

Air4EU

Air Quality Assessment for Europe: from local to continental scale



6th Framework Programme- Policy oriented Research
Priority 8.1 Topic 1.5 Task 2

Individual case study report 9: Assessment of stationary source contribution to surface pollutant concentrations using a 3-D Eulerian Air Quality Model

| | |
|----------------------|-------------|
| Deliverable: | D7.1 Part 9 |
| Dissemination level: | PU |
| Editor: | Ye YU |
| Version: | Final |
| Date: | March 2007 |
| Contract: | 503596 |

LIST OF AIR4EU PARTNERS

| Partic. no. | Participant name | Participant short name | Country |
|--------------------|------------------------------------------------------------------------------------|-------------------------------|----------------|
| 1 | Netherlands Research Organisation | TNO | NL |
| 2 | Norsk Institut for Luftforskning | NILU | NO |
| 3 | Aristotle University Thessaloniki | AUT | GR |
| 4 | University of Stuttgart | IER | DE |
| 5 | University of Hertfordshire | UH | UK |
| 6 | Universidade de Aveiro | UAVR | PT |
| 7 | AIRPARIF | AIRPARIF | FR |
| 8 | Agenzia per i Trasporti Autoferrotramviari e la Mobilità del Comune di Roma S.p.A. | ATAC | IT |
| 9 | Environment Agency | EA | UK |
| 10 | City Development Authority of Prague | URM | CZ |
| 11 | Enveco | ENVECO | GR |
| 12 | Gemeentewerken Rotterdam | GW | NL |
| 13 | Milieudienst Rijnmond | DCMR | NL |
| 14 | City of Oslo, Public Health Authority | OPHA | NO |

Table of Content

| | |
|----------------------------------------------------------------------------------------|-----------|
| 1. EXECUTIVE SUMMARY | 2 |
| 2. CASE STUDY DESCRIPTION | 2 |
| 2.1 Background..... | 2 |
| 2.2 Aim and description..... | 3 |
| 2.3 Relevance to recommendations in Air4EU | 3 |
| 3. METHODOLOGY | 4 |
| 3.1 The Models-3/CMAQ | 4 |
| 3.2 Assessment of the results | 5 |
| 4 RESULTS..... | 6 |
| 4.1 CMAQ performance | 6 |
| 4.2 Contribution of industrial point source emissions to ambient pollution levels..... | 9 |
| 5 CONCLUSION AND DISCUSSION | 11 |
| 5.1 Assessment of the case study | 11 |
| 5.2 Improvements in assessment derived from case study..... | 11 |
| 5.3 Recommendations resulting from the case study | 12 |
| 5.4 Suitability for implementation in other cities | 12 |
| REFERENCES..... | 12 |

1. Executive Summary

Conventionally stationary source emissions are evaluated using dispersion models in the near-field, with background concentrations provided from measurements. Usually one ground-level upwind monitoring site is used to describe the composition of the background air coming into a region of a stationary source. Long-range deposition from stationary sources is treated by assuming the stationary source makes up part of the emissions within a grid square. More strict emission controls are already in place, or will be in place for most major European stationary sources, which would tend to focus attention on the interaction of stationary sources, in combination with other sources, rather than the behaviour of a single source on its own. The complex nature of the problem which is governed by multipollutant and multiscale interactions and coupling between atmospheric chemistry and dynamics suggest that improved methods are needed. This case study demonstrates the use of a 3-D Eulerian air quality model-CMAQ to estimate the contribution of UK stationary point source emissions to ground level pollution levels focusing on the uncertainty related to model resolution. The investigated area is the Southeast of England. Twenty-two monitoring stations with observation on O₃ and NO₂ are available within the studied domain. The model was assessed using statistical parameters including correlation coefficient, fractional bias, root mean squared error, normalized standard deviation and fraction of predictions within a factor of 2 of observations. Results indicate that the CMAQ model has better performance for O₃ than for NO₂. For NO₂ fine-resolution simulation presents better statistic scores. The performance of the model for ground-level O₃ was not improved when the horizontal grid spacing was reduced. The contribution of UK point sources to ground level O₃ and NO₂ concentrations in the presence of other sources, i.e. area, traffic and biogenic, was assessed using sensitivity analysis where emissions from UK point sources were removed. The dependence of results on model resolutions was discussed. This case study supports a number of recommendations concerning the uncertainty estimation of numerical models.

2. Case study description

2.1 Background

Industrial sources release pollutants such as sulphur dioxide (SO₂), Nitrogen Oxides (NO_x), Volatile Organic Compounds (VOCs) and Particulate Matter (PM₁₀) into the environment. The emitted SO₂, NO_x and VOCs in the atmosphere can undergo a complex series of chemical reactions resulting in the production of environment and human health harmful pollutants such as ozone (O₃), gaseous NO₂ and secondary aerosols containing sulphates and nitrates. Standards and limit values for these pollutants have been specified in the National Air Quality Strategy (DETR 2000) and European Air Quality Framework Directives (96/62/EC). As part of the Convention on Long-Range Transboundary Air Pollution, pollutants associated with industrial sources (sulphur, NO_x and VOCs) are also subject to emission ceilings set for 2010 in the Gothenburg Protocol (1999). Many specific sources, such as, combustion plants and electricity power stations, are controlled by the Protocol through strict emission limit values.

Conventionally stationary source emissions are evaluated using dispersion models in the near-field, with background concentrations provided from measurements. Usually one ground-level upwind monitoring site is used to describe the composition of the background air coming into a region of a stationary source. Long-range deposition from stationary sources is treated by assuming the stationary source makes up part of the emissions within a grid square. More strict emission controls are already in place, or will be in place for most major European stationary sources, which would tend to focus attention on the interaction of stationary sources, in combination with other sources, rather than the behaviour of a single source on its own.

A big challenge for the control of secondary pollutants, such as O₃, and their precursors is the non-linearity between the emissions of primary pollutants and the resulting concentrations. This makes the assessment of the size of the contribution of an industrial plume to the removal or formation of a secondary pollutant particularly difficult. The complex nature of the problem which is governed by multipollutant and multiscale interactions and coupling between atmospheric chemistry and dynamics suggest that improved methods are needed. Eulerian grid models are being a common way to investigate the chemistry and physics among pollutants from different sources on urban and regional scales. They can treat the detailed distribution of emissions and represent the horizontal and vertical transport of precursors and products. The Models-3/ CMAQ (Community Multi-scale Air Quality model) (Byun and Ching, 1999) is one of such models.

This case study investigate the possible use of the 3-D Eulerian air quality model - CMAQ, for estimating the contribution of UK stationary point source emissions to ground level O₃ and NO₂ concentrations and the uncertainty related to model resolutions.

2.2 Aim and description

This case study is to assess the possible use of advanced models as a tool for regulating secondary pollutants from stationary industrial point sources. The main aim is to demonstrate the possible use of the 3-D Eulerian air quality model - CMAQ as a tool for regulating secondary pollutants, such as ozone and nitrogen dioxide, related to emissions from large stacks.

Modelling results from CMAQ model are presented and recommendations on methods are made. For regulatory applications, a model must demonstrate “acceptable” performance before contributions from different emission sources are estimated. Many evaluation approaches have been recommended in the literature (Chang and Hanna, 2004; M 6.5). In this case study the performance of CMAQ model was evaluated quantitatively using five statistical parameters, i.e. correlation coefficient, fractional bias, root mean squared error, normalized standard deviation and fraction of predictions within a factor of 2 of observations. Observations from 22 monitoring stations are available in the studied domain for model evaluations. Uncertainties resulting from model resolutions are investigated and further research needs are identified.

2.3 Relevance to recommendations in Air4EU

This case study tests a number of recommendations concerning model resolution, uncertainties assessment and their dependence on model resolution. This case study in particular, touches on the following recommendations:

- The uncertainty of model results should be determined, and should form an integral part of the AQ-assessment.
- The definition and standardisation of validation procedures is essential for the soundness of use of model results and a clear statement of model uncertainties is indispensable in any AQ assessment study.
- Further research is needed to improve the model's capability for NO₂, including the effect of horizontal model resolution on model performance.

3. Methodology

3.1 The Models-3/CMAQ

The CMAQ model is a comprehensive Eulerian grid model designed to support air quality modelling applications ranging from regulatory issues to scientific research on atmospheric processes. The model can address tropospheric ozone, acid deposition, visibility, fine particulate and other air pollutant issues in the context of “one” atmosphere perspective where complex interactions among multiple atmospheric pollutants and between regional and urban scales are confronted (Dennis et al. 1996, Byun and Ching 1999). In this case study, the model was configured to have four nesting levels with spatial resolutions down to 3 km (see Figure 3.1). The information on grid spacing and number of grid points is provided in Table 3.1. The outermost model domain has been set-up to be large to reduce the possible effect of model boundary conditions on the overall model predictions. Twenty-six vertical σ -levels extend from the surface to about 14 km. Vertical layers are unevenly distributed with fifteen layers in the lowest kilometre, and a surface layer of approximately 14 m above ground level (AGL).

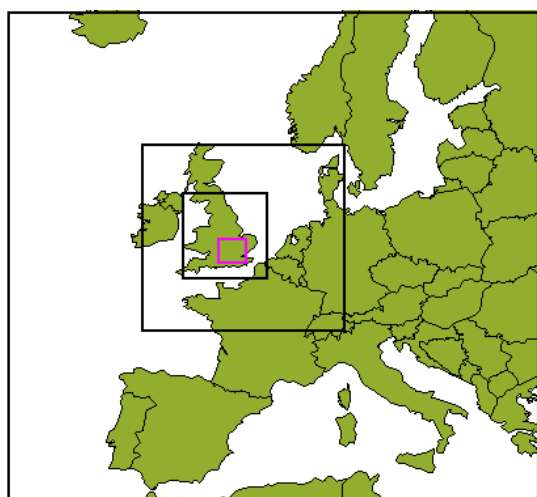


Figure 3.1 The nested CMAQ modelling domain. Note that the MM5 domain is at least five grid cells larger than the CMAQ domain.

Table 3. 1 Grid structure for model runs

| Grid | Δx (km) | N_x | N_y |
|------|-----------------|-------|-------|
| 1 | 81 | 42 | 42 |
| 2 | 27 | 8 | 48 |
| 3 | 9 | 60 | 66 |
| 4 | 3 | 60 | 54 |

CMAQ model currently includes three chemical mechanisms, i.e. CB-IV (Gery et al. 1994), RADM2 (Stockwell et al. 1990) and SAPRC99 (Carter 2000). In the present study CMAQ was applied with the modified version of the CB-IV chemical mechanism, named ‘cb4_ae3_aq’, along with the ‘smvgear’ chemistry solver and the ‘aero3’ aerosol module. Meteorological fields, including wind speeds and directions, temperature, humidity, pressure, and solar radiation, were obtained from the fifth

generation Mesoscale Model (MM5) developed by Pennsylvania State University (PSU) and the National Center for Atmospheric Research (NCAR) (Grell *et al.* 1994). The boundary and initial conditions for the MM5 simulation was provided by the ECMWF 1°X1° reanalysis data available at every 6 h.

The initial and boundary conditions for model species for the coarsest domain is generated based on monthly mean data from the UK Meteorological Office global Lagrangian tropospheric 3-D chemical transport model (STOCHEM). STOCHEM outputs concentrations of 26 species with a horizontal resolution of 5° latitude X 5° longitude and 9 vertical layers extending from surface up to the 150 mb (Collins *et al.* 1997). The initial and boundary conditions for the inner three domains are provided by the coarser domain. To reduce the effect of initial conditions, all the simulations have a spin-up time of 48 hrs.

CMAQ requires hourly emissions for model species related to chemical mechanism (CB-IV in this study) for each grid cell of the model domains. Annual anthropogenic emissions for six pollutants, i.e. nitrogen oxides (NO_x), nonmethane volatile organic compounds (NMVOCs), sulfur dioxide (SO₂), carbon monoxide (CO), ammonia (NH₃) and particulate matter (PM₁₀), were taken from the EMEP 50 km (Vestreng *et al.* 2005, see also website <http://www.emep.int>) and UK NAEI (National Atmospheric Emissions Inventory) 1 km (<http://www.naei.org.uk/>) gridded data for the year 2002. Annual emissions and information on locations for point sources were extracted from the EPER (European Pollutant Emission Register)(<http://www.eper.cec.eu.int/>) and NAEI. These annual emissions data were processed by the Sparse Matrix Operator Kernel Emissions Modelling System (SMOKE) (Carolina Environmental Programs, 2003) to obtain gridded, hourly, speciated emission inputs for CMAQ. Temperature-dependent biogenic emissions of isoprene and monoterpenes were estimated following the method of Guenther *et al.* (1995) and Sanderson (2002), using the 100-m resolution CORINE Land Cover data and the hourly temperature and solar radiation values from MM5.

3.2 Assessment of the results

Model simulations for a summer high O₃ and NO₂ episode during the period of 22 -28 June 2001 were conducted using CMAQ to investigate the contribution of stationary industrial sources to near surface pollutant concentrations. During this episode there was a ridge of high pressure over the Western Europe, which led to the advection of oxidant-rich air over Southern England from the Europe continent. The warm weather (with maximum temperatures reached 30°C on 26 June 2001) and still conditions prevailed over much of southern and eastern Britain during this episode.

The impact of industrial sources was evaluated using a sensitivity analysis approach. Firstly concentrations were estimated for a base-case (Run A), which uses real emissions. Then concentrations from a sensitivity run (Run B) with adjusted emissions were calculated. In the base-case, simulation was accomplished base on the emissions inventory data from the EMEP, EPER and NAEI as described in section 3.1. In the sensitivity simulation, emissions from all UK point sources were removed.

The total model uncertainty and the uncertainty resulting from using different model resolutions are determined by comparing observations and model predictions from the base case through the application of several statistical parameters including correlation coefficient, fractional bias, root mean squared error, normalized standard deviation and fraction of predictions within a factor of 2 of observations. Definitions of these parameters are summarized in Table 3.2. The information from the evaluation helps to understand model limitations and provide a support for further model development.

The analysis will focus on the comparison of model predictions for the finest grid domain of the simulation, namely, the region overlapping the 9 km and 3 km domains. All the statistics were calculated for the whole simulated period excluding the first 48 h spin-up time. Model values used in statistic calculations were extracted from the first model level (about 14 AGL).

Table 3. 2 Definitions of statistical parameters. N is the product of the number of simulation hours and the number of ground-level monitoring stations providing hourly observational data. M_i or O_i represents the model predicted or observed value.

| Statistics | Definition |
|--------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------|
| Mean observation (O_0) | $\frac{1}{N} \sum_{i=1}^N O_i$ |
| Mean model prediction (M_0) | $\frac{1}{N} \sum_{i=1}^N M_i$ |
| Fractional bias (FB) ^c | $\frac{M_0 - O_0}{0.5(O_0 + M_0)}$ |
| Normalized Standard deviation (NSD) ^c | $\sigma_{\text{obs}} / \sigma_{\text{mod}}$ |
| Root Mean Square Error (RMSE) ^a | $\left\{ \frac{1}{N} \sum_{i=1}^N (M_i - O_i)^2 \right\}^{1/2}$ |
| Correlation coefficient (R) ^b | $\frac{\frac{1}{N} \sum_{i=1}^N \{(M_i - M_0)(O_i - O_0)\}}{\sigma_{\text{mod}} \times \sigma_{\text{obs}}}$ |
| Fraction of predictions within a factor of 2 of observations | $0.5 \leq M_i/O_i \leq 2.0$ |

^a McNally and Tesche (1993), ^b Hanna (1994), ^c Air 4EU report on Cross Cutting Issues (M 6.5).

4 Results

4.1 CMAQ performance

In this section model results from the base case were compared to observations from 22 monitoring stations for different grid resolutions.

Qualitatively, the model simulates the diurnal O_3 and NO_2 concentration patterns very well at all sites. Figure 4.1 compares the measured O_3 and NO_2 time series with the modelled results at two representative sites (i.e. a rural site – Harwell and a suburban site - London Bexley). At both sites, the model captures the O_3 night time lows quite well, but it tends to underpredict daytime peaks during high ozone days, for example on 24th and 26th June at London Bexley and on 25th and 26th June at Harwell.

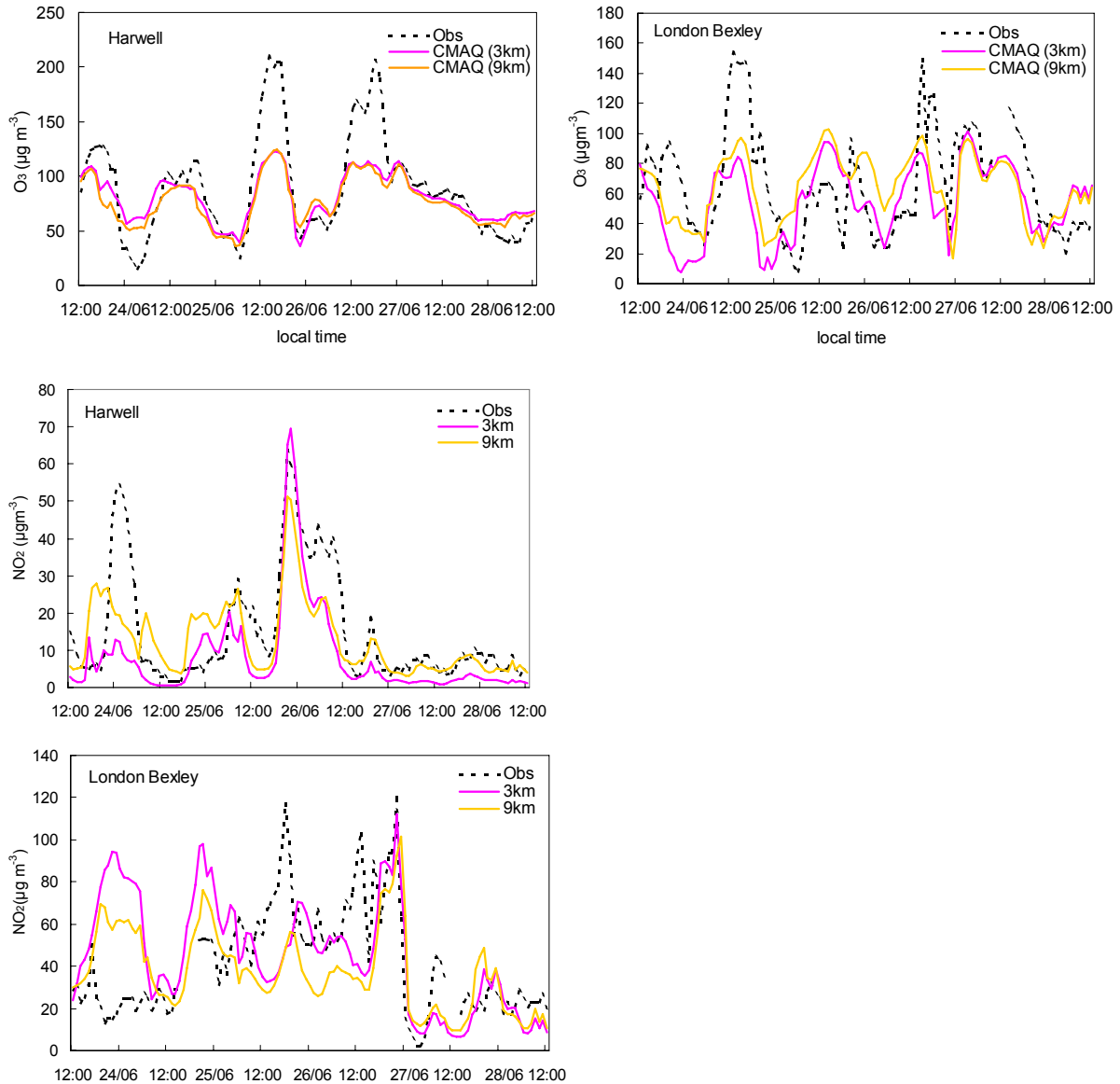


Figure 4.1 Comparison of measured and modelled time series of O_3 and NO_2 concentrations at London Bexley (suburban site) and Harwell (rural site). Modelled O_3 and NO_2 concentrations were extracted from the first model level (about 14 m AGL).

For NO_2 , the model gives better predictions after 4-day spin-up time (model simulation started on 22 June 2001). Both the lows and highs are captured fairly well.

Figure 4.2 shows scatter plots of measured–modelled O_3 pairs (left panel) and NO_2 pairs (right panel) for all the modelling hours and sites for the 3-km resolution simulation. Tables 4.1 and 4.2 summarize the corresponding O_3 and NO_2 performance statistics for both the 9-km and 3-km resolution simulations for different types of stations.

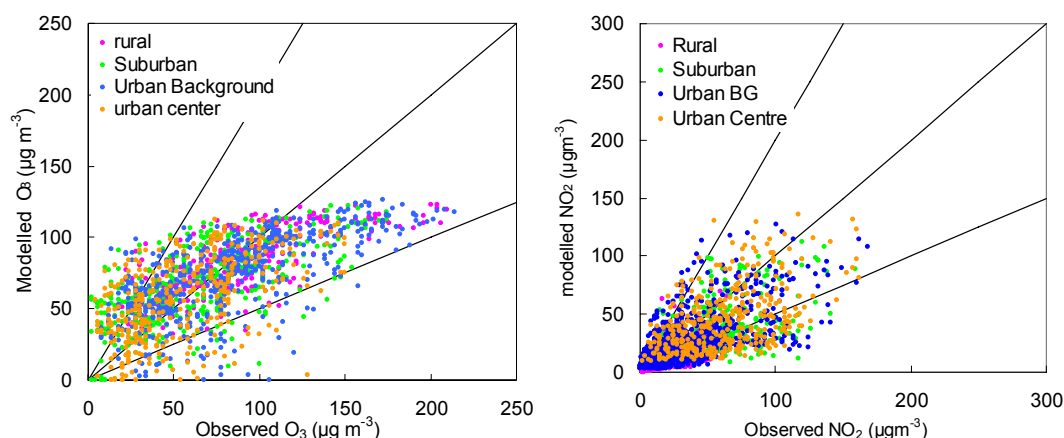


Figure 4. 2 Measured versus modelled O₃ (left) and NO₂ (right) concentration for all data pairs.

Table 4. 1 Quantitative performance statistics for near surface O₃ predictions with both 9-km and 3-km resolution for the innermost domain, i.e. domain 4.

| Statistics (µg m ⁻³) | All sites | | rural | | Sub urban | | Urban BG | | Urban centre | |
|-------------------------------------|-----------|-------|-------|-------|-----------|------|----------|-------|--------------|------|
| | 3km | 9km | 3km | 9km | 3km | 9km | 3km | 9km | 3km | 9km |
| O ₀ | 73.3 | | 88 | | 64.6 | | 81.8 | | 56.5 | |
| M ₀ | 69.6 | 72.4 | 82.8 | 81.7 | 66.3 | 71.7 | 73 | 74.3 | 57 | 62.7 |
| FB | -0.05 | -0.01 | -0.06 | -0.07 | 0.03 | 0.1 | -0.11 | -0.09 | 0.01 | 0.1 |
| R | 0.69 | 0.66 | 0.76 | 0.73 | 0.65 | 0.63 | 0.69 | 0.69 | 0.56 | 0.53 |
| RMSE | 31.2 | 31.9 | 28.7 | 29.8 | 30.6 | 31.7 | 33.3 | 33.0 | 30 | 31.7 |
| NSD | 1.51 | 1.58 | 1.8 | 1.8 | 1.47 | 1.54 | 1.64 | 1.65 | 1.21 | 1.24 |
| Fraction of 2 (%) | 81.8 | 80.3 | 94.2 | 92.4 | 75.9 | 75.3 | 84.3 | 84.4 | 73.2 | 68.2 |

Table 4. 2 Quantitative performance statistics for near surface NO₂ predictions with both 9-km and 3-km resolution for the innermost domain, i.e. domain 4.

| Statistics (µg m ⁻³) | All sites | | rural | | Sub urban | | Urban BG | | Urban centre | |
|-------------------------------------|-----------|-------|-------|-------|-----------|-------|----------|-------|--------------|-------|
| | 3km | 9km | 3km | 9km | 3km | 9km | 3km | 9km | 3km | 9km |
| O ₀ | 39.8 | | 17.8 | | 50.2 | | 34.1 | | 61.9 | |
| M ₀ | 28.0 | 25.3 | 10.0 | 12.8 | 32.8 | 28.1 | 25.2 | 22.5 | 44.6 | 39.5 |
| FB | -0.56 | -0.64 | -0.56 | -0.32 | -0.42 | -0.56 | -0.3 | -0.41 | -0.32 | -0.44 |
| R | 0.68 | 0.63 | 0.54 | 0.52 | 0.62 | 0.56 | 0.66 | 0.61 | 0.67 | 0.64 |
| RMSE | 25.9 | 28.4 | 14.1 | 12.9 | 30.1 | 33.7 | 23.1 | 25.3 | 34.5 | 37.6 |
| NSD | 1.31 | 1.4 | 1.25 | 1.26 | 1.3 | 1.52 | 1.36 | 1.34 | 1.2 | 1.24 |
| Fraction of 2 (%) | 59.7 | 54.4 | 40.9 | 55.7 | 62.9 | 54.9 | 62.4 | 51.8 | 63.0 | 59.3 |

On average, the model slightly under-predicts O₃ concentrations with a FB of -0.05 and -0.01 for 3 km and 9 km resolution, respectively. In total, 81.8 % of all modelled O₃ concentrations are within a factor of 2 of the corresponding measured concentrations. The 9-km and the 3-km resolution simulations give comparable model performance for most of the stations but slightly better performance is achieved by the 3-km resolution simulation at sub-urban and urban centre stations, where pollutants may have significant chemical gradients. When looking at R and RMSE, one may conclude that the model performs better for rural area than for urban areas. However, it should be treated with caution as the statistics depend on the availability and distribution of monitoring stations.

Simulations for NO₂ tend to show much larger bias and error than the similar statistics for O₃. Overall the model significantly under-predict NO₂ concentrations with a FB of -0.56 and -0.64 for 3 km and 9 km resolution, respectively. Over all the sites, 59.7 % of modelled concentrations are within a factor of 2 of the corresponding measured values. In general the 3-km resolution gives better predictions than the 9-km resolution simulation, especially for urban areas but the improvement on model performance by using 3-km instead of 9-km grid spacing is not very significant.

While high-resolution modelling has scientific merit and potential uses, the current availability of monitoring networks, the lack of highly resolved spatial and temporal input data and the limitations of model formulation (e.g. urban area representation in meteorological models), do not allow us to draw a firm conclusion on the superiority of the high-resolution model predictions. More research is needed to examine the proper model resolution for different applications.

The above analyses indicate that CMAQ model was able to reproduce the observed temporal and spatial variations of O₃ and NO₂. Statistic parameters indicate satisfactory overall model performance for O₃ and NO₂ with better performance for O₃ predictions. However, when CMAQ is used as a regulatory tool, caution should be taken as it may miss highest O₃ episodes.

4.2 Contribution of industrial point source emissions to ambient pollution levels

The contribution of point source emissions to ambient pollution levels presented in this section is defined as the difference between base case simulation (Run A) and the sensitivity simulation (run B), i.e. $C_A - C_B$, where C is the concentration of O₃ or NO₂ at the first model level. All the results shown in this subsection are from the 9-km domain, i.e. domain 3 in Figure 3.1, unless otherwise stated. The contributions from all the UK point source emissions to near surface concentrations of two main pollutants, i.e. O₃ and NO₂, are investigated using the difference fields of the daily maximum 8-h running mean O₃ concentration and the daily maximum 1-hr NO₂ concentration.

Figures 4.3 and 4.4 show the contribution from all the UK point sources to the daily maximum 8-hr running mean O₃ and the daily maximum 1-hr NO₂ concentration, respectively, on two representative days, i.e. on episode day and after episode day. It can be seen from these figures that the extent and magnitude of the contribution are different on different simulation days. There is a tendency for point sources to have more noticeable effect on surface O₃ concentrations on episode day, for example 25 – 26 June 2001. However, overall point source emissions are not a significant contributor to surface ozone concentrations (see also Figure 4.5), with a maximum domain-wide averaged contribution of 0.49 ppb on 25 June, accounting for about 1% of the domain-wide averaged ground level O₃ concentration on that day.

It is noted that negative contributions occur in some areas in North and southeast England, indicating O₃ is suppressed when adding pollution to those areas. One common characteristic of these areas is that there are large anthropogenic NO_x emissions and the ratios of total VOC to total NO_x are very low (less than 4), suggesting a VOC-limited regime for O₃ production. Adding point source emissions to those already polluted areas, e.g. Greater London and Greater Manchester, results in even lower VOC to NO_x ratios. Under this condition, NO_x serves as sink of OH, a key radical to oxidize VOCs, and

reacts with peroxy radicals to generate peroxy-nitrate (e.g., PAN). Both of these effects act to limit ozone build-up.

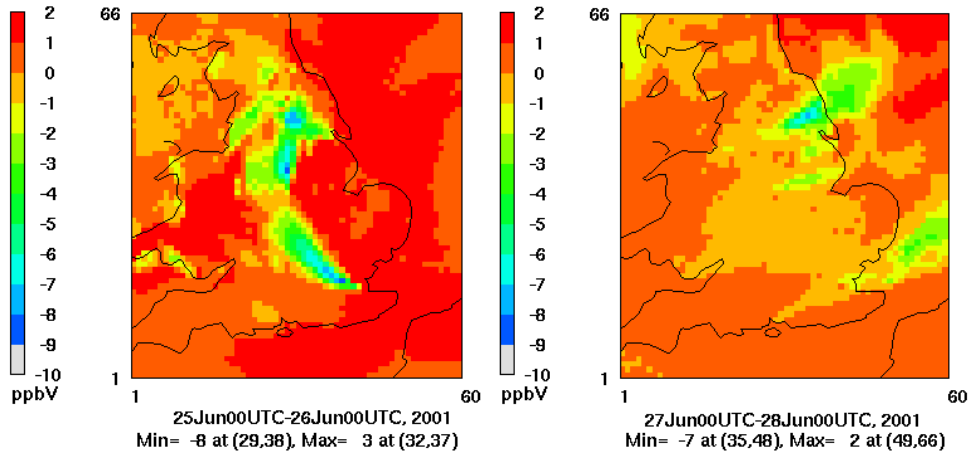


Figure 4.3 Difference field of daily maximum 8-hr running mean O₃ concentration on episode day (left) and after the episode day (right).

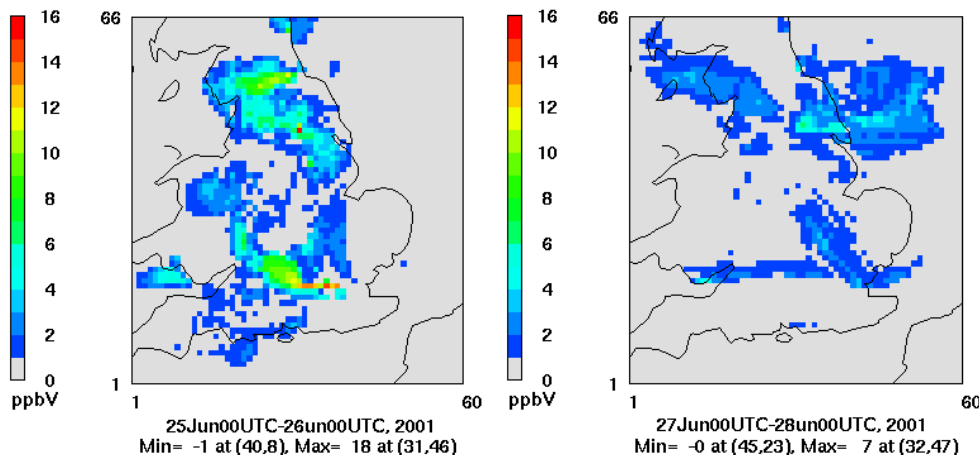


Figure 4.4 Difference field of daily maximum 1-hr NO₂ concentration on episode day (left) and after the episode day (right).

Point source emissions are a positive contributor to surface NO₂ concentrations in the domain studied as seen from Figure 4.4 and Figure 4.5. It is obvious from Figure 4.4 that the contribution from point source emissions to the ground level NO₂ concentrations was greater on high pollution days, e.g. 25 – 26 June 2001. Point source emissions account for about 7.7% of the domain-wide averaged daily maximum 1-hr NO₂ concentrations. Geographically, point source emissions have the greatest influence in areas where large power plants are located, for example the Trent Valley, Northeast of England.

Figure 4.5 summarises the percentage contributions from point source emissions to ground level O₃ and NO₂ concentrations estimated for the finest domain, i.e. domain 4, based on results from 9-km and 3-km resolution simulations. All the values shown in Figure 4.5 are based on averages over the innermost model domain. On average, point sources contribute about 7.1% and 8.6% to surface NO₂ concentrations for 9-km and 3-km resolution simulation, respectively.

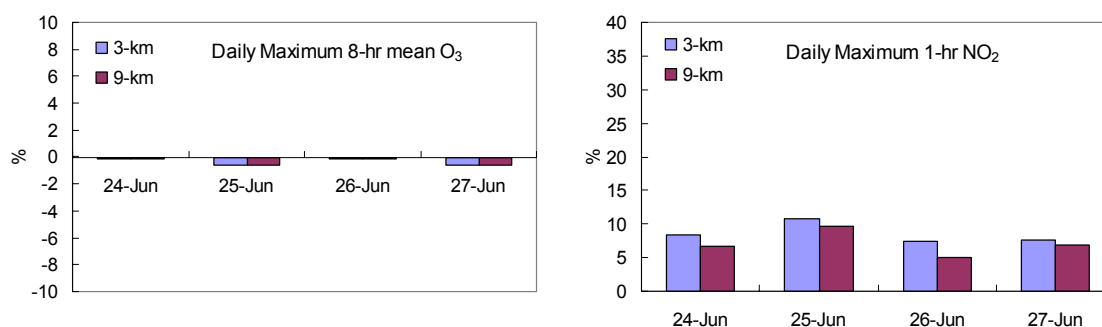


Figure 4.5 Percentage contribution of point source emissions to near surface O₃ (left) and NO₂ (right) concentrations for the innermost model domain for 9-km and 3-km resolution simulations. The dates shown in the figure are the start date for the calculation of the daily maximum 8-hr running mean or daily maximum 1-hr concentrations.

The overall contribution of point source emissions to ground level O₃ is very low and negative (showing ozone titration) on most of the simulation days, indicating VOC-limited regime for O₃ production in the studied domain during the simulation period. Further studies are needed to examine the possible effect of UK point source emissions on areas outside of the UK through, e.g. long-range transport. There is no significant difference between 3-km and 9-km resolution simulations. Considering the model's tendency to underpredict NO₂ concentrations, these percentage values can be considered conservative for regulatory use.

5 Conclusion and discussion

5.1 Assessment of the case study

This case study draws the attention on the importance of model evaluation before its regulatory use. Different statistical parameters recommended in an earlier Air4EU Cross Cutting Issue report (M6.5) were used to evaluate model performance. Overall CMAQ model is able to reproduce the observed temporal and spatial variations of O₃ and NO₂. Statistic parameters indicate satisfactory overall model performance. For most stations the model performs better for ozone than for primary pollutant such as NO₂.

This case study also investigated how model resolution can affect model performance and the results on the contribution of point source emissions to near surface pollutant concentrations. In the case of O₃ it is found that the 9-km and the 3-km resolution simulations give comparable model performance. For NO₂, generally the 3-km resolution gives better predictions than the 9-km resolution. However no significant difference was found in the calculated contribution of point source emissions to near surface O₃ and NO₂ concentrations by using different model resolutions.

These results can serve as guidance when models of the similar type are to be used for regulatory purposes.

5.2 Improvements in assessment derived from case study

This case study demonstrated the use of more advanced models, i.e. the Models-3/CMAQ, for the evaluation of stationary point source contributions to near surface pollution levels. By including interactions between different pollutants from different source categories, the case study has brought improvements in the following areas:

1. Give information on both spatial distribution and temporal variation of point source contributions to near surface pollutant concentrations. This information will help end users to identify the most affected area and the most important pollutant to regulate.
2. The effect of model resolution on the assessment has been discussed. It is suggested that for the area and resolution considered (i.e. 9-km vs. 3-km) the uncertainty resulting from using coarser resolution is not significant.

5.3 Recommendations resulting from the case study

The following recommendations for assessing stationary point source contributions to near surface pollution levels using advanced 3-D models, such as Models-3.CMAQ, can be identified.

1. When an assessment involves complex interactions among multi-pollutant and multi-scales more advanced models should be used. 3-D Eulerian air quality models, such as CMAQ, are able to give valuable information on magnitude of point source contributions, and their spatial distribution. This information can help end users to identify the most affected area and the most important pollutant to regulate.
2. When a model is applied to a new study area, a thorough evaluation should be performed and model performance should be documented. Both qualitative and quantitative methods should be used to get a full picture of model performance before its regulatory use.
3. Model resolution needed in an assessment depends on the application and pollutants concerned. Because of the lack of highly resolved spatial and temporal input data and the limitations of model formulation, more research is needed to examine the proper model resolution for different applications.
4. Based upon the results from this study, future studies may consider using results from 9-km modelling to assess stationary point source contributions to near surface O₃ and NO₂ concentrations and then incorporate grid-resolution uncertainties into the computed results for NO₂ for areas, such as urban area, where model results tend to be more sensitive to model resolution.

5.4 Suitability for implementation in other cities

The method used in this case study can be easily repeated for other areas. Users can select their own domain of interest and carry out the analysis for a different area. The method can be easily extended to evaluate other emission sources, e.g. biogenic, transport, and domestic heat, and to examine emissions from individual stacks or pollutants. End users can use the information presented in this paper to aid in their decision as to whether or not a model application is acceptable for regulatory purposes.

The method presented here also allows considering future changes of climate and its effect on landcover change and biogenic emissions.

References

Byun D W, Ching J K S , 1999 Science algorithms of the EPA Models-3 Community Multiscale Air Quality (CMAQ) Modeling System. EPA Report No. EPA-600/R-99/030.

Carolina Environmental Programs, 2003. Sparse Matrix Operator Kernel Emission (SMOKE) Modeling System. University of Carolina, Carolina Environmental Programs, Research Triangle Park, NC.

- Carter W, 2000 Documentation of the SAPRC-99 chemical mechanism for voc reactivity assessment. Final report to California Air Resources Board, contract no. 92-329, University of California-Riverside, 8 May.
- Chang, J.C. and Hanna, S.R., 2004. Air quality model performance evaluation. *Meteo. Atmos. Phys.* 87, 167-196.
- Collins W J, 1997, Tropospheric ozone in a global-scale three-dimensional Lagrangian model and its response to NO_x emission controls. *Journal of Atmospheric Chemistry*, 26 (3), 223-274.
- Dennis R L, Byun D W, Novak J H, Galluppi K J, Coats C J, Vouk M A, 1996 The next generation of integrated air quality models: EPA's Models-3. *Atmospheric Environment*, 30, 1925–1938.
- DETR et al., 2000 Department of the Environment, Transport and the Regions, The Scottish Executive, The National Assembly for Wales and The Department of the Environment in Northern Ireland. The Air Quality Strategy for England, Scotland, Wales and Northern Ireland January 2000. <http://www.defra.gov.uk/environment/airquality/strategy/index.htm>
- Gery M W, Whitten G Z, Killus J P and Dodge M C, 1994 A photochemical kinetic mechanism for urban and regional scale computer modeling. *Journal of Geophysical Research*, 94, 12925–12956.
- Gothenburg Protocol, 1999 http://www.unece.org/env/lrtap/multi_h1.htm.
- Grell G A, Dudhia J, Stauffer D R, 1994 A description of the Fifth-Generation Penn State/NCAR Mesoscale Model (MM5). NCAR Technical Note, NCAR/TN-398+STR, National Center for Atmospheric Research, Boulder, CO.
- Guenther A, Hewitt C N, Erickson D, Fall R, Geron C, Graedel T, Harley P, Klinger L, Lerdau M, McKay W A, Pierce T, Scholes B, Steinbrecher R, Tallamraju R, Taylor J and Zimmerman P, 1995 A global model of natural volatile organic compound emissions. *Journal of Geophysical Research*, 100 (D5), 8873-8892.
- Hanna S R, 1994: Mesoscale meteorological model evaluation techniques with emphasis on needs of air quality models. In Pielke R A, Pearce R P (eds), *Mesoscale modeling of the atmosphere*. Meteorological Monographs, Vol 25. American Meteorological Society, Boston, MA 02108, pp 47-58.
- M 6.5, Cross-Cutting 2: Uncertainties of Models & Monitoring, Air4EU Milestone Report 6.5.
- McNally D. and Tesche T W, 1993: MAPS sample products. *Alpine Geophysics*, 16225 W. 74th Dr., Golden, CO 80403.
- Sanderson M G, 2002 Emission of Isoprene, Monoterpenes, Ethene and Propene by Vegetation, Hadley Centre technical note 40, UK Meteorological Office.
- Stockwell W R, Middleton P and Chang J S, 1990) The second generation regional acid deposition model chemical mechanism for regional air quality modeling. *J. Geophys. Res.* 95(d10), 16,343-16,367.
- Vestreng V, Breivik K, Adams M, Wagener A, Goodwin J, Rozovskaya O and Pacyna J M, 2005 Inventory Review 2005, Emission Data reported to LRTAP Convention and NEC Directive, Initial review of HMs and POPs. Technical report MSC-W 1/2005, ISSN 0804-2446.