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Global sensitivity analysis of chemistry-climate model budgets of tropospheric ozone and OH: Exploring model diversity

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Abstract. Projections of future atmospheric composition change and its impacts on air quality and climate depend heavily on chemistry-climate models that allow us to investigate the effects of changing emissions and meteorology. These models are imperfect as they rely on our understanding of the chemical, physical and dynamical processes governing atmospheric composition, on the approximations needed to represent these numerically, and on the limitations of the observations required to constrain them. Model intercomparison studies show substantial diversity in results that reflect underlying uncertainties, but little progress has been made in explaining the causes of this or in identifying the weaknesses in process understanding or representation that could lead to improved models and to better scientific understanding. Global sensitivity analysis provides a valuable method of identifying and quantifying the main causes of diversity in current models. For the first time, we apply Gaussian process emulation with three independent global chemistry transport models to quantify the sensitivity of ozone and hydroxyl radicals (OH) to important climate-relevant variables, poorly-characterized processes and uncertain emissions. We show a clear sensitivity of tropospheric ozone to atmospheric humidity and precursor emissions which is similar for the models, but find large differences between models for methane lifetime, highlighting substantial differences in the sensitivity of OH to primary and secondary production. This approach allows us to identify key areas where model improvements are required while providing valuable new insight into the processes driving tropospheric composition change.

1 Introduction

Atmospheric photochemistry and transport processes play important roles in the Earth system by controlling the impact of natural and anthropogenic trace gas emissions on air quality and global climate. Methane (CH_4) and ozone (O_3) are the second and third most important greenhouse gases contributing to climate change since the preindustrial era (IPCC, 2013).

The atmospheric abundance of both gases has increased substantially due to anthropogenic activity, and their fates are strongly coupled through the short-lived hydroxyl (OH) radical. CH_4 is an O_3 precursor and O_3 is a major source of OH, which controls the oxidation of CH_4 and many other trace gases. At the surface O_3 contributes to poor air quality and is damaging to human health, crop yields and natural ecosystems (Monks et al., 2015). The relatively short lifetime of these gases makes them attractive targets for emission controls (Shindell et al., 2012), but scientific uncertainties associated with the processes that govern their abundance and distribution has hindered implementation of effective control policies.

Current global chemistry-climate models representing the co-evolution of atmospheric O_3 and CH_4 show differences in CH_4 lifetime of almost a factor of two (Wild, 2007; Voulgarakis et al., 2013). This prevents them from simulating the observed atmospheric build-up of CH_4 correctly or attributing its causes reliably, and leads to substantial uncertainty in the impact of future emission changes on global climate (Stevenson et al., 2013; IPCC, 2013). The underlying cause is differences in OH, which depends on humidity, sunlight, O_3, and on a wide range of chemical and dynamical processes. For O_3, on the other hand, the abundance, seasonality and spatial variation are represented relatively well in models under present-day conditions, but observed changes in surface O_3 since the preindustrial era are systematically underestimated (Stevenson et al., 2013). Models have difficulty reproducing recent observed trends in surface O_3 driven by changes in precursor emissions, natural sources, stratospheric influx and transport patterns (Parrish et al., 2014). This is a major concern because changes in the tropospheric abundance of O_3 influence our assessment of radiative forcing and also attainment of air quality objectives on regional and urban scales (e.g., Akimoto, 2003). These discrepancies suggest that there are major weaknesses in our fundamental understanding of the chemical, dynamical, and emission processes controlling the distribution, interaction and fate of O_3, CH_4 and OH, or in how these processes are represented in global chemistry and climate models.

Global sensitivity analysis provides a valuable approach to determine the major drivers of model behaviour, and has been applied to atmospheric chemistry schemes to explore uncertainties in tropospheric O_3 (Derwent and Murrells, 2013; Christian et al., 2017; Ridley et al., 2017; Newsome and Evans, 2017). These studies have typically used Monte Carlo-based ensemble approaches for simple models (e.g., Ridley et al., 2017) or structured random-sampling approaches for more computationally intensive models (e.g., Christian et al., 2017), and have focussed on sensitivities in a single model framework. In this study we demonstrate the use of Gaussian process emulation for global sensitivity analysis, applied previously to models of aerosol processes (Lee et al., 2011, 2013) and air quality (Beddows et al., 2017; Aleksankina et al., 2019), and apply it to explore the sensitivity of global tropospheric O_3 and CH_4 lifetime to uncertainty in key model processes and inputs. We investigate how the sensitivities differ across three independent chemistry-transport models, and demonstrate how this approach may be used to explore the diversity in model responses and to identify where model results differ.

2 Approach

We consider here two important global diagnostics of model performance, the tropospheric O_3 burden and the chemical lifetime of CH_4 in the troposphere. The tropospheric O_3 burden is the annual mean mass of O_3 below the tropopause, defined here by the 150 ppb isopleth of monthly mean O_3. The chemical lifetime of CH_4 reflects the lifetime of CH_4 to removal by OH in

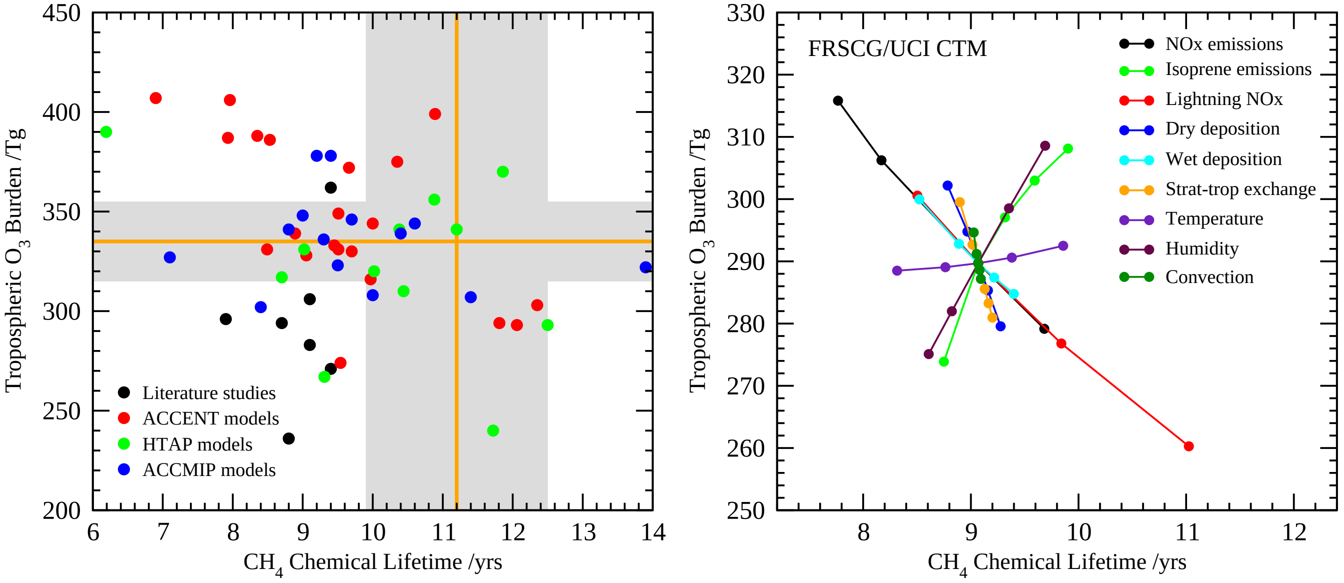

Figure 1. Tropospheric oxidant budgets from previous published studies and model intercomparisons (left panel, **a**), along with measurement-based estimates of the tropospheric O_3 burden and CH_4 lifetime (shaded regions). The right panel **(b)** shows results from one-at-a-time sensitivity studies with a single model revealing the extent to which individual processes can influence the budgets (see Wild (2007) for details). Note that the right panel covers only part of the parameter space shown in the left panel.

the troposphere, and provides a useful proxy for global tropospheric oxidizing capacity. Global model studies in the literature and previous model intercomparisons show a large diversity in modelled budgets (see Fig. 1), where the range in O_3 burden and CH_4 lifetime both span about a factor of two. There is no clear relationship between the budget terms on an annual basis, highlighting the relatively complex relationship between tropospheric O_3 and OH that reflects physical and dynamical processes as well as photochemistry.

Observation-based determination of these global quantities is difficult. However, assessment of three global O_3 climatologies derived from ozonesonde measurements over the 1980s and 1990s indicates an annual mean tropospheric O_3 burden of 327–344 Tg when applying the same 150 ppb isopleth definition of the tropopause used in model analysis (Wild, 2007), suggesting a burden of about 335±20 Tg. Ensemble mean O_3 burdens from recent model intercomparisons lie close to this: 344±39 Tg from ACCENT (Stevenson et al., 2006), 328±41 Tg from HTAP (Fiore et al., 2009) and 337±23 Tg from ACCMIP (Young et al., 2013), see Table 1, but about half of published studies lie outside the observationally-constrained range (see Fig. 1). A thorough observation-based sensitivity analysis of the factors contributing to CH_4 removal gave a whole-atmosphere lifetime of 9.1±0.9 yr, and a corresponding CH_4 chemical lifetime of 11.2±1.3 yr (Prather et al., 2012). The latter is substantially longer than that derived from model intercomparisons: 9.6±1.4 yr from ACCENT (Stevenson et al., 2006), 10.2±1.7 yr from HTAP (Fiore et al., 2009) and 9.8±1.6 yr from ACCMIP (Voulgarakis et al., 2013), and two thirds of the model studies shown

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Table 1. Global tropospheric metrics from previous model studies

Studies	Number	O_3 burden	CH_4 lifetime	References
Early literature studies	33 studies	307±38 Tg		Wild (2007)
ACCENT intercomparison	21 models	344±39 Tg	9.6±1.4 yr	Stevenson et al. (2006)
HTAP intercomparison	12 models	328±41 Tg	10.2±1.7 yr	Fiore et al. (2009)
ACCMIP intercomparison	14 models	337±23 Tg	9.8±1.6 yr	Young et al. (2013); Voulgarakis et al. (2013)
Observational estimates		335±20 Tg	11.2±1.3 yr	Wild (2007); Prather et al. (2012)

in Fig. 1 lie outside this range. However, it is difficult to judge the validity of existing model results without a clearer idea of the uncertainties involved and how they contribute to the corresponding biases.

The sensitivity of the budget terms to individual processes has been explored in previous studies using the Frontier Research System for Global Change version of the University of California Irvine Chemical Transport Model (FRSGC/UCI CTM) in Wild (2007). One-at-a-time sensitivity runs were performed varying surface NO_x emissions (30–60 $\mathrm{TgN\,yr^{-1}}$), isoprene emissions (0–650 $\mathrm{TgC\,yr^{-1}}$), lightning NO_x emissions (0–7.5 $\mathrm{TgN\,yr^{-1}}$), convective lifting, stratospheric influx and deposition processes (all ±50%), temperature (±5^o C) and humidity (±20%), and results are summarised in Fig. 1. This study highlighted the responses of a single model to particular processes, but the variations spanned relatively little of the parameter space defined by previous model studies, suggesting that substantial additional uncertainties were not accounted for here, including process interactions, neglected processes, and structural differences between models.

To explore the sensitivity of tropospheric budgets to uncertainty in several processes at once, we perform a global sensitivity analysis using Gaussian process emulation, following the approach of Lee et al. (2011). This allows us to reproduce the model response across a multidimensional parameter space based on a small ensemble of model runs at points representing a combination of inputs that are optimally chosen to fill the space. We select eight key variables that influence global oxidant budgets substantially, and that span a range of model inputs (e.g., emissions), processes (e.g., deposition) and meteorological variables, see Table 2. These are loosely based on the earlier one-at-a-time studies, and while they do not encompass all sources of uncertainty, they are chosen to represent key uncertainties while ensuring that the study remains computationally tractable. We select surface emissions of NO_x from natural and anthropogenic sources, the dominant precursor for O_3 in the troposphere; lightning emissions of NO, which are highly uncertain and have a disproportionately large impact on O_3 and OH due to the altitude of the source; and biogenic emissions of isoprene, which dominate global sources of volatile organic compounds. We include dry deposition, which is important for uptake of O_3 and other species at the surface, and wet deposition which is important for removal of soluble precursors. We vary the atmospheric humidity used by the model photochemistry, which plays an important role in O_3 chemistry and OH formation, but leave it untouched for other processes to avoid perturbing model dynamical processes. We vary cloud optical depth, an uncertain variable which has a major influence on photolysis rates in the lower troposphere. Finally, we vary turbulent mixing in the planetary boundary layer (PBL), which has an important role in lifting and dispersing surface oxidants, but which remains poorly constrained. For each variable, we define a range that

Table 2. Variables and uncertainty ranges used in this study

Variables	Range
Surface NOx emissions	30–50 $\mathrm{TgN\,yr^{-1}}$
Lightning NO emissions	2–8 $\mathrm{TgN\,yr^{-1}}$
Biogenic isoprene emissions	200–800 $\mathrm{TgC\,yr^{-1}}$
Dry deposition rates	± 80%
Wet deposition rates	± 80%
Atmospheric humidity	± 50%
Cloud optical depth	× 0.1–10
Boundary layer mixing	× 0.01–100

encompasses the maximum and minimum likely values that is loosely based on published studies from the literature, and these are presented in Table 2.

Following Lee et al. (2011), we use maximin Latin hypercube sampling to optimally select 80 points from across the eight-dimensional parameter space. Each point represents a combination of values chosen from the range for each variable, and specifies the values to use for a full model simulation. An additional 24 points are selected to provide an independent test of the validity of the emulators that are built. This defines a set of 104 model simulations to perform. For this study, we use three independent global chemistry-transport models: the FRSGC/UCI CTM (Wild, 2007), the Goddard Institute for Space Studies Global Climate Model, GISS GCM (Shindell et al., 2013), and the Community Atmosphere Model with Chemistry, CAM-Chem (Lamarque et al., 2012). The models differ in their sources of meteorology, but are run for a full year (following 6–12 months spin-up) under conditions that are broadly consistent with 2001 meteorology, a year without strong climate phenomena such as El Niño. Natural and anthropogenic emissions differ somewhat across the models, reflecting different assumptions and online generation of natural emissions, but we scale the magnitude of global annual emissions to 40 $\mathrm{TgN\,yr^{-1}}$ for surface $\mathrm{NO_x}$, 5 $\mathrm{TgN\,yr^{-1}}$ for lightning NO and 500 $\mathrm{TgC\,yr^{-1}}$ for isoprene in the control run, accepting that differences in emission distributions in the models contribute to structural uncertainty. We assume uncertainty ranges of ±25% for surface $\mathrm{NO_x}$ and ±60% for lightning NO (Schumann and Huntrieser, 2007) and isoprene emissions (Ashworth et al., 2010). Other variables are scaled according to the factors shown in Table 2 without further standardization between models. Dry and wet deposition rates are scaled for all species considered; atmospheric water vapour is scaled for the model photochemistry scheme only and cloud optical depth based on prescribed or calculated cloud fields is scaled for the model photolysis scheme only. Boundary layer mixing is perturbed through scaling the effective vertical diffusion coefficient so that turbulent mixing of tracers between layers varies from negligible to almost complete every model time step.

Emulators are then built for each model for each output of interest using the methods described in Lee et al. (2011) and Ryan et al. (2018). We focus here on global annual mean tropospheric $\mathrm{O_3}$ burden and $\mathrm{CH_4}$ chemical lifetime for simplicity. The emulators are tested through use of the additional 24 validation simulations to evaluate their performance. For the outputs

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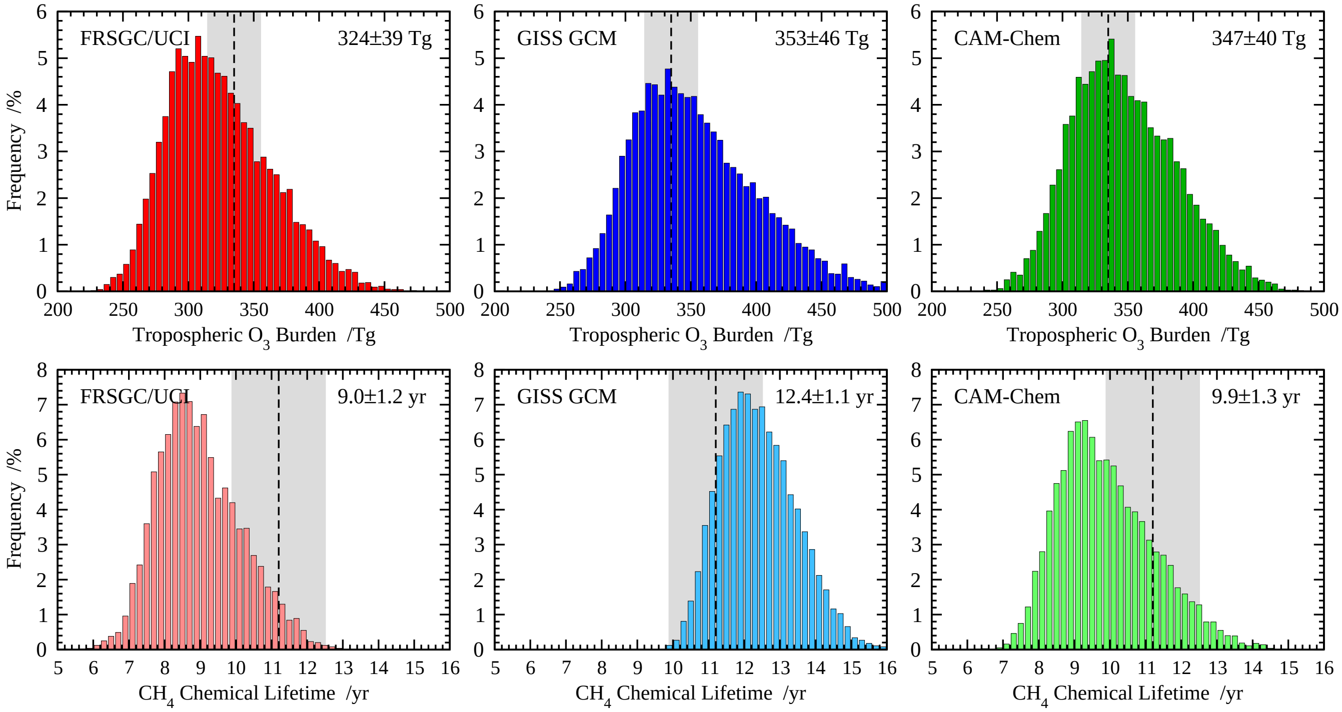

Figure 2. Probability distributions for the global annual mean tropospheric O_3 burden (top row) and tropospheric chemical lifetime of CH_4 (bottom row) for each model. The mean and standard deviation over 10,000 realizations are shown on the upper right of each panel, and observation-based estimates of O_3 burden and CH_4 lifetime are shown shaded.

considered here, the model response surfaces are relatively smooth, reflecting the stable behaviour of the global O_3 burden and CH_4 lifetime, and the emulators fit the validation runs very closely with a correlation coefficient $r > 0.99$ (see Ryan et al., 2018). The emulators reproduce the response of the full model within the variable ranges defined, and can be used in place of the model for intensive analysis such as uncertainty propagation through the use of Monte-Carlo approaches that would not be computationally feasible with the full model. This allows us to define formal error bars for the response of each model, and to carry out global sensitivity analysis by determining the contribution of each variable to the overall variance in modelled O_3 burden and CH_4 lifetime.

3 Model responses and contributions to variance

We first use the emulators built for each model to propagate the uncertainty in the selected variables to uncertainty in O_3 burden and CH_4 lifetime. We use a Monte Carlo approach to randomly select 10,000 points from across the response space for each model, sampling uniformly across the full input range of each variable, and use this to generate the probability distribution for each model. Figure 2 shows the distribution in global O_3 burden and CH_4 lifetime from each model. The

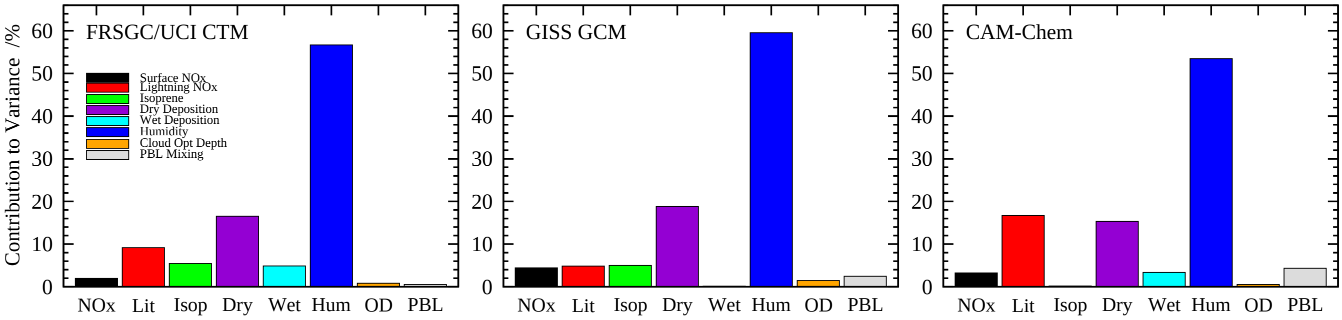

Figure 3. Contributions of each variable to the total variance in the simulated tropospheric O_3 burden in each model.

behaviour of the models is similar, with a normalised standard deviation of 12–13% for O_3 burden and 9–13% for CH_4 lifetime, and the distributions are slightly skewed, reflecting the nonlinear response of these budget terms to the governing processes. The 1σ uncertainty in each budget term is comparable in magnitude to that seen between different models in recent model intercomparisons (see Table 1); while this may be fortuitous, it demonstrates that process uncertainty contributes substantially to model diversity.

For each model, the mean O_3 burden lies within the observational uncertainty range, along with 32–38% of the distribution. A large proportion of each distribution lies outside the observational range, suggesting that the uncertainty ranges adopted for some of the variables were larger than needed, or that a normal distribution of uncertainty could have been assumed across each range in place of a uniform distribution. For mean CH_4 lifetime, agreement with observations is less good, with the GISS GCM and CAM-Chem lying at opposite boundaries of the observed range and the FRSGC/UCI CTM lying outside it. For the GISS GCM, 53% of the distribution lies inside the observed range, while for the FRSGC/UCI CTM it is only 24%. These discrepancies highlight that uncertainty in chemistry and transport processes not considered here may play a substantial role in governing the CH_4 lifetime.

The sensitivity for each variable is determined by variance decomposition, which quantifies the contribution of each variable to the variance in the model output, and is shown in Fig. 3. We neglect the contribution of interactions between variables, which can be identified through this approach but which remain below 4% of the variance for the model responses examined here. For the global O_3 burden, the models show relatively similar sensitivities, with atmospheric humidity contributing 55–60% of the variance in all three models, and dry deposition processes contributing 15–20%, see Fig. 3. The models show different sensitivities to lightning NO_x, isoprene emissions and wet deposition, but the contribution of these terms is relatively small. While a large sensitivity to humidity is expected, given the important role of atmospheric water vapour as a chemical sink of O_3, it is notable that humidity has not been prescribed in previous model intercomparison studies and is rarely diagnosed except where changes in climate are expected (e.g., Lamarque et al., 2013). It may therefore constitute a substantial fraction of the diversity in tropospheric O_3 burden seen across these studies.

For the tropospheric CH_4 lifetime, the models show markedly different sensitivities, with humidity contributing nearly 60% of the variance for the FRSGC/UCI CTM and CAM-Chem, but less than 5% for the GISS GCM, where emissions of NO_x and

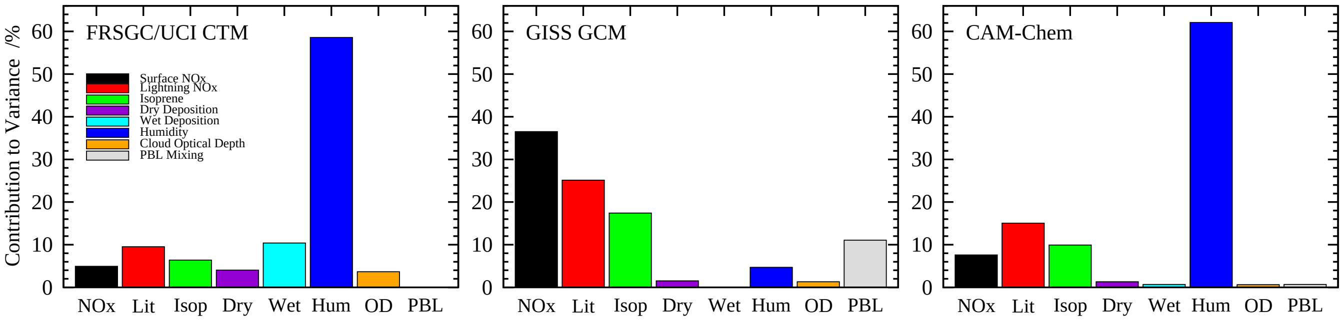

Figure 4. Contributions of each variable to the total variance in the simulated annual mean CH_4 chemical lifetime in each model.

isoprene are responsible for nearly 80% of the variance, see Fig. 4. It is clear that the factors governing tropospheric OH are very different in the models, highlighting differences in chemical environment and transport patterns that affect the location and magnitude of CH_4 oxidation. Sensitivity to humidity suggests that primary sources of OH dominate through photolysis of O_3 and subsequent reaction of O^1D with water vapour. Sensitivity to NO_x emissions suggests that secondary sources of OH dominate through oxidation of NO, and sensitivity to isoprene highlights the importance of VOC as a source and sink of OH and as a mechanism for locking up and transporting NO_x. Interestingly, the GISS GCM also shows substantial sensitivity to boundary layer mixing, highlighting the importance of transport of fresh emissions from the surface for secondary OH formation.

These differences have important implications for assessment of future composition change. Future scenarios projecting increased emissions of greenhouse gases and reduced emissions of O_3 precursors (e.g., RCPs 4.5, 6.0 and 8.5) are likely to lead to increased future humidity and reduced surface NO_x. The FRSGC/UCI CTM and CAM-Chem would be expected to show a reduction in CH_4 lifetime due to greater OH concentrations associated with higher water vapour, while the GISS GCM would show an increase in CH_4 lifetime due to lower secondary production of OH associated with reduced NO_x emissions. Analysis of future changes in CH_4 lifetime for models contributing to the ACCMIP intercomparison suggests that this is indeed the case, with the GISS GCM one of three models showing increased lifetime by 2100 for the RCP6.0 pathway, and four models showing decreased lifetime (Voulgarakis et al., 2013). An understanding of the causes of this differing sensitivity is thus important for explaining the different model responses.

4 Investigating model differences

The sensitivity of modelled O_3 burden and CH_4 lifetime to two key variables, humidity and surface NO_x emissions, is shown for the FRSGC/UCI CTM and GISS GCM in Fig. 5. These response surfaces are generated using the emulator for each model assuming that the other six variables are unchanged. While the O_3 burden is slightly higher in the GISS GCM, the gradients across the response surfaces are similar in the models. High O_3 burdens occur as the gradient steepens at low humidity values, explaining the greater skew in the distribution evident for the GISS GCM in Fig. 2 and suggesting that atmospheric humidity

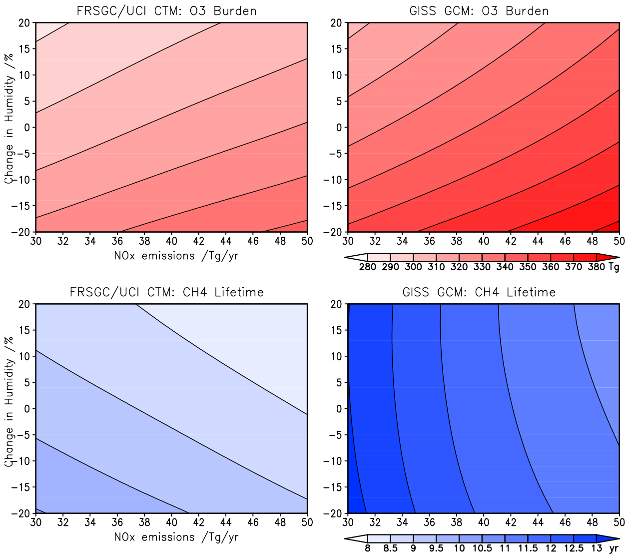

Figure 5. Sensitivity of tropospheric O_3 burden and CH_4 chemical lifetime to changes in surface NO_x emissions and humidity in the FRSGC/UCI CTM and GISS GCM.

may be lower in this model. However, the relative changes in burden with NO_x emissions and humidity are very similar across all three models, see Fig. 6. The responses for CH_4 lifetime show notably different behaviour, with much greater sensitivity to NO_x and very little sensitivity to humidity in the GISS GCM compared to the other models. At high humidities the CH_4 lifetime appears almost insensitive to humidity, suggesting a saturation in OH formation in this model. In contrast, the other models show a very similar degree of sensitivity to humidity in both O_3 burden and CH_4 lifetime that ranges from +7% to -5% across the humidity range considered here. This indicates a much stronger coupling between O_3 and OH formation, and highlights the dominance of the primary OH source in these models.

The response surfaces shown here allow us to estimate the impact of changes in future humidity and surface NO_x emissions in the absence of other changes. A reduction in NO_x emissions from 40 to 30 $\mathrm{TgN\,yr^{-1}}$ and increase in humidity of 15%, corresponding loosely to the changes between 2000 and 2050 expected along the RCP8.5 pathway (van Vuuren et al., 2011), would lead to an increase in CH_4 lifetime of 1.3 yr in the GISS GCM (from 11.7 to 13.0 yr), an increase of 0.2 yr in CAM-Chem, and no change in the FRSGC/UCI CTM. While this neglects the influence of other emission and climate changes, particularly the increase in CH_4 concentrations which would extend the lifetime in all models, it demonstrates the very different sensitivities anticipated for different models under future climate scenarios.

To help identify the cause of the differing model responses, we show the contribution of the dominant variables to the variance in the annual mean tropospheric column CH_4 chemical loss rate at each model grid point in Fig. 7. This shows how the contribution of the different processes governing CH_4 removal varies geographically and reveals further differences between the models. For the FRSGC/UCI CTM and CAM-Chem, humidity dominates the variance at most locations, particularly downwind of midlatitute emission regions, and is minimum in equatorial regions where there is a greater sensitivity to lightning

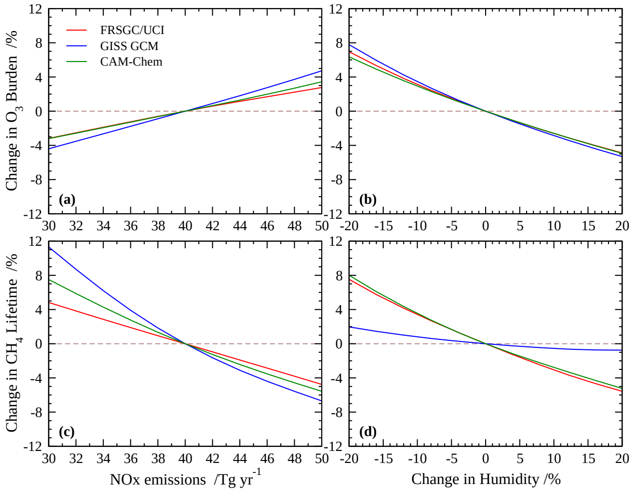

Figure 6. Relative changes in tropospheric O_3 burden **(a, b)** and CH_4 chemical lifetime **(c, d)** to changes in surface NO_x emissions and humidity alone in each model.

NO_x. The effect of isoprene and surface NO_x emissions are largely limited to the source regions of these gases, indicating that their impacts on OH are relatively localised. For the GISS GCM, humidity dominates at mid-latitudes, but contributes little in tropical regions, where primary emissions of NO_x and isoprene dominate. Of particular interest is the widespread impact from surface NO_x emissions, which contribute substantially to CH_4 lifetime over remote ocean regions. The effect of NO_x on OH in these locations suggests that substantial nitrogen is transported to these regions in the form of reservoir species such as peroxyacetyl nitrate (PAN), and this is supported by the patterns of transport seen in the isoprene contribution, where the effects are not localised to source regions as they are for the other two models. It is therefore likely that differing treatments of NO_y chemistry are one cause of the different model sensitivities. However, a more detailed exploration of the sensitivity to chemical processes would be needed to confirm this. Our analysis provides a valuable guide to locations where model responses are likely to differ most, such as in tropical oceanic regions, and further investigation of OH sensitivity in these regions should bring improvements in our understanding of atmospheric processes and in their representation in current global-scale models.

5 Conclusions

We have demonstrated the value of Gaussian process emulation in performing global sensitivity analysis of computationally-intensive global atmospheric chemistry transport models, and in applying this across a number of models to investigate model diversity. The approach provides a simple way of exploring the sensitivity of key terms in the tropospheric oxidant budget to governing processes and inputs, and we show that it can provide substantial new insight into the causes of differences between models.

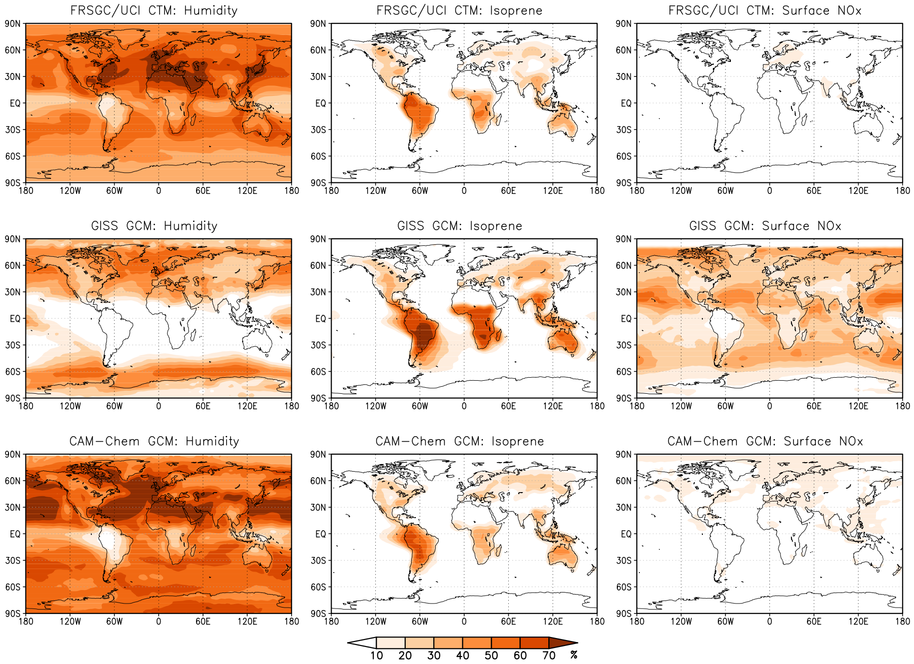

Figure 7. Contributions (in %) to the total variance in the annual tropospheric column CH_4 chemical loss rate in each model from the dominant variables: humidity, isoprene emissions and surface NO_x emissions.

Our study has highlighted the large sensitivity of the tropospheric O_3 burden to atmospheric water vapour, suggesting that this variable should be diagnosed or perhaps constrained in future model intercomparisons to permit clearer characterization of differences in model chemistry. More surprisingly, we find that the drivers of variability in global OH can be very different between models, and this may contribute to the large diversity in modelled tropospheric CH_4 lifetimes seen in recent model intercomparisons. Given the importance of atmospheric oxidising capacity for both air quality and climate change, this difference in OH behaviour is a major cause for concern and is a clear priority for further investigation.

While we have shown the value of emulation approaches for exploring model behaviour much more thoroughly than through simple one-at-a-time sensitivity studies, this study has been largely exploratory in nature, investigating the effects of a very limited number of variables. A more detailed global uncertainty analysis is required that considers a wider range of model processes and inputs and incorporates a more rigorous assessment of uncertainty in each variable. Application of observation-based constraints is then needed to restrict the size of the response space to calibrate the models and identify specific processes in need of refinement. Applying this approach across different models accommodates the structural uncertainties in model formulation, permitting a more robust assessment of process understanding. This would provide a strong evaluation framework for improving understanding of the physical and chemical processes driving atmospheric composition change and its effects on air quality and climate.

Data availability. The monthly mean output from each model for the ensemble of runs performed in this study will be made available from the CEDA data archive, and can be accessed by request to the corresponding author.

Author contributions. OW, LL, FO and AV designed the study. OW, AV and JL ran model simulations, and ER and LL performed the analysis. OW prepared the manuscript with contributions from all co-authors.

Competing interests. The authors declare that they have no conflict of interest.

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