Final response to comments on "Analysis of CO₂, CH₄ and CO surface and column concentrations observed at Reunion Island by assessing WRF-Chem simulations"

First and foremost, we want to express our gratitude to the referees for taking the time to read our article and provide constructive feedback. We will reply to their comments below. The comments given by the referees are written in black, while author comments are in blue. Modifications to the manuscript are written in *italic*.

Referee 1

Main comment:

I would recommend that the discussion of those findings the authors would assign the highest importance should be enhanced. Currently, the conclusions read like a long listing of various minor items – all correct, most of them interesting and worth to mention – but altogether leaving the reader somewhat with the impression that the study lacks a scientific focus.

We propose to rewrite the complete conclusion section such that the questions from the introduction are explicitly addressed. Below is our alternative conclusion (to replace lines 529 - 560).

Nevertheless, the results enable us to answer the scientific questions posed in the introduction.

(1) To what extent are the observations influenced by local and nearby sources and sinks, or long-range transport of emitted gases?

At both Saint-Denis and Maïdo, the in situ observations are heavily influenced by local and nearby sources and sinks, especially for CO_2 . However, the in situ observations at Maïdo can detect both signals from the coastal regions and from afar at night, when the Observatory is located in the boundary layer and the free troposphere, respectively. On the other hand, the column-averaged mole fractions describe different air masses than those near the surface and are not or only very slightly influenced by local activities. As shown by previous studies and confirmed here, these measurements at Reunion Island are influenced by processes from distant areas such as Africa and Madagascar. This is further evidenced by the fact that a model resolution of 50 km appears to be sufficient to simulate these observations.

(2) What are the different tracer contributions to the observed concentrations, both at the surface and in the total column?

The surface CO_2 mole fractions in Saint-Denis follow a distinct diurnal cycle with values up to 450 ppm at night, driven by local anthropogenic emissions, planetary boundary layer dynamics and accumulation due to low wind speeds. Additionally, the signal includes respiration from vegetation that is carried by eastern winds from more rural regions. At the Maïdo Observatory on the other hand, a similar diurnal cycle of CO_2 is found but with much smaller amplitude. There, the surface CO_2 mole fractions are essentially driven by the surrounding vegetation that take up CO_2 during the day and release CO_2 during the night through respiration. The different model tracers of XCO_2 show contributions from fire emissions during the biomass burning season, but also positive biogenic enhancements associated with the dry season. For CH_4 , tracer contributions reveal that the emission sources within the model domain have only a minimal effect on the overall signal. Besides the background, local anthropogenic fluxes are the major source influencing the in situ observations at Reunion Island. However, the comparisons between the model fields and observations at Saint-Denis show that the anthropogenic emissions from EDGAR are likely largely overestimated; this is even more evidenced at Maïdo. Some (minor) impacts from Africa and Madagascar can be seen in the XCH_4 observations: fire plumes during the biomass burning season and wetland emissions during the rainy season. For XCO, the importance of biomass burning plumes from Africa and elsewhere for the observed variability is confirmed. These plumes can also be detected by the in situ observations at Maïdo at night, while local anthropogenic signals are the main influence during the day.

(3) How accurate is WRF-GHG in simulating the different observation types of the three gases (CO₂, CH₄ and CO) in the Southern Indian Ocean region, in particular at Saint-Denis and at the Maïdo Observatory? What are its strengths and weaknesses?

In general, WRF-GHG shows great skill in simulating the different in situ surface and column observations of GHG. The simulations of XCO₂ and XCO show a high correlation with the TCCON data with coefficients of 0.9 and 0.89, respectively. Similarly, a Pearson correlation coefficient of 0.9 and low errors are found between the model and NDACC XCO time series. Further, WRF-GHG is able to adequately reproduce the in situ CO observations at Maïdo, and consequently to some extent the anabatic winds that are typical for the northwest part of the island, despite the differences in modeled and observed wind directions. The high model resolution of 2 km is needed to accurately represent local fluxes and small-scale processes that affect the in situ observations. However, because of the complex topography and the unique local wind patterns, an even higher resolution might be needed to simulate more precisely the observations at Maïdo. In addition, certain model flaws were discovered in this study. Due to an overestimation of local wind speeds in the capital, WRF-GHG underestimates the nocturnal CO_2 buildup, leading to a correlation coefficient of only 0.62 between the model and surface CO_2 measurements at Saint-Denis. Further, we found a small model underestimation of the amplitude of the diurnal cycle of surface CO_2 at Maïdo which might indicate that the VPRM parameters could be improved for this region. Finally, WRF-GHG fails to reproduce accurately the different CH_4 observations at Reunion Island due to a seasonal bias in the background arising from the CAMS reanalysis.

Minor comments:

Paragraph starting line 33 – this paragraph details a bit on strengths and weaknesses of remote sensing (NDACC and TCCON) and in-situ measurements. The more recent COCCON network should be mentioned here, as it aims to fill in the gap between global and small-scale measurements (allowing arrangements of portable spectrometers for observing dedicated areas of interest as, e.g., metropolitan areas). See

https://amt.copernicus.org/articles/8/3059/2015/

https://acp.copernicus.org/articles/19/3271/2019/

https://amt.copernicus.org/articles/14/1047/2021/

The following sentence was added into this paragraph at line 41: "Recently, this kind of observations from mobile low-cost FTIR spectrometers within the Collaborative Carbon Column Observing Network (COC-CON) has been used to constrain fluxes in urban regions (Hase et al. (2015), Makarova et al. (2021), and Vogel et al. (2019))."

line 54: "near the surface, winds . . . originate in" \rightarrow "near the surface, air masses . . . originate in" Done

Line 59: "However" \rightarrow "However," Done

Line 69ff: Here, WRF studies in the context of city emissions conducted by other groups should be mentioned here as well, e.g.,

https://acp.copernicus.org/articles/19/11279/2019/

https://acp.copernicus.org/articles/21/13131/2021/acp-21-13131-2021.pdf

The first article by Zhao et al. (2019) suggested by the referee is already mentioned in the text (line 71). The second article by Jones et al. (2021) which assesses urban methane emissions using FTIR observations is indeed very interesting. However, the study uses the Stochastic Time-Inverted Lagrangian Transport (STILT) model and not WRF. Therefore, it won't be added to the references in the introduction.

Paragraph starting line 128: this explanation has some redundancy (FTIR observations ... providing mole fractions in an atmospheric column The spectra are used ... to retrieve the total column-averaged dry-air ...)

It is correct that both sentences are very similar. However, they were written for a different purpose.

The first sentence aims to indicate in general the difference with respect to the in situ measurements: the FTIR observations measure a signal along the solar path of an atmospheric column. The second sentence aims to be more specific about the product that is retrieved from these FTIR observations and further used in this study: the total column-averaged dry-air mole fraction (the so-called Xgas).

We deleted the first sentence in the manuscript and additionally mentioned the location of the FTIR instrument next to the PICARRO analyzer. The paragraph starting at line 129 now becomes:

"In September 2011, BIRA-IASB installed a high-resolution Bruker IFS 125HR FTIR at STD, next to the PICARRO analyzer. This instrument is primarily dedicated to measuring the near-infrared (NIR: 4000 - 16000 cm⁻¹) spectra and contributes to TCCON (Wunch et al. (2011)). The solar spectra are used to retrieve the total column-averaged dry-air mole fraction of CO_2 , CH_4 and CO (De Maziere et al. (2017))".

Paragraph starting at line 147: The authors correctly stressed earlier the tying of TCCON results to WMO standards. The systematic error budget of NDACC vs TCCON should be mentioned here explicitly (as both data sets are mixed together later in the discussion) – which amount of bias between the two remote sensing data sets might be expected?

The differences between NDACC and TCCON data are mentioned later in the article, in the results section. First the TCCON results are discussed, and afterwards the NDACC results are examined. At the latter, we tried to refer back to the TCCON results and justify the observed differences. For XCH₄, this is done at lines 478-479 by referring to Zhou et al. (2018). They showed that XCH₄ at Reunion Island from TCCON is about 10 ppb lower than NDACC XCH₄ due to differences in vertical sensitivity. A more comprehensive study about the differences between TCCON and NDACC including 6 sites was made for XCO by Zhou et al. (2019). This is referred to on lines 502-504. They found that for XCO the bias between the two networks was below 2 % in the Southern Hemisphere, and that taking into account the smoothing error is important. More specifically, a systematic bias of 2.5 % was found between NDACC XCO and TCCON XCO at Reunion Island. We will adapt these lines in the article to be more explicit: Lines 478-479: "Zhou et al. (2018) showed that NDACC XCH₄ is generally about 10 ppb lower than TCCON XCH₄ at Reunion Island due to their difference in vertical sensitivity."

Lines 502-504: "This is probably linked to biases between the TCCON and NDACC data sets. Zhou et al. (2019) showed that there is a bias of 2.5% between TCCON XCO and NDACC XCO at Reunion Island, due to differences in the retrieval algorithm and data corrections."

Line 309: "agree less" \rightarrow "agree less well" Done

Figure 6 and associated discussion: Given the elevated altitude of the MA station, it might be worth to appropriately filter the in-situ data (selecting those data which are sampling free tropospheric air) and specifically discuss the resulting subset, see a study following this approach for the Izana station on Tenerife here: https://amt.copernicus.org/articles/7/2337/2014/

The study of Sepúlveda et al. (2014) mentioned by the referee compares the CH_4 mole fractions of several NDACC sites with co-located in situ observations, which are first filtered to represent regional-scale signals (of the free troposphere). The proposed filter for Izaña (which is a subtropical high mountain observatory, like Maïdo) is averaging the nighttime in situ observations to obtain a nighttime mean. The solar FTIR measurements, which can only be performed during the day, are averaged to obtain a daily mean mole fraction. This daily mean is then compared with the mean of the two in situ nighttime averages around that day.

We can recalculate the statistical metrics and remake the correlation plots of Figure 6 in the manuscript for the nighttime data at Maïdo, as suggested by the referee. Like this, the model performance of those observations that should be representative of the free troposphere can be evaluated. Figures A1, A2 and A3 here below show these results. For CO, the model performs equally well for all observations (day or night): both the root mean square error (RMSE) and mean bias error (MBE) show similar values when taking only the nighttime pairs (compared to all or only daytime). The correlation coefficients are all equal to 0.83.

As mentioned in the manuscript, a low correlation coefficient and larger errors are found for the model performance of CH_4 because of a seasonal bias in the background information. Here again, similar statistics are found when comparing daytime observations, nighttime observations or both together. However, the metrics seem to be slightly better for the daytime points compared to the nighttime points. This might be related to the incorrect spike at night in the anthropogenic tracer of WRF-GHG as shown in Figure 15(a) in the manuscript, linked to EDGAR emissions. It is not clear why WRF-GHG simulates higher anthropogenic values for nighttime points compared to daytime points, leading to a larger overestimation.

The plots for CO_2 in Fig. A3 also show similar statistics for all subsets. There is a small overestimation of the daytime points and a small underestimation of the nighttime points. In the manuscript we linked this to the biogenic tracer and suggested this might be caused by inaccurate VPRM parameters. The higher correlation during the day might suggest that the photosynthesis parameters are more representative than the respiration parameters for the biosphere at Reunion Island. Because of the large impact of local surface fluxes on CO_2 , the nighttime in situ observations at Maïdo are less representative of the free troposphere than the other two gases.

We added a few sentences in the manuscript (around line 440) clarifying that the analysis was made on the full dataset because no significant differences were found on any subset:

"Remark that all statistical analyses of in situ observations at MA are performed on the complete dataset. Studies at other high-altitude stations often filter only those measurements which are representative for the free troposphere (Sepúlveda et al. (2014)). However, analyses comparing only day- or nighttime data at MA showed no significant differences in the results."



Figure A1: Scatter plot of observed and simulated in situ CO at Maïdo, using (a) all paired data points, (b) only daytime points and (c) only nighttime points. The red line is the regression line while the black dashed line is the identity line.



Figure A2: Similar as Fig. A1 but for CH_4 .

Para starting line 373 discussing long-range transport. The findings reported by Frey et al. for the Gobabeb station (see section "influence of African biosphere") might be of special relevance here: https://amt.copernicus.org/articles/14/5887/2021/.

This is indeed a very interesting and relevant article. We would like to thank the referee for pointing out this research. We included the following paragraph linked to this study at line 379: "A recent study using a COCCON spectrometer at Gobabeb in Namibia showed that the African biosphere can impact the



Figure A3: Similar as Fig. A1 but for CO_2 .

observed XCO_2 signal there due to medium and long-range transport (Frey et al. (2021)). More specifically, they demonstrated that the carbon sink of the African biosphere during Austral Summer can be observed in their XCO_2 measurements while it is not (or to a lesser extent) visible in the timeseries at Reunion. Backward trajectories for one specific day in February 2017 revealed that this contrast comes from the sampling of different air masses. This might suggest that the influence of fluxes from the African continent on the air above Reunion is seasonally dependent: a high impact during the dry season and a lower impact during wet season. Backward trajectory simulations over a longer time period performed by Zhou et al. (2018) might suggest this as well, however more research is needed to proof this statement."

Statement line 380: Does this imply that remote sensing measurements are more useful than in situ measurements for studying long-range transport (possibly also reflected by the fact that lower model resolution is sufficient for achieving good correlation between model and measurements)?

This is definitely the case at Reunion, but elsewhere it is probably dependent on the specific location of the site. In general, long-range transport takes place in the free troposphere which is not sampled by in situ observations (which usually sample the planetary boundary layer). Only on rare occasions, such as during nighttime on mountain tops like Maïdo, do in situ observations sample the lower free troposphere and possibly long-range transport (Baray et al. (2013)). Remote sensing measurements provide mole fractions averaged over a complete atmospheric column and are therefore less impacted by local surface exchanges and variations in planetary boundary layer height, and representative for a larger area than in situ measurements.

Line 391: "in more details" \rightarrow "in more detail" Done

Referee 2

Main comment:

Some results warrant a more detailed discussion, especially the issue of the low correlation of CH_4 . Looking at Figure 6 (b) and (d) seems to suggest that there could be two apparent distributions for CH_4 that, if fitted separately, could produce much more reasonable slopes and improved coefficients. Have you attempted to separate the data based on external drivers that could explain the two distributions? The two apparent distributions visible in the scatter plot of in situ CH_4 can be linked with its seasonal cycle: observations show minimum values in December–February and maximum values in August–September. We attempt to split the data into two subsets: from November until April, and from May until October. This roughly corresponds with the wet and dry season, respectively. Observed CH_4 values of the first subset fall generally between 1780 ppb and 1800 ppb, while values in the second subset are generally larger than 1800 ppb. Furthermore, due to the seasonal bias in the background data, the error between WRF-GHG and the observations is larger in the months November until April. All this is visible in Fig. A4 and A5, where the scatter plots like in Fig.6 (b) and (d) are shown for both subsets separately.

The root mean square error (RMSE) and mean bias error (MBE) are larger for the November-April data compared to the May-October data. This is caused by the bias in the background data.

In the scatter-plot of the complete dataset as in the manuscript (Fig. 6), the slope of the regression line was 0.34 at Saint-Denis and 0.31 at Maïdo. The slopes of the subsets are slightly larger, see Fig.A4 and A5. (Remark that the limits on the x- and y-axis are now different for every panel.) We have not found such large slopes as one would visually expect (as indicated by the red lines in the figure of the referee comment). This is because the linear regression line minimizes the least-squares error and is often confused with the first principal component, which represents an orthogonal regression and would likely correspond with the suggested red lines.

We will add the following sentence in the manuscript at line 386: "Figure 6(b) and (d) show two apparent distributions in the scatter-plot. This is linked with the seasonal cycle of CH_4 : observations show minimum values in December–February and maximum values in August–September."



Figure A4: Scatter plot of observed and simulated in situ CH_4 at Saint-Denis, using (a) only data from the months November until April or (b) only data from the months May until October. The solid black line is the regression line while the dashed black line is the identity line.



Figure A5: Similar as Fig. A4 but at Maïdo.

Minor comments:

L1: The authors should consider changing the title as they report modelling result using WRF-GHG (passive tracer) rather than the version of WRF with active chemistry (WRF-CHEM). We have considered using WRF-GHG in the title instead of WRF-Chem. However, as WRF-Chem is actually the collective name for the many chemistry mechanisms that are available (the so-called chem_opt),

we decided to choose the more general term to increase the visibility of the article.

L14: please add that the Pearson's correlation coefficient was used here. Done

L67: please correct "etc..." Done

L83: please elaborate on what "..." refers to or just give the elements in the brackets as an example. Done

L225, Figure 2: please consider adding the information on the vertical resolution and top of the domain in the caption of Figure 2.

We added the following sentence to the caption of Figure 2: "All domains have 60 (hybrid) vertical levels extending from the surface up to 50 hPa.".

L309: please change to "... agree less well ..." Done

L337: Please elaborate on the assumption that there is "no vegetation within the city". A simple search of aerial photos of St. Denis reveals multiple parks and vegetation along the shoreline. Maybe the assumption is rather that the impact of local vegetation is negligible compared to fossil fuel combustion? We agree that this sentence might be confusing. We aim to say that within the model, there is no vegetation at the grid cell of the city and consequently no biogenic CO_2 flux. We don't say that there is no vegetation in reality but rather that the model sees it that way. The VPRM model used within WRF-GHG includes 8 vegetation classes: evergreen forest, mixed forest, deciduous forest, crops, grasses, shrubland, savanna and barren. These classes are mapped from 48 vegetation categories from the global land cover dataset SYNMAP (Jung et al. (2006)). The WRF grid cell at Saint-Denis is classified as "barren", which is the collective name for all SYNMAP vegetation categories without vegetation: "Barren, Urban and built-up, Permanent snow and ice". It is the only VPRM vegetation class that results in a zero biogenic flux. The next sentence in the manuscript, at line 338, points to the assumption within the model as being realistic because indeed the impact of local vegetation within the city is negligible compared to fossil fuel combustion, as the referee noted. We will clarify this in the manuscript as follows (line 337): "The biogenic CO_2 flux at the grid cell of STD is zero because the corresponding VPRM vegetation class is 100 % "Barren, urban and built-up": the model assumes that there is no vegetation within the city. Given that Saint-Denis is the capital city of Reunion Island and has plenty of anthropogenic activities, the impact of local vegetation is probably very small and these WRF-GHG fluxes appear realistic.".

L368: Is the statement related to the nighttime respiration true in general or here specifically for a local imbalance in the boundary layer?

The statement is true in general. However, we admit that the sentence might be confusing as respiration is not only limited to the night. We will drop the nighttime adjective. The statement is reasoned as follows: for a given grid cell, the biogenic CO_2 flux is positive when the respiration is larger than the gross ecosystem exchange. Such a positive flux will add a certain CO_2 concentration to the bottom layer of the biogenic tracer within that model cell. In the manuscript it is shown that the column-average of that biogenic tracer above Reunion is positive. This value is the result of the transport of this biogenic tracer all over the model domain. A positive value at a certain time suggests that in general, across the entire domain, the respiration is larger than the gross ecosystem exchange (in reality only across the upstream influence region, but as WRF-GHG is an Eulerian transport model, this kind of information is not provided).

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