

# **CMAQ Development for UK National Modelling:**

## **Technical Report on the Influence of Boundary Conditions on Regional Air Quality**

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### **1. Introduction**

The initial and boundary conditions are two important requirements in the regional chemical transport modelling system to simulate regional air quality realistically. In any regional chemical transport model, boundary conditions are required for the entire simulation period, while initial conditions are only necessary at the start of the simulation. Berge et al. (2000) suggested that the effect of initial conditions could be reduced by using adequate spin-up time prior to the actual model simulations in photochemical oxidant modelling. However, Seinfeld and Pandis (1997) reported that upwind boundary conditions significantly impact on the evolution of chemical species throughout the spin-up process.

Recent studies (Heald et al., 2003; Chin et al., 2007; Lin et al., 2008) have revealed that the prerequisite of accurate boundary conditions in regional air quality models has become very important as intercontinental transport of air pollutants strongly influences regional air quality. The methods to generate chemical boundary conditions for regional air quality models varies from the use of a constant background concentrations of selected tracers, through use of idealized vertical profiles, to dynamically varying boundary conditions from global chemical transport models. In the past years, regional chemical transport models have to be dependent on climatological averages as boundary conditions as well as fixed value concentrations for most of the species. The use of invariant boundary conditions in regional chemical transport models does not simulate the temporal and spatial distribution of air pollutants accurately especially during some of the extreme pollution events due to transportation of air pollutants from other regions.

The performance of regional chemical transport models is influenced by the availability of high resolution (temporal and spatial) input data. In recent years, the advancement of computational technologies makes it possible to generate reasonably high resolution (spatially as well as temporally) tracer species using global chemical transport models. Previous studies suggested that using dynamic tracer species concentrations from global chemical transport models as boundary conditions improves the prediction of air pollutants in the regional chemical transport models. Tang et al. (2007) studied the sensitivity of regional air quality models to various lateral and top boundary conditions from three different global models and found that simulations are sensitive to the boundary conditions especially for relatively long-lived species such as carbon monoxide and ozone. Schere et al. (2012) compared the sensitivity of European regional air quality to boundary conditions from GEOS-Chem and GEMS global models. They found that both model simulations worked well in reproducing the ozone profile at higher altitudes. However, in the lower troposphere simulation with GEMS boundary conditions showed degradation in the performance of regional models as compared to the simulation with GEOS-Chem boundary conditions.

In this study we evaluate the sensitivity and performance of CMAQ-UK to various boundary conditions. We have employed CMAQ-UK with boundary conditions from four global models: STOCHEM, GEMS, GEOS-Chem and MACC. We will recommend to DEFRA the most suitable boundary conditions for CMAQ-UK model runs. The following section explains the MACC boundary conditions and its mapping onto CMAQ chemical species. The description of other boundary conditions such as STOCHEM, GEMS and GEOS-Chem were explained in the Phase 1 project report "*CMAQ Development for National Modelling: Development of a Provisional CMAQ-UK Configuration*".

## 2. General description of MACC

The MACC/II (Modelling Atmospheric Composition and Climate, [www.gmes-atmosphere.eu](http://www.gmes-atmosphere.eu)) is a research project funded by the European Union under the FP7 programme. The main aim of the project is to establish the core global and regional atmospheric environmental service that cover European air quality, global atmospheric composition, climate and UV, and solar energy delivered as a component of Europe's GMES (Global Monitoring for Environment and Security) initiative. The global model and data assimilation system used in MACC was based on the European Centre for Medium-Range Weather Forecasts' (ECMWF) Integrated Forecast System (IFS). Inness et al, (2012) described the main modelling components as well as the assimilation of satellite data in the global model to generate the MACC reanalysis atmospheric composition data sets. The global MACC reanalysis service provides a reanalysis for the years 2003-2012 of trace gas and aerosol concentrations.

The experience gained by generating the reanalysis of atmospheric composition as part of the GEMS project assisted in the development of MACC reanalysis data. MACC used a later version of the modelling system as compared to the one used in the GEMS project, and benefited from the assimilation of more and reprocessed satellite data. The modelling system was used in the MACC project to produce reanalysis of atmospheric composition data for the period 2003 to recent years, by assimilating satellite data to constrain O<sub>3</sub>, CO, NO<sub>2</sub>, CO<sub>2</sub>, CH<sub>4</sub>, and aerosol optical depth.

Table 1 shows the main features of the MACC modelling system.

## 3. Mapping species onto CMAQ-MACC

There are 13 gas phase species that can be mapped from MACC directly onto the CB05 chemical scheme in CMAQ-UK. These are: NO<sub>2</sub>, NO, O<sub>3</sub>, HNO<sub>3</sub>, H<sub>2</sub>O<sub>2</sub>, CH<sub>2</sub>O, CH<sub>3</sub>CHO, CO, PAN, C<sub>5</sub>H<sub>8</sub>, SO<sub>2</sub>, OH and C<sub>2</sub>H<sub>6</sub>. Particulate species available from MACC are primary organics, elemental carbon, sodium, chloride, sulphate and desert dust. Desert dust is not treated explicitly in CMAQ and is therefore added to the non-speciated PM mass carried by the model. The methodology developed for mapping species from GEMS to CMAQ have been utilized for CMAQ-MACC mapping.

**Table 1. Main features of the MACC modelling system**

| MACC                    | Details                                                                                                                                                                                                                              |
|-------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Meteorology             | ECMWF                                                                                                                                                                                                                                |
| Resolution              | T255 (85 KM), 60 layers (MOZART), 3 hours                                                                                                                                                                                            |
| Anthropogenic emissions | MACC D-EMIS (MACCcity) in monthly resolution, A modification of GFEDv3 : Within each month and 0.5 deg grid cell the GFED3.0 emissions are redistributed to daily and 0.1 deg resolution according to gridded MODIS FRP observations |
| Natural emissions       | MACC D-EMIS (MACCcity) in monthly resolution                                                                                                                                                                                         |
| Chemical species        | 115 (MOZART-3)                                                                                                                                                                                                                       |

## 4. Modelling setup

In this study, we have developed a methodology to provide boundary conditions to the CMAQ modelling system with the outputs from global chemical transport models STOCHEM, GEMS, GEOS-Chem and MACC. The Weather Research Forecasting (WRF) tool was used to drive the meteorology for CMAQ modelling. To study the seasonal variations of air pollutants due to the use of different boundary conditions in CMAQ, one winter (January) and one summer (July) month of year 2006 were selected. In this study we have run WRF-CMAQ modelling system with 50 and 10 km horizontal resolutions. The 50 km horizontal resolution model domain covers the whole of Europe with 87 x 97 grid cells and the 10 km horizontal resolution domain covers mainland UK and Ireland with 133x93 grid cells. The vertical resolution includes 23 sigma layers and contains 7 layers below 1km height. The parameterization

options and other model options are described in Table 2 and are as proposed for the CMAQ-UK configuration in Phase 1 of this project. The following results and discussions are based on the 10km horizontal resolution model domain that covers UK.

**Table 2: WRF and CMAQ model configurations proposed for CMAQ-UK configurations**

| WRF                |                            | CMAQ                          |                                 |
|--------------------|----------------------------|-------------------------------|---------------------------------|
| Parameter          | Assumption                 | Parameter                     | Assumption                      |
| WRF version        | 3.4                        | CMAQ version                  | 4.7.1                           |
| Grid resolution    | 50km (Europe) to 10km (UK) | Grid resolution               | 50km (Europe) to 10km (UK)      |
| Spatial projection | ETRS89-LCC                 | Spatial projection            | ETRS89-LCC                      |
| Vertical layers    | 23 (7 below 1km)           | Vertical layers               | 23 (7 below 1km)                |
| IC/BC              | EMWF/GFS                   | IC/BC                         | STOCHEM, GEOS-Chem, GEMS/MACC2  |
| Nudging            | Grid (T, WS, Q)            | Chemical Scheme               | CB 05                           |
| PBL                | MYNN 2.5 level TKE         | Temporal emissions profiles   | FMI-TNO-KCL                     |
| Microphysics       | WSM 3-class simple ice     | Point source details          | Include plume rise calculations |
| Cumulus            | Kain-Fritsch               | Emissions processor           | Smoke (v3.X)                    |
| Radiation (SW/LW)  | RRTM/Dudhia                | Area anthropogenic emissions  | EMEP/NAEI                       |
| Land surface       | NOAH                       | Point anthropogenic emissions | NAEI/EPRTTR                     |
| Land use           | WPS IGBP MODIS (30s+20m)   | Natural emissions             | MEGAN/Biomass burning           |
| Surface layer      | MYNN                       |                               |                                 |

## 5. Results and discussions

The influence of four different boundary conditions used in CMAQ on O<sub>3</sub>, NO<sub>x</sub>, PM<sub>2.5</sub>, and PM<sub>10</sub> concentrations are quantified in terms of a) differences in the spatial distribution of concentrations of O<sub>3</sub>, NO<sub>x</sub> and PM, b) differences in the vertical distribution of ozone at one site, and c) differences in hourly concentrations during the winter and summer periods at AURN monitoring sites compared with observations.

### 5.1 Comparison of Spatial distribution of O<sub>3</sub>, NO<sub>x</sub>, PM<sub>2.5</sub>, and PM<sub>10</sub> concentrations

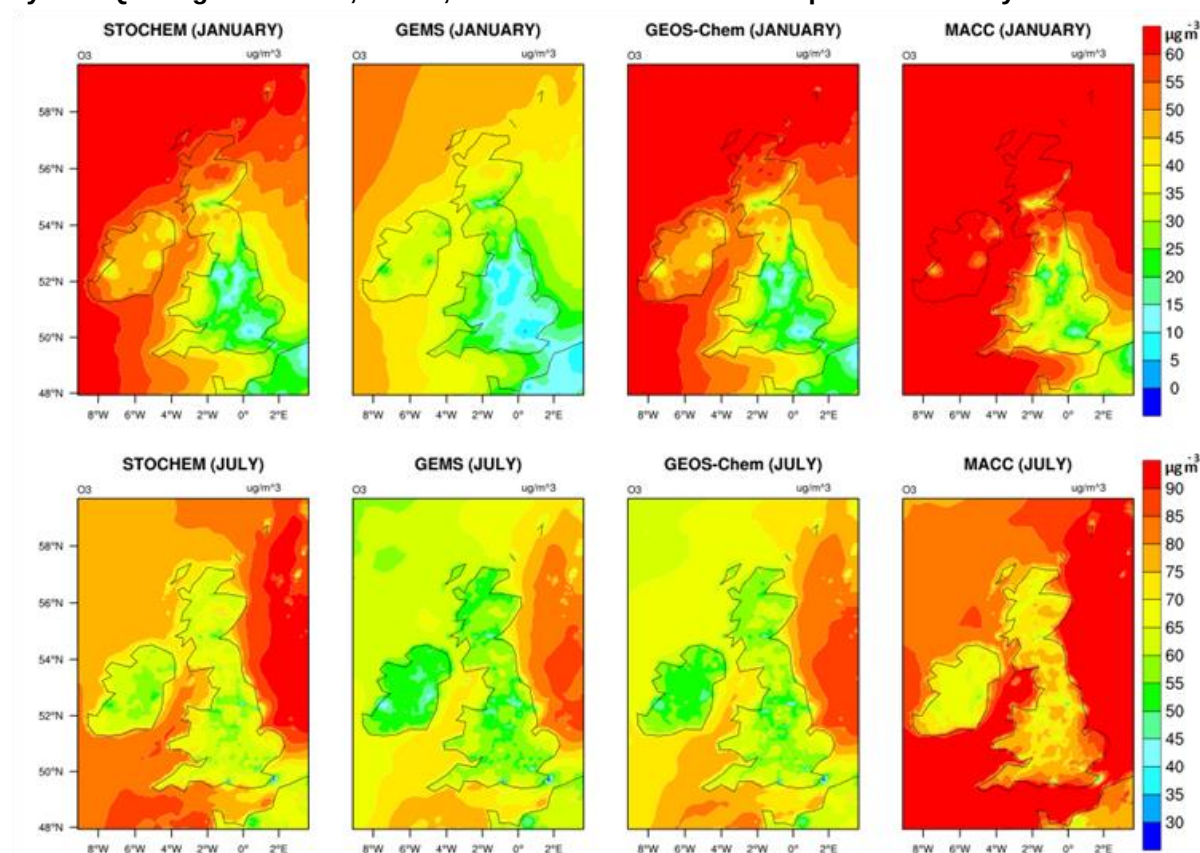
Figure 1 shows the simulated monthly averaged spatial distribution of ozone from CMAQ using STOCHEM, GEMS, GEOS-Chem and MACC output as boundary conditions during January and July 2006. The spatial distribution of ozone shows that CMAQ simulated higher ozone concentration during the summer (July) period than in the winter (January) period due to seasonal variation in temperature and solar radiation. The average difference in the ozone concentration between winter and summer months is about 30-50 µgm<sup>-3</sup> over the domain.

The CMAQ simulation using MACC boundary condition shows higher concentrations of ozone than CMAQ simulations with other boundary conditions in both seasons. All the model simulations show less ozone concentrations over urban agglomerations in the UK especially during winter months. This indicates that the chemical reactions of ozone with primary emissions of NO<sub>x</sub> influences the concentration of ozone during the winter season.

The results show that CMAQ runs with GEMS boundary conditions simulated lower concentrations of ozone compared with runs using other boundary condition sources. This feature using GEMS boundary conditions during winter months is also reported by Schere et al (2012). Figure 1 shows a similar pattern in the spatial distribution of ozone concentrations from CMAQ simulations with GEMS and GEOS-Chem

during the summer period. CMAQ simulations with the MACC boundary conditions led to higher ozone concentration (more than  $25 \mu\text{g m}^{-3}$ ) as compared to CMAQ simulations with

**Figure 1: Spatial distribution of  $\text{O}_3$  during January and July 2006 (monthly averaged) simulated by CMAQ using STOCHEM, GEMS, GEOS-Chem and MACC output as boundary conditions.**



GEMS and GEOS-Chem. CMAQ simulation with STOCHEM boundary conditions predicted less surface ozone concentration than other CMAQ simulations.

Figure 2 shows the simulated monthly averaged spatial distribution of  $\text{NO}_x$  from CMAQ using STOCHEM, GEMS, GEOS-Chem and MACC output as boundary conditions during January and July 2006. The spatial distribution of  $\text{NO}_x$  from CMAQ simulations with STOCHEM, GEMS, GEOS-Chem and MACC shows that the use of different boundary conditions in CMAQ is not significantly influencing the surface  $\text{NO}_x$  concentrations. This shows that primary emissions from different source sectors over a region impact more on the surface  $\text{NO}_x$  concentrations as compared to the boundary conditions.

**Figure 2: Spatial distribution of NO<sub>x</sub> during January and July 2006 (monthly averaged) simulated by CMAQ using STOCHEM, GEMS, GEOS-Chem and MACC output as boundary conditions.**

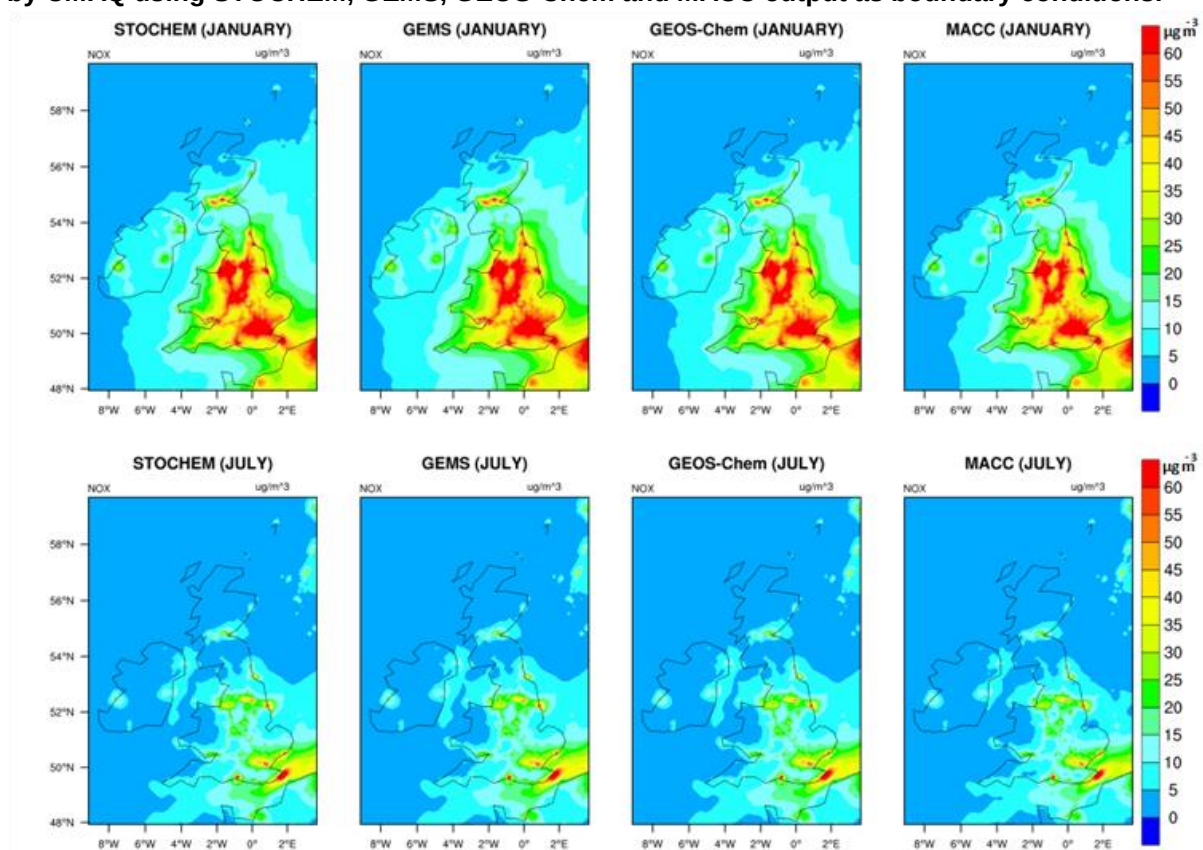
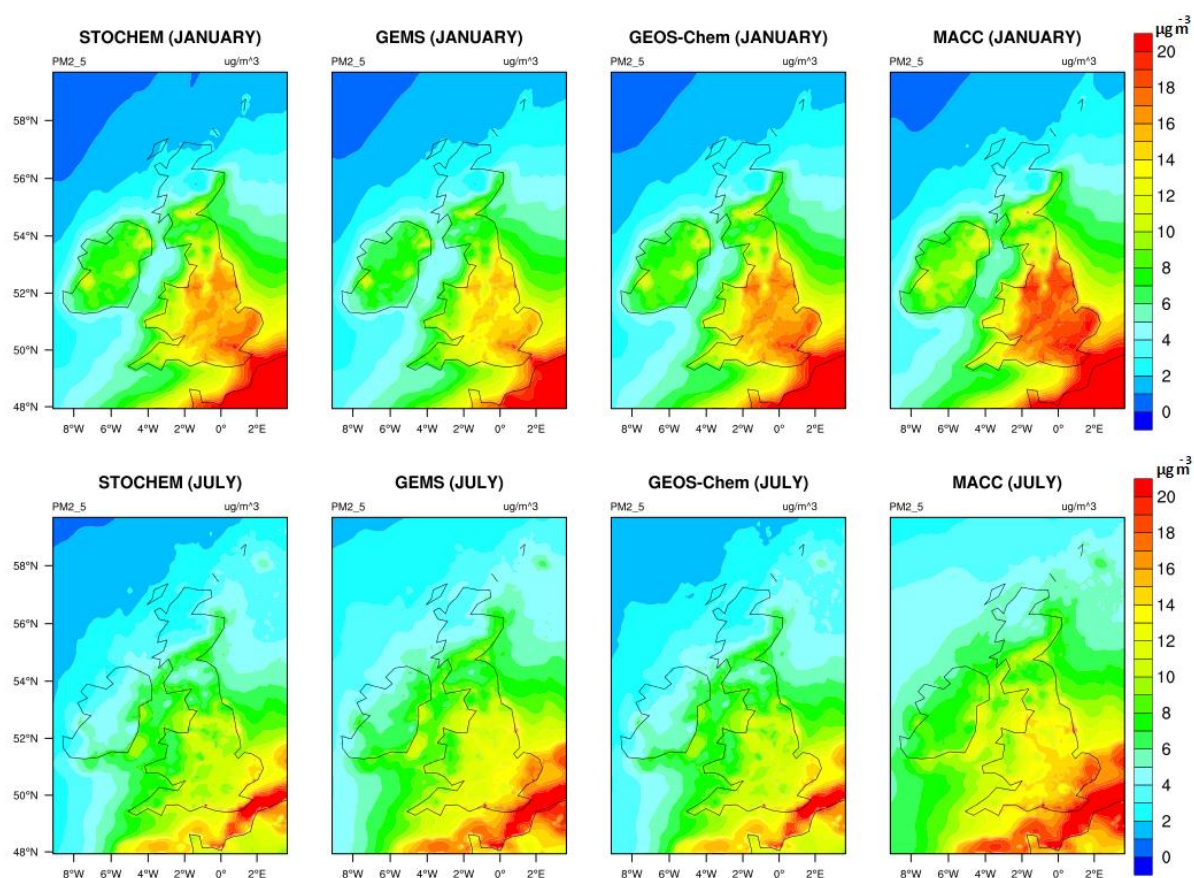


Figure 3 depicts the simulated monthly averaged spatial distribution of PM<sub>2.5</sub> from CMAQ using STOCHEM, GEMS, GEOS-Chem and MACC output as boundary conditions during January and July 2006. The spatial distribution of PM<sub>2.5</sub> from CMAQ simulations with different boundary conditions shows that during the winter season CMAQ predicts higher PM<sub>2.5</sub> concentration than in the summer months by about 8-10  $\mu\text{g m}^{-3}$ . As found for ozone, the CMAQ simulation with GEMS boundary conditions predicted less concentration of PM<sub>2.5</sub> during the winter month than predicted using other boundary condition sources. Figure 3 shows that the CMAQ simulation using MACC boundary condition gives higher concentrations of PM<sub>2.5</sub> than CMAQ simulations with other boundary conditions.

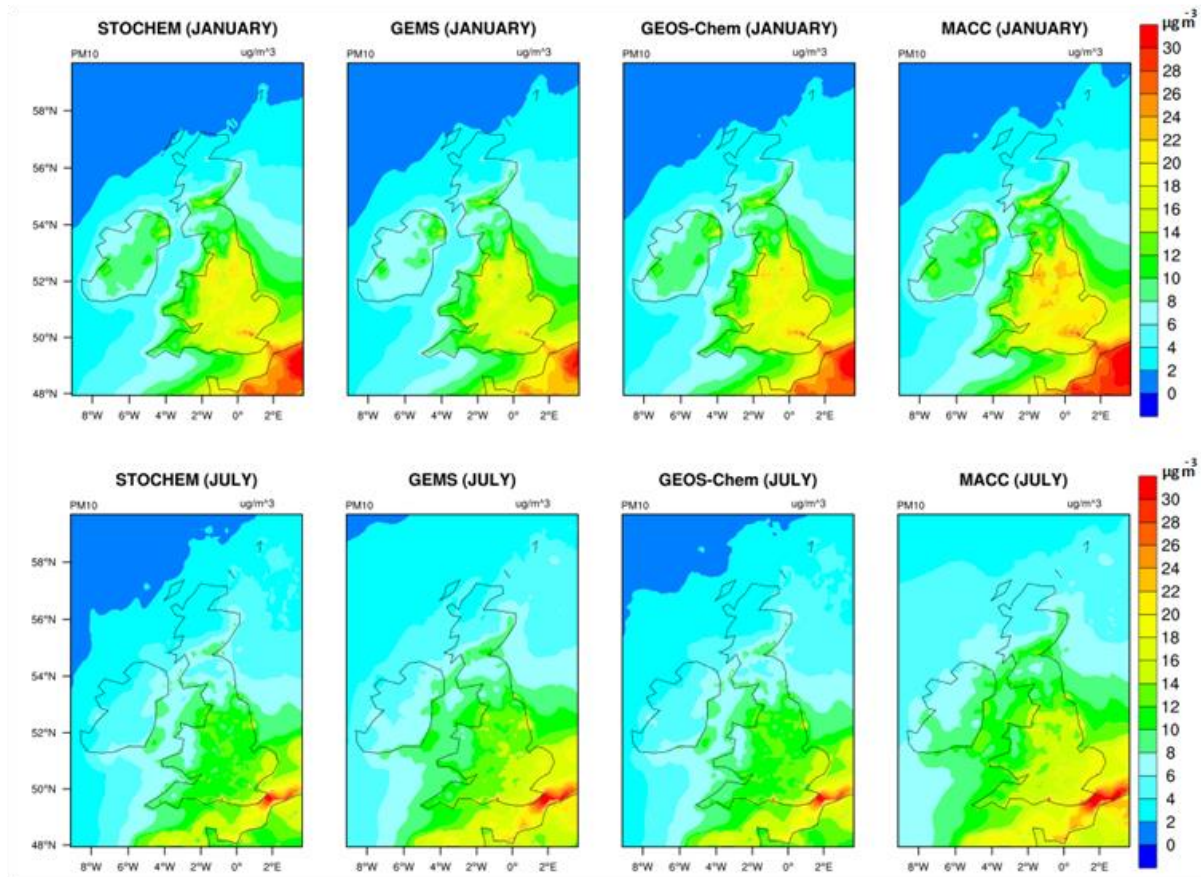
Figure 4 shows the simulated monthly averaged spatial distribution of PM<sub>10</sub> from CMAQ using STOCHEM, GEMS, GEOS-Chem and MACC output as boundary conditions during January and July 2006. The figure shows that the prediction of surface PM<sub>10</sub> concentrations follows the same pattern as PM<sub>2.5</sub> concentrations with regard to the effect of each boundary condition source.



**Figure 3: Spatial distribution of PM<sub>2.5</sub> during January and July 2006 (monthly averaged) simulated by CMAQ using STOCHEM, GEMS, GEOS-Chem and MACC output as boundary conditions.**



**Figure 4: Spatial distribution of PM<sub>10</sub> during January and July 2006 (monthly averaged) simulated by CMAQ using STOCHEM, GEMS, GEOS-Chem and MACC output as boundary conditions.**



## 5.2 Vertical distribution of ozone at Lerwick, Shetland

Figure 5 presents the observed and modelled vertical distribution of ozone concentrations at Lerwick, Shetland on 4th and 11th January 2006 at 11 GMT. The x-axis represents ozone concentration in  $\mu\text{g m}^{-3}$  and the y-axis represents height in hPa. The ozonesonde measurements at Lerwick were taken by the UK Met Office using Electrochemical Concentration Cell methods (Komhyr et al., 1995).

Figure 5 shows that CMAQ simulations with STOCHEM, GEMS and GEOS-Chem boundary conditions underpredicted ozone concentrations from surface to 575 hPa level on 4 January 2006 whereas simulations using MACC overpredicted ozone compared with the measurements. Use of STOCHEM leads to a significant overprediction of ozone concentrations at the 550 to 275 hPa height, by about  $50 \mu\text{g m}^{-3}$  compared with observed values.

The vertical distribution profile on 11 January 2006 shows that CMAQ with GEMS boundary conditions underestimated the ozone concentration throughout the vertical levels. The CMAQ simulation with MACC boundary conditions agrees fairly well with the observation throughout the vertical levels. The profile using GEMS shows less of an underestimation of ozone than GEOS-Chem near the upper troposphere (above 300 hPa). The CMAQ model simulations with GEMS, GEOS-Chem and MACC boundary conditions well predicted the tropopause height (level where sudden increase of ozone concentration occurs) as observed.

**Figure 5: Vertical distribution of observed and modelled ozone concentration on 4 and 11 January 2006 at Lerwick, Shetland. Ozone concentration in  $\mu\text{g m}^{-3}$  are shown on the x-axis and height represented in hPa is shown on the y-axis**

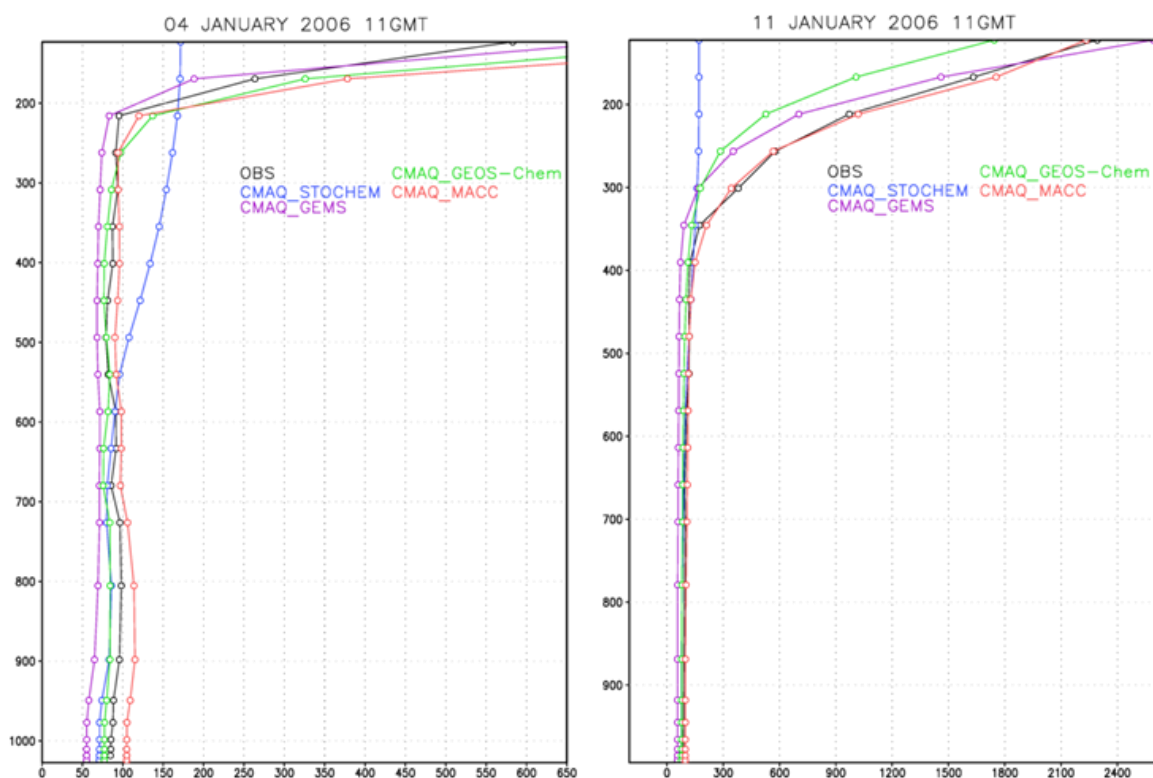
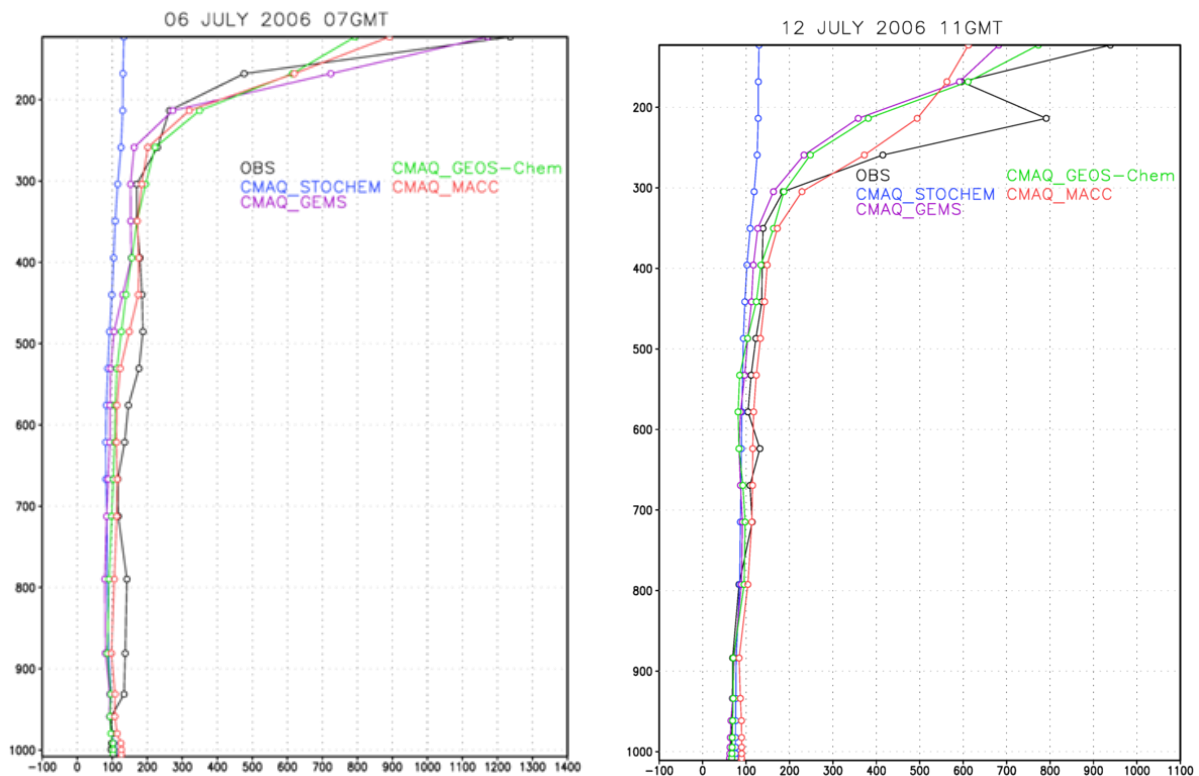


Figure 6 shows the observed and modelled vertical distribution of ozone at Lerwick, Shetland on 6 and 12 July 2006 at 11 GMT. All model simulations underestimated the ozone concentration as compared to the observation at heights from 950 hPa to 450 hPa level on the 6 July 2006. The vertical structure in ozone concentrations in the 450-200 hPa height range is well reproduced by CMAQ using MACC and GEOS-Chem boundary conditions. For the 12 July 2006 simulation, the vertical distribution of ozone simulated by CMAQ using MACC boundary conditions agrees well with the observed values especially in the mid troposphere, but is rather underestimated in the upper troposphere.



**Figure 6: Vertical distribution of observed and modelled ozone concentration on 6 and 12 July 2006 at Lerwick, Shetland. Ozone concentration in  $\mu\text{gm}^{-3}$  are shown on the x-axis and height represented in hPa is shown on the y-axis**



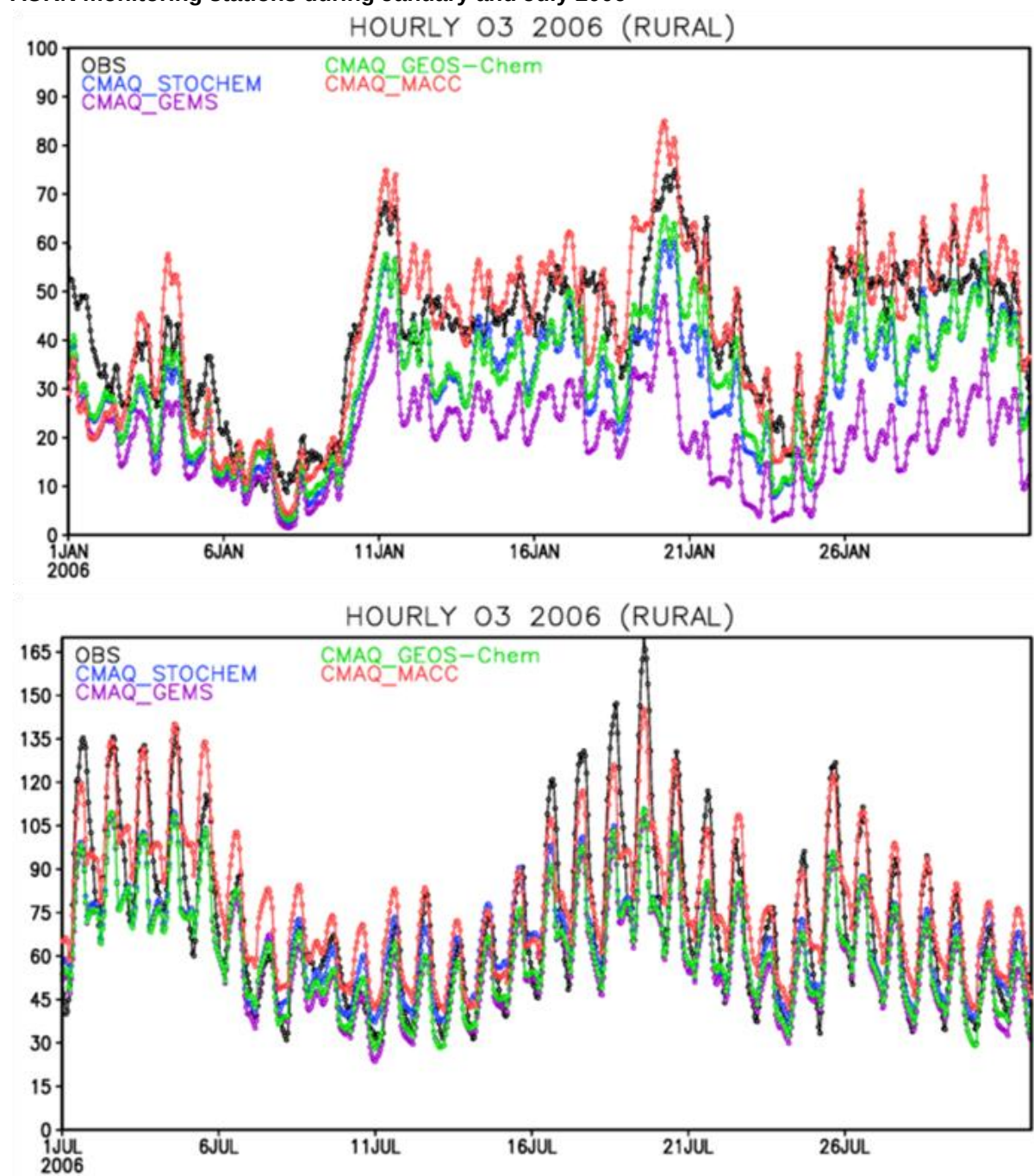
### 5.3 Comparison of model performance with observations from AURN sites

The performance of CMAQ simulations with different boundary conditions were carried out for January and July 2006 by comparing with available AURN observations.

Figure 7 shows the measured and modelled surface ozone concentration averaged over all available observations from rural AURN stations during January and July 2006. It shows that the simulated ozone concentration from CMAQ simulation with STOCHEM, GEMS, and GEOS-Chem underestimated as compared to the observations during both winter and summer seasons. However, CMAQ simulations with MACC boundary conditions overestimated surface ozone in almost all days in January 2006. The CMAQ simulation with MACC boundary conditions shows fairly good agreement with observations over the rural stations during the summer season and outperformed compared to other model simulations.

Figure 8 shows the measured and modelled surface ozone concentration averaged over all available urban AURN monitoring stations during January and July 2006. The performance of CMAQ with MACC boundary conditions for the urban stations is similar to that shown for the rural sites as shown in Figure 7. However, CMAQ with GEOS-Chem boundary conditions shows better agreement with the observations during January 2006.

Figure 7: Measured and modelled surface ozone concentration averaged over all available rural AURN monitoring stations during January and July 2006



**Figure 8: Measured and modelled surface ozone concentration averaged over all available urban AURN observations during January and July 2006**

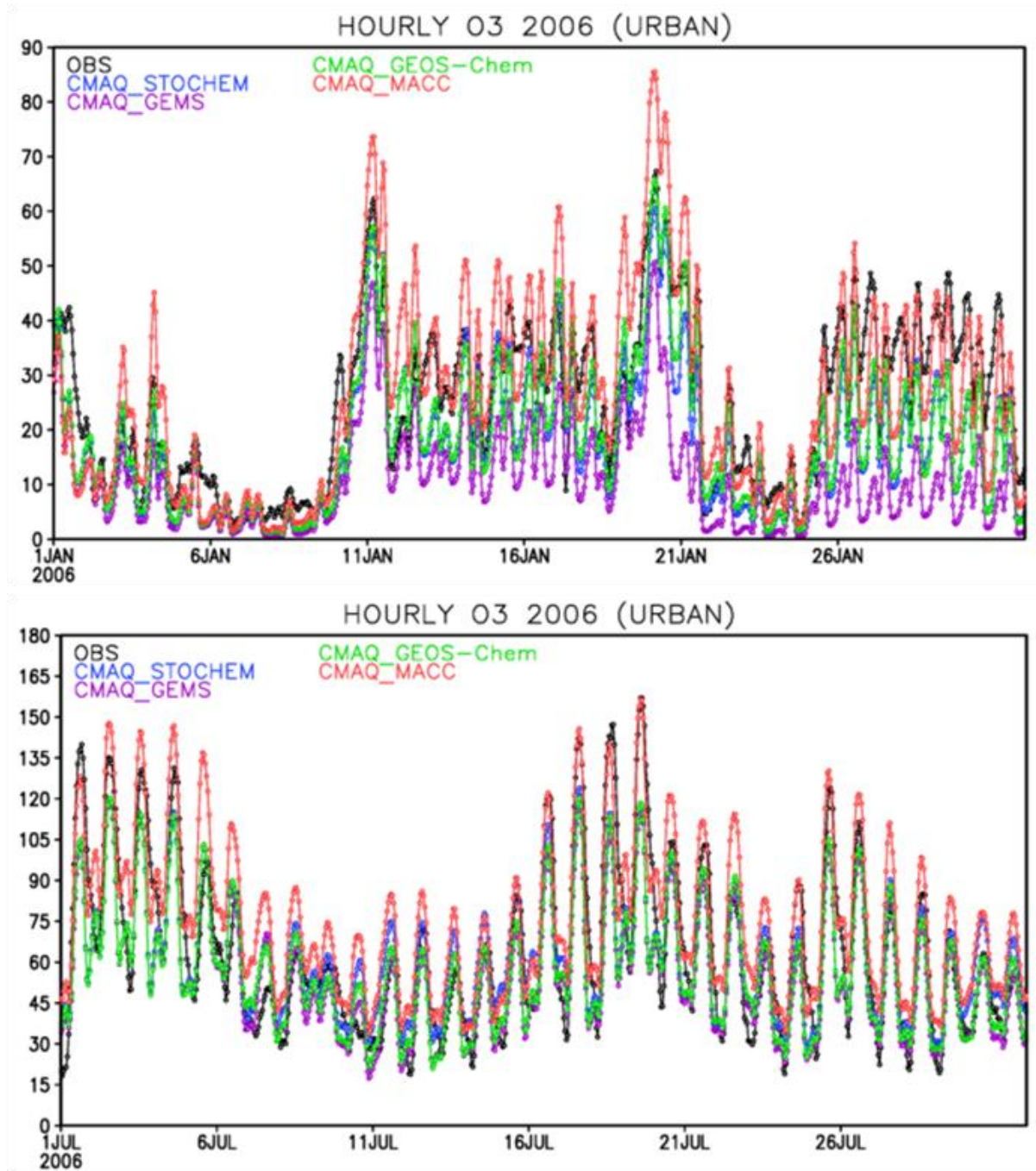


Figure 9 shows the measured and modelled surface  $PM_{2.5}$  concentration averaged over all available observations from rural AURN stations during January and July 2006. The figure shows that all the model simulations underestimated the surface  $PM_{2.5}$  concentration during winter and summer periods and the CMAQ simulation with GEMS underestimated to a greater extent compared with other boundary condition runs for January. The hourly variation of  $PM_{2.5}$  concentration during the summer period shows that all the model runs underpredict  $PM_{2.5}$  in a similar manner. The CMAQ simulation with MACC boundary conditions performed better than the other model simulations especially during winter periods.



**Figure 9: Measured and modelled surface PM<sub>2.5</sub> concentration averaged over all available rural AURN observations during January and July 2006**

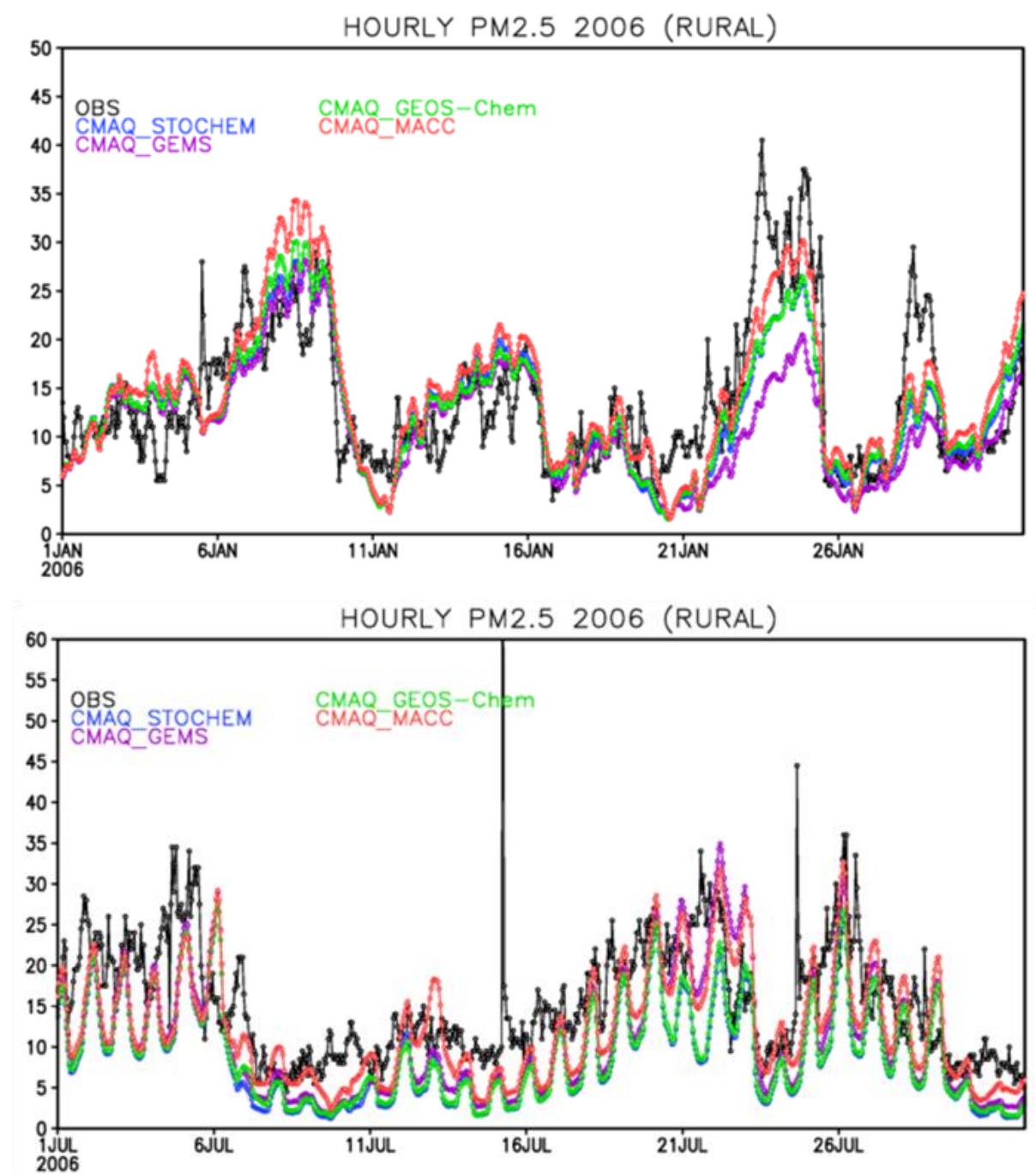


Figure 10 shows the measured and modelled surface PM<sub>2.5</sub> concentration averaged over all available observations from urban AURN stations during January and July 2006. Figure 10 shows that the CMAQ simulation with MACC boundary condition performed better than other model simulations. The urban modelling results shows a similar performance pattern to that shown for the rural modelling results in Figure 9.



**Figure 10: Measured and modelled surface PM<sub>2.5</sub> concentration averaged over all available urban AURN observations during January and July 2006**

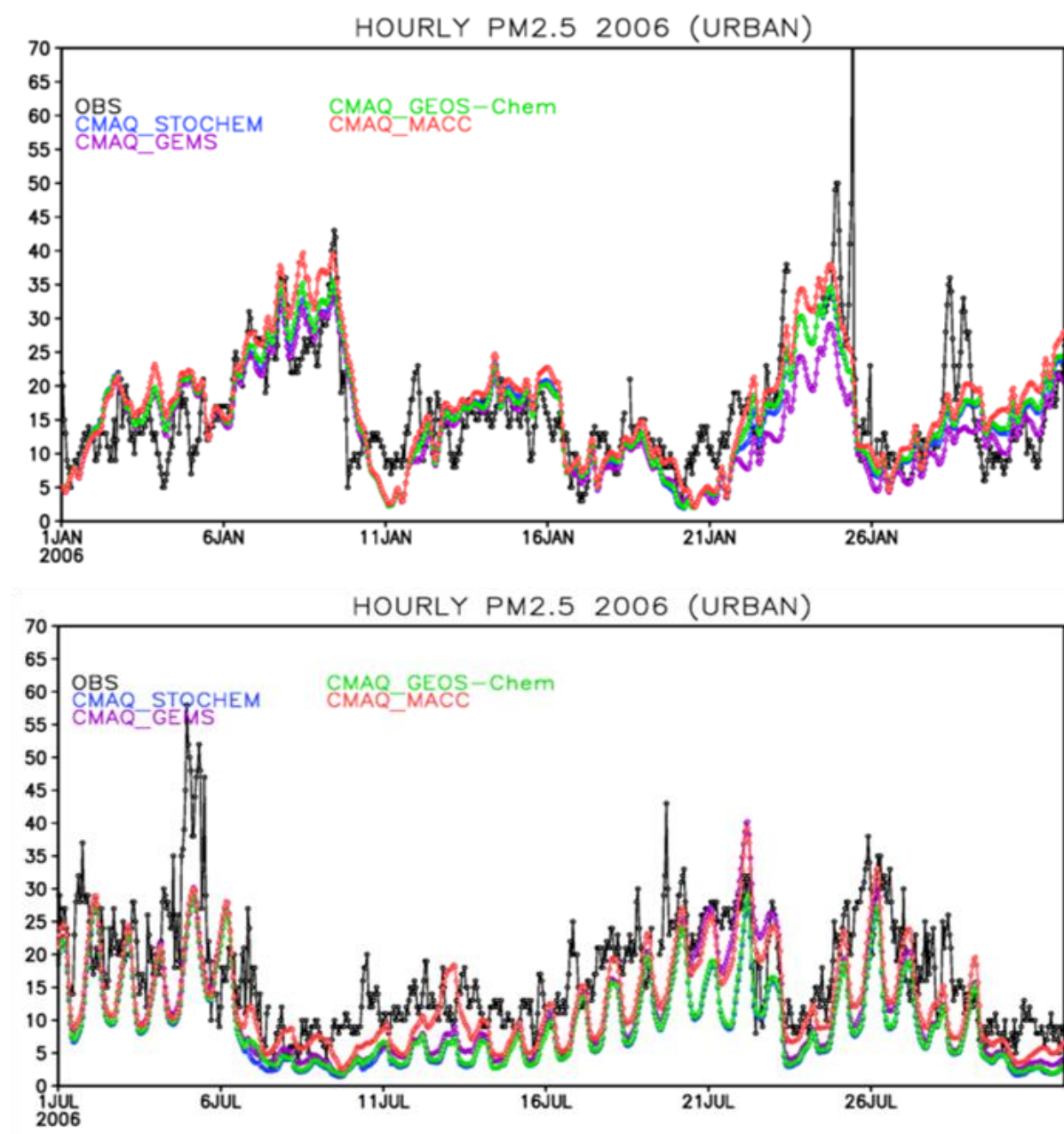
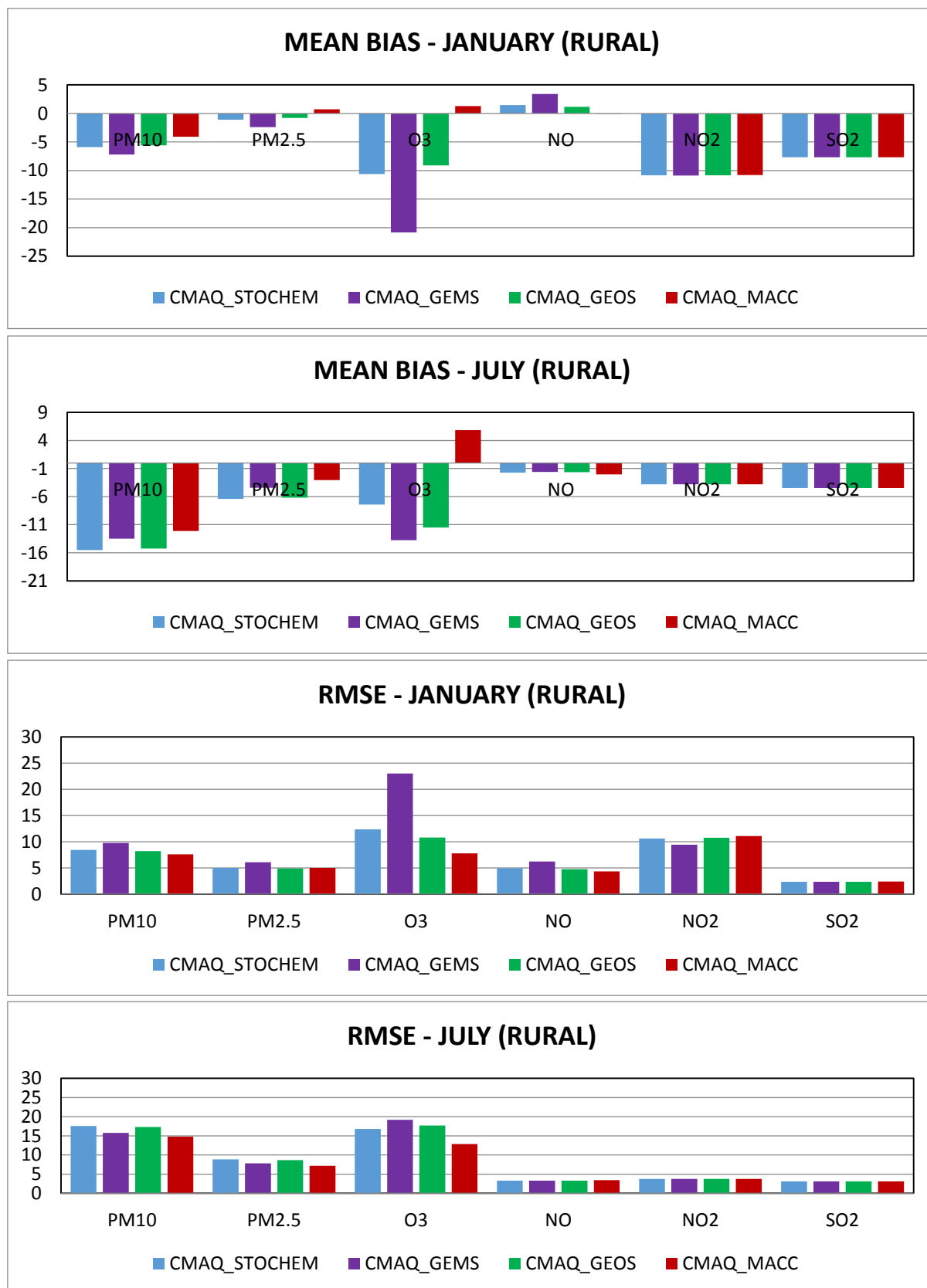
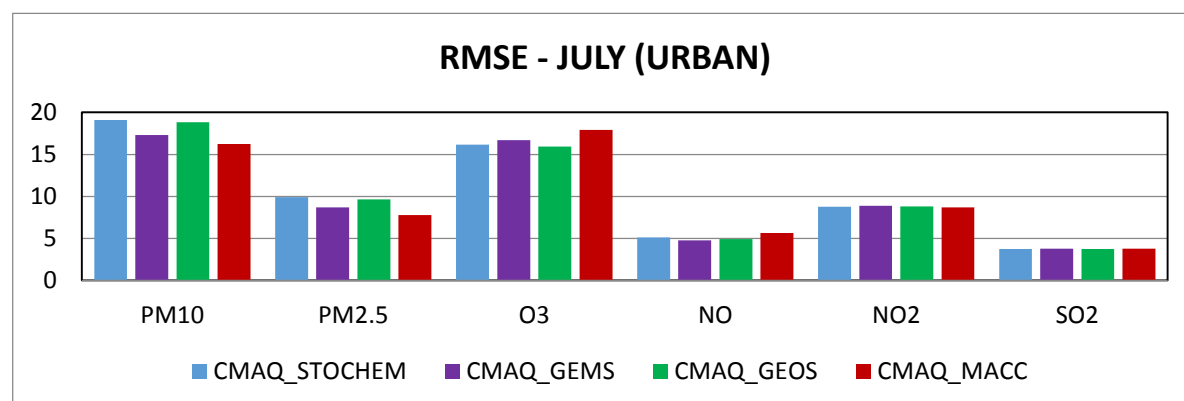
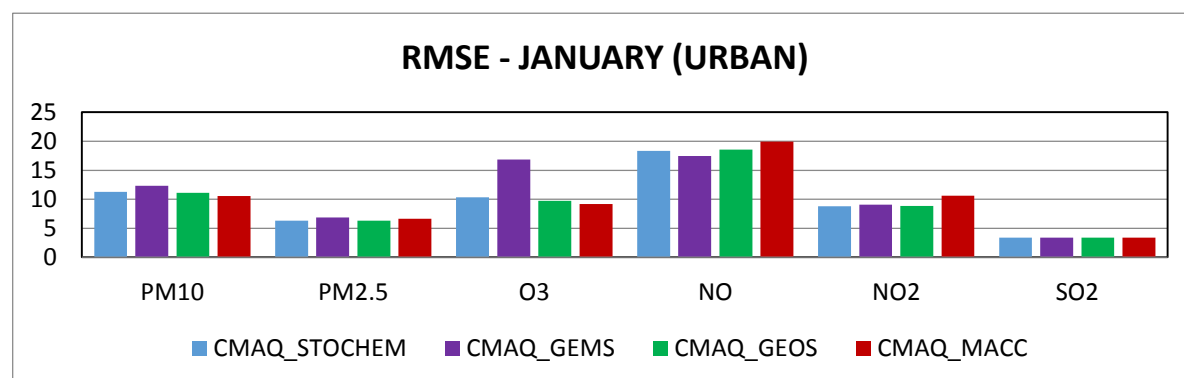
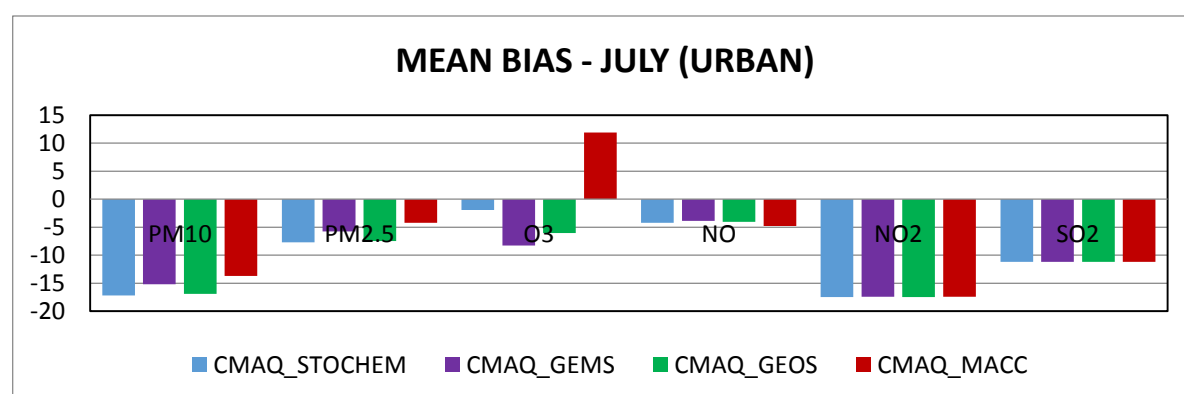
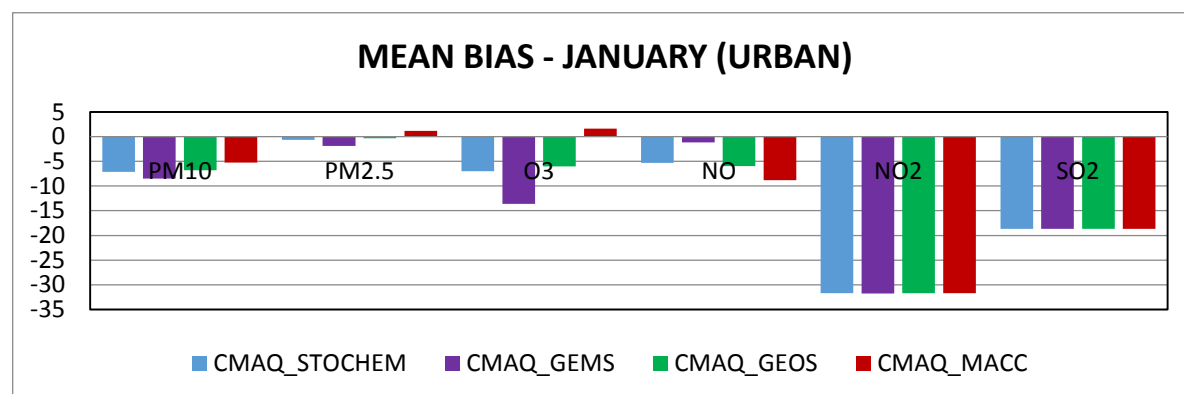


Figure 11 and Figure 12 show the mean bias and root mean square error of PM<sub>10</sub>, PM<sub>2.5</sub>, O<sub>3</sub>, NO, NO<sub>2</sub>, and SO<sub>2</sub> from CMAQ simulations with different boundary conditions compared with the observations from rural and urban background stations in the UK during January and July 2005. The result shows that CMAQ simulations with different boundary conditions did not impact significantly on the concentrations of NO, NO<sub>2</sub>, and SO<sub>2</sub>. However, as found in previous studies the use of boundary conditions in CMAQ does considerably influence modelled ozone and PM concentrations. The CMAQ simulation with MACC boundary conditions performs better than other model simulation in terms of less mean bias and less RMSE. As apparent from the 2006 simulations, CMAQ with GEMS boundary condition shows large mean bias and RMSE especially during the winter period.

**Figure 11: Mean bias and Root Mean Square Error (RMSE) of PM<sub>10</sub>, PM<sub>2.5</sub>, O<sub>3</sub>, NO, NO<sub>2</sub>, and SO<sub>2</sub> from different CMAQ simulations compared with the observations from rural background stations in the UK during January and July 2005**



**Figure 12: Mean bias and Root Mean Square Error (RMSE) of PM<sub>10</sub>, PM<sub>2.5</sub>, O<sub>3</sub>, NO, NO<sub>2</sub>, and SO<sub>2</sub> from different CMAQ simulations compared with the observations from urban background stations in UK during January and July 2005.**



## 6. Summary

The present study investigated the influence of four different boundary conditions from the STOCHEM, GEMS, GEOS-Chem and MACC global models on UK regional air quality using the WRF-CMAQ modelling system. To study the impact of different boundary conditions on the predicted seasonal variations, the WRF-CMAQ modelling system has been used to simulate January and July as representative of winter and summer seasons.

This study supports the findings of Schere et al (2012) such that the region where internal forcing by emissions and chemistry in the models is weaker is fairly sensitive to the specification of the boundary conditions. A comparison of the spatial distribution of ozone and PM concentration modelled in different CMAQ simulations suggested the choice of boundary conditions from different global models influences the overall chemistry and transport of air pollutants (horizontally as well as vertically) in the regional models, affecting the overall results. The results showed that CMAQ simulations with MACC boundary conditions predicted comparatively high concentrations of ozone and particulate matter compared with CMAQ simulations using STOCHEM, GEMS, and GEOS-Chem boundary conditions.

The comparison of modelled and observed vertical distribution of ozone at the Lerwick ozonesonde station showed that all the model simulations reproduced the vertical structure of ozone fairly well, with some underestimations apparent. CMAQ simulations using MACC boundary conditions generally reproduced the vertical structure better than simulations with other boundary conditions.

Overall the hourly time series comparison and statistics revealed that CMAQ simulations with MACC boundary conditions performed better than CMAQ simulations with STOCHEM, GEMS, and GEOS-Chem boundary conditions.

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