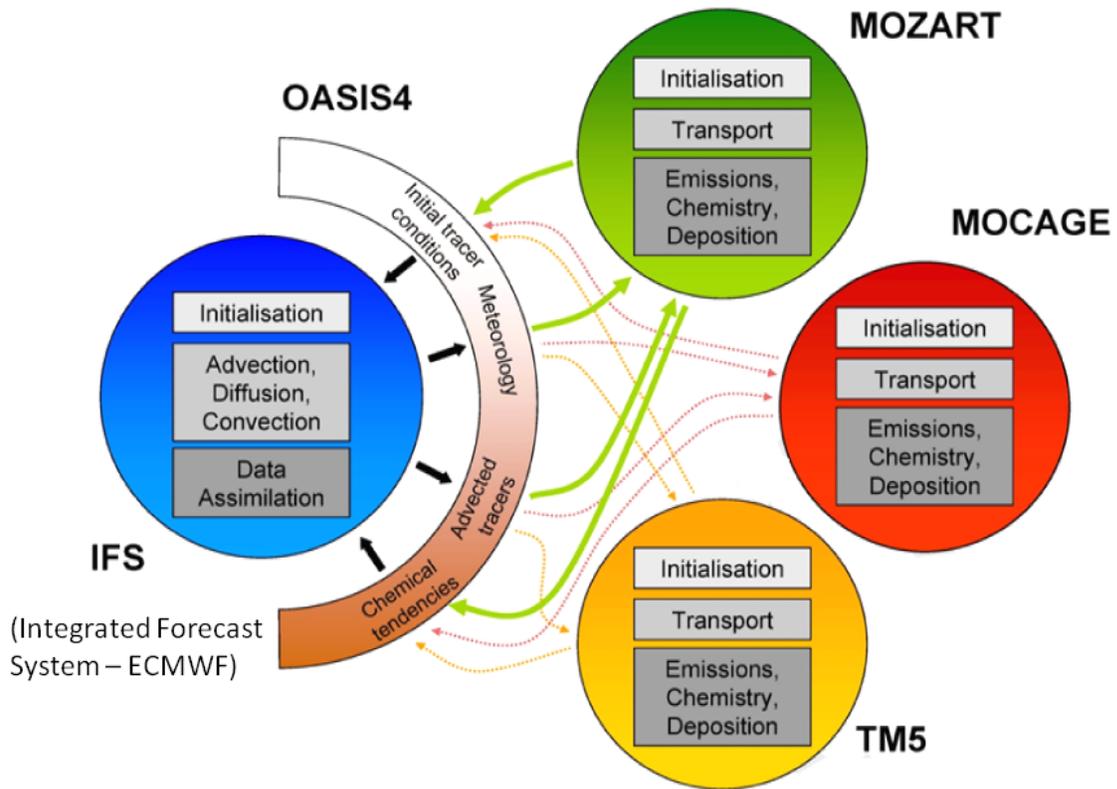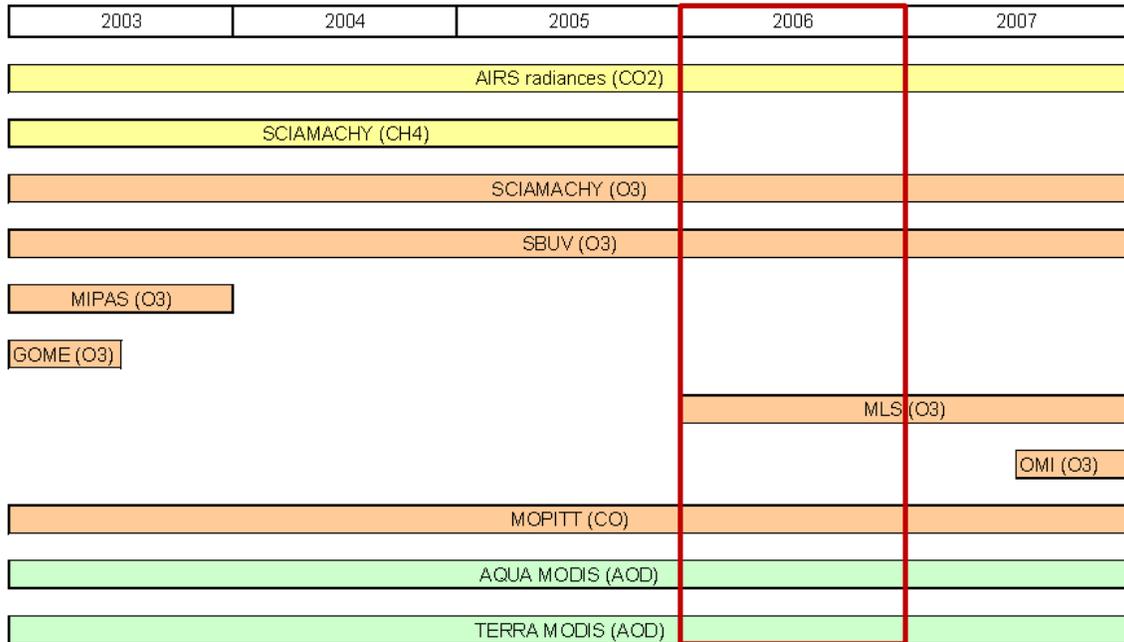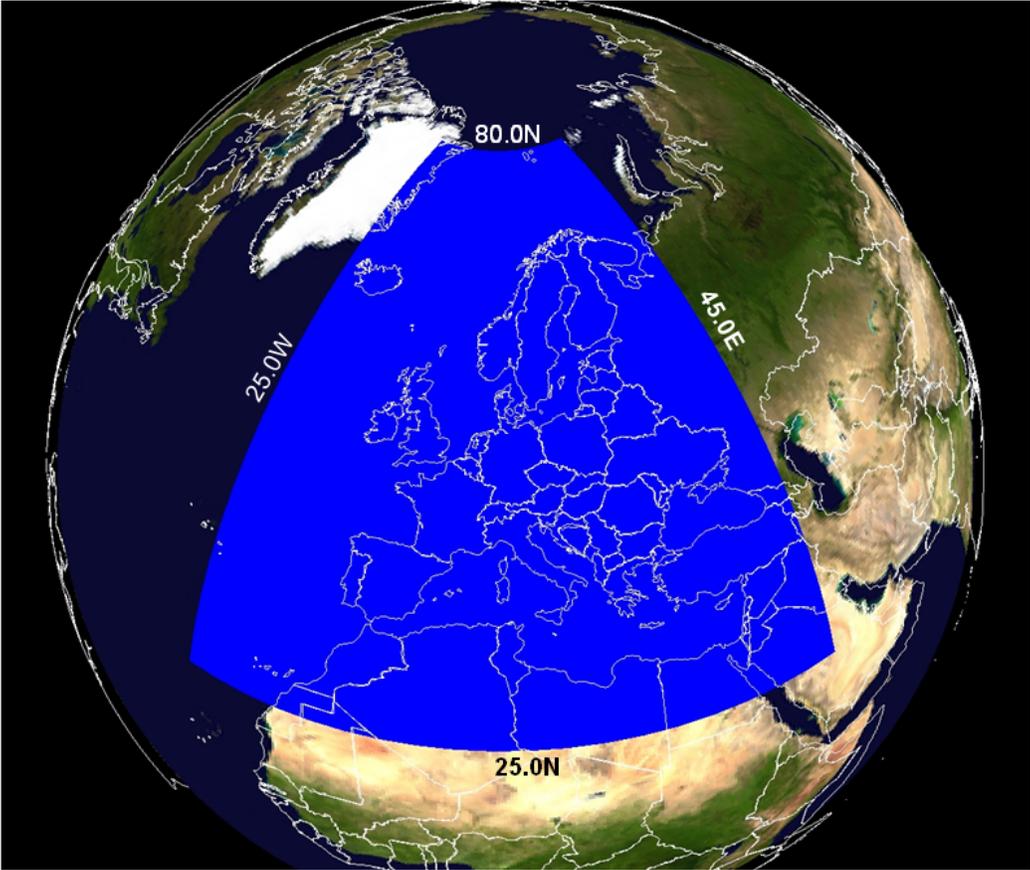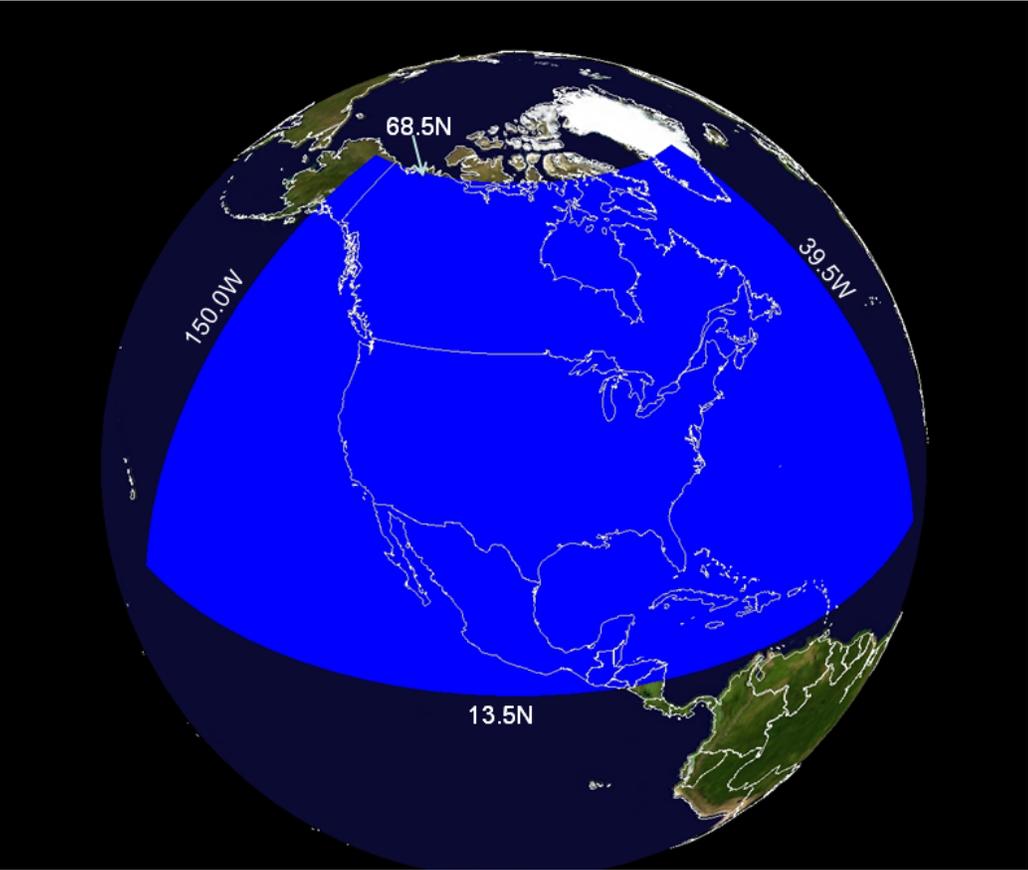
Figure 3 (a)

Figure 3b

Figure 4.
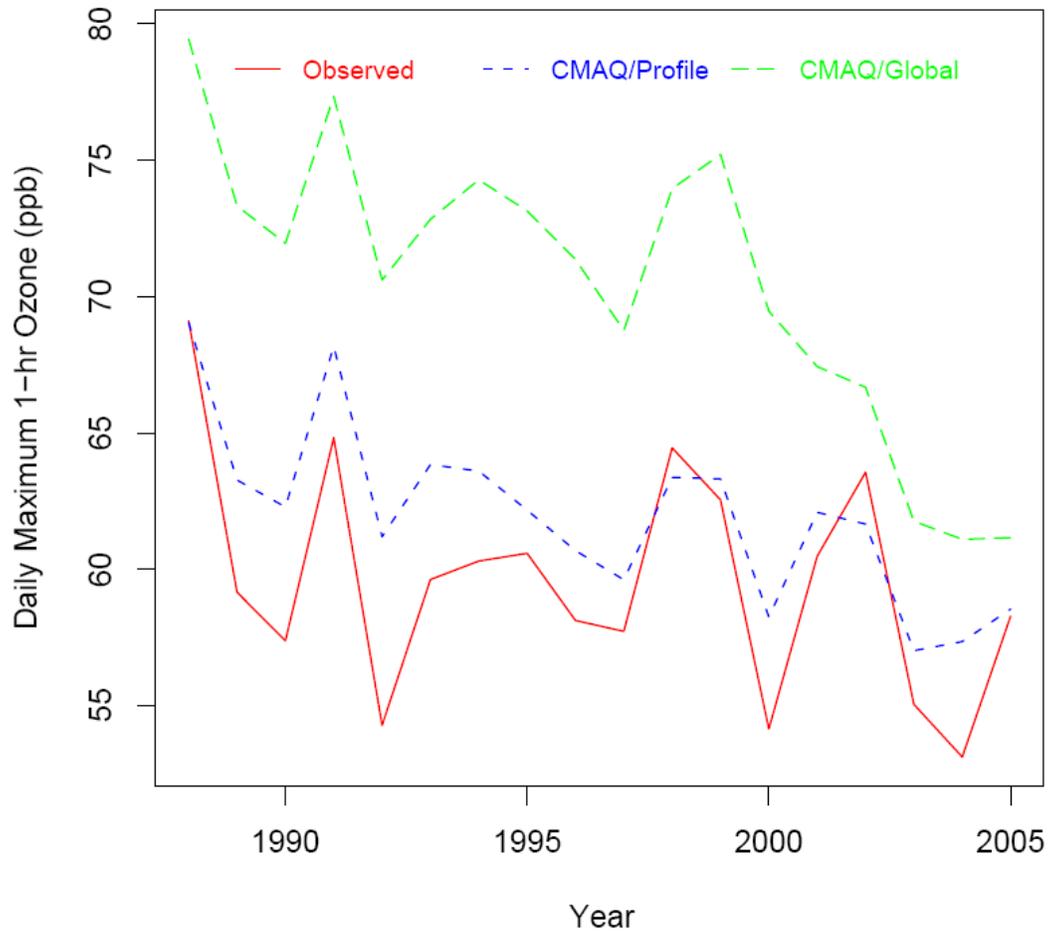
Figure 5

Figure 6a

Figure 6b

trinidad 200608 mean

Figure 7

Figure 8a

kelowna 200603 mean

Figure 8b

kelowna 200608 mean
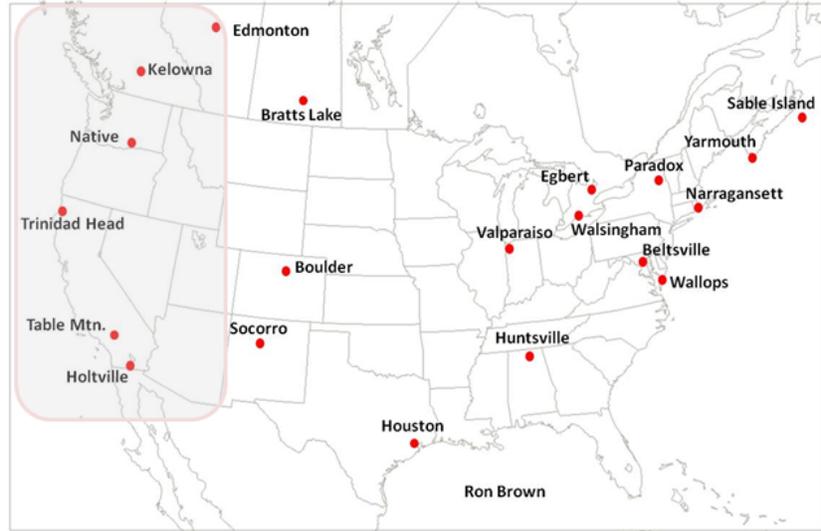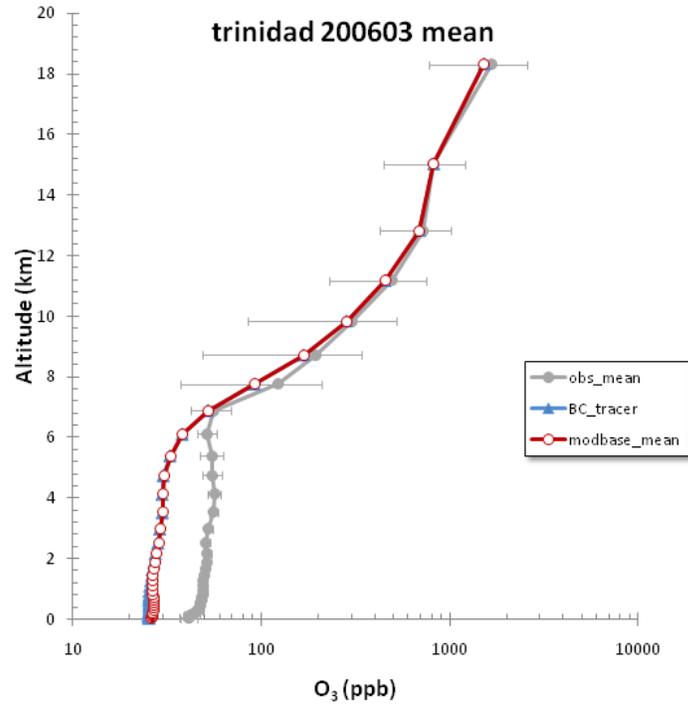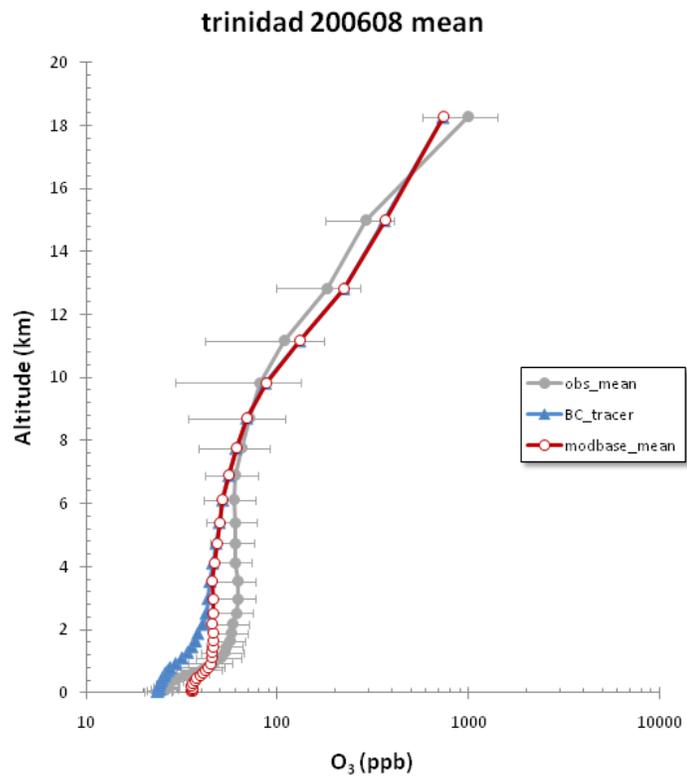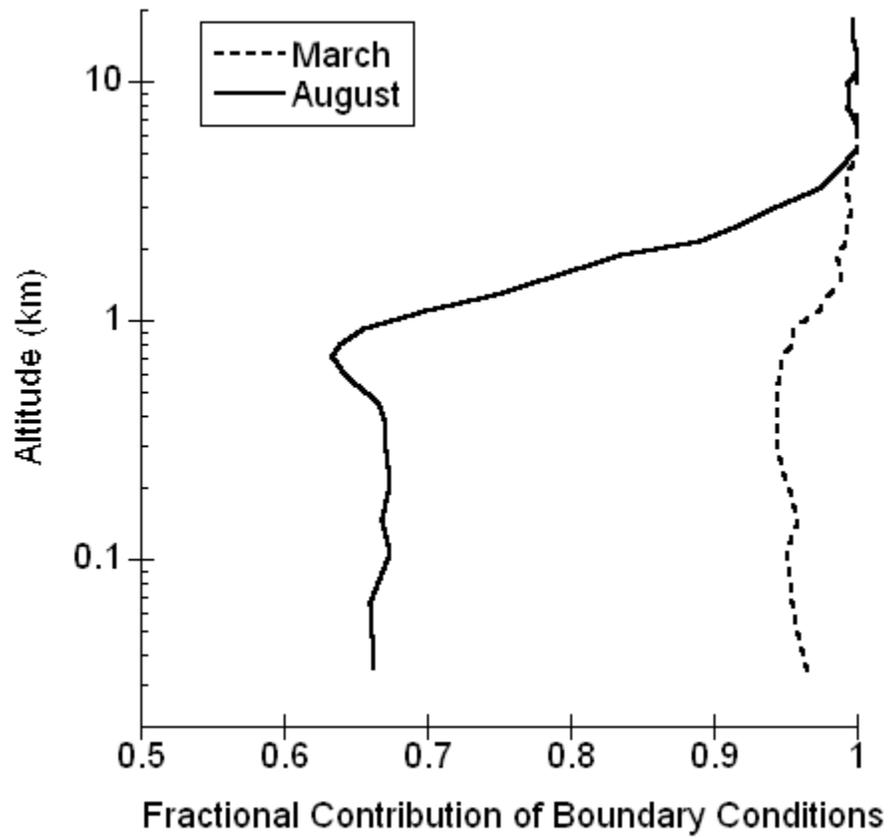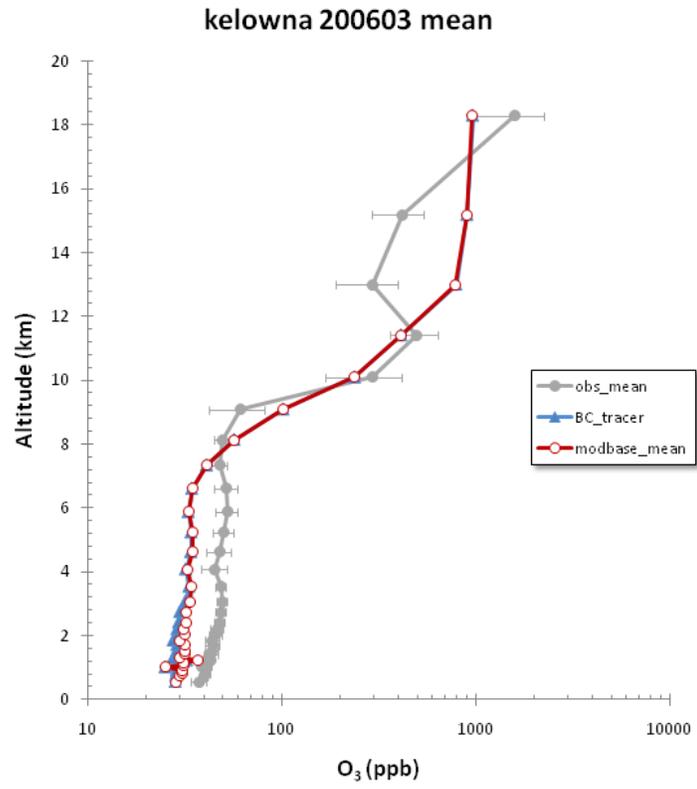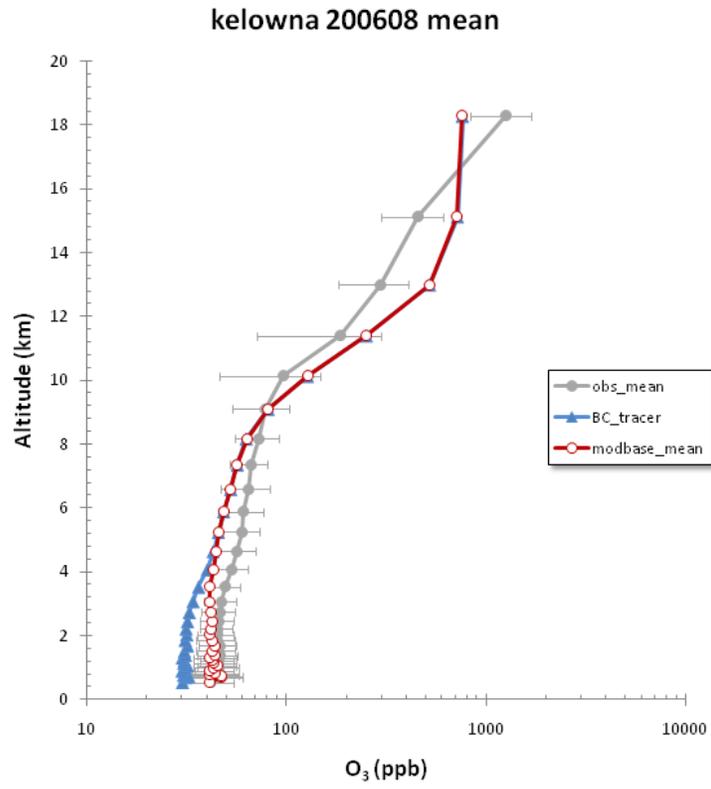
Figure 9

Figure 10

Figure 11

a) Summer Season b) Fall Season c) Winter Season d) Spring Season
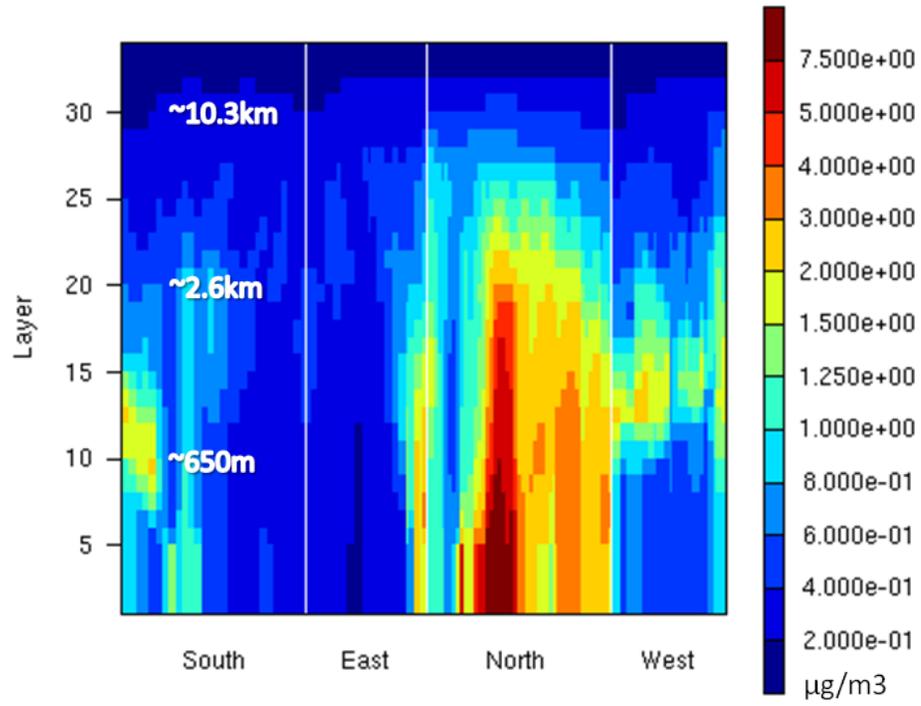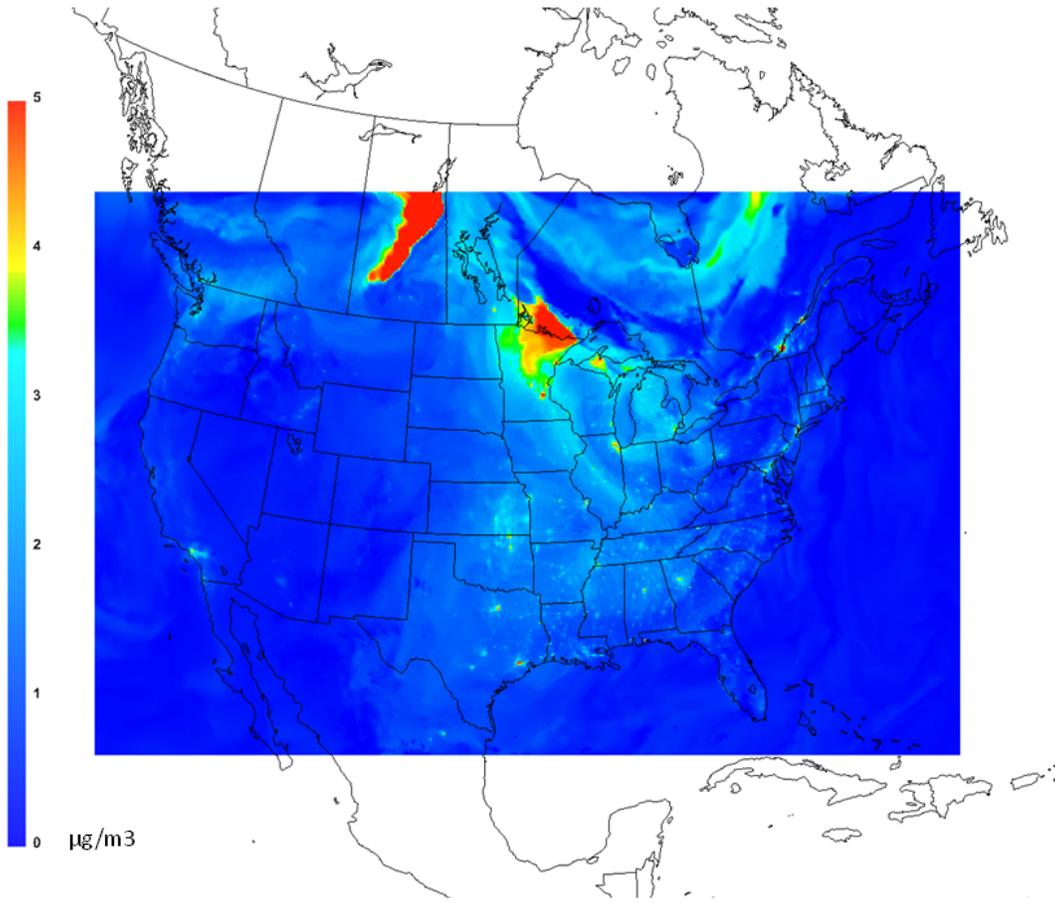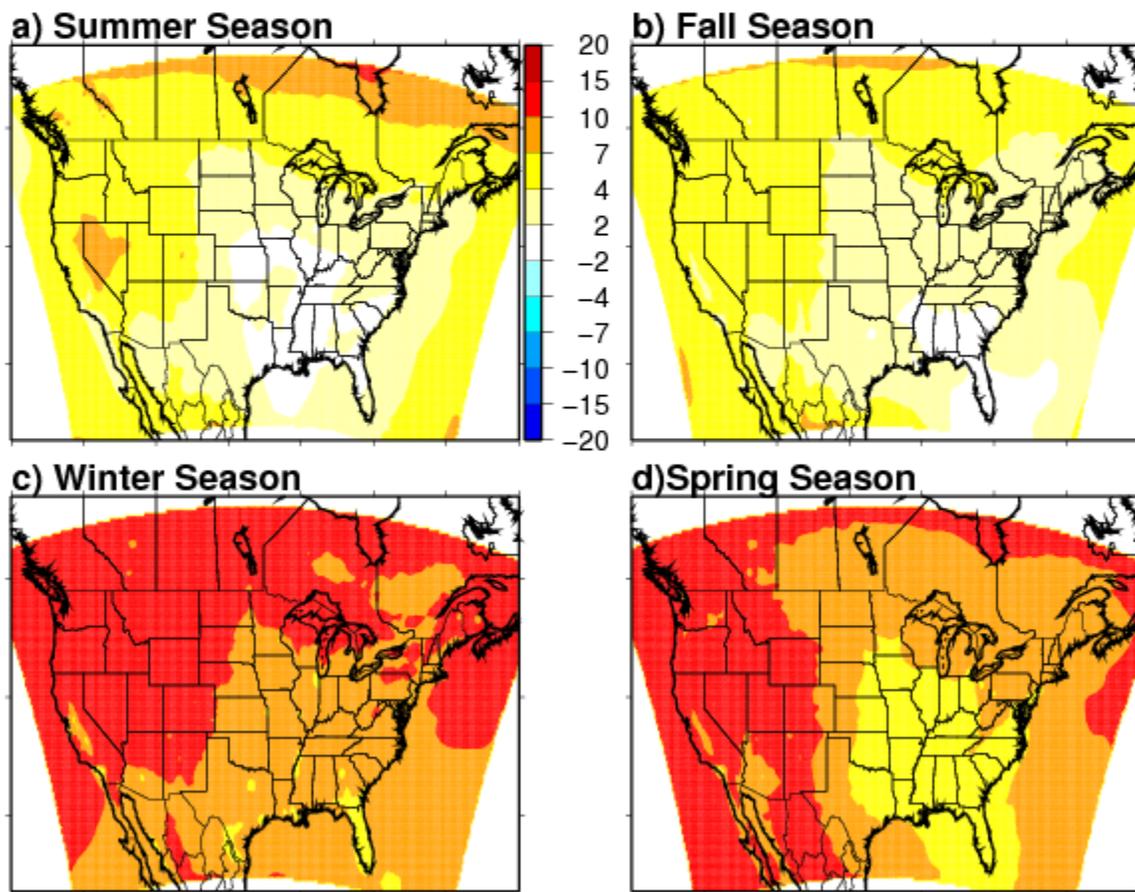
Figure 12.

Figure 13.

Figure 14.

(a) Observed O$_3$ mixing ratio — Lerwick (UK), 2006

(b) Simulated O$_3$ mixing ratio, 2006 — GEMS

(c) Simulated O$_3$ mixing ratio, 2006 — GEOS-Chem

Figure 15.

(a) Mace Head, 2006 — GEMS

(b) Mace Head, 2006 — GEOS-Chem