

Author response:

We thank the editor for their comments, which have helped us to improve our manuscript. All comments have been addressed. The editor comment is shown in black, our reply is shown in blue and extracts from the revised manuscript text are shown in 'commas', with altered text in red and deleted text with strikethrough.

(1) A very technical suggestion, but your statement: "Regions are defined as NO<sub>x</sub>-limited when increasing VOCs or OH acts to reduce O<sub>3</sub> concentrations..." seems a bit too categorical in my view. NO<sub>x</sub>-limited regimes can certainly include situations O<sub>3</sub> production is not sensitive to perturbations in VOC abundance (see for example Figure 12-4 in Jacob (1999), Introduction to Atmospheric Chemistry). I would suggest wording along the lines of:

"In NO<sub>x</sub>-limited regimes where O<sub>3</sub> production is proportional to NO<sub>x</sub> concentrations, increasing VOCs or OH can also act to reduce O<sub>3</sub> concentrations..."

We thank the editor for this suggestion and have modified the sentence:

'In NO<sub>x</sub>-limited regimes where O<sub>3</sub> production is proportional to NO<sub>x</sub> concentrations, increasing VOCs or OH can also act to~~Regions are defined as NO<sub>x</sub>-limited when increasing VOCs or OH acts to~~ reduce O<sub>3</sub> concentrations through oxidation and formation of organic peroxides (Pacifico et al., 2012). In this NO<sub>x</sub>-limited case, increasing NO<sub>x</sub> will lead to greater O<sub>3</sub> formation.'

(2) One of the reviewers asks, "Please discuss also how the models deal with the soil NO<sub>x</sub> emissions in the simulations.". In your response you state:

"NO<sub>x</sub> emissions, including biomass burning emissions, are prescribed based on the SSP3-7.0 scenario but lightning NO<sub>x</sub> and soil NO<sub>x</sub> differs between the models based on the chosen parameterisation of individual models"

Should this sentence be, "Anthropogenic NO<sub>x</sub> emissions, including biomass burning emissions are prescribed based on..."? Otherwise it's confusing since you first state that NO<sub>x</sub> emissions are prescribed, but then state certain sources that are not. Please clarify.

We thank the editor for identifying this mistake and the sentence has been corrected:

'Anthropogenic NO<sub>x</sub> emissions, including biomass burning emissions, are prescribed based on the SSP3-7.0 scenario, soil NO<sub>x</sub> is prescribed by each model and-but lightning NO<sub>x</sub> and-soil NO<sub>x</sub> differs between the models based on the chosen parameterisation of individual models. Compared to the present-day, NO<sub>x</sub> emissions in biomass burning areas decrease in Africa to follow projected trends, but do not change in South America. NO<sub>x</sub> emissions increase in cities and Nigeria especially has major growth in urban areas. Compared to the scenario without climate change, total lightning NO<sub>x</sub> emissions increase in all models, and the increases occur during the wet season (Fig. S4). MRI predicts much larger increases than GISS and UKESM1, and UKESM1 shows a decrease in lightning NO<sub>x</sub> over the Amazon basin in December–February (Fig. S4a) although the net effect over all seasons is positive. Peroxyacetyl nitrate (PAN) decreases in all models (–94 ppt, –61 ppt, –30 ppt for UKESM1, GISS and MRI respectively) due to increased thermal decomposition. In GISS and UKESM1, the increase in isoprene emissions can increase removal of NO<sub>x</sub> via formation of isoprene nitrates. ~~Soil NO<sub>x</sub> does not change in response to climate change in any model.~~'

On the topic of this reviewer's same comment, in your Model Descriptions Section 2.1, I suggest that you include very brief descriptions of the isoprene and soil NO<sub>x</sub> parameterization schemes used in the models where these emissions are calculated interactively (in addition to just including the relevant citations). In this section, I would also encourage you to be explicitly clear as to what natural emissions will (and will not) respond to the climate component of the simulations (for example, I would almost recommend a short table). Furthermore, when these emissions are prescribed, please consider including the time period over which these emissions are prescribed (e.g., "based on a climatology from Year X to Year Y").

A table has been included and greater detail has been added to the descriptions for each model including the years of the climatologies where available.

## 2.1 Model descriptions

A comparison among models of natural emissions that may respond to the climate are shown below (Table 1). Where emissions are prescribed, the source is provided. Emissions that are interactive will respond to climate change. Further details on each of the Earth system models, including descriptions of the interactive emissions schemes and the tropospheric chemistry schemes are provided below.

	<u>Isoprene</u>	<u>Terpenes</u>	<u>Other VOCs</u>	<u>Soil NO<sub>x</sub></u>	<u>Lightning NO<sub>x</sub></u>
<u>UKESM1</u>	<u>Interactive (Pacifico et al., 2011)</u>	<u>Interactive (Guenther, 1995)</u>	<u>MACCity-MEGAN (Sindelarova et al., 2014)</u>	<u>Yienger &amp; Levy (1995)</u>	<u>Interactive (Price &amp; Rind, 1992)</u>
<u>GISS</u>	<u>Interactive (Guenther, 1995)</u>	<u>Lathiere et al. (2005)</u>	<u>Lathiere et al. (2005)</u>	<u>GEIA (Guenther et al., 1995),</u>	<u>Interactive (Price &amp; Rind, 1992)</u>
<u>MRI</u>	<u>GEIA (Guenther et al., 1995),</u>	<u>GEIA (Guenther et al., 1995),</u>	<u>Müller et al. (1992)</u>	<u>Yienger &amp; Levy (1995)</u>	<u>Interactive (Price &amp; Rind, 1992)</u>

**Table 1: Sources of natural emissions of ozone precursors. Where emissions are prescribed, the source is provided. Interactive emissions respond to climate change.**

Changes to UKESM1 description:

'Interactive emissions include isoprene, monoterpenes and; lightning NO<sub>x</sub> and soil NO<sub>x</sub>. Isoprene and monoterpene emissions respond to light and temperature and the isoprene scheme also includes CO<sub>2</sub> inhibition (Archibald et al., 2020a; Mulcahy et al., 2018). Isoprene emissions are calculated from vegetation productivity and increase in response to light and temperature (with an optimum at 40 °C). Emissions of isoprene are inhibited by CO<sub>2</sub> following the emission model of Pacifico et al. (2011). Lightning NO<sub>x</sub> is calculated using the parameterisation of Price and Rind (1992), which calculates a lightning flash density based on cloud-top height. Nitrogen oxide molecules produced per flash is 7.5 x10<sup>26</sup> for cloud-to-ground flashes and 2.25 x10<sup>26</sup> for cloud-to-cloud flashes. Secondary organic aerosols (SOA) are calculated as a fixed yield of 26% from gas-phase oxidation reactions involving monoterpene sources. Soil NO<sub>x</sub> is prescribed as an annual flux of 12 Tg, according to Yienger and Levy (1995) and other biogenic emissions are prescribed as monthly mean climatologies based on the years 2001–2010 (Guenther et al., 2012).'

Changes to MRI description:

‘Lightning NO<sub>x</sub> is interactive and based on a lightning flash density parameterisation (Price & Rind, 1992). A cloud-to-ground flash produces  $6.7 \times 10^{26}$  molecules per flash and a cloud-to-cloud flash produces  $6.7 \times 10^{25}$  molecules per flash. ~~but~~ Other natural emissions from land and ocean are prescribed as monthly climatologies, including isoprene and soil NO<sub>x</sub> (Deushi & Shibata, 2011). 15 % of natural terpene emissions at the surface form SOA and SOA have identical properties to POA.’

Changes to GISS description:

‘Lightning NO<sub>x</sub> is interactive as described by Kelley et al. (2020). The NO yield is  $1.75 \times 10^{26}$  molecules per flash ~~Natural emissions include soil NO<sub>x</sub>~~ Soil NO<sub>x</sub> is prescribed as a monthly mean from GEIA. ~~and isoprene emissions are interactive and, which~~ respond to light and temperature (Shindell et al., 2006) following the algorithm defined by Guenther et al., (1995). Monoterpenes are prescribed as monthly means from Lathiere et al. (2005) based on the year 1990. SOA are calculated using the CBM4 chemical mechanism to describe the gas phase tropospheric chemistry together with all main aerosol components including SOA formation and nitrate, and is calculated using four tracers in the model. Isoprene (VOCs) contribute to the formation of SOA (Tsigaridis et al., 2018).’

References:

Müller, J. F. : Geographical distribution and seasonal variation of surface emissions and deposition velocities of atmospheric trace gases. *J. Geophys. Res.: Atmos.*, 97(D4), 3787-3804, 1992

Yienger, J. J., & Levy, H.: Empirical model of global soil-biogenic NO<sub>x</sub> emissions. *J. Geophys. Res.: Atmos.*, 100(D6), 11447-11464, 1995

Sindelarova, K., Granier, C., Bouarar, I., Guenther, A., Tilmes, S., Stavrakou, T., Müller, J.-F., Kuhn, U., Stefani, P., and Knorr, W.: Global data set of biogenic VOC emissions calculated by the MEGAN model over the last 30 years, *Atmos. Chem. Phys.*, 14, 9317–9341, <https://doi.org/10.5194/acp-14-9317-2014>, 2014

Lathiere, J., Hauglustaine, D. A., De Noblet-Ducoudré, N., Krinner, G., & Folberth, G. A.: Past and future changes in biogenic volatile organic compound emissions simulated with a global dynamic vegetation model. *Geophys. Res. Lett.*, 32(20), 2005