

Dear Editor, and referees,

We appreciate the time and effort that you dedicated to providing feedbacks on our manuscript. We have addressed the comments and incorporated suggestions made by Reviewers 1 and 2. Please see below, in blue, for a point-by-point response.

Best regards,

Minqiang Zhou and all co-authors

Referee #1:

The manuscript by Minqiang Zhou et al., presents the three weeks of ground-based remote sensing observations of XCO<sub>2</sub>, XCH<sub>4</sub>, XCO and XN<sub>2</sub>O using the EM27/SUN FTIR spectrometer, along with in situ surface measurements of CO<sub>2</sub> and CH<sub>4</sub> at the Qomolangma Station. This study provides valuable total column greenhouse gas measurements in a region where observational data are scarce, particularly over the Qinghai-Tibetan Plateau. The manuscript is generally well written. However, further discussion and clarification in parts of the data analysis and interpretation would strengthen the overall impact and scientific value of the work. I consider the study suitable for publication after the authors have thoroughly address the specific technical comments outlined below.

Specific comments:

Line 109: The manuscript states that co-located measurements of EM27/SUN and TCCON were performed to derive calibration factors. However, these are two spectrometers with different spectral resolutions. How strong is the correlation between the measurements from the two instruments? Were side-by-side comparisons performed both before and after the campaign? If so, were there any noticeable changes in instrument performance before or after the campaigns, especially considering the long-distance transportation?

Thanks for asking these important questions.

The EM27/SUN instrument was operated at Xianghe TCCON site directly after the campaign in July 2022. Unfortunately, we had no time to operate the side-by-side comparisons before the campaign due to the tight schedule. Instead, we have observed the EM27/SUN instrument line shape (ILS) parameters before and after the campaign in the lab (Table A1). Both the modulation efficiency and phase error keep almost unchanged before and after the campaign, indicating that the instrument was well protected during the long-distance transportation.

The EM27/SUN and TCCON do have discrepancy due to their different spatial resolutions and instrumental parameters. Therefore, in this study, we use the side-by-side comparisons after the campaign to correct the EM27/SUN systematic biases. Figure A1 shows the time series correlation plots between the EM27/SUN and TCCON at Xianghe. The correction coefficients are 0.96, 0.97, 0.99, and 0.55 for XCO<sub>2</sub>, XCH<sub>4</sub>, XCO, and XN<sub>2</sub>O respectively. The scaling factors of 1.001,

0.995, 0.970, and 1.004 are applied to correct the systematic uncertainties of the EM27/SUN XCO<sub>2</sub>, XCH<sub>4</sub>, XCO, and XN<sub>2</sub>O retrievals, respectively.

Regarding the EM27/SUN XN<sub>2</sub>O measurements, it is noted that R value of XN<sub>2</sub>O between EM27/SUN and TCCON measurements is relatively low. It is likely due to the uncertainty of XN<sub>2</sub>O retrieval is relatively large and the variation of XN<sub>2</sub>O is very low during this side-by-side period. To better understand the performance of the EM27/SUN XN<sub>2</sub>O measurements, we have operated another EM27/SUN instrument at Xianghe and made side-by-side comparison between 18 October 2021 and 1 March 2022. Figure A2 shows the time series and scatter plot of EM27/SUN and TCCON XN<sub>2</sub>O measurements. The R value between the two datasets reaches up to 0.81 during this time period, indicating that the EM27/SUN can observe the relative large day-to-day XN<sub>2</sub>O variation. Therefore, we keep the EM27/SUN XN<sub>2</sub>O measurements in this study, but we limit ourself to only use the daily XN<sub>2</sub>O variation when calculating the correlation among these species (Figure 3 in the AMTD).

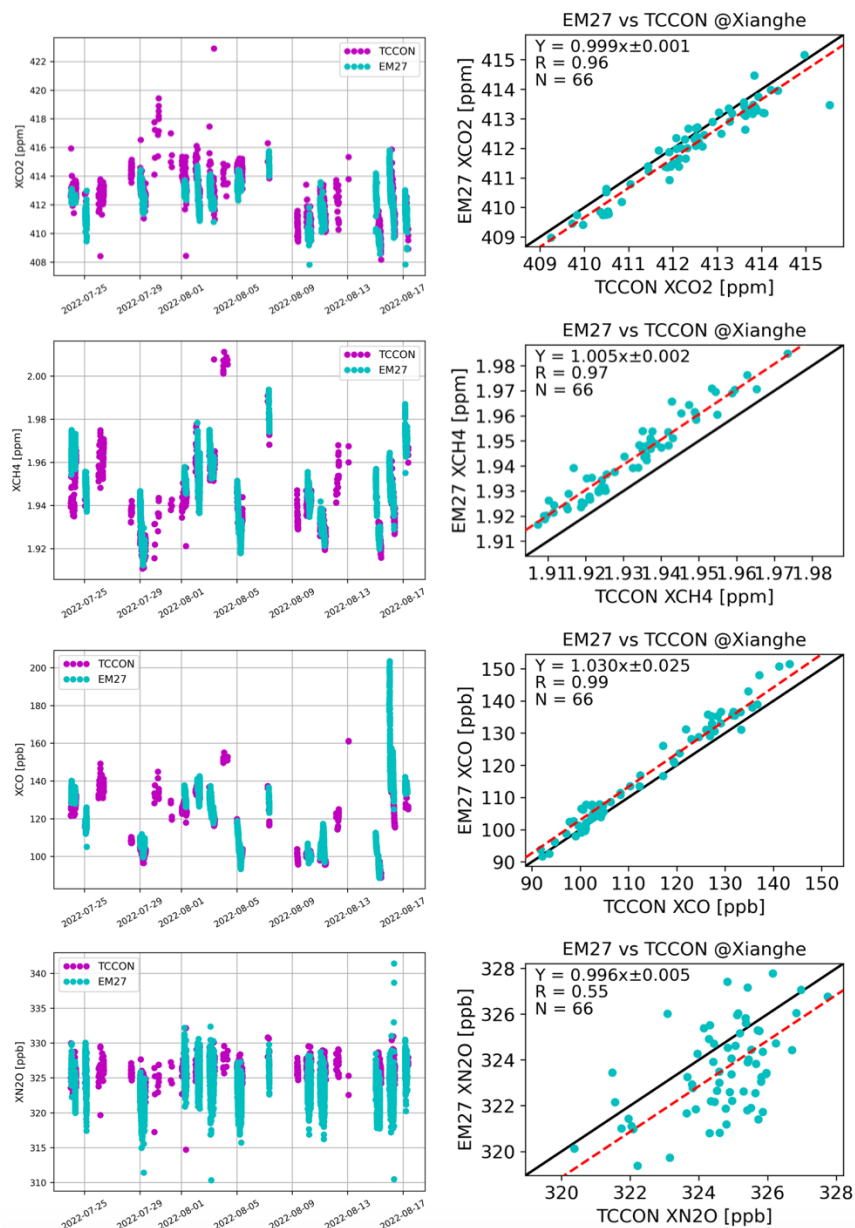


Figure A1. The time series (left panels) and hourly co-located scatter plots (right panels) of EM27/SUN and TCCON XCO<sub>2</sub>, XCH<sub>4</sub>, XCO, and XN<sub>2</sub>O at Xianghe between 24 July and 16 August 2022 just after the campaign.

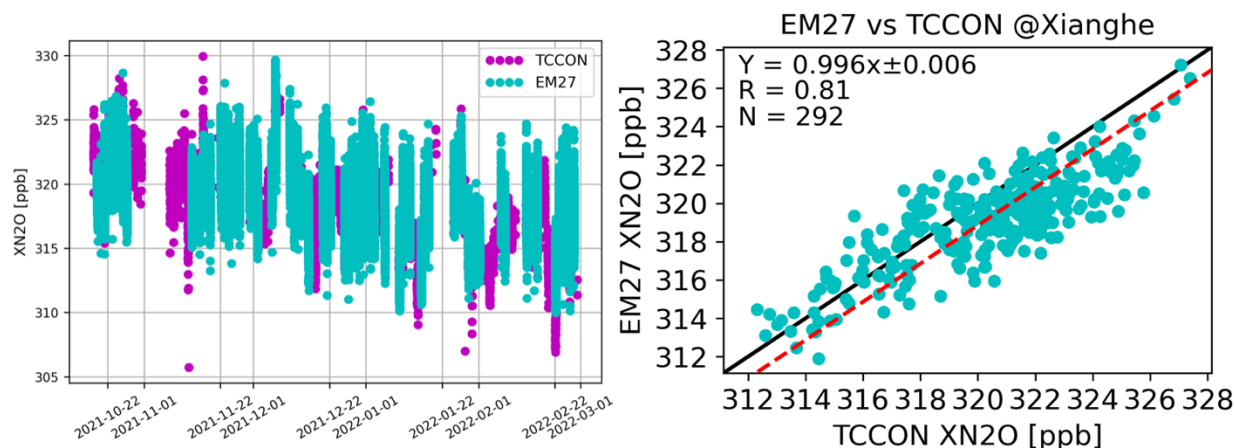


Figure A2. The time series (left panel) and hourly co-located scatter plots (right panel) of EM27/SUN and TCCON XN2O at Xianghe between 18 October 2021 and 1 March 2022.

Table A1. The modulation efficiency and phase error of the EM27/SUN instrument line shape.

Date	Modulation efficiency	Phase error
In 2019 from Bruker company	0.9792	0.0026
23 March 2022; before campaign	0.9773	0.0033
04 August 2022; after campaign	0.9771	0.0035
03 July 2023; one year after campaign	0.9762	0.0036

In the revised version, we add texts to clarify the instrument status before and after the campaign.

Line 148: XN<sub>2</sub>O is not a primary target gas retrieved by the EM27/SUN spectrometer. Could the authors clarify how the precision of XN<sub>2</sub>O measurements from EM27/SUN compares with that from TCCON, in terms of variability, biases or overall data quality? Additionally, are there any limitations or uncertainties associated with retrieving XN<sub>2</sub>O using EM27/SUN?

Thanks for the comments.

The retrieval uncertainty of EM27/SUN XN<sub>2</sub>O is relatively large, because of the weak absorption lines of N<sub>2</sub>O in the short-wave infrared band as compared to CO<sub>2</sub>, CH<sub>4</sub> and CO. This is also the reason that XN<sub>2</sub>O is not included as a primary target gas of EM27/SUN.

Based on the GGG2020 retrieval algorithm according to the optimal estimation method, the estimated retrieval uncertainty of EM27/SUN XN<sub>2</sub>O is about 1.5ppb (the retrieval uncertainty of TCCON XN<sub>2</sub>O is about 0.8ppb). By looking at the EM27/SUN XN<sub>2</sub>O measurements, we select the EM27/SUN measurements between 11:00-13:00 (LST;  $\pm 1$  hour around noon time), and we assume the XN<sub>2</sub>O is stable in this short temporal window (an example shown in Figure A3). Then we calculate the standard deviation of EM27/SUN XN<sub>2</sub>O measurements, which is about 1.6 ppb close to the GGG2020 estimated uncertainty. Regarding the systematic uncertainty, based on the two side-by-side campaigns, the systematic uncertainty of EM27/SUN XN<sub>2</sub>O measurement is about 0.5%. This systematic bias could be corrected by applying a scaling factor.

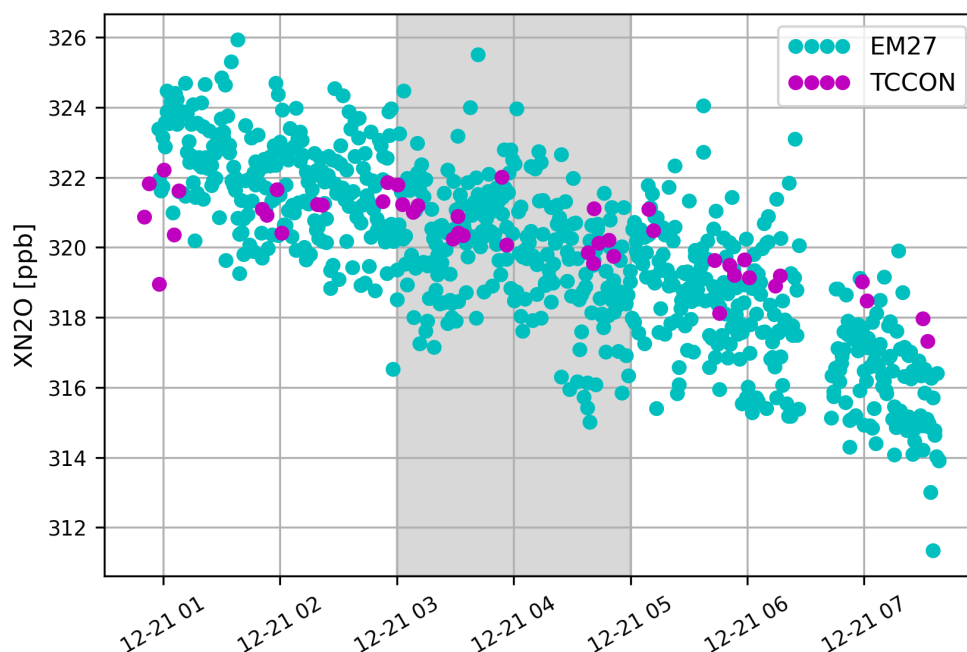


Figure A3. The time series of EM27/SUN and TCCON XN2O at Xianghe on 21 December 2021 (UTC). The grey shade (11:00-13:00 LST) indicates the temporal window to calculate the standard deviation of EM27/SUN XN2O measurements.

Line 195-205: In this paragraph, the authors discuss the varying correlations among XCO<sub>2</sub>, XCH<sub>4</sub>, and XCO across three different weeks. Could the authors elaborate on the possible reasons behind these differences – such as the influence of different emission sources or atmospheric processes? Specifically, during the second week, the correlations among Xgases are notably higher. Were there any changes in meteorological conditions, such as wind direction or speed, that may have contributed to the transport of polluted air masses from source regions during that period? Incorporating a meteorological model, such as WRF or back-trajectory analysis, could help better interpret these variations and strengthen the conclusions.

Thanks for the suggestions.

As shown in the Figure 8 (AMTD), we used the TROPOMI XCO measurements together with ERA5 wind data to understand the Xgas enhancement in the second week. The relatively high XCO values at QOMS on 16, 22, and 24 May correspond to south winds, which bring the air mass with high CO mole fraction to QOMS. On 18 and 21 May, the wind direction was from the west to the east along the southern edge of the Himalayas mountains and did not bring air mass to QOMS so low XCO values are observed in all regions of the southern Tibetan Plateau. On 23 May, relatively high XCO values are observed at approximately 200 km west or east of QOMS, but low XCO values are observed at QOMS. Based on the TROPOMI satellite measurements and the wind data, we conclude that the day-to-day variation of XCO observed at QOMS is largely influenced by atmospheric transport, and the air mass transported from southern Asia can enhance the CO mole fractions over the Tibetan Plateau.

In addition, following your suggestion, we now add the FLEXPART backward simulations to better understand the sources of the air mass coming from during this campaign. The main settings of the FLEXPART model are listed in Table A2. Figure A4 shows the backward sensitivities on 7, 16 and 21 May 2022. Compared to 7 and 21 May, there is a significant polluted air mass at QOMS on 16 May from North India. It is indicated that the enhancement of the measurements on 16 May 2022 is mainly due to the atmospheric transport.

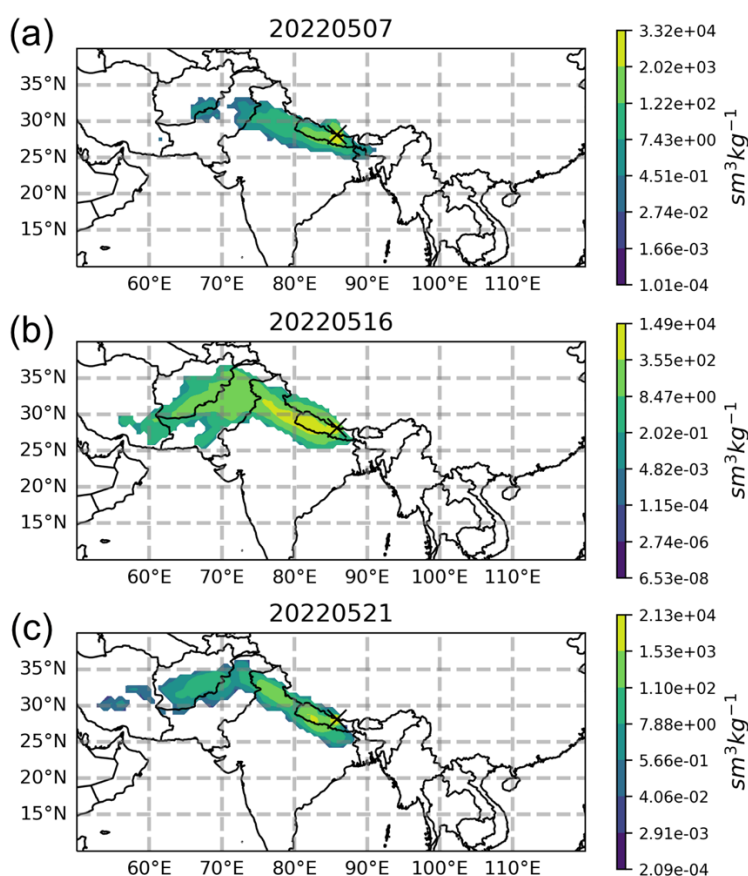


Figure A4. The spatial distribution of air backward sensitivities at QOMS between 04:00-05:00 (UTC) on 7, 16, 21 May 2022. The QOMS site is marked with a cross symbol.

Table A2. The key settings of FLEXPART model backward run.

Parameter	Settings
Tracer	Air
Release location	$\pm 0.05^\circ$ around QOMS
Release height	0-1000 m a.g.l.
Release time	04:00-05:00 (UTC)
Number of backward running days	3 days
Number of releasing particles	20000
Meteorological data	NCEP CFSv2 with $0.5^\circ \times 0.5^\circ$ horizontal resolution and 64 vertical levels

Line 215: Significant enhancements of CO<sub>2</sub> and CH<sub>4</sub> were also observed in the surface measurements, particularly on 16 May, consistent with the FTIR observations. It would strengthen the manuscript to include further discussion on the potential causes of these enhancements on that specific day. For example, were there any known emission events, changes in local activity, or meteorological conditions (e.g., atmospheric stability, wind patterns) that could explain the observed peak in both gases?

During the entire campaign period, there are no special local activities. The enhancement in the second week should not be due to emission events. According to the TROPOMI XCO measurements and FLEXPART back simulations, we believe the variation of the measurements is mainly due to the atmospheric transport (meteorological conditions). More information are added in the revised version.

Line 226: It might also be helpful to clearly state the criteria used to define collocation between the EM27/SUN and surface measurements.

Thanks for the comment. We use the co-existed hourly means to select the EM27/SUN and surface measurement co-located data pair.

“To select the co-located data pair, we use the co-exited FTIR and surface hourly means.”

Line232-233: “The CH<sub>4</sub> mole fractions in the stratosphere are much lower than those in the troposphere due to the chemical reaction and atmospheric dynamic transport”. The sentence does not appear to be closely related to the content presented here. It looks it better fit with next paragraph. Are the authors suggesting that the lower CH<sub>4</sub> amounts in the stratosphere reduce the total amount of XCH<sub>4</sub> measured by the EM27/SUN compared to higher surface measurements? Please elaborate further on the behavior of CH<sub>4</sub> in this context.

Thanks for the comment. We have moved the lines to the next paragraph and restructure the texts to make it more easy to read.

Line 240: Although surface measurements are generally higher than those from EM27/SUN on average, the biases between the two vary over time and show opposite patterns for CO<sub>2</sub> and CH<sub>4</sub>, as illustrated in Figure 5. EM27/SUN reports higher XCO<sub>2</sub> than surface CO<sub>2</sub> before around 18 May, while it shows relatively lower XCH<sub>4</sub> compared to surface CH<sub>4</sub>. It shows the other way around after May 18. Could the authors provide further explanation for these discrepancies?

In Figure 5, the EM27/SUN measurements are shown in the right y-axis, which has a different range as compare to the left y-axis. In general, the EM27 reports lower XCO<sub>2</sub> and XCH<sub>4</sub> than surface measurements all through the campaign period. To avoid confusion, we now modify the caption of the Fig.5.

“Similar to Figure 4, but adding the EM27/SUN FTIR XCO<sub>2</sub> and XCH<sub>4</sub> hourly means and stds (right y-axis) between 13 and 24 May 2022.”

Technical comment:

Line 74: using a the ... >>> using a ...

Line 221: Fiugre 4. The small panel shows the daily variation ( $\Delta X_{\text{gas}}$ ), please denote how the background was removed.

Line229: Figure 7 >>> Figure 5

Done.