Anonymous Referee #2

The paper compares wind and EM27/SUN data at 5 sites taken as part of the Munich Urban Carbon Column Network in August 2018 with a ¿400m resolution WRF model with emissions. The goal is top down verification of CO2 and CH4 emissions, that are challenging given that these are long lived species that are influenced by long range transport and also local sources. The analysis is detailed analysis is presented well and valuable particularly in identifying conditions of uniformity ion regional air masses when a "gradient" method is explored, that may be useful operationally for top down verification. The paper should advance GHG verification strategies.

I do have the following questions and concerns that demand further clarifications by the authors:

We thank the anonymous Referee #2 for their time and valuable comments to improve this manuscript. The questions posted are addressed in point-by-point replies below. The referee's comments have been repeated in black. The authors' replies are marked in blue and the edited contents of the manuscript are documented in red in tables below each comment. Moreover, we set the numbers of the figures in this revision as 'R' plus the numbers (e.g., Figure R1), while the figures in the manuscript are numbered with 'M' plus its numbers (i.e., Figure M1).

1. A more careful explanation of the CO2 bias would be useful as it appears to be constant and obviously a statement that it cancels out.

Response: Thank you for this comment. We have extended our discussion of the CO_2 bias, adding to the supplement (see Sect. S14). In this study, we consider cases of CO_2 and CH_4 separately, as even though they are both simulated by the WRF and CAMS models, differences in their flux spatio-temporal patterns will lead to different biases when comparing to our measurements.

The modelled total column concentration of CO_2 (XCO_2) is made up of three parts, the background contribution (Model.(X)CO_2_BCK, see Fig. R1), the enhancements induced by human activities (Model.(X)CO_2_ANT) and biogenic activities (Model.(X)CO_2_BIO). Each of these components could contribute to the model-measurement discrepancy. As discussed in the manuscript, the bias in the model-measurement comparison of XCO_2 could be attributed to three main causes: i) overestimation of the modelled background concentration from CAMS, ii) errors in concentration enhancements brought by anthropogenic fluxes, and iii) errors in simulated biogenic fluxes.

To understand the background-related cause in depth, we analyzed the variations in the time series of CAMS itself, and compared the modelled and measured values. This has been included in the edited supplement (see Sect. S14). As seen from the red and orange curves in Fig. R1, the day-to-day magnitude and variations in Model.XCO_2_BCK are mostly determined by its initialization (CAMS.XCO_2). For the simulations of background concentrations of tracer gas in WRF-Chem, it begins with initializing the 3-D concentration field at the very beginning of the simulation cycle (i.e. 30th July in our study) and it is updated via the lateral boundary conditions from the global fields at a 3-hour interval (using CAMS fields for both). On the basis of Model.XCO_2_BCK (orange), the daily-mean total column concentrations (green) vary slightly with the positive anthropogenic fluxes (Model.XCO_2_ANT) and the carbon sink from biogenic activities during the daytime (Model.XCO_2_BIO). The mean bias between CAMS.XCO_2 and Obs.XCO_2 ($\pm$ its standard deviation) is 4.8 $\pm$ 0.7 ppm. Even though the overestimation of anthropogenic emission fluxes from the inventory and the uncertainty in the estimation of biogenic fluxes by the model could contribute to the model-measurement bias, this shows that the overestimation of CAMS overall plays a dominant role in the magnitude of the model-measurement bias of XCO_2.

We also checked the vertical distribution of the model values from CAMS and WRF-Chem on 16, 17, and 22 August at 12 UTC (see Fig. R2). In general, the vertical distributions of CAMS CO_2 and the modelled CO_2_BCK from WRF are quite similar but differ slightly close to the ground level. This also indicates that the magnitudes and the vertical structure of background initialization of CO_2 (CAM.CO_2) play a decisive role in the modelled background (WRF.CO_2_BCK) and total concentrations. Furthermore, emissions caused by human activities (blue, Fig. R2) contribute to the total concentration (green) in the planetary boundary layer (PBL, below approx.

2 km). For the enhancements associated with biogenic activities (green curve), carbon sources from respiration contribute significantly to the total concentration of CO_2 near ground level, while air masses heavily influenced by photosynthetic uptake (with less CO_2) and coming from the outer domain play a key role at higher altitudes, especially on 22 August. This could explain the dip seen on this date (see the pink box in Fig. R1). The animation of biogenic concentrations over D01 attached in the supplement provides a visual perspective of this phenomenon.

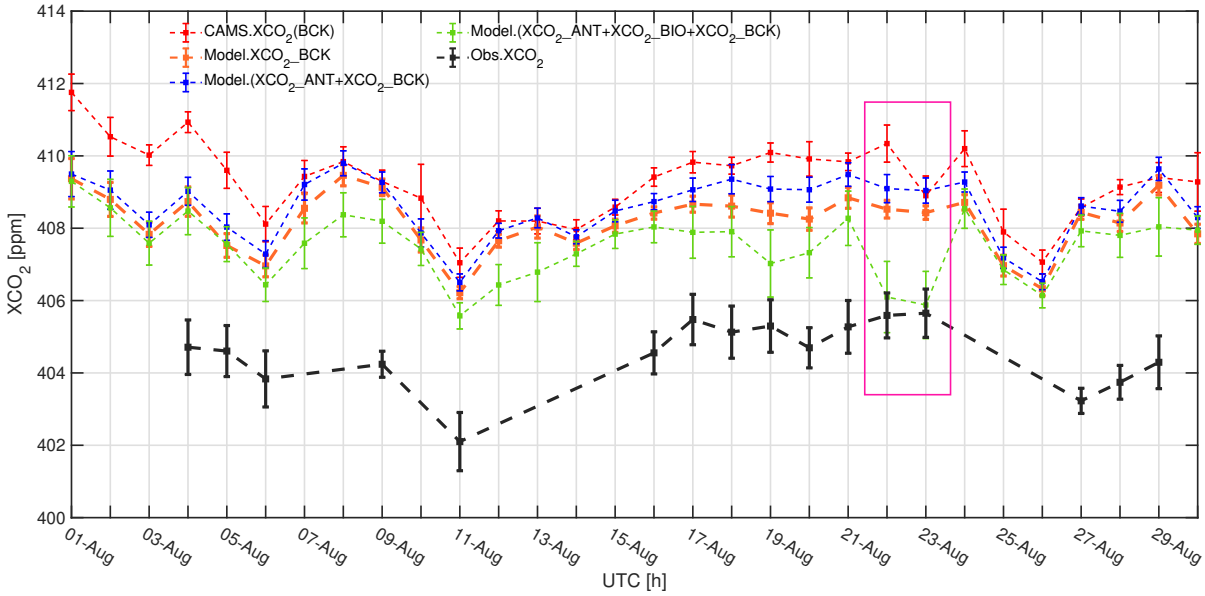

Figure R1: Time series of the daily mean measured values over five sites of MUCCnet (black) and the averaged modeled XCO_2 from CAMS (red, CAMS.XCO_2) and WRF over D03 during the daytime (i.e., 6:00 UTC to 17:00 UTC). The modelled column concentrations are pressure-weighted means (see Eq. 1 in the manuscript). The error bars represent the standard deviation of the simulated values over D03 and the measured over the five sites of MUCCnet. The orange curve represents the mean modelled column background concentration (Model.XCO_2_BCK). The green curve shows the averaged total column concentration (Model.XCO_2_BCK+Model.XCO_2_ANT+Model.XCO_2_BIO) and the blue curve shows the averaged column concentrations considering only the background and anthropogenic activities (Model.XCO_2_BCK+Model.XCO_2_ANT), without biospheric fluxes.

Owing to the relatively large bias of CO_2 brought in by CAMS, we considered using the model-measurement MB over all the measurement dates (i.e., 3.7 ppm) to "correct" the modelled values. This could help to see if the model could reproduce similar variations to those seen by the measurements. These variations are determined by the modelled biogenic effects, initial emission fluxes from the inventory, the modelled advection of air masses influenced by human and biogenic fluxes, etc.

However, this is not the case for CH_4, since no significant model-measurement bias can be found in the daily-mean XCH_4 (cf. Fig. 4(c) & (d) of the updated manuscript). Due to the quite weak biogenic activities of CH_4 in and around Munich (cf. Fig. 6 of the updated manuscript), the model-measurement bias of CH_4 is mostly caused by the uncertainties in human-related emissions.

To eliminate the CO_2 bias which could be mostly caused by the overestimation of background and to better observe the day-by-day variations in XCO_2 (see Fig. M4), we subtract the MB over the entire available measurement period (i.e., the 15 dates shown in Fig. M4; 3.7 ppm) from the modelled values for all sites and for each

available measurement date. In the manuscript, we chose to show 7 continuous days (from 16 to 22 August) as our key study period (see Sect. M4.3.2), while the rest is included in the supplement (see Sect. S7). Therefore, the MB discussed in of Sect. M4.2.3 is the remaining mean bias over these 7 consecutive days, after correcting with the value derived from the full 15 days.

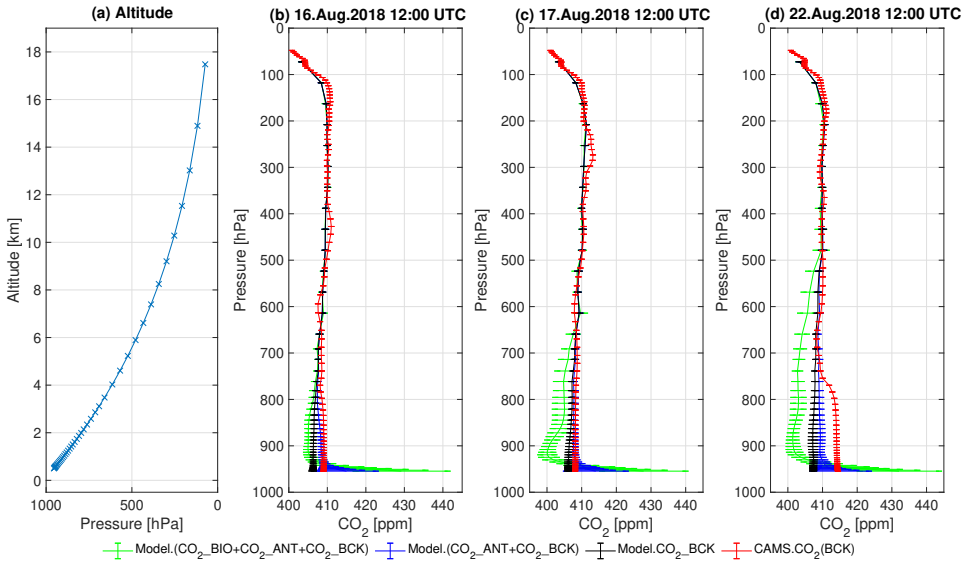

Figure R2: Vertical profiles of (a)Altitudes, averaged modelled CO_2 over D03 on (b)16, (c)17, and (d)22, August 2018 from CAMS and WRF-Chem. The red curve represents the values from CAMS, and the others stand for our model results, with green for the total values, blue for the sum of the background and the human-related enhancements, and red for the background.

To clearly state this cancellation in Sect. 4.2.3, we have added the following sentence:

Lines 331∼335	*As described in Sect. 4.2.2., a MB of 3.7 ppm in CO_2 has been found over all the available measurement dates (see Fig. 4), which is defined to be the difference between the smoothed and measured daily mean XCO_2 and the modelled values. To eliminate the bias (too high modelled background CO_2) and focus on the model-measurement differences due to other causes, this MB is subtracted from the modelled XCO_2 in the day-by-day model-measurement comparison for all sites and for each simulation date.*

2. Was CO measured with the EM27 as this would provide an independent constraint? If not then this should be mentioned as an additional valuable data to collect as new EM27's can do this together with CO2 and CH4.

Response: Thank you for this comment. CO is also measured by MUCCnet, but we did not include it in our modeling framework, and is outside the scope of the current study. In an ongoing project, we consider studying CO in the following step, and we have added this information in the conclusion and outlook of this study.

Lines 595∼596	*A study into the use of the simultaneously measured total column carbon monoxide (XCO) to constrain emissions from combustion processes can be carried out.*

3. For methane the EM27 measures the total column, including the stratosphere where it falls off. TCCON does correct for this using HF that unfortunately the EM27 does not measure. The gradient method and analysis

assumes this is constant and this should be clearly stated with citations (Saad et al). If this correction is not made the observations should be biased a low.

Response: Thanks you for this valuable suggestion and the recommendation of Saad's study. In this study, the HF correction was not implemented in the retrieval process of EM27/SUNs in MUCCnet. The HF correction is applied to correct the tropopause heights of the a-priori CH_4 profiles used in the retrieval and the effect of this corrections on the XCH_4 coefficient is verified to be small and well within the error bars (Geibel et al., 2012). Many thanks for your recommendations and we have added the information of this correction to the content.

Lines 236	*Additionally, the hydrogen fluoride (HF) correction (Saad et al., 2014) is not applied in our retrieval process of CH_4.*

The authors find a slight +ve bias "while in general the observed values are slightly higher, with a linear regression slope of 0.73 and a negative MB (-1.8 ± 4.0 ppb). This small bias could be caused by the initial and lateral boundary conditions from CAMS, or due to unknown or underestimated emissions" The possible reasons for this should be explained more clearly.

Response: From the perspective of the model itself, the bias in CH_4 concentrations is mainly attributable to the uncertainties in human activities. Bottom-up multinational emission inventories of CH_4 are generally compiled by scaling emissions using activity data and emission factors, which results in relatively large uncertainties (Bergamaschi et al., 2022). For the TNO_GHGco_v1.1 emission inventory used in this study, its point source information was collected on the location of power plants, large industrial installations, oil and gas production sites, airports and waste treatment locations (e.g. landfills), mostly from the E-PRTR (European Pollutant and Transfer Register) database. The data are valid for 2015. Therefore, in addition to the uncertainties due to the quantification of emissions in the inventory mentioned above, inconsistencies between emission information collected in 2015 or even earlier and actual emissions during the study period in 2018, could result in differences. Despite having chosen a high-resolution, state-of-the-art emission inventory, these uncertainties could contribute to the model-measurement differences. We have added the discussion related to causes of the uncertainties in the inventory to the manuscript as follows,

Lines 550∼557	*In this study, the modelled contributions from human activities are initialized with the emission fluxes from the emission inventory TNO_GHGco_v1.1 for the year 2015. The multinational bottom-up emission inventory holds large uncertainties, due to the large variability in spatiotemporal distributions of CH_4 emissions from different sectors in different regions that have not yet been fully captured by the emission inventory (Bergamaschi et al., 2022), the disaggregation from annual emissions to hourly values using temporal profiles and the temporal inconsistency of emission information from 2015 or even earlier than the study period in 2018. This could result in missing or underestimated emissions in the inventory, as suggested by the measurements. After delineating the areas where the uncertain sources could be located, they were further pinpointed based on the updated database and local knowledge.*

In addition to the uncertainties in the human emissions, we also extended our discussion of other potential causes in the CH_4 biases. The modelled meteorological fields could bring about errors in the advection, which would contribute to the biases of absolute methane concentrations (XCH_4) and further to discrepancies in the methane gradients. Furthermore, due to lags in the time it takes for air to reach the boundary, the variation of the background signals at each instrument could be large. This is definitely a key point to be considered when the concentration gradients are used for inversions to optimize the inventories (as in Jones et al., 2021), but it presents significant complexities for our study and its implementation within the WRF-based framework. In addition to the emission-related causes that lead to the model-measurement biases of concentrations and their gradients, Hedilius et al.(2017) pointed out that non-emission factors (like the mixed layer height and

topography) would further cause biased results. The importance of topography is verified as a significant factor in the variations of concentrations beyond the urban area. In our case, even though the elevations over our innermost domain (DO3, Munich) are rather consistent, i.e. around 550 m above sea level, the area around this domain's boundaries contains the complex topography of the Alps. It should be noted, that while it still plays a role, column measurements are less sensitive to mixing layer height than are in-situ measurements. The causes discussed here could contribute to errors in the concentration and thus, the gradients. We have extended the discussion other causes which could contribute to the biases in the model-measurement gradients as such:

Lines 540∼544	*In addition to the errors caused by the uncertainties in the initial emission inventory, other potential causes could contribute to errors in the concentration and thus, the gradients as well. The bias brought by the modelled meteorological fields can contribute to the bias of the modelled XCH$_4$, further to discrepancies in $\Delta XCH^S_{4,sla}$, by influencing on the advection (Wu et al., 2017). The variations in background concentrations at each site due to the lags in time when it takes for the in-flowing air to reach the boundary (as discussed in Jones et al. (2021)). Moreover, the non-emission factors (e.g., the mixed layer height and topography) could also introduce biased results results (Hedelius et al., 2017).*

4. There are many EM27 model studies of optimized fluxes such as Jones et al, Viatte et al that are cited. Another very relevant study Heerah et al JGR Atm 2022 that uses distributed EM27 data and WFR model to do systematic comparison with winds and inverse modeling for dairies should also be cited.

Response: Thank you for this recommendation. It is indeed a relevant study, and we have included a reference to it in the manuscript as follows:

Lines 86∼89	*For instance, highly-resolved meteorological fields can drive particle transport models (Fasoli et al., 2018). These Lagrangian footprints can then be used for inversion studies, similar to Heerah et al. (2021), who optimized dairy CH$_4$ emissions across the San Joaquin Valley using WRF-STILT inversions. Currently, an adapted Bayesian inversion model based on Jones et al. (2021) is being developed to infer anthropogenic CO$_2$ emissions, with the consideration of biogenic fluxes.*

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