Reply to reviewers

We thank the reviewers for their careful comments. We address their individual comments below.

One of the common comments was for a wider exploration of the observations using the back trajectories. This was not the motivation for the paper. The back trajectories were included as a tool to summarise the meteorology, and the emphasis was on using the observations to evaluate the CEDS emissions using the observations using a chemical transport model. There is currently other work ongoing in our group looking at these and other observations at the site through the lens of trajectories, but for this paper we have focused on exploring the consistency between the observations and anthropogenic emissions of ethane and propane. We have included comparisons between the model simulation and other species as a way of assessing the model performance in other areas however the emphasis is on the 2 hydrocarbons. We believe that this results in a useful conclusion that contributes to our understanding of this problem. We have updated the title and some aspects of the paper to reflect that emphasis.

REVIEWER 1

Page 2:

I recommend adding in a scale bar. Also, I recommend adding in another panel that details the island itself. This can include the size of the island (which is critical as the model simulations are coarse), where the site is, any local sources (?)

Response: We agree with the reviewer and have included an additional map for the area around the island and I have included a scale bar to indicate size.

Page 3:

I think the introduction doesn't motivate the full study yet. I would recommend including more information on the site and its importance and what has been found there. I would also recommend some discussion on biomass burning and anthropogenic emissions that are a focus of the study. This expansion would help to put the study in context of what is already known, what the gaps are, and how this study aims to fill the gaps. This point is one reason that I marked the scientific quality as "fair" as there isn't much of putting this work in context. I believe this can be strengthened in the next version.

Response: We agree with the reviewer and have included a paragraph of text outlining the relevance of the site to the wider Asian context.

Page 3:

These two paragraphs to me are results and could fit better in that section. I understand that the point of what impacts this site is important and can be needed in this introduction, however could that be done with previous literature in the introduction? I see there have been other studies,

perhaps could their findings be used in the introduction and then this shown in the results? Also, it would then help to have more details on the assessment in the methods (for example,. what is input met data and its spatial resolution?). Also, 10 days is quite long - how was that selected? etc.

I would then recommend adding in to the literature more information on what is known from the measurements at the site. This ties in my comment above.

Response: We've moved the discussion of the trajectories to the result section and have referenced that through the text.

Page 3:

I would recommend that you explicity describe why this is so important and what measurements here can tell about the region. That the measurements can be used to understand changes off the island is stated by referencing the COVID study, but I believe this point should be strengthened in the introduction.

Response: We've added some sentences to this paragraph highlighting the importance to the observatory and highlighting the work done to understand the impact of COVID.

Page 4:

What is used for electricity generation?

Response: Diesel generators (950 kW), Hateruma Wind Power (2x245 kW). We expect limited local anthropogenic emissions on the Island but power generation from diesel would influence NOx. We have updated the text accordingly.

Page 4:

I would recommend adding in additional information on these observations. What time resolution did you get? What time period did they measure? What QC/QA were applied? What is data capture rate (i.e. how many missing points)?

Response: We have included the fraction of data coverage in the table. All of the data was made available to us at a 1 hour resolution and we have included this in the caption. We have referred the reader to the references for QA/QC information.

Page 5:

This is only for some of the gases. Were the results similar for species that are more impacted by local emissions? I don't know these sources on the island, but as there is some electricity generation and people, perhaps then for NOx?

Response: We have assessed the differences for NOx and found only small changes between the simulations using grid resolution 0.5 x 0.625 and 0.25 x 0.3125. We have included this information.

Page 5:

Were these available at the same spatial resolution as the model or did you have to do anything to get on the same grid?

Response: THe CEDS emissions were available at 0.1 degree resolution. The HEMCO (Lin et al, 2021) module interpolates to the grid resolution of the model (0.5 degree x 0.625 degree). We have included this information in the text.

Page 5:

Is this of particular importance for this region? Or is it just to work towards more comprehensive emission inventory?

Response: We don't believe that this is of specific importance for the region but might help with the global underestimate in ethane and propane seen in other studies. We have updated the text to explain that.

Page 5:

Which emissions inventory from GEIA? I assume this is one on ECCAD, but it would help to state which one.

Response: Natural sources of NH3 are adapted from Global Emission InitiAtive (GEIA) as described by Bouwman et al. (1997). We have updated this in the text.

Page 6:

As the maps aren't very detailed, I googled to see the size of the island - it is tiny! Google says 6km lax length and 3km max width. The spatial resolution of the model is then ~25km x 30km. So the grid box is much larger than the size of the land even. I really think this needs to be explained more. Are the emissions then also at that resolution? If so, is the one grid cell then mostly emissions from the ocean and a little from the land? What about the met? As I noted below, the difference in the temp variability could also be that the air temp over the ocean would be simulated to have less diurnal variability - if the met was done at a higher spatial resolution?
I think the findings that the a lot of the measurements of this tiny island can be simulated with such a coarse resolution model is interesting and could be highlighted. But I do also think that this is a key part of the experimental set-up and a possible limitation that isn't discussed yet at all.

Response: We have made comments about the size of the island in the model configuration and have highlighted the potential problems.

Page 6:

I see that the time-resolution (e.g. hourly-averages, daily-averages?) for the comparisons isn't listed below nor in appendix. I recommend adding that to all. Also, I would recommend adding in information on the treatment of the observed data before the comparison. I noted above that I recommend adding in the method for QA/QC. However, I also recommend adding in here more on the observation data capture and if you had to use a data capture threshold (e.g. 80%) for comparing to the model if there were many missing points. Or did you have high data capture and so didn't need it?

Response: We have compared hourly averaged values from the observations and from the model in most species and have done daily means for NOx and NOy due to the high variability. We have included this information more clearly in the text

Page 6:

Just confirming that these are the same and not a typo?

Response: Those values are correct but we have changed the text for clarity.

Page 6:

It is hard to see the modelled in this graph. I also know it is hard to make such graphs look good. The green can't really be seen. Does it help to move it to the front? Or perhaps for the seasonal aspect would a wind rose perhaps show that they agree clearer? This is just an idea for authors to consider.

Response: We have updated the figures to make the model lines (green brought to front) clearly seen. We have also included a windrose in the appendix.

Page 6:

Could this also be due to the coarse resolution? Does the grid box for the site also go over water that would have less variation in air temp?

Response: We agree with the reviewer and have made it clear that this could be due to resolution impacts at different scales since the island is smaller than the model grid box at Hateruma - reflecting local heating and cooling at the observatory, and more regionally from the impact of the island.

Page 6:

What is the local environment? So more explanation of this may also help. Also, as I noted above, what about the coarse resolution and the other parts of the grid box that you are using for this comparison.

Response: We have updated the text to refer to the sub-grid issues of the meteorology being on a fairly coarse scale and the observations being local.

Page 8:

It could help to colour code by month to see this point more clearly.

Response: We have updated the images with colour code to show this.

Page 10:

I recommend updating this caption and below to "mean daytime values" or similar, as to me daily means 24-hr average. Also, why are the observed dots and the modelled lines? They are the same averaging time, correct? It is harder for me to compare the dots to the lines.

Response: We apologise for the confusion and we have updated the text to clarify this.

Page 10:

Is it daily or day-time? I recommend being specific with words so readers don't get confused.

Response: For NO_2 we have compared the 24hr-averages between the model and observation and have updated the text appropriately.

Page 10:

Should this be "the observed values"?

Response: We have updated the text with the suggestion.

Page 11:

Which species does the model not include?

Response: NO_y described here does not include NO3, Halogen nitrates and nitrites, organic nitrates which are not significant at the site. The model overestimates NOy so this small inaccuracy in the model comparison is not going to cause a change in the understanding of the problem.

Page 11:

Do you have any idea how much this could impact the observations?

Response: We don't have a clear understanding for this instrument. But this can be an important impact for some instruments (see for example https://acp.copernicus.org/articles/16/4707/2016/)

Page 11:

As worded, this seems to be a hypothetical statement. But if this simulation was run, then you did see the change, correct? If so, I recommend rewording.

Response: We have reworded this statement for clarity.

Page 12:

missing parenthesis

Response: We have updated the text.

Page 13:

I recommend expanding on this. Is it referring to the quality of the observations?

Response: We have updated the text to clarify that we are referencing the quality of the NASA generated meteorological field used to drive the model and their use of data assimilation to provide global meteorological information which is as close as possible to the observations.

Page 13:

I recommend adding here what time period this is for. 1 Jan 2018-31 Dec 2018 I believe?

Response: We have made that update.

Page 13:

I believe this is "overestimated"

Response: We have changed the text to reflect this.

Page 14:

Isn't the largest related to NOx and NOy? I understand why the experiments focus on biomass burning, but I would recommend re-writing this transition as the largest deviations are in NOx and NOy.

Response: We have updated the text to say that "although the largest model failures are for NOx and NOy species there is uncertainty about the accuracy of these measurements at such low concentrations. We therefore focus on the hydrocarbons which we have more confidence in."

Page 14:

This would impact on all species, not just ethane and propane. I think the transition and motivation could be improved to motivate why these were selected.

Response: Given the short lifetime of NOx (~ day) and the distance between the biomass burning sources and the sites (100s to 1000s of km), NOx concentrations will be less sensitive to biomass burning than say CO, ozone, ethane and propane.

Page 14:

What are the burning seasons for these? When/what seasons would you expect these to have an impact on the site?

Response: We have added some sentences to the text to outline the seasonality of biomass burning in the North and the South of Asia.

Page 14:

either

Response: Corrected

Page 14:

I recommend adding the coordinates of these domains in the text or caption. Also, it would help to keep the star for the location of the island - is it in N Asia? Do you also turn off its biomass burning emissions (are there local emissions?)

Response: We have added these coordinates to the text. The site represents the mid-latitude between the North and South Asia in our domain. Yes, the local emissions in the high resolution domain were also turned off. We have zoomed in a bit on the map and included a star for the island.

Page 14:

Why did you only look at these pollutants? NOx is also impacted by biomass burning.

Response: As mentioned earlier, we have more confidence in the observations of the hydrocarbons. And given the short lifetime of NOx, the site is likely less sensitive to biomass burning NOx than it is to CO, ozone, ethane and propane.

Page 14:

"at the site"

Response: We have updated the text to correct this.

Page 14:

From figure 2 is seems like the majority of the back trajectories in summer are from South Asia domain, correct? Looking at South Pacific and Philippines and SE Asia. Yet, the N Asia also has a large impact. I would recommend adding in the discussion the links between these findings and the prevailing region where the air masses come from during the seasons.

Response: The North Asia impact is consistent with periods when strong winds are passing over the island which draws air rapidly from the North. While these events happen in a shorter period, Figure 2 (b) only shows a weekly averaged contribution of air masses. However, we have included Figure A7 in the Appendix from the back-trajectory analysis to show the period where North and South both made significant contributions and explained this in the text.

Page 15:

I am not following this. If N Asia is turned off and we see a spike (so large decrease) that means a typhoon would have rapidly brought in more air from N Asia but we are missing it because it is turned off? Could it also be that there was an increase in fire activity? I recommend expanding on this statement as it seems key, but at least for me, I am not fully understanding the link.

Try and explain this in more detail.

Response: Yes, the large decrease represents the biomass burning emission that was missing when N Asia was switched off which could have rapidly been brought to Hateruma. Also, there might be an increase in fire activity in the same period. We have updated the text to explain this.

Page 15:

The scale is quite large in Figure 5, it is not really easy to tell how much higher the green is to the black in Figure 5 itself.

Response: We have updated the plots in the figures with smaller linewidths to make it easy to see the gap in plots.

Page 15:

"...the overestimate during the summer...", correct?

Response: Yes, the paragraph (line 238) is about the overestimate during the summer. We have updated the text to include this.

Page 15:

The overestimate in the summer, correct?

Response: Yes, we have updated the text to include this.

Page 16:

the contributions from these countries (esp Russia and inland china) seem to decrease dramatically in ~March. Why include April - June here when it seems from Figure 2 that their contribution is smaller in that period?

Response: Here, we aim to choose two periods in the year when there was almost a complete switch in wind flow from north to south and vice versa. Contributions from Russia decreased in March, however in the text we are referring to the period when the impact of polluting regions in Northern Asia fades and southerly wind dominates (this only happened after June from Fig 2). Similarly, for the period where northern-flow dominates, the regions in Southern Asia have significantly less impact only after September (Fig 2).

Page 16:

Why redefine the seasons here? They are defined differently in Figure 2. I recommend they are defined similarly across the manuscript. Here, you can use different words rather than seasons.

Response: We have updated the text in this section to avoid inconsistencies and used different words to describe the two periods in this section as wintry-months (October - June) and summery-months (July - September) which is different to winter and summer in earlier sections.

Page 16:

And not in the parent domain? This then limits which "Asian" emissions you are looking at and doesn't then include much of Russia (looking at Figure 1). This is a key point and a limitation that I think should be expanded on.

Response: We agree that our definition of 'Asian' here isn't correct. We have changed the text in to redefine this as 'in-domain' rather than 'Asian'

Page 17:

Is this really "background" isn't it "everything else" as the "Asian" domain is only part of Asia, and then there are anthropogenic emissions more locally and from outside of the high-resolution domain, and then also natural sources.

Response: We agree with the review that the term 'background' here might be misinterpreted and so have renamed this 'Rest of the world'.

Page 17:

I recommend this is explained more. If you are solving for A, then to me it looks like you have 2 equations with 3 unknowns as only the simulated X is known in each. Perhaps one more sentence to explain this would help readers to fully follow this.

Response: We've added more explanation into the text here.

Page 18:

insert ","

Response: Updated in text

Page 18:

Is this scaled experiment 1 or 2? I recommend adding this to the caption and also changing the name of the green line to also note which scaled experiment.

Response: This is the scaled experiment 2. We have updated the label and caption in the text.

Page 18:

Is this overestimate more than in the baseline simulation shown in Table 2? As they are different time periods, it is hard to compare if it makes the overestimate worse. Also, do the anthropogenic emissions have a seasonal cycle or are they more or less constant over the year? If they were constant, then this overestimate would have been expected, correct?

Response: The reviewer is correct. During the summer months, the model after the correction, now overestimates the concentration more than in the base model. However for most of the year and notably in the periods when the concentrations are high the model now gives a better simulation of the hydrocarbons.

The CEDS anthropogenic emission files provide annual gridded emissions data that were assigned seasonal patterns by month following Hoesly et al (2017). However, this seasonality is relatively small and so as the reviewer says this overestimate in the summer is to be expected. We have updated the text to reflect this.

Page 18:

scale experiment 2?

Response: Yes. We have updated this in the text

Page 19:

What was the seasonality of the emissions used here? Also, what about different contributions from anthropogenic vs biomass burning during different seasons? The summer overestimate does make it seem like a few things are overestimated (e.g. biomass burning and anthropogenic from this small domain, plus more??) in summer.

Response. The seasonality for the anthropogenic emissions is given by Hoesly et al. (2017). With higher emissions in winter than in summer due to increased combustion. We have updated model description text. The biomass burning emissions are given (GFED provides daily emissions based on satellite observations of fires). The reviewer is right that the fraction contributions from anthropogenic vs biomass burning varies during the year but the summer time VOCs overestimate appears to be a consistent feature. Given the change in air mass origin between the summer and the winter it is hard for us to distinguish a problem with the annual mean anthropogenic emissions from the North and the South of the domain vs error in the seasonality North and South of the domain. We have made this clearer in the text.

Page 19:

I recommend being very precise in the discussion here. As it seems this is only the high resolution part of the "North Asia" and not the full "North Asia" domain defined in the biomass burning section. To avoid confusion and to be precise with the results, I strongly recommend using very precise language to describe exactly what you are referring to.

Response: We've updated the text with a more precise description. "North Asia" replaced with "North Asia in our high resolution simulation domain"

Page 19:

Also here in Asia?

Response: Dalsøren et al. (2018) focused on the Northern hemisphere but doesn't make specific comments about regions.

Page 19:

Also here? Or are you pointing out this is a global problem? But all with the same order of magnitude?

Response: Yes, Dalsøren et al. (2018) identified this as a global problem with the current generation of bottom up inventories. The results here are roughly consistent with the corrections that they suggested to these inventories. We have updated the text to reflect this.

Page 19:

Why did you look at July? Was the emission impact the largest? Also, perhaps would you see a larger impact in peak ozone as average ozone also has all the night in it?

Response: We have added text to say that this is the period of highest ozone concentration over China. We could go hunting for a larger impact in the model but we believe that discussing the monthly mean ozone provides the simplest metric for the impact.

Page 20:

average July concentrations

Response: We have updated this in the text.

Page 20:

But not in all. NOx doesn't seem to have the same seasonal cycle (the others seem to have low levels in summer) nor does it seem to have a seasonal cycle at all (Table 2).

Response: We have updated the text to clarify that NOx do not have a seasonal cycle from our observation.

Page 20:

Was NOx not impacted?

Response: We have updated the text to clarify the impact of biomass burning on NOx is small <10%.

REVIEWER 2

Major Comments:

1. The number of figures in the paper is excessive for the analysis presented. Section 3 is the area that needs the most work; moving the time series to the appendix, consolidating the scatter plots into a single figure, and summarizing the quartile comparisons between the model and observations as a figure will focus the discussion considerably. I would also encourage a seasonal analysis of the data presented in the model/observation comparisons, which is interesting scientifically and will better motivate your use of a seasonal split in your later analysis.

Response: As this is the first time these observations have been presented we would suggest that there is some utility in showing these as a time series in the main body of the paper as we believe them to be informative to a wider audience.

Table 2 presents a seasonal analysis of the model performance and we have extended the discussion here to provide some more discussion in section 3 to ensure that there is more discussion of the seasonality.

2. Figure 2 and the back trajectory calculations are presented in the introduction but should be moved to the results. As the paper currently stands this figure is only used to motivate analyzing the winter data separately from the summer data in the VOC scaling simulations. However, leveraging these back trajectory groupings to get at source signatures can be very interesting for this type of receptor site analysis. For instance, when you get airflow from the oceans (North and South Pacific), does the model capture the background ozone adequately? Do you see ratio changes between ethane and pentane based on air mass source?

Response: We have moved the trajectory discussion to a results section as suggested.

We agree that another way to analyse the observational data would be through a more detailed analysis of the trajectories as suggested by the reviewer. A number of potential avenues exist here, ratios of Ethane and Propane to each other (as suggested) but also the their ratios to CO and the ratios of NO_y to CO etc. However, we think this is outside of the scope of this paper which is primarily aimed at understanding the concentrations from the perspective of a chemistry transport model and whether they are consistent with the current generation of emissions inventories. The trajectories are primarily there to provide meteorological context rather than as an analysis tool. We have made some comments on the underestimate of ozone in the summertime in the text of the paper and we have now tied this more strongly to the trajectories.

3. The Biomass burning analysis is interesting but feels incomplete. Is there a reason that there wasn't a run with no biomass burning at all? It seems like it would have helped clarify the behavior in Figure 12. This section is also very focused on the ethane disagreement, but biomass burning sources will influence all your chemical variables; does the biomass burning have any influence on the NOy modeling issues? Aging of smoke will also change the chemical characteristics of the measured air masses, and you have a dataset that allows some investigation on the impact of aging during transport. Motivating this section more thoroughly in the introduction will help structure the analysis here.

Response: Biomass burning impacts all of the species (Figure 7) however there are significant differences in the magnitude of the impact of these emissions due to their lifetimes. We have extended Figure 7 (based on comments from Review 1) to include other species and to highlight this.

4. I struggled with section 5 (scaling the anthropogenic emissions) and would like to see this section clarified. This is another area where leveraging the back trajectory groupings would have been useful in the calculations (i.e. defining the background). While the overall outcome of scaling up the emissions seems to have solved some of the magnitude issues in the model comparison, there's quite a bit more here that can be probed with respect to sources. This is an area where trace gas ratios coupled with back trajectories can clarify some of weirdness in the model/observation mismatches and help really get at where the

model is struggling. I was also curious if the % changes in OH and Ozone you show in Figure 16 match observations.

Response: We have re-written this section to clarify the mathematical approach used here. Whilst we agree that more work could be done in the future using the trajectories we believe it is outside of the scope of this paper which is attempting to provide an analysis oft he consistency of the current generation of atmospheric emissions used in atmospheric chemistry transport model. There are significant problems with lagrangian methods (difficulties in representing mixing processes, difficulties in the consideration of background etc) and in the future an adjoint approach to these data may allow for a more sophisticated methodology to assess the spatial error in the emissions more rigorously. We have included a discussion of this as a potential future direction in the conclusions.

We are unaware of how we could assess the % changes from observations. There are limited observations available (especially of OH) and it is unclear how the available observations could be used to undertake the analysis suggested.

5. In general I encourage the authors to look at the use of trace gas ratios in their analysis. The ethane/pentane ratio is a published metric, and even ratios to CO can be very helpful for understanding transport and aging. If CO2 is available for this time period, CO to CO2 ratios can be extremely useful in receptor site analysis and has been used previously in East Asia air quality research.

Response: We agree that there is additional information available from the ratio of these species but the objective of the paper is the evaluation of the consistency of the emissions datasets with the observations through the use of the GEOS-Chem model. We have changed the title and abstract of the paper to reflect this.

Technical Comments:

1. In general, I would like to see more details in the experimental section. You need more information in your measurement table (frequency of measurement, uncertainties, LOD), and I would have liked to have seen a summary table of the simulations run. For instance, I had missed that there was a no-shipping traffic run.

Response: We have included such a table into the Paper.

2. Check the layer order on figures; in the time series figures you occasionally swap which line is on top.

Response: We have gone through the paper and ensured that the model line is always the top line.

3. There are superfluous figures in this paper; I recommend moving all of the time series comparisons to the appendixes and consolidating scatter plots into single figures (ie,

section 3). I would also recommend consolidating your two map figures into a single map or moving the biomass burning domains to the appendix.

Response: As suggested by reviewer 3 we have combined Figure 4,5,6,7 and 8 into a single figure.

We have moved the biomass burning domains into the appendix as suggested.

4. The paper needs a brief editing pass for grammar and spelling.

Response: We have been through the paper again for grammar and spelling and apologise for any difficulties here.

5. The scatter plots need to have labels for the 1:1 lines.

Response: We have included a description of the two lines into the figure captions.

6. I would also recommend adding subfigure lettering, following the journal guidelines.

Response: We have included subfigure lettering where appropriate.

7. In Figure 12 I recommend adjusting the legend labels for clarity.

Response: We have made the legend labels larger and included information into the figure caption for clarity.

8. Please be explicit about your time period of study.

Response: We have been more explicit about the time period of the study in the text of the paper.

9. One of the points made in the introduction is the use of geologic sources ethane and propane; was this a novel addition to the model? It isn't clear from the text, but this seems like a neat addition that wasn't probed further in the modeling.

Response: The geological source is a new addition to the model but the contribution to the global total is small (3.0 Tg and 1.7 Tg annually for ethane and propane respectively). We have clarified this in the text.

REVIEWER 3

1. How do the observed levels and seasonality at Hateruma compare with other background sites in Asia (oceanic regions or high-altitude mountains)?

Response: We have included some additional information about the comparison with other clean oceanic sites in the introduction. High altitude mountain sites are unlikely to provide a useful comparison as they have a distinctly different set of processes

controlling their concentration; we have thus not provided any additional information here.

2. Air mass trajectories and observations should be analyzed more deeply to identify regions where emissions are underestimated, instead of considering a general increase across Asia.

Response: We have re-emphasised in the paper that the objective is to assess whether the current generation of anthropogenic emissions inventories, and specifically for the small hydrocarbons, are consistent with the observations made at the site. As suggested by the reviewers there are other approaches that could be made to extract information from these observations but they lay outside of the objectives of this paper.

3. How well the atmospheric dynamics/transport has been captured by the model (e.g., in comparison to ERA5 reanalysis). Describe any nudging that has been considered to limit the errors.

Response: GEOS-Chem is an offline model and thus directly uses reanalysis data (the NASA Goddard GMAO MERRA reanalysis in this case) as the meteorological input. The MERRA reanalysis is the GMAO equivalent product to the ECMWFs ERA5. There is no 'nudging' as the model doesn't calculate its own meteorology. We have emphasised this point in the model description. Figure 3 compares the local observed meteorological variable against those in the model.

4. Section 3.1: l.129: does the high-resolution simulation tend to better capture the local environmental effect?

Response: The high resolution simulation is not better. The limitation in size of model grid box compared to the tiny island has been discussed in the text as suggested by reviewer 1.

5. Figures 4-5-6 can be combined into a single figure with 3 rows, similarly figures 7-8-9.

Response: We have combined figures 4,5,6,7 8 and 9 into a single figure as suggested.

6. Manuscript needs a language check/improvement; e.g., replace "NOy observations are in overestimate by model" by "NOy is overestimated by the model"

Response: We have been through the paper again for grammar and spelling and apologise for any difficulties here.

7. How much the correlations between the model and observations (r values) change when emissions are tuned?

Response: We have included this information in the paper.

8. NOx/NOy bias seems the major limitation of the model at Hateruma island; is this also the case with other stations in Asia!

Response: We have included a discussion of the previous literature on this topic into the paper.