

Evidence

Comparison of simple and advanced regional models (CREMO)

Chemical mechanism choice: Impacts of ozone precursor emission reductions in the United Kingdom on episodic peak ozone

Report – SC060037/e

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Miranda Kavanagh
Director of Evidence

Executive summary

Ozone is a secondary pollutant formed in the lower atmosphere from the sunlight-initiated oxidation of volatile organic compounds (VOCs) in the presence of oxides of nitrogen (NO_x). Exposure to ground-level ozone has effects on human health, crops and vegetation, and materials such as rubber, paints and plastic. Air quality standards have been established to mitigate effects on human health and vegetation, but these standards are widely exceeded across the UK and Europe.

UK measurements of ambient ozone concentrations show substantial variability from hour to hour, day to day and from season to season. In addition, the three major factors controlling ozone concentrations operate at different spatial scales. Chemical mechanism choice has been examined within an uncertainty analysis framework. Allowance can be made for likely systematic biases in model input data by rerunning calculations many times. Six widely used and accepted chemical mechanisms produce little difference in policy-relevant model output results across the UK, suggesting that choice of chemical mechanism is not critical in policy-relevant ozone model evaluations.

This report is part of a study undertaken within the CREMO project. Overall outcomes for the Environment Agency are presented in

FISHER, B.E.A., 2012. *Comparison of simple and advanced regional models (CREMO): Outcomes for the Environment Agency*. Report SC060037/R. Bristol: Environment Agency.

The study of chemical mechanisms is especially relevant to the two studies within CREMO relating to ozone:

HAYMAN, G., SOKHI, R., CHEMEL, C., GRIFFITHS, S., VINCENT, K., DORE, A.J., SUTTON, P. and WRIGHT, R., 2012. *Comparison of simple and advanced regional models (CREMO): Ozone diagnostics*. Report SC060037c/R. Bristol: Environment Agency.

HAYMAN, G., SOKHI, R., CHEMEL, C., GRIFFITHS, S., VINCENT, K., DORE, A.J., SUTTON, P. and WRIGHT, D.R., 2012. *Comparison of simple and advanced regional models (CREMO): Model evaluation: Ground-level ozone*. Report SC060037d/R. Bristol: Environment Agency.

Contents

1	Introduction	6
2	Ground-level ozone	7
2.1	Assessment of six chemical mechanisms against observations for July 2006	9
2.2	Ozone responses to 30% reductions in VOC and NO _x emissions across the UK and Europe	10
2.3	NO _x versus VOC limitation for UK ozone precursor sources	14
3	Conclusions	16

1 Introduction

Model calculations with the Photochemical Trajectory Model PTM have generally shown that ozone precursor emission reductions in the United Kingdom alone have a much reduced impact on UK episodic peak ozone levels compared with the same percentage emission reductions carried out across Europe as a whole. In this study, this result is tested with a range of chemical mechanisms to see whether it is robust or whether it is a consequence of chemical mechanism choice.

Chemical mechanism choice is a difficult policy issue for regulators. If model results for certain policy-relevant outcomes turn out to be chemical mechanism dependent, how are they to decide which set of results are correct? This is a particular issue when policy formulation is based on large, sophisticated 3-D Eulerian air quality grid models with their enormous requirements for computing resources for input data preparation and for running. These resource requirements generally mean that it has not been possible to explore the sensitivity of policy outcomes to the full uncertainty in model input parameters, chemical mechanism choice included.

Here we use the PTM model to examine the impacts of ozone precursor emission reductions in the UK on episodic peak ozone levels in the UK. This study represents an extension of previous work which examined the impacts of ozone precursor emission reductions across the UK and the rest of Europe. In this previous work, Monte Carlo sampling was used to generate a large number of input data sets which sampled the full uncertainty ranges for all the parameters in the PTM model. Broadly speaking, this work showed that when the full uncertainty range in model input is taken into account, there was little to choose between the chemical mechanisms for their representation of the impacts of 30% NO_x and 30% VOC across-the-board emission reductions on episodic peak ozone levels in the UK.

Whilst our previous study was relevant to the Defra interest in developing a modelling capacity for their air quality policy development, it is not of immediate direct interest to the Environment Agency who is responsible for the regulation of large, industrial air pollution sources in England and Wales. Hence, here we have focussed on 30 per cent NO_x and 30 per cent VOC reductions in the UK only and their impact on episodic peak ozone levels in the UK using the same Monte Carlo sampling methodology.

2 Ground-level ozone

In principle, it should be a straightforward task to compare a set of chemical mechanisms with respect to their suitability for representing accurately photochemical ozone formation. In an ideal world, a simple or a complex photochemical air quality model would be configured to run each candidate mechanism and the version that most accurately predicted ozone levels compared with observations would be chosen. However there is no guarantee that the mechanism that gives the most accurate prediction of base case ozone levels would give the most accurate prediction of policy responses to ozone precursor emission reductions, say of VOC or NO_x. There is also the difficulty associated with configuring complex models to run with all the different chemical mechanisms because of the large amount of computer resources required in running them, particularly with the more complex mechanisms, such as the CRI mechanism. Finally, any model would need to be supplied with a large amount of 'best estimate' meteorological and emissions data, in addition to the chemical mechanism, for whatever time period over which any comparison would need to be carried out. Although there are such 'best estimate' input data covering emissions and meteorological data, we have no way of guaranteeing that these 'best estimate' input data are not free from systematic biases. Each chemical mechanism has its own traits and peculiarities which could make its performance against observations especially favourable or unfavourable under certain circumstances depending on the presence of any systematic biases in the 'best estimate' input data. We need to be sure that the chosen chemical mechanism is performing correctly for the correct reasons. The cancellation of errors could give good performance if for example relatively unreactive emissions were compensated by an over-reactive chemical mechanism and vice versa.

The selection of chemical mechanism has therefore been performed within a Monte Carlo uncertainty framework rather than using single 'best estimate' input data. It has also been performed using the PTM model because this is the only European policy model with the capacity to handle all the candidate chemical mechanisms, independent of their complexity. The six candidate chemical mechanisms included: CB05, CBM-4, OSRM, SAPRC-99, SAPRC-07 and the CRI mechanisms, the latter acted as the 'gold standard' mechanism. The assessment has been completed using July 2006 as test period, a period that contains elevated ozone levels above 100 ppb at the Harwell, Oxfordshire AURN monitoring station. Uncertainty ranges were assigned subjectively to all elements of input data to the PTM model, see Table 2.1, and these uncertainty ranges were randomly sampled to provide tens of thousands of sets of model input parameters.

Table 2.1 Representation of the uncertainties in the PTM model input parameters in the Monte Carlo study of parametric uncertainties^{a,b,c}

Input parameter	Representation	Range
CO emissions	multiplicative scaling	x 0.5 – 2.0
CH ₄ emissions	multiplicative scaling	x 0.5 – 2.0
C ₅ H ₈ emissions	multiplicative scaling	x 0.25 – 4.0
NH ₃ emissions	multiplicative scaling	x 0.5 – 2.0
NO _x emissions	multiplicative scaling	x 0.5 – 2.0

Comparison of simple and advanced regional models

Chemical mechanism choice

SO ₂ emissions	multiplicative scaling	x 0.5 – 2.0
VOC emissions	multiplicative scaling	x 0.5 – 2.0
VOC speciation	multiplicative scaling	x 0.5 – 2.0
Air parcel longitude	additive	± 0 – 0.45 °
Air parcel latitude	additive	± 0 – 0.28 °
Boundary conditions	multiplicative scaling	
Boundary layer depth	multiplicative scaling	x 0.5 – 2.0
Choice of mechanism	random	
Choice of trajectory	random	
Dry deposition velocity	multiplicative scaling	x 0 – 1.0
Photolysis rate coefficient	multiplicative scaling	x 0.7 – 1.3
Rate coefficient	multiplicative scaling	x 0.7 – 1.3
Relative humidity	multiplicative scaling	x 0.5 – 2.0
Temperature	additive	± 0 – 3 °C

Notes:

^a all assignments in this table are subjective;

^b a scaling factor of unity represents 'best estimate' model input;

^c uncertainties in longitudes and latitudes have 'Gaussian' shapes, the remainder have 'top hat'.

The six chemical mechanisms have been harmonised in such a way as to facilitate comparison and evaluation. This means that our evaluation does not address the actual chemical mechanism produced by the mechanism developers. Our study addresses a harmonised version of each chemical mechanism produced by the implementation of the four steps detailed below. For simplicity, each chemical mechanism is still referred to by its original name and it is implicitly understood that reference is being made to the harmonised version. The following steps were taken to harmonise each chemical mechanism:

Step 1: the fast photochemical reactions involving O³P, O¹D, OH, H, HO₂, NO₃ with N₂, O₂, H₂O, CO, O₃, NO, NO₂, HNO₃, HO₂NO₂, N₂O₅, SO₂, sulphate and nitrate aerosol, (the so-called inorganic reactions), were replaced by a common set of 47 reactions, of which 35 were thermal reactions, 8 were photochemical and 4 were aerosol formation reactions as specified in MCMv3.1.

Step 2: all complex temperature, pressure and humidity dependent rate coefficients were replaced by a common set of 17 rate coefficients as specified in MCMv3.1.

Step 3: the formation and thermal decomposition of all PAN-type molecules were replaced by a common pair of temperature and pressure-dependent reaction rate coefficients as specified in MCMv3.1.

Step 4: all photolysis rate coefficients were replaced by a common set as specified in MCMv3.1.

As a result of implementing the above harmonisation steps, the chemical mechanisms have been distorted significantly away from the actual versions produced by the mechanism developers. Our aim has been to understand the likely impact on ozone air quality policy development that has resulted from the different techniques and parameterisations that have been applied by the mechanism developers to synthesise and simplify understanding of the basic atmospheric chemistry involved in the formation of O₃ from the oxidation of the emitted VOCs in the presence of NO_x. Our results may well have been different if we had adopted an 'as is' implementation of the chemical mechanisms compared with our 'harmonised' approach. Equally well, they would have been different if we had adjusted the mechanisms to rematch smog chamber data, following the adjustments made to the fast photochemical reactions.

Each randomly-generated parameter set was then run through the PTM and the model predictions for Harwell, Oxfordshire were compared with the observations for each day of July 2006. If the comparison was found 'acceptable', the parameter set was retained for further use as a base case. If not, the parameter set was rejected and no further use was made of it. A total of 186,000 parameter sets were randomly selected and evaluated against observations, of which about 30% were found 'acceptable'. Those parameter sets that were found 'acceptable' were then reused with 30% reductions in NO_x and VOC emissions applied and the scenario results were paired up with the corresponding base case results. In this way, the single 'best estimate' results were replaced by a distribution of results based on Monte Carlo sampling. Each set of results could be ranked in ascending order and various percentiles estimated representing the distribution of predictions about a central or median value.

2.1 Assessment of six chemical mechanisms against observations for July 2006

Figure 2.1 presents the PTM model predicted 50 percentiles of the 'acceptable' 15:00z ozone concentrations for each day of July 2006 with each of the six chemical mechanisms together with the mid-afternoon AURN observations for the Harwell, Oxfordshire site. The PTM model was well able to account for the pattern of elevated and baseline levels throughout the month and the day-to-day variability as shown by the observations, independent of the choice of chemical mechanism. Over the entire month, model performance is excellent as shown by the mean fractional biases which were in the range -0.04 – -0.09, well below the Defra target of ± 0.2 , for the 50 percentile concentrations. Note that all six mechanisms slightly underestimate the observations as illustrated in Figure 2.1.

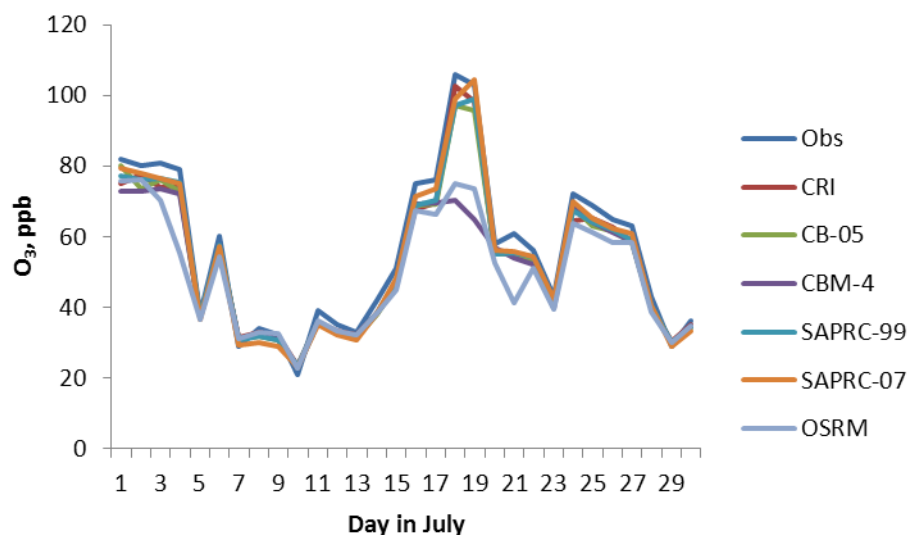


Figure 2.1 Model predicted mid-afternoon ozone concentrations using six different chemical mechanisms and the AURN observations for Harwell, Oxfordshire for each day of July 2006.

The only point of departure between the six chemical mechanisms was shown by the model performance on the highest ozone days: 18th and 19th July 2006. The CBM-4 and OSRM mechanisms were unable to account for the episodic peak ozone concentrations which were over 100 ppb on these two days, by margins of over 30 ppb. The remaining four chemical mechanisms also underestimated the peak concentrations but by margins that were significantly smaller, 2 – 9 ppb, compared with those found with the CBM-4 and OSRM mechanisms. Of all of the mechanisms considered, model performance was best for the CRI mechanism across the whole month and for 18th July. Having said that, model performance was only marginally better for the CRI mechanism compared with that for the CB-05, SAPRC-99 and SAPRC-07 mechanisms. Model performance on July 19th was best for the SAPRC-99 and SAPRC-07 mechanisms. Model performance was poorest for the CBM-4 mechanism for all the metrics studied.

2.2 Ozone responses to 30% reductions in VOC and NOx emissions across the UK and Europe

In the next phase of the assessment, attention is turned to the model responses to reductions in the emissions of the ozone precursors: NOx and VOCs. Each of the model runs with the 'acceptable' parameter sets was repeated with a 30% reduction in NOx emissions and then a 30% reduction in VOC emissions. These emission reductions were applied only to man-made sources and were applied across-the-board, that is to say, equally to all sources and countries. They were applied to UK sources alone and to all sources across UK and Europe. The 'acceptable' runs were paired up between the base case and emission reduction cases so that for each run, distributions were obtained of the model responses, defined as $[O_3]_{\text{base case}} - [O_3]_{\text{scenario case}}$. These ozone responses are defined so that any decrease in ozone relative to the base case appears as a positive quantity and any increase in ozone appears as a negative quantity. If the model response to the 30% reduction in VOC emissions is greater than

the reduction due to 30% reduction in NO_x emissions, then that model run is defined as VOC-limited and vice versa.

Figure 2.2 presents a scatter plot of the 50 percentile O₃ responses on each day of July 2006 showing the responses to 30% VOC reductions carried out across the UK alone versus 30% VOC reductions carried out across the UK and the rest of Europe. The plotted points cover 6 chemical mechanisms and the 31 days of July 2006. Each mechanism generated a set of points on the 1:1 line and a set of points that fell below this line. The points on the 1:1 line imply that the 50%-ile O₃ responses to the VOC reductions were identical for the UK alone and for UK and the rest of Europe. On these days, therefore, the UK was the only important source of VOC precursors and so VOC reductions across the rest of Europe made no significant difference to the O₃ levels. These days included 24th July, 27th July, 22nd July and 23rd July. Back trajectories confirmed that on these days, the air mass trajectories did not pass over any significant pollution sources in the rest of Europe and that the only significant precursor sources were in the UK. Each chemical mechanism showed this behaviour in equal measure for exactly the same days.

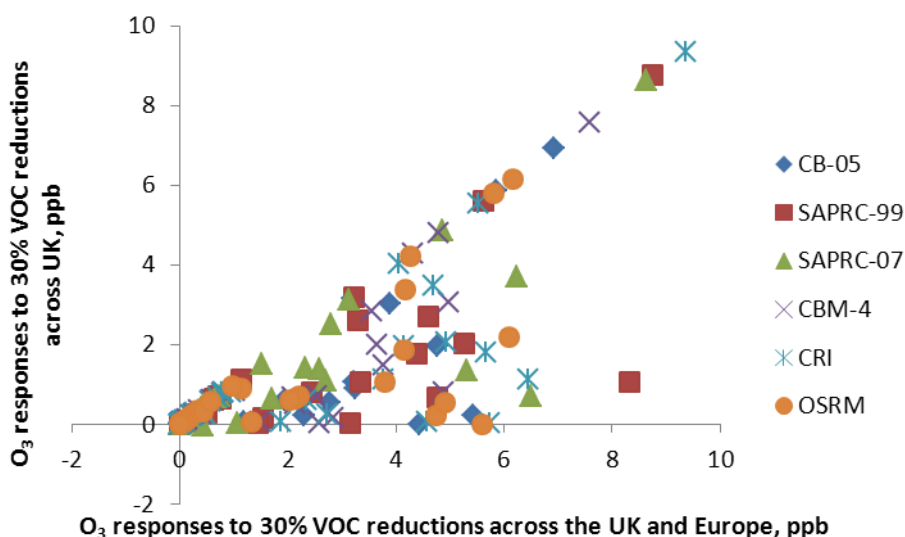


Figure 2.2 Scatter plot of the 50%-ile O₃ responses to 30% VOC reductions across the UK versus those to 30% reductions across the UK and the rest of Europe for 31 days and 6 chemical mechanisms

Table 2.2 50 percentile O₃ responses using 6 chemical mechanisms to 30% VOC reductions across the UK on 4 days during July 2006 when 30% VOC reductions across the rest of Europe made no significant impact

Date	Observed O ₃ , ppb	CB-05	CBM-4	SAPRC-99	SAPRC-07	CRI	OSRM
24 th	69	6.92	7.59	8.75	8.64	9.33	5.80
27 th	43	5.85	4.78	5.61	4.86	5.53	6.17
22 nd	43	3.15	4.29	3.18	3.12	4.01	4.22
23 rd	72	0.68	0.81	0.64	0.36	0.82	0.57

Table 2.2 provides a detailed comparison of the 50 percentile O₃ responses to 30% VOC reductions across the UK for the six chemical mechanisms for those days when the UK was the only significant source of VOC precursors. The table entries have been ranked in order of the highest UK contribution according to the CRI chemical mechanism. Also shown is the observed mid-afternoon hourly maximum O₃ level on each day.

The OSRM mechanism, in agreement with the 5 other mechanisms, was able to confirm that there were four days when the 50 percentile O₃ responses to 30% VOC reductions across the UK were significantly greater than those to 30% VOC reductions across the rest of Europe. The OSRM responses on two of the four days that were well within the range indicated by the other five mechanisms. However on the other two days 24th and 27th July, the OSRM O₃ responses fell slightly outside of the range indicated by the other five mechanisms.

Consideration of the complete uncertainty distributions of the O₃ responses for the 24th July presented in Figure 2.3, shows that the differences in 50 percentile response exhibited by the OSRM mechanism from those given by the other five mechanisms are not statistically significant. An examination of Figure 2.3 reveals that the interquartile uncertainty ranges for all six mechanisms overlap each other and there is no evidence to think that the OSRM mechanism is statistically different from any of the others.

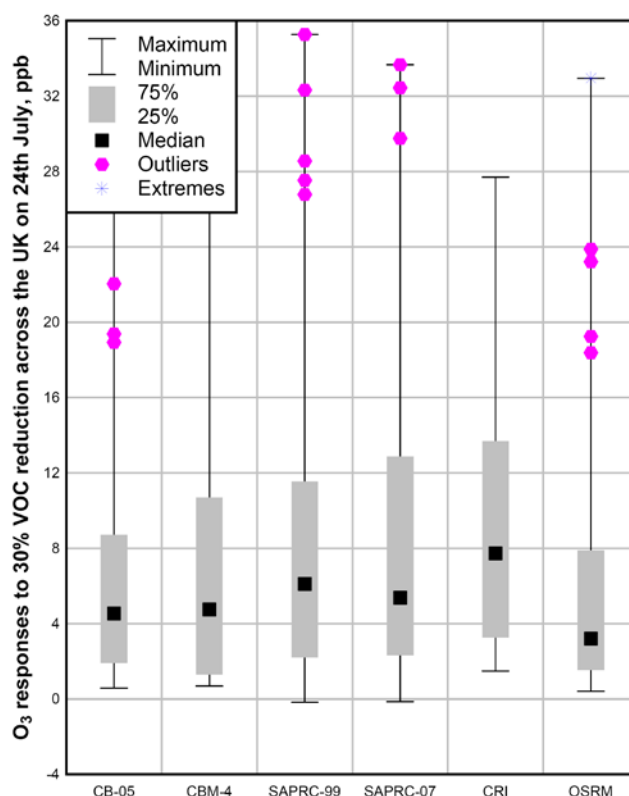


Figure 2.3. Box-and-whisker plots of the O₃ responses to 30% VOC reductions across the UK predicted for 24th July 2006 using the six chemical mechanisms. Key: 50 percentile responses are shown as black squares, interquartile ranges as grey rectangles, filled hexagons as outliers, stars as extremes and error bars covering the maxima and minima.

The remainder of the points in Figure 2.2 that fell away from the 1:1 line represent those days when the ozone responses to 30% VOC reductions across UK and the rest of Europe were greater than the responses to 30% VOC reductions across the UK alone. That is to say, there was a contribution to ozone from sources in the rest of Europe in addition to that from the UK alone. This was the situation on the days with the highest episodic peak ozone levels, July 18th and 19th. On those days with the highest episodic peak ozone levels, the responses to 30% VOC reductions across the rest of Europe were significantly greater than those to 30% reductions across the UK alone. 50 percentile O₃ responses to 30% VOC reductions in the rest of Europe were between 5.1 and 69.6 times larger than those to 30% reductions across the UK alone on the 18th July depending on chemical mechanism choice. With the OSRM mechanism, the 50 percentile O₃ responses were 8.2 times larger for the rest of Europe compared with the UK alone, well within the range of the other five mechanisms.

2.3 NO_x versus VOC limitation for UK ozone precursor sources

An important issue for policy-makers has been whether to control NO_x or VOC precursor emissions to achieve cost-effective reductions in episodic peak ozone levels. Here we examine whether 30% reductions in NO_x or VOC emissions carried out across the UK give greater reductions in episodic peak ozone levels. An air parcel can be characterised as NO_x-limited if the 50 percentile O₃ response to 30% NO_x emission reductions is greater than that generated by 30% VOC reductions and vice versa. There is an issue for policy-makers then if the different chemical mechanisms give different assignments for each day depending on chemical mechanism choice.

Table 2.3 presents the NO_x- versus VOC-limitations assigned to each day of July 2006 based on the 50 percentile O₃ responses to 30% NO_x or VOC emission reductions carried out across the UK. There is clearly significant day-to-day variability between the assignments and between the different chemical mechanisms but there are a number of common features. There was agreement on the NO_x- versus VOC- limited assignments given by the 'gold-standard' CRI mechanism and the OSRM mechanism on 25 of the 31 days. This is well within the range of 24 – 27 days agreement found between the other four mechanisms and the CRI mechanism. The OSRM gave assignments that accorded well with the other five chemical mechanisms throughout July.

Table 2.3 NO_x- versus VOC limitations assigned on the basis of 50 percentile O₃ responses

Date in July	Date in July	CB-05	CBM-4	SAPRC-99	SAPRC-07	CRI	OSRM
1 st	82	VOC	VOC	VOC	VOC	VOC	VOC
2 nd	80	NO _x	NO _x	NO _x	NO _x	NO _x	NO _x
3 rd	81	VOC	VOC	VOC	VOC	VOC	VOC
4 th	79	NO _x	VOC	VOC	VOC	VOC	NO _x
5 th	38	VOC	VOC	VOC	NO _x	VOC	VOC
6 th	60	VOC	VOC	VOC	VOC	VOC	VOC
7 th	29	NO _x	VOC	NO _x	NO _x	VOC	VOC
8 th	34	NO _x	NO _x	NO _x	NO _x	NO _x	NO _x
9 th	32	NO _x	NO _x	NO _x	NO _x	NO _x	NO _x
10 th	21	NO _x	NO _x	NO _x	NO _x	VOC	NO _x
11 th	39	NO _x	NO _x	NO _x	NO _x	NO _x	NO _x
12 th	35	VOC	VOC	VOC	NO _x	VOC	VOC

13 th	33	VOC	VOC	VOC	VOC	VOC	VOC
14 th	42	NOx	NOx	NOx	NOx	NOx	VOC
15 th	51	NOx	VOC	NOx	NOx	NOx	VOC
16 th	75	VOC	VOC	VOC	VOC	VOC	VOC
17 th	76	VOC	NOx	VOC	VOC	VOC	VOC
18 th	106	NOx	VOC	VOC	VOC	NOx	NOx
19 th	103	NOx	NOx	NOx	NOx	NOx	NOx
20 th	58	NOx	NOx	NOx	NOx	NOx	NOx
21 st	61	NOx	NOx	VOC	NOx	VOC	NOx
22 nd	56	VOC	VOC	VOC	VOC	VOC	VOC
23 rd	43	VOC	VOC	NOx	NOx	VOC	VOC
24 th	72	VOC	VOC	VOC	VOC	VOC	VOC
25 th	69	NOx	NOx	NOx	NOx	NOx	NOx
26 th	65	VOC	VOC	VOC	VOC	VOC	VOC
27 th	63	VOC	VOC	VOC	VOC	VOC	VOC
28 th	43	NOx	NOx	NOx	NOx	NOx	NOx
29 th	29	NOx	NOx	NOx	NOx	NOx	NOx
30 th	36	NOx	NOx	NOx	NOx	NOx	NOx
31 st	43	NOx	VOC	NOx	NOx	NOx	VOC

3 Conclusions

Chemical mechanism choice inside the PTM model has been examined within a Monte Carlo input parameter uncertainty analysis framework. Within this framework, maximum allowance can be made for likely systematic biases in model input data by rerunning the PTM model with thousands of randomly-generated 'acceptable' input parameter sets. There turns out to be little difference in policy-relevant model output results using any of six different chemical mechanisms: CB-05, CBM-4, SAPRC-99, SAPRC-07, CRI or OSRM. We have found no statistically-significant differences between policy-relevant outcomes generated with the OSRM mechanism compared with the other five mechanisms when assessing the impacts of NO_x or VOC precursor emission reductions carried out across the UK.

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