Predictions of UK Regulated Power Station Contributions to Regional Air Pollution and Deposition: A Model Comparison Exercise

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ABSTRACT

Contributions of the emissions from a UK regulated fossil-fuel power station to regional air pollution and deposition are estimated using four air quality modeling systems for the year 2003. The modeling systems vary in complexity and emphasis in the way they treat atmospheric and chemical processes, and include the Community Multiscale Air Quality (CMAQ) modeling system in its versions 4.6 and 4.7, a nested modeling system that combines long- and short-range impacts (referred to as TRACK-ADMS), and the Fine Resolution Atmospheric Multi-pollutant Exchange (FRAME) model. An evaluation of the baseline calculations against UK monitoring network data is performed. The CMAQ modeling system version 4.6 dataset is selected as the reference dataset for the model footprint comparison. The annual mean air concentration and total deposition footprints are summarized for each modeling system. The footprints of the power station emissions can account for a significant fraction of the local impacts for some species (e.g. more than 50% for SO_2 air concentration and non-sea-salt sulfur deposition close to the source) for 2003. We calculate the spatial correlation and the coefficient of variation of the root mean square error (CVRMSE) between each model footprint and that calculated by the CMAQ modeling system version 4.6. The correlation coefficient quantifies model agreement in terms of

spatial patterns, and the CVRMSE measures the magnitude of the difference between model footprints. Possible reasons for the differences between model results are discussed. Finally, implications and recommendations for the regulatory assessment of the impact of major industrial sources using regional air quality modeling systems are discussed in the light of results from this case study.

IMPLICATIONS

Modeling tools are required to assess the contribution of industrial sources to ambient levels of air pollution, acid deposition, and eutrophication. This study evaluates the performance characteristics of regional air quality modeling systems in predicting contributions of the emissions from a UK regulated fossil-fuel power station to regional air pollution and deposition. It contrasts acid deposition modeling approaches used in the UK and demonstrates the sensitivity of the modelling systems to large emission changes. This work suggests considering an ensemble average of model calculations to provide an estimate of the uncertainty associated with an industrial source footprint.

INTRODUCTION

Despite large reductions in terms of absolute emission levels since the 1990s, the power generation sector remains a significant contributor to pollutant emissions in the UK.[1] The pollutants emitted by power stations include sulfur dioxide (SO_2), oxides of nitrogen (NO_x), and particulate matter smaller than 10 µm in aerodynamic diameter (PM_{10}). The power generation sector contributed 48%, 24%, and 7% to the UK emissions of SO_2, NO_x, and PM_{10} respectively, in 2007.[1] These three air pollutants are associated with negative effects on human health (e.g. respiratory problems) and damage to the environment. Deposition of sulfur and nitrogen can lead to critical loads for acidity levels being exceeded in sensitive terrestrial and aquatic ecosystems. Additionally, nitrogen deposition can cause eutrophication of ecosystems. Emissions of SO_2, NO_x, and PM_{10} from power stations are regulated by the 'EC Directive 2001/80/EC on the limitation of emissions of certain pollutants into the air from large combustion plants.[2] The European Union (EU) has set air quality limit values for a range of air pollutants, which are specified by the 'EC Directive 2008/50/EC on ambient air quality and cleaner air for Europe.[3] In the UK, national air quality standards and objectives have been set to meet these legal limit values.[4] Recognizing that air pollutants cross national borders, national ceilings for emissions of key pollutants (including SO_2 and NO_x) have been put in place at the EU level as part of the Convention on Long-Range Transboundary Air Pollution, and set for 2010 in the 1999 Gothenburg Protocol. In this context, the regulatory assessment of power stations (and more generally large industrial sources) is an important factor to include in the design of a cost effective strategy to meet emission-ceiling targets and to reduce air pollution, acidification and eutrophication of ecosystems, and climate change impacts. Such an assessment requires appropriate modeling tools.

A number of air quality modeling systems have already been applied for regulatory purposes in the UK. These modeling systems include a nested modeling system (referred to as TRACK-ADMS,[5] hereafter), used for national annual audits,[5] and the Fine Resolution Atmospheric Multi-pollutant Exchange (FRAME) model,[6] used for national assessment of acid deposition.[6,7,8] Recently, the UK Environment Agency has been considering using more advanced (in the way they treat atmospheric and chemical processes) air quality modeling systems, such as the Community Multiscale Air Quality (CMAQ) modeling system,[9,10] as one of its primary regulatory assessment tools. Hence, a model comparison exercise has been setup to examine the performance characteristics of regional air quality modeling systems in relation to regulatory use, and more specifically the response of those modeling systems to large emission changes. For the purpose of this exercise, contributions of the emissions from a UK regulated fossil-fuel power station to regional air pollution and deposition are quantified using the CMAQ modeling system in its versions 4.6 and 4.7, TRACK-ADMS, and FRAME, for the year 2003.

BASELINE CALCULATIONS

Setup of the Modeling Systems

The formulations of the four air quality modeling systems (the CMAQ modeling system in its versions 4.6 and 4.7, TRACK-ADMS, and FRAME) as regards the treatment of atmospheric and chemical processes are quite different, as are the requirements in terms of input datasets (e.g. meteorology, emissions).

The CMAQ modeling system is a state-of-the-science Eulerian air quality modeling system,[11] which has been used extensively for a variety of applications (e.g. retrospective, forecasting, regulatory,[12,13] process-level applications). It can simulate the dynamics and composition of the atmosphere over a broad range of spatial and temporal scales in a consistent framework based on first-principles solutions. The setup and operational evaluation of the CMAQ modeling system, version 4.6, at a horizontal resolution of 5 km for the UK is detailed by Chemel et al.[10] The calculations performed with the CMAQ modeling system, version 4.7, have been configured to be as close as possible to those of version 4.6 (e.g. same grid coordinates, chemical schemes, meteorological fields, similar treatment of chemical initial and boundary conditions for the outer domain). Foley et al.[14] documented the major changes from version 4.6 to 4.7 and their impact on model performance characteristics.

TRACK-ADMS is a modeling system used to produce annual high-resolution maps of air concentration of SO_2, NO_x, and PM_{10}, and of deposition of non-sea-salt (nss) sulfur (SO_x) and nitrogen across the UK. It combines the Trajectory Model with Atmospheric Chemical Kinetics (TRACK),[15] for long-range impacts and the Atmospheric Dispersion Modelling System (ADMS),[16] for short-range impacts. The setup of TRACK-ADMS is as that used for national annual audits.[5] The horizontal resolution is 20 km for the long-range Lagrangian chemistry-transport model, TRACK, at distances greater than 50 km from the source, and 1 km for the short-range dispersion model, ADMS, at distances less than 50 km from the source. Model outputs are adjusted by calibration factors used either to adjust modeled values based on measurements or to account for transport and sources not directly modeled.[5] It is worth noting that TRACK-ADMS does not discriminate between oxidized nitrogen (NO_y) and reduced nitrogen (NH_x) deposition and provides only total deposition (i.e. sum of wet and dry depositions) as a standard output. The modeled wet deposition of nss sulfur is calculated as the wet deposition from long-range sources of sulfur alone. Short-range wet deposition of sulfur was assumed to be small compared with its short-range dry deposition,[17] and was not modeled. Basically, the travel time from the source is not long enough for significant oxidation of SO_2 to take place, so that wet deposition is not an effective removal process. The modeled wet deposition of nitrogen is calculated as the sum of the wet deposition from long-range sources of nitrogen and the short-range wet deposition of ammonia (NH_3). The modeled dry deposition of nitrogen is calculated in the same way but also includes the short-range dry deposition of NO_x. Dry deposition is estimated from modeled ground-level concentration assuming a constant dry deposition velocity, except for the short-range dry deposition of NH_3, which is derived on an hour-by-hour basis throughout the year with a time-varying dry deposition velocity.

FRAME is a Lagrangian chemistry-transport model used to simulate annual mean air concentrations of SO_2, NO_x, and NH_3, and depositions of nss SO_x, NO_y, and NH_x, along straight-line trajectories at a horizontal resolution of 5 km in the UK. The setup of FRAME is as that used for national assessment of acid deposition.[6] A detailed description of the original version of FRAME and its development to improve the representation of sulfur and oxidized nitrogen are given elsewhere.[18,19]

All the model grids cover the UK (see Figure 1) and model results are presented for the 'UK domain'. The horizontal resolution of the CMAQ modeling system and FRAME is 5 km. For TRACK-ADMS, it is 20 km far from the source and 1 km close to the source. Outputs of the modeling systems have been reprojected on a common grid with an effective horizontal resolution of 5 km in order to accommodate the different grids and horizontal resolutions of the models and to minimize the effects of interpolation due to the reprojection. Note that interpolating outputs of TRACK-ADMS from a horizontal resolution of 1 km to 5 km results in a smoothing effect that will not significantly change its overall performance (since it will perform either slightly better or worse depending on location). The vertical resolution is different for each modeling system. In the present study, we focus on ground surface air concentration and deposition. The assessment of the impact of vertical resolution is being considered for future work.

Figure 1 here

TRACK-ADMS and FRAME use annual mean observational data to derive the meteorological fields (incl. precipitation map, and wind frequency and wind speed roses) for the chemistry-transport model,[5,6] while the CMAQ modeling system uses outputs from a meteorological model, the Weather Research and Forecasting (WRF) model, version 3.0.1.1, nudged towards analyses in the present study.[10] A wind rose is used in FRAME to give the appropriate weighting to directional air concentration and deposition for calculation of mean air concentration and total deposition. It is not practical to harmonize input data for the meteorology in the present model comparison exercise, so each modeling system has used its own input meteorological dataset.

Chemical initial and boundary conditions were derived from calculations of larger-scale chemistry-transport models for the CMAQ modeling system[10] and FRAME calculations,[7] while TRACK-ADMS used data from remote sites to estimate the contributions of sources not directly modeled to air concentration and deposition.[5]

For an effective model comparison, input emissions datasets for the modeling systems have been kept as consistent as possible. The four air quality modeling systems used the same annual anthropogenic emissions data as that used by the CMAQ modeling system.[10] The distribution of the emissions in time was not prescribed. Since the focus of the present work is on regulated industrial sources, a detailed emission inventory for point sources in the UK including stack parameters and emissions data by source sectors is required.[20] Such a detailed emission inventory was specifically compiled for the purpose of the model comparison exercise. For the CMAQ modeling system, emissions from point sources were mixed instantaneously in the entire grid cell indentified at the level of sources plume rise. The Lagrangian plume-in-grid approach, which is implemented in the CMAQ modeling system to resolve the spatial scale of large point sources plumes,[21] was not used in the present study, as while this option is available in version 4.6, it is not supported in version 4.7. Although large point source plumes cannot be represented explicitly by Eulerian air quality modeling systems, their representation can be approximated by using fine grid spacings,[22] as is the case in our work. TRACK-ADMS and FRAME are designed to track plumes in a Lagrangian reference frame, so that there is no need to further resolve their spatial scale.

Biogenic gas emissions were included in the CMAQ modeling system and TRACK but not in ADMS and FRAME. They are important for studying regional ozone pollution, but this topic is out of the scope of the present study. Sea-salt emissions contribute to PM_{10} and sulfur deposition. They were included in the CMAQ modeling system but considered only for PM_{10} in TRACK-

ADMS and not considered at all in FRAME. In order to compare like to like, we did not consider the sea-salt contribution to sulfur deposition in our work.

Evaluation of the Modeling Systems

An evaluation of the model baseline calculations against UK monitoring network data for the year 2003 is performed in order to gain insights into the performance characteristics of each modeling system and to provide some guidance as regards the selection of a reference dataset for the model footprint comparison. Modeled air concentrations of SO_2, NO_x, and PM_{10} are compared with measurements from monitoring sites of the UK Automatic Urban and Rural Network (AURN) and those run by the major power plant companies in the UK, information from which is collated by JEP, comprising 82 and 34 sites, respectively. Airport, kerbside, roadside, urban center, and urban industrial AURN monitoring sites were excluded as being non-representative of typical background concentrations, while all remote, rural, suburban, and urban background sites were kept for the model evaluation. All the JEP monitoring sites are located in the vicinity of power stations and can be classified as rural or urban background sites. Modeled wet depositions of nss SO_x, NO_y, and NH_x are compared with observational data derived from the Secondary Acid Precipitation Monitoring Network (SAPMN), comprising 38 sites providing collection of precipitation and measurements of ion concentrations. Precipitation was collected at those sites using bulk precipitation samplers. The limitations of using this data for the evaluation of wet deposition should be discussed. Previous experience has indicated that bulk collectors do not measure precipitation very well because not all the rainwater is collected. Measurements from such bulk samplers can be tainted by the dry deposition of gas and particles on the funnel surface, which are washed into the sample and thus included in it.[23,24] Dry deposition was found to contribute around 20% for sulfate (SO_4^{2-}), 20-30% for nitrate (NO_3^-), and 20-40% for ammonium (NH_4^+) ion concentrations in the UK.[24] The dry contribution to wet deposition is not quantified for each sample and is thus part of the observational error. In addition, wet depositions derived from site-specific measurements may not be representative of grid cell averages, which may be affected by the spatial variability of rainfall amounts and ion concentrations due to orographic enhancement effects.[23,25,26] In order to examine the effects of spatial variability in rainfall amounts on wet deposition data, we also consider using the UK Met Office precipitation observations gridded at a 5-km horizontal resolution to be compared with that of the bulk collectors. A quantification of the effects of spatial variability of ion concentrations on wet deposition requires further research, which is kept in mind for future work. The spatial coverage of the monitoring networks is displayed in Figure 1, along with the type (e.g. urban, rural) of the AURN sites.

The fraction of the model predictions, within a factor of two of the observations (FO2), the correlation coefficient, and the normalized mean bias (NMB) are calculated considering all monitoring sites, for the annual mean air concentration and deposition of the measured species. A summary of the values of these statistical metrics is provided in Tables 1 to 3. Model budgets for nss sulfur and nitrogen deposition are given in Table 4. It should be noted that the year 2003 was very dry with the lowest annual precipitation of the last two decades. This resulted in lower than average wet deposition and higher dry deposition.

Table 1 here

Table 2 here

Table 3 here

Table 4 here

Model acceptance criteria for 'operational' evaluation have recently been defined in the UK.[27] It is recommended that an air quality modeling system is considered acceptable if the FO2 values are greater than 50% and if the NMB values lie within the range -20 – 20% for both air concentration and deposition. Correlation coefficients are not recommended as evaluation metrics because they can be strongly influenced by the presence of outliers when there are a small number of pair values. However, they turn out to be informative in the present study to investigate how sensitive model performance is to the derivation of wet deposition from the measurements of precipitation and ion concentrations.

It is worth noting that all the models fulfill the first criterion (namely, FO2 > 50%) for SO_2, NO_x, and PM_{10} air concentrations, and for nss SO_x, NO_y, and NH_x wet depositions (see Table 1). Using site-specific measurements of precipitation rather than the gridded UK Met Office precipitation observations leads to larger FO2 values for all the species and modeling systems considered, with the exception of NH_x wet deposition for FRAME. Interestingly, the correlation coefficients for wet deposition are increased significantly for all the modeling systems when using the precipitation collected by the bulk collectors (see Table 2). The low correlation coefficients obtained for SO_2 and PM_{10} do not indicate per se poor performance but are the result of a narrow range of concentrations and the presence of outliers for SO_2. This result suggests that the rainfall amounts collected by the bulk collectors provide a better spatial representation of what was measured at the sites than the gridded UK Met Office precipitation observations. In contrast to the first criterion, all models fail to fulfill the second criterion (namely, NMB in the range -20 – 20%, see Table 3). Indeed, the NMB for one or more of the species air concentration and/or wet deposition is outside the range for all the modeling systems. All modeling systems tend to under estimate the annual mean air concentrations of SO_2, NO_x, and PM_{10} (as indicated by a negative NMB in Table 3). The NMB values for wet deposition indicate that the precipitation collected by the bulk collectors is less than that derived using the gridded UK Met Office precipitation observations. The NMB absolute values are smaller when using the precipitation collected by the bulk collectors for the CMAQ modeling system, version 4.6. Conversely, these values are larger for FRAME. This result is to be expected since FRAME is using an annual mean precipitation map derived from the gridded UK Met Office precipitation observations. As for the CMAQ modeling system, version 4.7, no clear pattern is evident in the wet deposition results with the NMB absolute values increasing for nss SO_x and NH_x, and decreasing for NO_y.

The ranges of variation in the UK wet, dry, and total deposition budgets around the mean values calculated across the modeling systems are -33 – 19%, -40 – 21%, and -16 – 24%, respectively, for nss sulfur, and -26 – 24%, -19 – 13%, and -6 – 10%, respectively, for nitrogen (see Table 4). Overall, there appears more variability in the output of the modeling systems in terms of mass deposited in the UK for nss sulfur than for nitrogen. The annual total deposition of nss sulfur, as calculated by each modeling system for the year 2003, is presented in Figure 2. In the CMAQ modeling system precipitation is calculated explicitly by the WRF model while in the other modeling systems it is derived from an annual mean precipitation map derived from the gridded UK Met Office precipitation observations, and an enhanced washout rate is assumed over hilly areas due to the scavenging of cloud droplets by the seeder-feeder effect.[28] Interestingly, the CMAQ modeling system, version 4.7, produces 72% more nss sulfur wet deposition than version 4.6 (see Table 4), while the precipitation field is the same as in version 4.6. Foley et al.[14] incrementally evaluated the effect of the major changes from version 4.6 to 4.7 on model performance characteristics. The changes most relevant to deposition, namely those changes to the resolved cloud model and to the coarse particle treatment, were not found to have a significant impact on sulfur and nitrogen deposition when averaged across monitoring stations. We found that the difference between the two model calculations with the CMAQ modelling system, in terms of nss sulfur wet deposition, is associated with more nss sulfate aerosols formed in version 4.7 than in version 4.6, especially in Scotland. We keep this point in mind for future work. Outside of Scotland, the spatial patterns of nss sulfur total deposition from the different modeling systems are very similar with high deposition simulated over the North of England, the Midlands, the hilly areas of Wales, and the Thames Estuary.

No single modeling system among those considered in the model comparison exercise provides the overall best performance but we would emphasize that the purpose of the comparison exercise is not to identify and select the best performing modeling system.

FOOTPRINT CALCULATIONS

We have decided to select the CMAQ modeling system version 4.6 dataset as the reference dataset for the model footprint comparison. The main reasons for the selection are summarized below:

- As opposed to TRACK-ADMS outputs, the CMAQ modeling system and FRAME outputs are not adjusted by calibration factors used either to adjust modeled values based on measurements or to account for transport and sources not directly modeled. Selecting the CMAQ modeling system or FRAME for the reference dataset is scientifically preferable because it does not involve any calibration of the outputs.

- The CMAQ modeling system is the most sophisticated modeling system among those considered in the model comparison exercise. It can simulate complex physical processes that transport and transform multiple pollutants in a physically realistic process-based way in a dynamical environment. It has been applied to short-term episode modeling as well as the production of annual statistics in a variety of places around the globe, including the UK. Conversely, TRACK-ADMS and FRAME treat some of the chemical processes in a more simplistic way, are limited in the species considered, have a simple representation of meteorology, and have been applied essentially to produce annual statistics in the UK.
- Selecting the CMAQ modeling system version 4.6 rather version 4.7 enables one to appreciate changes made to the CMAQ modeling system from one release version to the next release version.

The annual mean air concentration and total deposition footprints are calculated for a fossil-fuel power station located in the South-East of England (see Figure 1) for the year 2003. The choice of the power station is fairly arbitrary but it is not close to the coast, nor near other power plants so that its plume is isolated and directional analysis could be applied to monitoring sites around it. The method used to calculate the footprint of this source consists of calculating the difference between the baseline calculation and that with the source removed.

The power station emissions can account for a significant fraction of the local impacts for some species for 2003 (see Tables 5 and 6), even though their contributions to the UK annual mean air concentrations of SO_2, NO_x, and PM_{10}, and total deposition budgets are small (see Table 7). The mean contributions calculated across the modeling systems are 2.45% for SO_2, 0.60% for NO_x, 0.30% for PM_{10}, 2.13% for nss sulfur deposition, and 0.22% for nitrogen deposition. These values are comparable to those reported for similar power stations elsewhere.[29] There are rather large differences in the predicted maximum contributions of the power station to regional air pollution and deposition (see Table 5). Overall, results from the footprint calculations suggest that the power station contributes, locally, more to SO_2 air concentration and nss sulfur deposition than to NO_x and PM_{10} air concentrations, and nitrogen deposition. The maximum contributions for SO_2 air concentration and nss sulfur deposition are more than twice those of the other species reported in Table 5, for all modeling systems except TRACK-ADMS, which also predicts a relatively large maximum contribution for PM_{10}. This result reflects the large contribution of the power generation sector to SO_2 emissions in the UK (70% in 2003).[1] In comparison, its contribution to NO_x and PM_{10} emissions in the UK in 2003 were 22% and 6%, respectively.[1] The contribution of the power station to regional total deposition of nss sulfur, as calculated by each modeling system for the year 2003, is presented in Figure 3. The spatial extent of the nss sulfur total deposition footprint is consistent across the modeling systems and is limited to the South of England. The contribution of the power station to nss sulfur total deposition is most significant close the source. While the footprints from the two calculations with the CMAQ modeling system appear to be very similar, those from TRACK-ADMS and FRAME show some differences. In contrast to the other modeling systems, TRACK-ADMS does not predict the maximum contribution at the location of the source but at some distance downwind of the source, and attaches more importance to the northeast sector. Also, the calculations by TRACK and ADMS look to be loosely coupled. As for FRAME, it tends to give more weight to the southeast direction than the CMAQ modeling system. These differences indicate that the wind fields used by TRACK-ADMS and FRAME differ appreciably from those used by the CMAQ modeling system, which are for the year 2003.

Table 5 here

Table 6 here

Table 7 here

The maximum distance from the power station at which its contribution is half of its maximum contribution depends strongly on the modeling system and species considered (see Table 6). This is partly explained by the shape of the distributions of the contributions of the power station to regional air pollution and deposition. Indeed, we found that those distributions for the CMAQ modeling system are more skewed (larger skewness), and more sharply peaked (larger kurtosis) than for TRACK-ADMS and FRAME. This indicates that the contributions are more localized in space for the CMAQ modeling system than for TRACK-ADMS and FRAME. This result can also be inferred from Table 6. Further work is required to identify the reasons for the differences between the shapes of the models distributions.

We calculate the spatial correlation and the coefficient of variation of the root mean square error (CVRMSE) between each model footprint and that calculated by the CMAQ modeling system version 4.6, in the area indicated by a dashed polyline in Figure 3 (see Table 8). The correlation coefficient quantifies model agreement in terms of spatial patterns, and the CVRMSE measures the magnitude of the difference between model footprints. The CVRMSE is a dimensionless measure that is extremely useful when comparing between datasets with different mean values. A CVRMSE value of 10% for a modeling system would indicate that the mean variation in air concentration (or deposition) between this modeling system and the reference modeling system (the CMAQ modeling system version 4.6) is 10% of the mean value of the air concentration (or deposition) calculated by the reference modeling system. Table 8 indicates that the two calculations with the CMAQ modeling system are in good agreement with each other, both in terms of spatial patterns and magnitude of the footprints. Larger differences are found between the footprints produced by the CMAQ modeling system, TRACK-ADMS, and FRAME. At some locations the magnitude of the footprints can differ by more than a factor of two.

Table 8 here

SUMMARY AND CONCLUSIONS

Contributions of the emissions from a UK regulated fossil-fuel power station to regional air pollution and deposition are estimated using four air quality modeling systems for the year 2003. The modeling systems vary in complexity and emphasis in the way they treat atmospheric and chemical processes, and include the CMAQ modeling system in its versions 4.6 and 4.7, TRACK-ADMS, and FRAME. An evaluation of the baseline calculations against UK monitoring network data has revealed that all modeling systems tend to under estimate the annual mean air concentrations of SO_2, NO_x, and PM_{10}, and that there is a high variability in the output of the modeling systems for nss sulfur and nitrogen deposition. No individual modeling system was found to provide the overall best performance. One needs caution in making regulatory or policy decisions on the basis of one model. However, the agreement is good enough to make broad, general decisions, but this will become more difficult as emissions reductions become harder to implement. The CMAQ modeling system version 4.6 dataset was selected as the most appropriate reference dataset for the model footprint comparison.

The annual mean air concentration and total deposition increments due to the power station were summarized for each modeling system and compared using a range of diagnostic metrics. Differences between model results depend, inter alia, on the treatment of plume chemistry,[30] and emissions data processing. For instance, for the CMAQ modeling system, emissions from point sources were mixed instantaneously into the entire grid cell indentified at the level of sources plume rise, while for TRACK-ADMS and FRAME point sources plumes are tracked in a Lagrangian reference frame. In addition, the current theoretical understanding of the processes leading to acid deposition is limited.[31] Detailed process-level studies are needed to pinpoint deficiencies in acid deposition modeling. This wide area of research is kept for future work.

There are large uncertainties in the assessment of contributions of industrial sources to regional air pollution and deposition. A critical question that remains to be examined is whether uncertainties such as those reported in the present work still render such model footprints meaningful for policy applications. Quantifying the uncertainty associated with a single modeling system is extremely difficult given the range of inputs and process calculations.[32,33] Hence, an ensemble average of model calculations could be used to provide an estimate of the uncertainty associated with an industrial source footprint. It has to be recognized that air quality modeling systems such as TRACK-ADMS and FRAME still have run times much faster than those of advanced systems such the CMAQ modeling system. For this reason, such modeling systems are attractive for source-receptor calculations involving a large number of model calculations.

Other modeling systems have been used extensively to map sulfur and nitrogen deposition in the UK, namely the Concentration Based Estimated Deposition (CBED)[34] modeling system and the Hull Acid Rain Model (HARM).[35] CBED is the operational observation-based modeling system used to inform policy makers about current levels of sulfur and nitrogen deposition in the UK. HARM has been used to support the development of emissions abatement strategies for reducing acid deposition in the UK.[36] A comparison of the model deposition budget predictions reported in our work with those of these modeling systems will be undertaken in a future study.

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TABLES

Table 1. Percentage fraction of predictions, within a factor of two of observations (FO2), considering all monitoring sites within the Automatic Urban and Rural Network (AURN) and the Joint Environmental Programme (JEP) monitoring sites, for the annual mean air concentrations of SO_2, NO_x, and PM_{10}, and within the Secondary Acid Precipitation Monitoring Network (SAPMN) for non-sea-salt (nss) SO_x, NO_y, and NH_x wet depositions, for each modeling system for the year 2003. The figures for wet deposition that are given in brackets correspond to observational data derived using the gridded UK Met Office precipitation observations (see text for details).

	CMAQ V4.6	CMAQ V4.7	TRACK-ADMS	FRAME
SO_2	87.7	87.7	69.2	78.5
NO_x	72.6	58.9	91.8	84.9
PM_{10}	88.2	100.0	100.0	NA
Nss SO_x wet deposition	100.0 (86.5)	83.8 (83.8)	NA	81.1 (81.1)
NO_y wet deposition	97.3 (86.5)	100.0 (89.2)	NA	91.9 (83.8)
NH_x wet deposition	97.3 (75.7)	86.5 (81.1)	NA	62.2 (70.3)

Table 2. Same caption as Table 1 for the correlation coefficient.

	CMAQ V4.6	CMAQ V4.7	TRACK-ADMS	FRAME
SO_2	0.27	0.29	0.40	0.28
NO_x	0.76	0.77	0.76	0.76
PM_{10}	0.09	0.00	0.45	NA
Nss SO_x wet deposition	0.82 (0.43)	0.75 (0.41)	NA	0.83 (0.44)
NO_y wet deposition	0.85 (0.51)	0.86 (0.54)	NA	0.77 (0.27)
NH_x wet deposition	0.78 (0.34)	0.67 (0.31)	NA	0.65 (0.19)

Table 3. Same caption as Table 1 for the normalized mean bias (NMB), as a percentage.

	CMAQ V4.6	CMAQ V4.7	TRACK-ADMS	FRAME
SO_2	-6.7	-8.6	-39.5	-11.7
NO_x	-41.5	-47.2	-15.5	-6.4
PM_{10}	-32.7	-8.9	-20.2	NA
Nss SO_x wet deposition	-2.7 (-12.6)	50.9 (35.5)	NA	70.8 (53.4)
NO_y wet deposition	-12.0 (-22.6)	-9.0 (-20.0)	NA	39.9 (23.0)
NH_x wet deposition	-13.3 (-22.8)	32.7 (18.2)	NA	67.8 (49.4)

Table 4. UK deposition budgets for non-sea-salt (nss) sulfur (in Gg S) and nitrogen deposition (in Gg N), as calculated by each modeling system for the year 2003.

	CMAQ V4.6	CMAQ V4.7	TRACK-ADMS	FRAME
Nss SO_x wet deposition	57	98	NA	102
Nss SO_x dry deposition	130	131	NA	65
Nss S total deposition	187	229	154	167
NO_y wet deposition	46	50	NA	67
NO_y dry deposition	75	79	NA	61
NH_x wet deposition	48	79	NA	90
NH_x dry deposition	97	103	NA	69
N total deposition	266	311	266	287

Table 5. Maximum percentage contribution of the power station to regional air concentration for SO_2, NO_x, and PM_{10}, and non-sea-salt (nss) sulfur and nitrogen total deposition, for each modeling system for the year 2003.

	CMAQ V4.6	CMAQ V4.7	TRACK-ADMS	FRAME
SO_2	70.2	68.1	22.7	38.7
NO_x	22.5	19.5	2.9	7.7
PM_{10}	6.0	3.0	10.1	NA
Nss S total deposition	67.1	60.2	15.6	32.0
N total deposition	7.3	6.0	1.1	2.7

Table 6. Maximum distance (in km) from the power station at which its contribution to regional air concentration for SO_2, NO_x, and PM_{10}, and non-sea-salt (nss) sulfur and nitrogen total deposition, is half of its maximum contribution, for each modeling system for the year 2003.

	CMAQ V4.6	CMAQ V4.7	TRACK-ADMS	FRAME
SO_2	10	15	115	60
NO_x	5	5	140	70
PM_{10}	5	35	10	NA
Nss S total deposition	15	20	115	35
N total deposition	5	5	115	55

Table 7. Percentage contribution of the power station to the UK annual mean air concentrations of SO_2, NO_x, and PM_{10}, and non-sea-salt (nss) sulfur and nitrogen total deposition budgets, for each modeling system for the year 2003.

	CMAQ V4.6	CMAQ V4.7	TRACK-ADMS	FRAME
SO_2	2.19	2.17	2.58	2.85
NO_x	0.67	0.63	0.47	0.62
PM_{10}	0.34	0.28	0.28	NA
Nss S total deposition	2.24	1.85	1.87	2.55
N total deposition	0.19	0.16	0.13	0.39

Table 8. Spatial correlation coefficient and coefficient of variation of the root mean square error (CVRMSE, in percents), reflecting similarities between the footprints of air concentrations of SO_2, NO_x, and PM_{10}, and total depositions of non-sea-salt (nss) sulfur and nitrogen, with respect to the reference modeling system (the CMAQ modeling system, version 4.6), for each modeling system in the area indicated by a dashed polyline in Figure 3, for the year 2003.

	Spatial correlation coefficient			CVRMSE		
	CMAQ V4.7	TRACK-ADMS	FRAME	CMAQ V4.7	TRACK-ADMS	FRAME
SO_2	0.98	0.36	0.61	16.7	116.4	98.4
NO_x	0.98	0.34	0.62	24.9	101.4	96.0
PM_{10}	0.82	0.61	NA	39.8	105.8	NA
Nss S total deposition	0.98	0.46	0.78	17.8	108.0	71.8
N total depostion	0.90	0.42	0.43	46.3	101.1	195.5

LIST OF FIGURE CAPTIONS

1. Location and type (remote, rural, suburban, urban background) of monitoring sites in the UK Automatic Urban and Rural Network (AURN) and the Joint Environmental Programme (JEP) monitoring sites, as filled circles, and the Secondary Acid Precipitation Monitoring Network (SAPMN), as open circles, used for the evaluation of the model baseline calculations. The grey-filled area corresponds to the ‘UK domain’ used for the model comparison exercise. The location of the fossil-fuel power station considered for the footprint calculations is marked by a cross symbol.
2. Annual total deposition of non-sea-salt (nss) sulfur, as calculated by each modeling system in the ‘UK domain’ for the year 2003: (a) CMAQ version 4.6, (b) CMAQ version 4.7, (c) TRACK-ADMS, and (d) FRAME.
3. Percentage contribution of the power station to regional non-sea-salt (nss) sulfur total deposition, as calculated by each modeling system in the ‘UK domain’ for the year 2003: (a) CMAQ version 4.6, (b) CMAQ version 4.7, (c) TRACK-ADMS, and (d) FRAME. Note that the color scale is not linear. The dashed polyline represents the area over which the statistics reported in Table 8 are calculated.