Response to referee comments on “Seasonal isoprene emission estimates over tropical South America inferred from satellite observations of isoprene”

Sun et al.

We thank referee #1 for the helpful comments. Below we have responded to all individual comments. Our responses are written in blue. Text from the manuscript is shown italicized with new text added to the manuscript <u>italicized and underlined</u>. Line numbers refer to those in the marked-up version of revised manuscript with track changes.

This is a revised version of a manuscript on a study deriving isoprene emissions over tropical South America using the RAL IMS CrIS isoprene product and linear regression of GEOS-Chem predictions. The authors have made a number of edits that improved the clarity of the presentation, and I thank them for that. However, I think the paper still needs some work to more fully demonstrate the utility of this study to the broader community. I have two major suggestions and a few minor ones listed below, after which I would recommend publication.

Major suggestions:

1. The addition of the NOx sensitivity studies is an important one, but as presented it feels like an add-on to the paper. The authors discuss the sensitivities in a general sense in Section 3.3, noting that negative biases result in an overestimate of the derived emissions, and vice versa. However, it doesn’t seem that the isoprene emissions presented earlier in the paper actually take these findings into account. Given that the authors document model NO2 column biases as compared to TROPOMI (which they note are generally negative in the wet season and positive in the dry season), they should estimate the impact of these on their seasonal emissions. Ideally this would also be done spatially, given that the NOx biases (particularly in the dry season) are not necessarily collocated with the isoprene emission hotspots. It looks like the authors opted to apply spatially invariant NOx scaling factors in their sensitivity tests, but perhaps they could interpolate between these tests to come up with seasonal maps that represent their best emission estimate? This would be much more useful to the community than simply reporting a 20% uncertainty on the regional average emissions due to NOx uncertainties (since the local uncertainties could be much higher).

We have now added discussion about monthly satellite derived isoprene emissions using grid-dependent scaled NOx emissions at line 420:

> “<u>*Figure 6 shows monthly isoprene emission rates inferred from CrIS isoprene column data for which grid-dependent NO_x emissions are scaled using TROPOMI tropospheric NO_2 columns. These grid-dependent scaling factors are determined by monthly NO_2 column differences between GEOS-Chem and TROPOMI (Fig. S2). For example, for a model grid where the monthly TROPOMI NO_2 column is 75% lower than the corresponding GEOS-Chem value, we scale the model NO_x emissions by a factor of 0.25 (EmisScale_NO = 0.25). Using this approach, we account for the spatial distribution of model biases in NO_x emissions. As a caveat, scaling the NO_x emissions in the model in this way does not necessarily reflect the real emission biases. For*</u>

example, the influence of convection and advection are not considered in the distribution of atmospheric NO_x. Moreover, these scaling factors are calculated at the satellite overpass time and consequently do not represent any time-dependence in model bias. We examine the monthly simulated isoprene mole fractions with scaled NO_x emissions at the ATTO site. The scaling factor is 1.25 for March to May, 0.75 for July to September, and 1 for June and October to December based on the monthly differences between model and satellite NO_2 columns at ATTO. The resulting model bias is reduced for wet and dry months. For wet months (March~May), the mean model isoprene mole fraction is reduced from 4.2 to 3.8 ppbv, corresponding to higher NO_x emission levels, compared with the observed value of 2.9 ppbv. For dry months (July~September), the mean model isoprene mole fraction is increased from 3.2 to 3.6 ppbv, corresponding to lower NO_x emission levels, closer to the observed value of 3.6 ppbv. "

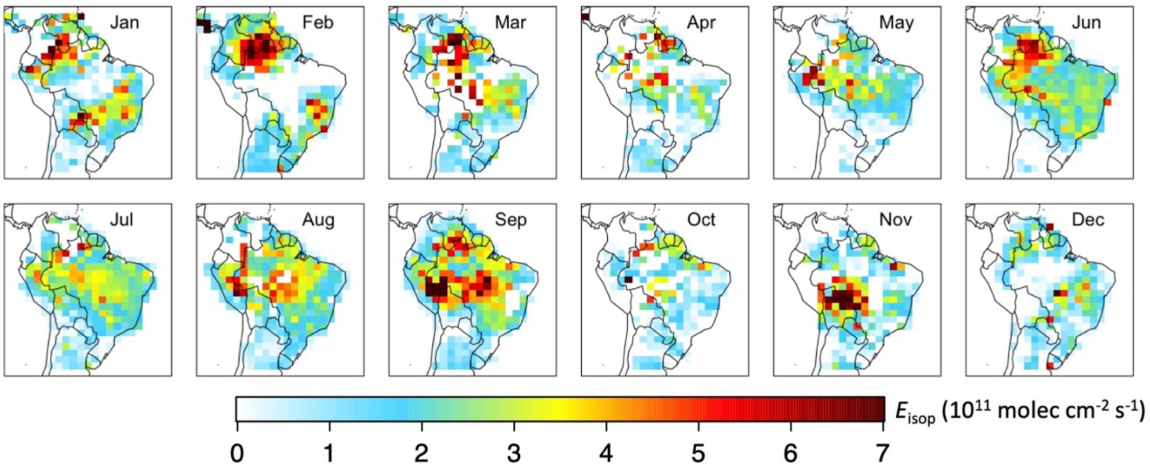

Figure 6: Monthly CrIS derived isoprene emission rates over tropical South America for 2019 using grid-dependent scaled NO_x emissions.

2. While I recognize the challenges associated with validating satellite measurements of isoprene, I still think this paper needs some type of evaluation of the actual CrIS columns, as opposed to only evaluating the model predictions with CrIS-based emissions. At the very least, the authors could add a secondary y-axis to Figure 3b and plot the CrIS columns sampled at the ATTO site? It would be good to know if the temporal variation of the CrIS data is the same as that observed in situ.

We have now updated Figure 3b in the main text where blue triangles denote CrIS isoprene columns sampled at the ATTO site. The monthly satellite retrieved isoprene columns generally follow the observed monthly variations at ATTO.

Minor suggestions:

Line 87-89: Fu et al. (2019) is also an optimal estimation retrieval approach.

We have revised accordingly:

> *"Fu et al. (2019) developed the first direct retrieval of isoprene using infrared radiance measurements from CrIS, using the MUSES algorithm which follows optimal estimation principles, while others have adopted other optimal estimation retrieval approaches (Palmer et al., 2022)."*

Line 158-160: The sentence added here is redundant to the one that precedes it. The main point that still needs to be added here is the fact that the RAL IMS columns are lower than ground-based retrievals does not indicate a low bias in the RAL IMS product.

We have revised accordingly:

> *"IMS column averages tend to be lower than those derived from surface-based observations where surface level concentrations are high as expected, which does not necessarily indicate a low bias from RAL IMS product."*

Figure 5b: As mentioned above, I find the presentation in this section to be too general, this figure included. I'm also a bit confused by the data in this plot. Are these examples for a single model grid box? It doesn't seem like the lines actually represent linear fits to the plotted points?

We have revised accordingly as below. We did check the linear regression models for the isoprene emission rate ~ isoprene lifetime relationships from each sensitivity test, and all the regression relationships have p-value less than 0.05 partly because the sample size is large enough. The majority of points are overlapped and thus the regression lines may help separate data points from different sensitivity cases. The regression lines here represent how the overall emission rate ~ lifetime relationships change under different NOx levels over the studied region:

> *"Figure 5b summarises the monthly E_{isop} and the corresponding isoprene lifetimes from all model grids over tropical South America, showing that lower NO_x emissions (EmisScale_NO < 1) generally have longer isoprene lifetimes and lower predicted isoprene emission estimates."*