

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 XCO₂, XCH₄, XCO and XN₂O using the EM27/SUN FTIR spectrometer, along with in situ surface measurements of CO₂ and CH₄ 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 XCO₂, XCH₄, XCO, and XN₂O 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 XCO₂, XCH₄, XCO, and XN₂O retrievals, respectively.

Regarding the EM27/SUN XN₂O measurements, it is noted that R value of XN₂O between EM27/SUN and TCCON measurements is relatively low. It is likely due to the uncertainty of XN₂O retrieval is relatively large and the variation of XN₂O is very low during this side-by-side period. To better understand the performance of the EM27/SUN XN₂O 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 XN₂O 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 XN₂O variation. Therefore, we keep the EM27/SUN XN₂O measurements in this study, but we limit ourself to only use the daily XN₂O variation when calculating the correlation among these species (Figure 3 in the AMTD).

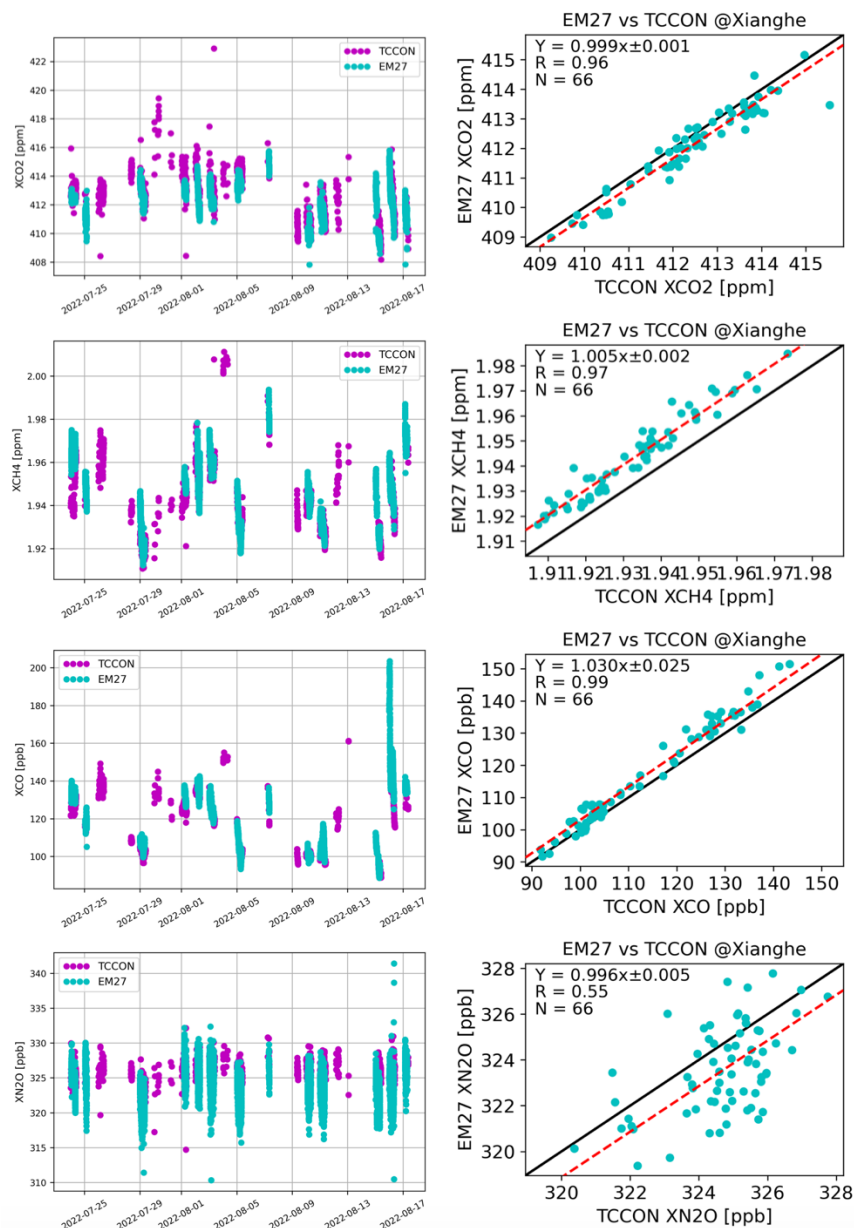


Figure A1. The time series (left panels) and hourly co-located scatter plots (right panels) of EM27/SUN and TCCON XCO₂, XCH₄, XCO, and XN₂O at Xianghe between 24 July and 16 August 2022 just after the campaign.

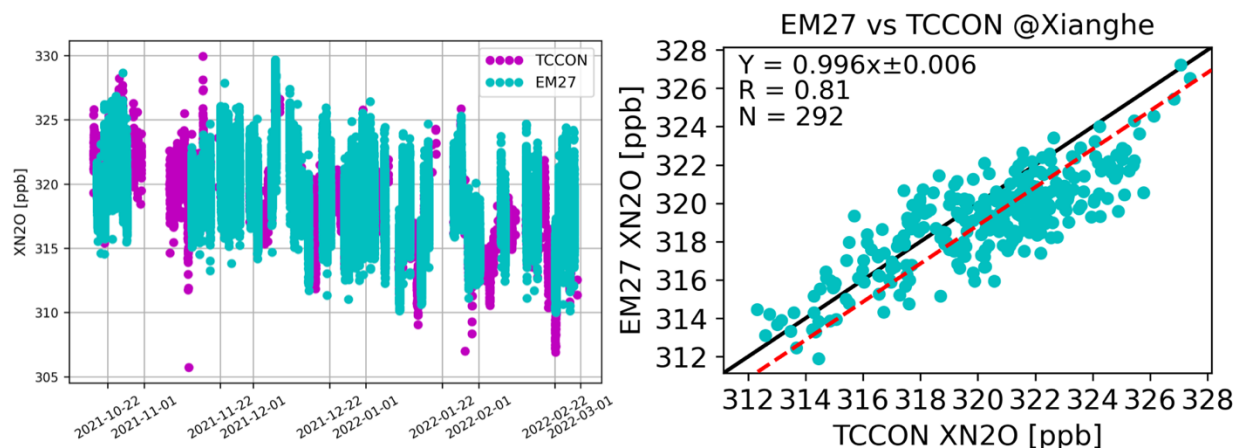


Figure A2. The time series (left panel) and hourly co-located scatter plots (right panel) of EM27/SUN and TCCON XN2O at Xianghe between 18 October 2021 and 1 March 2022.

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: XN₂O is not a primary target gas retrieved by the EM27/SUN spectrometer. Could the authors clarify how the precision of XN₂O 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 XN₂O using EM27/SUN?

Thanks for the comments.

The retrieval uncertainty of EM27/SUN XN₂O is relatively large, because of the weak absorption lines of N₂O in the short-wave infrared band as compared to CO₂, CH₄ and CO. This is also the reason that XN₂O 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 XN₂O is about 1.5ppb (the retrieval uncertainty of TCCON XN₂O is about 0.8ppb). By looking at the EM27/SUN XN₂O measurements, we select the EM27/SUN measurements between 11:00-13:00 (LST; ± 1 hour around noon time), and we assume the XN₂O is stable in this short temporal window (an example shown in Figure A3). Then we calculate the standard deviation of EM27/SUN XN₂O 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 XN₂O measurement is about 0.5%. This systematic bias could be corrected by applying a scaling factor.

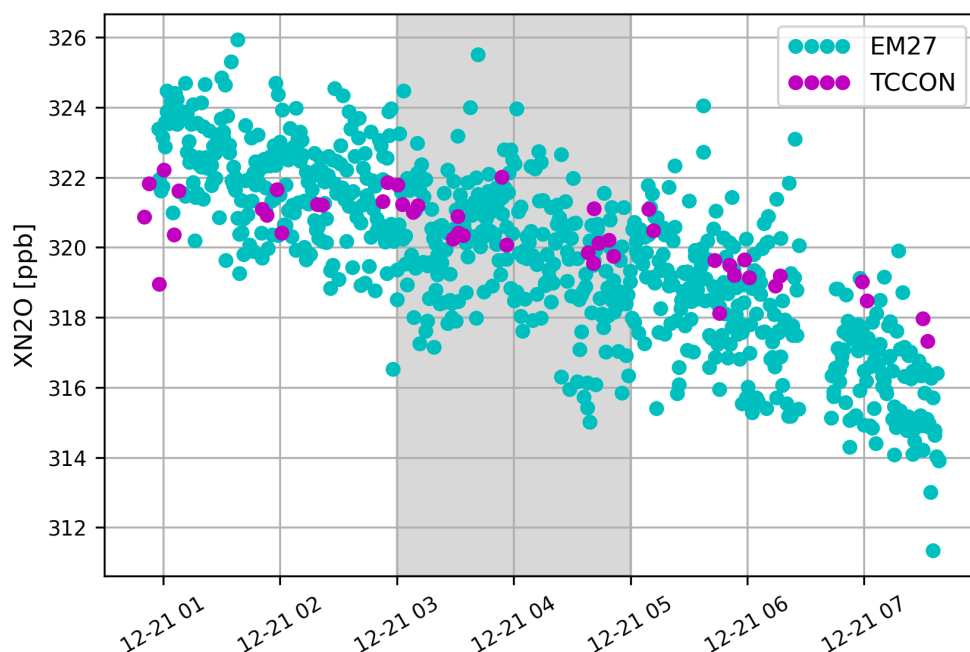


Figure A3. The time series of EM27/SUN and TCCON XN2O at Xianghe on 21 December 2021 (UTC). The grey shade (11:00-13:00 LST) indicates the temporal window to calculate the standard deviation of EM27/SUN XN2O measurements.

Line 195-205: In this paragraph, the authors discuss the varying correlations among XCO₂, XCH₄, 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.

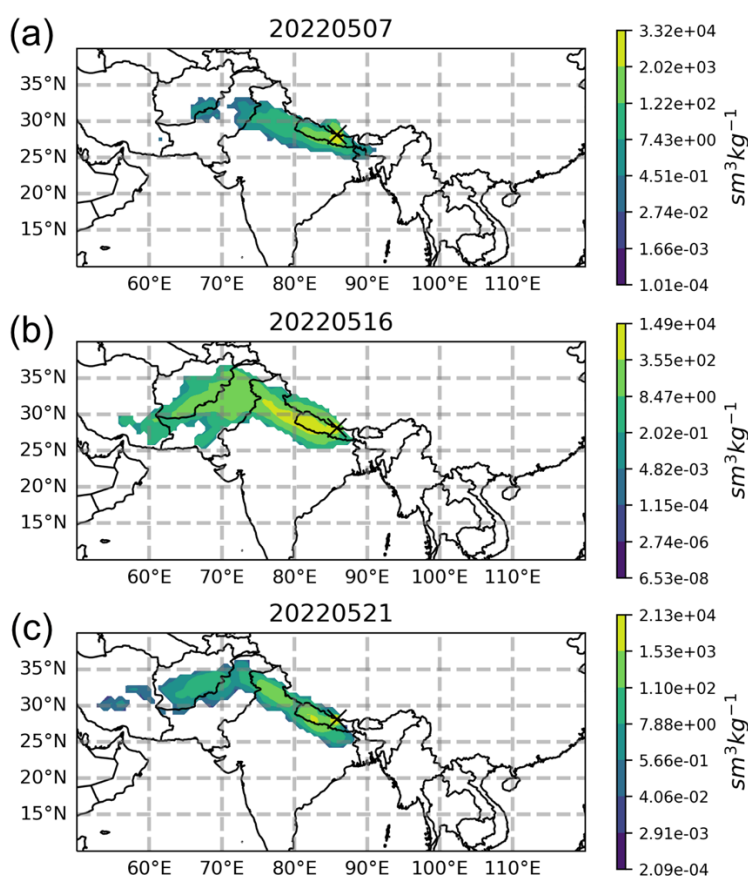


Figure A4. The spatial distribution of air backward sensitivities at QOMS between 04:00-05:00 (UTC) on 7, 16, 21 May 2022. The QOMS site is marked with a cross symbol.

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 CO₂ and CH₄ 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 CH₄ 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 CH₄ amounts in the stratosphere reduce the total amount of XCH₄ measured by the EM27/SUN compared to higher surface measurements? Please elaborate further on the behavior of CH₄ 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 CO₂ and CH₄, as illustrated in Figure 5. EM27/SUN reports higher XCO₂ than surface CO₂ before around 18 May, while it shows relatively lower XCH₄ compared to surface CH₄. 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 XCO₂ and XCH₄ 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 XCO₂ and XCH₄ 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 (ΔX_{gas}), please denote how the background was removed.

Line229: Figure 7 >>> Figure 5

Done.

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 #2:

In this contribution, the authors presented the first ground-based FTIR GHG observation data collected on Qinghai-Tibetan Plateau from a 20-day campaign and the inter- comparisons with in-situ measurements and satellite GHG products. This study fills an important gap in GHG observations over the Tibetan Plateau. The paper is well-structured and summarizes the key findings. While the core contribution is solid, the manuscript would benefit from revisions to improve clarity and strengthen certain aspects of the analysis.

Major comments

1. Calibration: The study employs standard GGG2020 retrieval procedures and X/O₂ normalization. However, it remains unclear whether additional calibrations were implemented to account for the unique high-altitude environment at QOMS. The manuscript should address whether Instrument Line Shape (ILS) characterization was performed under high-altitude conditions. Since GGG2020 a priori profiles are based on GEOS-FPIT, which may not fully capture local thermodynamic conditions on the plateau, it would strengthen the analysis to discuss whether alternative data sources (e.g., radiosonde data or ERA5 reanalysis) were considered, and how sensitive the retrievals, particularly XCH₄, are to such profile inputs in this setting.

Thanks for the comments.

Regarding the ILS characterization, unfortunately we did not have the lab resources at QOMS and could not perform the ILS measurements during the campaign. Instead, we did the ILS measurements before and after the campaign at IAP/CAS Beijing (Table 1). As the modulation efficiency and phase error keep almost unchanged before and after the campaign, indicating that the EM27/SUN instrument was well protected during the long-distance transportation. The observed ILS parameters have been used by the GGG2020 code. Since the ILS is mainly determined by the alignment of the instrument and not dependent on the surface pressure, the ILS parameters should be stable during this campaign.

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

Regarding the a priori inputs, we have no measurements, such as radiosonde data.

Following your suggestion, we have done a comprehensive uncertainty analysis of the FTIR retrievals. We test ERA5 data by using the temperature and humidity profiles from ERA5 reanalysis data. In addition, to estimate the uncertainty from the gas a priori profile, in the revised version, we manually add 2% random noise on the TCCON prior. More detail information could be found in Table A2.

2. Uncertainty: While the manuscript acknowledges limitations in GEOS-FPIT a priori profiles (Lines 156-162), a quantitative assessment of their impact is lacking. Given this study's contribution to satellite validation, it is recommended to include a comprehensive uncertainty analysis of the FTIR retrievals. Specifically, a decomposition of systematic and random error components — including contributions from spectroscopy, observation geometry, a priori dependence, and instrumental effects— would significantly enhance the robustness of the results and align with AMT community standards.

Thanks for the suggestion. In the revised version, we have added the uncertainty estimation (Table A2) in the appendix I. The uncertainties of the EM27/SUN CO₂, CH₄, CO and N₂O measurements at QOMS are estimated by perturbing the inputs using the GGG2020 code. In this study, we include contributions from instrumental effects (ILS), observation geometry (pointing offset), temperature profile, spectroscopy (line intensity), a priori dependence, and measurement noise.

Table A2. List of sources, values used for the uncertainty analysis, and CO₂, CH₄, CO and N₂O column retrieval uncertainties for all measurements at QOMS site using the GGG2020 code. (ME: modulation efficiency amplitude; SNR: signal-to-noise ratio). The third column provides the partitioning of the error values between random (ran) and systematic (sys) contributions. Note that an uncertainty less than 0.01% is indicated as '-'.

Error source	Uncertainty value	sys/ran contribution [%]	CO ₂ column uncertainty (sys/ran) [%]	CH ₄ column uncertainty (sys/ran) [%]	CO column uncertainty (sys/ran) [%]	N ₂ O column uncertainty (sys/ran) [%]
Prior	2%	50/50	0.01/0.01	0.02/0.02	0.04/0.04	0.08/0.08
ILS (ME and phase error)	1% and 0.01rad	50/50	0.1/0.1	0.1/0.1	0.1/0.1	0.1/0.1
Pointing offset	0.1°	10/90	-/0.1	-/0.1	-/0.1	-/0.2
Temperature profile	2 K	50/50	0.03/0.03	0.15/0.15	0.80/0.80	0.07/0.07
Spectroscopy	2%	100/0	2.0/-	2.0/-	2.0/-	2.0/-
Measurement noise	1/SNR	0/100	-/0.1	-/0.1	-/0.3	-/1.2
Total			2.0/0.2	2.0/0.2	2.0/0.9	2.0/1.2

Minor comments

1. Timing: The campaign was conducted in May, but it remains unclear whether this timing was chosen based on specific meteorological conditions and seasonal patterns of greenhouse gas concentrations in the QTP region, or if it was mainly due to logistical constraints such as resource availability and research team schedules. This raises questions about the representativeness of the May observations - to what extent can the findings represent annual or seasonal variations in greenhouse gas concentrations? Could the authors comment on whether the GHG levels during this period are climatologically representative based on past in situ or model records, or whether they might

reflect transient synoptic conditions? Are there any plans to conduct additional observations during other seasons to provide a more complete temporal coverage?

Thanks for the comment.

This GHG campaign is part of the Earth Summit Mission-2022, so the timing is mainly determined by the resource availability and research team schedules. As a key task of the Earth Summit Mission: a team climbed up to the top of Mt. Qomolangma to install several meteorological stations and brought back the ice core of the mountain. The best time to climb the Mt. Qomolangma is in spring (between April and May). The mountain is virtually unclimbable in summer due to the deadly monsoon season which generally brings mist to the area. In winter, this region is extremely cold.

Using the CAMS global greenhouse gas reanalysis (EGG4) model (<https://ads.atmosphere.copernicus.eu/datasets/cams-global-ghg-reanalysis-egg4?tab=overview>), we plot the XCO₂ and XCH₄ mole fractions at QOMS in 2020. Based on the CAMS model, the XCO₂ at QOMS keeps increasing in May and it starts decreasing in August. The XCH₄ at QOMS in May is relatively low, and the XCH₄ values become large in July-September.

In August 2024, we have installed one EM27/SUN at Nagqu, Tibet. Currently, the FTIR at Nagqu is continuously operating and we plan to keep the measurements at least for another year.

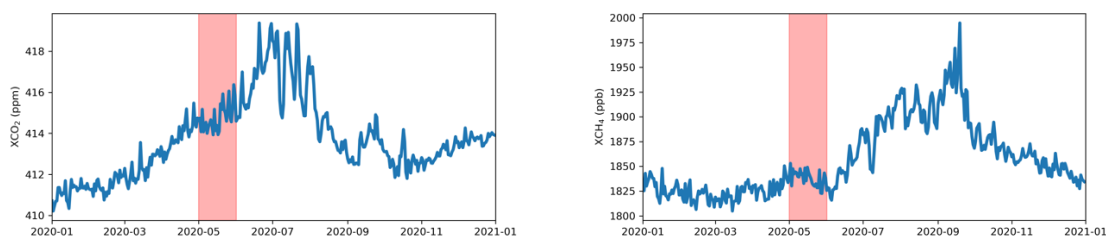


Figure A1. The time series of XCO₂ (left) and XCH₄ (right) from the CAMS greenhouse gas model simulations in 2020.

2.OCO-2 Distance (Line 132): The spatial representativeness of QOMS FTIR measurements, particularly when comparing with OCO-2 data at distances up to 500 km, requires further discussion. Has the spatial variability of XCO₂/CH₄ over the Tibetan Plateau been evaluated through regional modeling or high-resolution simulations? A discussion of potential spatial representativeness error would be helpful here.

Thanks for the suggestion.

We now use the CAMS model simulations in May 2020 to estimate the spatial variability of XCO₂ between the QOMS and OCO-2 footprints in this study.

The mean and std of the differences in CAMS XCO₂ between the QOMS and OCO-2 footprint around 29.2°N, 84.5°E (~250 km away) are 0.02 ppm and 0.27 ppm, respectively. The mean and

std of the differences in CAMS XCO₂ between the QOMS and OCO-2 footprint around 30.2°N, 82.4°E (~480 km away) are 0.10 ppm and 0.34 ppm, respectively.

In the revised version, we have add the texts:

“As the footprint of OCO-2 is not exactly over the QOMS site, we the CAMS model simulations in May 2020 to estimate the spatial variability of XCO₂ between the QOMS and OCO-2 footprints in this study. The mean and std of the differences in CAMS XCO₂ between the QOMS and OCO-2 footprint around 29.2°N, 84.5°E (~250 km) are 0.02 ppm and 0.27 ppm, respectively. The mean and std of the differences in CAMS XCO₂ between the QOMS and OCO-2 footprint around 30.2°N, 82.4°E (~480 km) are 0.10 ppm and 0.34 ppm, respectively. According to the CAMS model simulations, the XCO₂ spatial variability in this region is relatively small.”

3. Comparison to 6 sites (Line 164): The rationale for comparing QOMS to six mid- latitude urban/suburban TCCON sites is unclear. They are mostly urban or suburb sites while QOMS is far from anthropogenic sources. It could be imagined that the Xgas at QOMS is lower. Given QOMS is a remote high-altitude site, a more meaningful comparison may be with other background or mountain sites such as Izaña, which are more comparable in atmospheric conditions and anthropogenic influence.

Thanks for the suggestion. In the revised version, the measurements at Izaña are added in the Table 1, and related texts are modified in the paper.

4. 3 weeks (Line 196): The correlations between XCO₂ and XCH₄ are different in the three weeks. Are they driven by different weather/chemical conditions? It would help to link these changes in correlation patterns with meteorological conditions, e.g., changes in wind direction or boundary layer dynamics.

Thanks for the suggestions.

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. It is noted that the enhancement of the measurements on 16 May 2022 is mainly due to the transport. Compared to 7 and 21, on 16 May, there is a significantly greater air mass at QOMS from North India, with higher gas concentrations.

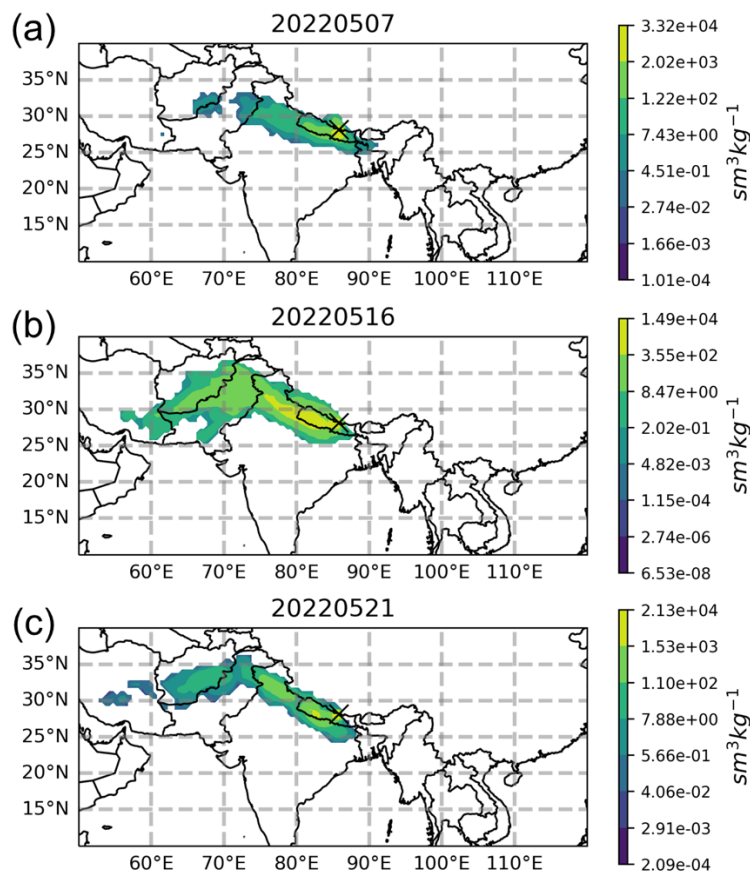


Figure A4. The spatial distribution of air backward sensitivities at QOMS between 04:00-05:00 (UTC) on 7, 16, 21 May 2022. The QOMS site is marked with a cross symbol.

5. in-situ VS TC (Line 230): The CH₄ surface mole fraction is **averagely** 97 ppb larger than the XCH₄, but Figure 5b appears to show contradictory patterns where surface measurements are lower than XCH₄ in 7 of 12 days. Could the “97 ppb higher” represent the real difference? This discrepancy between the daily mean bias and daily value distribution may suggest that outliers or skewed distributions contributed disproportionately. A histogram or scatter plot of daily differences might clarify this.

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 XCO₂ and XCH₄ than surface measurements all through the campaign period. To avoid confusion, we now add some texts in the caption of the Fig.5.

“Similar to Figure 4, but adding the EM27/SUN FTIR XCO₂ and XCH₄ hourly means and stds (right y-axis) between 13 and 24 May 2022.”

Technical comments

Line 2: and contribute to ... → contributing to/and contributions to

Line 8: end → and

Line 17: Qomolongma → Qomolangma

Line 227: mesurements → measurements

Line 328: OCO-3 → OCO-2

Line 366: CH₄mole → CH₄ mole

It is recommended to write out full form at first mention, e.g. “std”.

Thanks, and all these have been corrected.

Greenhouse gas measurement campaign of the Earth Summit Mission-2022: ground-based *in situ* and FTIR observations and contributes to satellite validation in the Qomolangma region

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Abstract

The Qinghai-Tibetan Plateau (QTP) is a key system that impacts the global carbon balance, but greenhouse gases (GHGs) mole fraction measurements in this region are limited due to the tough environment. Supported by the Second Tibetan Plateau Scientific Expedition Program, we carried out an integrated GHG measurement campaign in May 2022 as part of the Earth Summit Mission-2022 at the Qomolangma station for atmospheric and environmental observation and research (QOMS; 28.362°N, 86.949°E, 4276 m a.s.l.). In this study, the first GHG column-averaged mole fraction measurements (X_{gas}) at QOMS are presented, including XCO₂, XCH₄, XCO, and XN₂O, derived from a ground-based Fourier-transform infrared spectrometer (FTIR; Bruker EM27/SUN). We then compare them to surface *in situ* and satellite (TROPOMI and OCO-2) measurements. The

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mean FTIR XCO₂ and XCH₄ are 7.8 ppm and 97 ppb less than those near the surface, respectively. The difference between OCO-2 land nadir and EM27/SUN XCO₂ measurements is 0.21±0.98 ppm, which is consistent with OCO-2 retrieval uncertainty. However, a relatively large bias (1.21±1.29 ppm) is found for OCO-2 glint XCO₂ measurements, which is related to the surface albedos and surface altitudes. The EM27/SUN measurements indicate that the uncertainty of OCO-2 satellite XCO₂ measurements is relatively large in the QTP mountain region and its quality needs to be further assessed. The difference between FTIR and TROPOMI XCO measurements is -5.06±5.36 (1σ) ppb (-4.7±5.1%) within the satellite retrieval uncertainty. The XCO measurements at QOMS show the local air mass is largely influenced by atmospheric transport from southern Asia, and it is important to carry out long-term measurements to quantify the contribution of the cross-regional transport in this region.

Keywords: Qomolangma, greenhouse gas, EM27/SUN, OCO-2, TROPOMI

1 Introduction

The Qinghai-Tibetan Plateau (QTP) plays an important role in regional and global climate systems (Ge et al. 2017; Tada et al. 2016; Zhang et al. 2021). The high mountains in the QTP, including the Earth's highest summit (Mt. Qomolangma; 8848.86 m a.s.l.), strongly affect the atmospheric thermodynamic and dynamic conditions. On the other hand, environmental changes, including human activities, atmospheric warming, and cryosphere thaw, can in turn make significant impacts on the hydrology, ecosystems, and biogeochemistry in the QTP (Rui et al. 2011; Wu et al. 2021; Zhang et al. 2015). The QTP has abundant original forest and soil resources, and serves as a huge carbon storage (Ding et al. 2016; Wang et al. 2020; Jia et al. 2021). A small change in carbon storage of the QTP could make an impact on the global carbon balance. However, there is still a large uncertainty about the terrestrial ecosystem carbon sink in the QTP (Wang et al. 2021; Piao et al. 2022). The uncertainties associated with eddy covariance data processing may lead to an overestimation of the carbon sink (Wang et al. 2022). Using a 'top-down' approach can help us to estimate the carbon sink in the QTP (Jiang et al. 2016). However, limited greenhouse gases (GHGs) measurements are currently available in this region because of the tough environment (Guo et al. 2020; Liu et al. 2021; Zhou et al. 2023).

64 To better understand the level and temporal variation of atmospheric GHGs mole fraction over
65 the QTP, we carried out an integrated GHGs measurement campaign at the Qomolangma station
66 for atmospheric and environmental observation and research, Chinese Academy of Sciences
67 (QOMS) in May 2022 as part of the Second Tibetan Plateau Scientific Expedition Program (Earth
68 Summit Mission-2022). During this campaign, a compact Fourier-transform infrared spectrometer
69 (FTIR), Bruker EM27/SUN, was applied to retrieve the column-averaged dry air mole fraction of
70 CO₂, CH₄, CO, and N₂O (XCO₂, XCH₄, XCO, and XN₂O) at QOMS between 5 and 24 May 2022.
71 The FTIR measures the GHG columns, which are less affected by the local meteorological
72 parameters, such as the boundary layer height and wind turbulence (Wunch et al. 2011; Zhou et
73 al. 2018). The ground-based FTIR GHGs measurement is widely used to validate satellite
74 observation because of its high precision and similar measurement technique to the satellite (Zhou
75 et al. 2016; Wunch et al. 2017; Sha et al. 2021). In addition, a gas analyzer (ABB Ultra-Portable
76 Greenhouse Gas Analyzer; GLA132), using a off-axis integrated cavity output spectroscopy (OA-
77 ICOS) technique, was applied to measure the CO₂ and CH₄ mole fractions near the surface. In the
78 following sections, we will give an introduction to the observation site, and present the XCO₂,
79 XCH₄, XCO, and XN₂O derived from the EM27/SUN FTIR spectra at QOMS. The results from
80 the *in situ* and ground-based FTIR measurements are shown in Section 3. In Section 4, the *in situ*
81 surface measurements are compared to the FTIR column measurements. Moreover, the FTIR
82 measurements are then compared to independent satellite observations. Finally, the conclusions
83 are drawn in Section 5.

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84 2 Observation site and data

85 2.1 The QOMS site

86 The QOMS (latitude 28.362°N; longitude 86.949°E; with an elevation of 4276 m a.s.l.) is
87 situated approximately 30 km away from the Everest Base Camp and around 650 km from Lhasa
88 (Figure 1). The station is located about 3 km to the south of the local village, in an area with
89 minimal human influence. The surface of the station is flat, and it is mainly covered by sand and
90 gravel with sparse vegetation. More information about the QOMS refers to Ma et al. (2023).

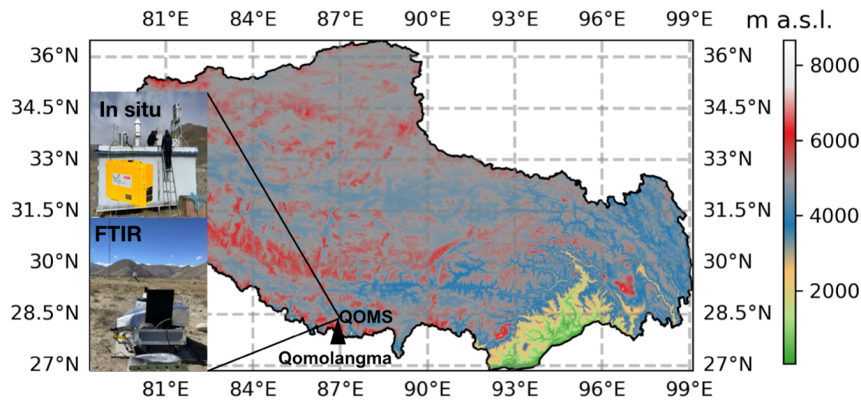


Figure 1. The integrated GHGs measurement campaign carried out at the Qomolangma station for atmospheric and environmental observation and research station (QOMS) in May 2022 including both the FTIR remote sensing and surface *in situ* measurements.

2.2 Ground-based FTIR

A Bruker EM27/SUN FTIR was operated at QOMS between 5 and 24 May. The measurement settings of the EM27/SUN follow the guidance of the COllaborative Carbon Column Observing Network (COCCON), which records the direct solar absorption spectra between 4000 and 12000 cm^{-1} with a spectral resolution of 0.5 cm^{-1} using two InGaAs detectors (Frey et al., 2019). The advanced retrieval algorithm GGG2020 is applied to retrieve the O_2 and GHGs (CO_2 , CH_4 , CO , and N_2O) total columns, and then X_{gas} is calculated as $0.2095 (TC_{\text{GHG}}/TC_{\text{O}_2})$. Using the ratio between target species and O_2 can reduce the same uncertainty from the instrumental and atmospheric parameters (Yang et al. 2002). GGG2020 is commonly used in the ground-based GHG remote sensing community, and it includes: 1) the conversion from interferogram to spectrum with the DC correction (Keppel-Aleks et al. 2012); 2) a non-linear least squares fitting code (GFIT); 3) a post-correction procedure to reduce the retrieval uncertainty resulting from spectroscopy and observation geometry (Laughner et al. 2023). We have observed the EM27/SUN instrument line shape (ILS) parameters before and after the campaign in the lab. Both the Modulation efficiency and phase error keep almost unchanged before and after the campaign, reflecting that the instrument was well protected during the long-distance transportation. Table A1 in appendix I shows the systematic and random uncertainties of CO_2 , CH_4 , CO and N_2O column

retrievals are 2.0%/0.2%, 2.0%/0.2%, 2.0%/0.9%, and 2.0%/1.2%, respectively. The systematic uncertainty is dominated by the spectroscopy. To further reduce the systematic uncertainty of the EM27/SUN retrievals, the EM27/SUN instrument was operated together with the TCCON site at Xianghe (Yang et al. 2020) for three weeks in July-August 2022 after the campaign. Based on the co-located measurements of EM27/SUN and TCCON, the scaling factors of 1.001, 0.995, 0.970, and 1.004 have been derived and applied to correct the systematic uncertainties of the EM27/SUN XCO₂, XCH₄, XCO, and XN₂O retrievals, respectively.

2.3 *In situ*

The near-surface CO₂ and CH₄ mole fractions were observed by the ABB GLA132 gas analyzer at QOMS continuously between 13 and 24 May 2022 (Figure 1). To ensure the accuracy of the GHG *in situ* measurements, we calibrated the gas analyzer using the standard gas once per day. The precisions of the *in situ* measurements (1 second) are within 0.3 ppm and 2.0 ppb for CO₂ and CH₄, respectively (<https://www.envicontrol.com/storage/app/media/uploaded-files/UGGA%20LGR%20GLA132-GGA.pdf>). Note that the drifts of CO₂ and CH₄ within 24 hours of the analyzer are within their measurement uncertainties.

2.4 *Satellite*

We use the Orbiting Carbon Observatory-2 (OCO-2) satellite level 2 bias-corrected XCO₂ retrospective processing v11.2r (Kiel et al. 2019). The OCO-2 XCO₂ is retrieved using the ACOS algorithm by the CO₂ absorption lines around 1.61 and 2.06 μm , together with the information about surface pressure, cloud and aerosol scattering constrained by the O₂-A band around 0.76 μm (O'dell et al. 2018). For more details about the OCO-2 XCO₂ data, we refer to the https://docsserver.gesdisc.eosdis.nasa.gov/public/project/OCO/OCO_L2_ATBD.pdf. The OCO-2 XCO₂ uncertainty has been accessed by comparing to the Total Carbon Column Observing Network (TCCON), and the absolute median difference across TCCON sites over the globe is found to be less than 0.4 ppm and the root mean square of the differences is less than 1.5 ppm (Wunch et al. 2017). The footprint size of each OCO-2 pixel is 2.25 $\times$ 1.29 km². However, the width of the swath is only about 16 km, leading to a very small spatial coverage. During the campaign, there were 2 days when the orbit of OCO-2 overpassed within 500 km around QOMS. These OCO-2 satellite measurements are compared to EM27/SUN FTIR measurements to assess their quality at QOMS.

145 Regarding CH₄ and CO, we use the ESA operational offline level 2 products from the Sentinel-
146 5 Precursor (S5-P) TROPOspheric Monitoring Instrument (TROPOMI). Unfortunately, there is
147 almost no TROPOMI CH₄ retrieval at QOMS, mainly due to the complex orography and high
148 cloud coverage in this region (Lorente et al. 2021). Therefore, in this study, we only look at the
149 TROPOMI XCO data. The TROPOMI XCO retrievals are derived from the reflected solar
150 radiation in 2.3 μm band and the stripes of erroneous XCO retrievals are corrected by the fixed
151 masked de-stripping method (Landgraf et al. 2016; Borsdorff et al. 2019). The spatial resolution of
152 TROPOMI measurements is 7.0×5.5 km². Thanks to the large swath of about 2600 km, TROPOMI
153 provides XCO measurements in this region around 15:30 China Standard Time (CST) every day.
154 According to the validation study made by Martínez-Alonso et al. (2022), the mean difference
155 between TROPOMI XCO data and AirCore measurements is 2.02±11.13 (1σ) %.

156 3 Results

157 3.1 Ground-based FTIR column measurements

158 The time series together of *a priori* and retrieved XCO₂, XCH₄, XCO, and XN₂O
159 measurements from the EM27/SUN FTIR at QOMS between 5 and 24 May are shown in Figure
160 2. The *a priori* columns of FTIR retrievals are derived from the global atmospheric chemistry
161 model (GEOS-FPIT), which provides 6-hourly simulations with a spatial resolution of about 50
162 km. For more information about the GGG2020 *a priori* profiles, please refer to Laughner et al.
163 (2023). Keep in mind that the FTIR provides the measurements only in the daytime and under a
164 clear sky condition.

165 The mean and standard deviation (std) of XCO₂, XCH₄, XCO, and XN₂O are 418.4±0.6
166 ppm, 1888.3±8.0 ppb, 106.2±8.3 ppb, and 321.6±3.2 ppb, respectively. The EM27/SUN retrieved
167 columns are larger than the *a priori* columns for all these four species, indicating that the TCCON
168 prior is systematically underestimated in this region. In addition, the amplitudes of the variations
169 of XCO₂ and XCH₄ derived from the EM27/SUN measurements are larger than the model
170 simulations. Moreover, the day-to-day variations of these species are not well captured by the
171 GEOS-PFIT model, for example, the maximum XCO value observed by the FTIR measurements
172 on 16 May is not well-simulated in the model. The mean of XCO₂, XCH₄, and XCO observed at
173 QOMS are also compared to seven TCCON sites (Hase et al. 2023; Té et al. 2022; Warneke et al.
174 2022; Wennberg et al. 2022a,b; Zhou et al. 2022; García et al., 2022) in the northern hemisphere

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during the same time period (Table 1). The mean XCO₂ at QOMS is lowest among these sites, which is about 1.5-3.5 ppm less than urban sites, and about 0.5-1.5 ppm less than suburban sites and the mountain site (Izaña). The XCH₄ and XCO observed at QOMS are less than those at Xianghe and Caltech, but larger than other TCCON sites.

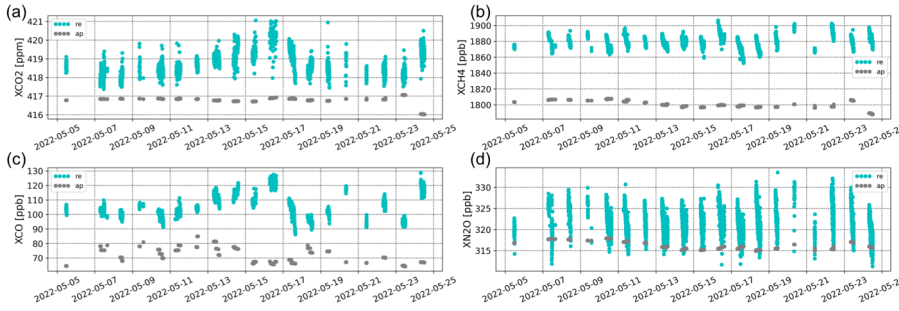


Figure 2. The time series of *a priori* (grey dots) and retrieved (cyan dots) XCO₂, XCH₄, XCO, and XN₂O measurements from the EM27/SUN FTIR between 5 and 24 May 2022 at QOMS.

Table 1. The mean and std of the XCO₂, XCH₄, XCO measurements at six TCCON sites in the northern hemisphere, together with our EM27/SUN measurements at QOMS. N is the number of measurements at each site between 5 and 24 May 2022.

Type	Site	Geolocation	XCO ₂ (ppm)	XCH ₄ (ppb)	XCO (ppb)	N
Urban	Paris	48.84°N;2.35°E;60 m	420.1±0.9	1870.4±4.2	97.9±7.2	685
	Xianghe	39.8°N;116.96°E;36 m	419.9±0.8	1906.7±12.	134.4±15.	268
	Caltech	34.13°N;118.12°W;230m	421.9±0.9	1899.2±6.8	117.6±6.4	1956
Suburb	Karlsruhe	49.1°N;8.439°E;116 m	418.9±0.8	1876.3±9.2	99.2±3.9	533
	Orleans	47.97°N;2.113°E;130m	418.9±0.8	1875.9±7.5	96.2±5.9	1641
	Lamont	36.6°N;97.486°W;320 m	419.9±0.7	1885.6±6.1	105.1±7.8	838
Mountain	Izaña	28.3°N;16.499°E;2367 m	419.8±0.4	1873.5±4.6	81.7±2.7	819
QTP	QOMS	28.3°N; 86.9°E; 4276 m	418.4±0.6	1888.3±8.0	106.2±8.3	5925

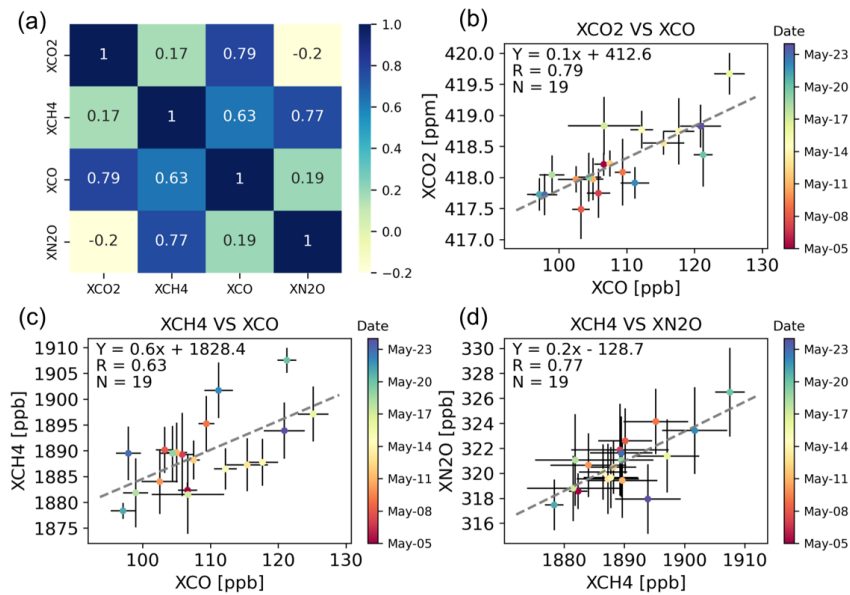
Figure 3 shows the covariance matrix among these four species observed by the EM27/SUN measurements. The good correlations are found between XCO₂ and XCO (R=0.79;

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195 $p_value < 0.001$), XCH_4 and XCO ($R=0.63$; $p_value < 0.01$), XCH_4 and XN_2O ($R=0.77$;
 196 $p_value < 0.001$). However, the correlations are relatively weak between XCO_2 and XCH_4 ($R=0.16$;
 197 $p_value=0.50$), XCO_2 and XN_2O ($R=-0.20$; $p_value=0.41$), XCO and XN_2O ($R=0.19$;
 198 $p_value=0.43$). The good correlation between XCH_4 and XN_2O is probably due to their similar
 199 physical and chemical process in the stratosphere (Wang et al. 2014; Ji et al. 2020).

200



201

202 **Figure 3.** The correlation matrix among XCO_2 , XCH_4 , XCO and XN_2O observed by the
 203 EM27/SUN measurements between 5 and 24 May 2022 at QOMS (a), together with the scatter
 204 plots between XCO_2 and XCO daily means (b), between XCH_4 and XCO daily means (c), and
 205 XCH_4 and XN_2O daily means (d). In each scatter plot, the error bar denotes the daily std, the dashed
 206 line is the linear regression, R is the Pearson correlation coefficient, and N is the number of the
 207 co-located measurements. The dot is colored by the measurement date.

208

209 The EM27/SUN measurements indicate that XCO is a good tracer for both XCO_2 and
 210 XCH_4 at QOMS, while the R between XCO_2 and CH_4 is only 0.17. To better understand this, we
 211 separate the time period into 3 weeks (Table 2). The R values are relatively low in the first week,
 212 especially between XCO_2 and XCH_4 . The day-to-day variations of these species are pretty low in
 213 the first week (Figure 2). In the second week, large enhancements of the three species on 16 May

are observed simultaneously, resulting in large R values. In the third week, strong vibrations in XCO₂, XCH₄, and XCO are observed, but unlike a single large enhancement in the second week, the enhancements in this week are discontinuous. The R values in the third week are larger than those in the first week, but less than those in the second week. Based on the three weeks, we understand that a good correlation (R=0.83) between XCO₂ and XCH₄ can be also observed when a large continuous enhancement occurs. However, the correlation between XCO₂ and XCH₄ becomes low and even negative (R=-0.26), when the variations of XCO₂ and XCH₄ are low.

Table 2. The correlations among XCO₂, XCH₄, and XCO in the first week (5-11 May), second week (12-18 May), and third week (19-24 May).

R	XCO ₂ and XCH ₄	XCO ₂ and XCO	XCH ₄ and XCO
Week 1 (5-11 May)	-0.26	0.60	0.46
Week 2 (12-18 May)	0.83	0.91	0.84
Week 3 (19-24 May)	0.40	0.89	0.79

3.2 *In situ* CO₂ and CH₄ measurements near the surface

The time series of CO₂ and CH₄ observed by the gas analyzer at QOMS near the surface between 13 and 24 May, together with their diurnal variations, are shown in Figure 4. The mean and std of CO₂ and CH₄ are 424.2±2.1 ppm and 1985.2±19.7 ppb, respectively. A good correlation between CO₂ and CH₄ is observed, with a Pearson correlation coefficient (R) of 0.82. Similar to the FTIR XCO₂ and XCH₄, the surface CO₂ and CH₄ mole fractions are the highest on 16 May. The mean CO₂ mole fraction in the daytime is about 0.9 ppm higher than that during the night. Contrary to CO₂, we observe the minimum CH₄ value at around 13:00, which is about 12 ppb less than that at midnight.

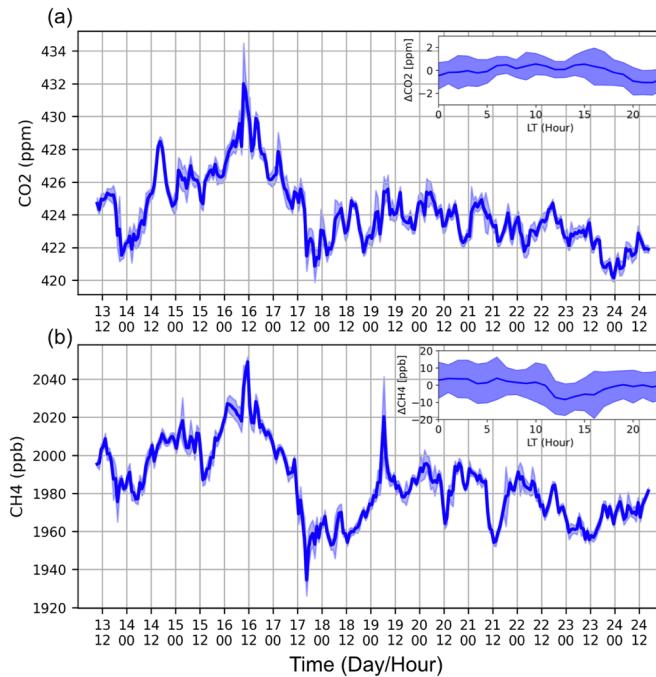


Figure 4. The time series of hourly mean and std of CO₂ (a) and CH₄ (b) mole fraction observed by the gas analyzer at QOMS near the surface. The small panel in the right-upper corner shows the daily variation of CO₂ and CH₄, and the Δgas is derived from the measurements by subtracting the daily median.

4 Inter-comparisons and discussions

4.1 Comparison between FTIR column and surface *in situ* measurements

The FTIR observes the CO₂ and CH₄ columns, while the *in situ* provides the surface mole fractions. Do the variations of XCO₂ and XCH₄ differ from the surface measurements? Here, CO₂ and CH₄ surface mole fractions are compared with the FTIR XCO₂ and XCH₄ measurements (Figure 7). To select the co-located data pair, we use the co-existed FTIR and surface hourly means. The mean CO₂ surface mole fraction is 7.8 ppm larger than the XCO₂, and the amplitude of the variation of CO₂ surface variation during this period is 4.8 times larger than the amplitude of the variation of XCO₂. According to the GEOS-PFIT model, CO₂ mole fraction decreases with altitude, especially above the tropopause height (Laughner et al. 2023). A good correlation between the

~~Deleted:~~ and the mean CH₄ surface mole fraction is 97 ppb larger than the XCH₄.

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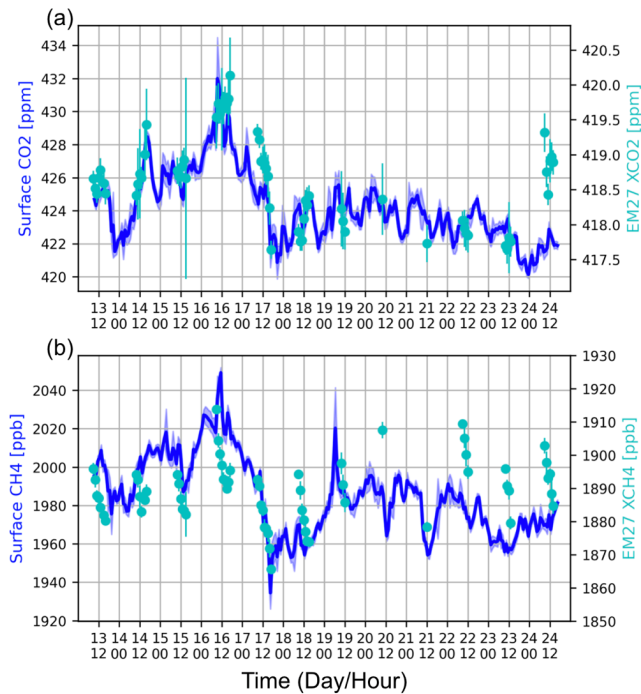
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261 surface CO₂ and XCO₂ is found, with an R of 0.74 (p_value<0.001). The CO₂ enhancement on 16
 262 May was observed both near the surface and in the column. We also notice that relatively high
 263 XCO₂ was observed on 24 May but with low CO₂ mole fractions at the surface. It is indicated that
 264 CO₂ enhancement occurs at high altitudes but not at the surface.



265
 266 **Figure 5.** Similar to Figure 4, but adding the EM27/SUN FTIR XCO₂ and XCH₄ hourly means
 267 and stds (right y-axis) between 13 and 24 May 2022.
 268

269 Similar to CO₂, the mean CH₄ surface mole fraction is 97 ppb larger than the XCH₄, and
 270 the amplitude of the variation of CH₄ near the surface during this period is 2.9 times larger than
 271 the amplitude of the variation of XCH₄. The CH₄ mole fractions in the stratosphere are much lower
 272 than those in the troposphere due to the chemical reaction and atmospheric dynamic transport
 273 (Sepúlveda et al. 2014; Wang et al. 2014). The correlation between the surface CH₄ and XCH₄ is
 274 relatively weak as compared to that between CO₂ and XCO₂ but still statistically significant
 275 (R=0.41; p_value<0.01). The weak correlation in CH₄ is probably due to that CH₄ has a much

larger vertical gradient than the CO₂ between the troposphere and the stratosphere (Sepúlveda et al. 2014). Therefore, vertical transport in the upper troposphere and lower stratosphere (UTLS) and horizontal transport in the stratosphere both make strong impacts on CH₄ column. Nevertheless, the CH₄ enhancement on 16 May was observed both near the surface and in the column.

To better understand the enhancement of CO₂ and CH₄ at QOMS on 16 May, we use the FLEXPART v10.4 backward simulations (Pisso et al., 2019) to show where the sources of the air mass are coming from during this campaign. The main settings of the FLEXPART model are listed in Table 3. Figure 6 shows the backward sensitivities on 7, 16 and 21 May 2022. Compared to 7 and 21 May, there is a significant air mass at QOMS on 16 May coming from North India, with higher gas concentrations. It is inferred that the enhancement of the measurements on 16 May 2022 is mainly due to the atmospheric transport.

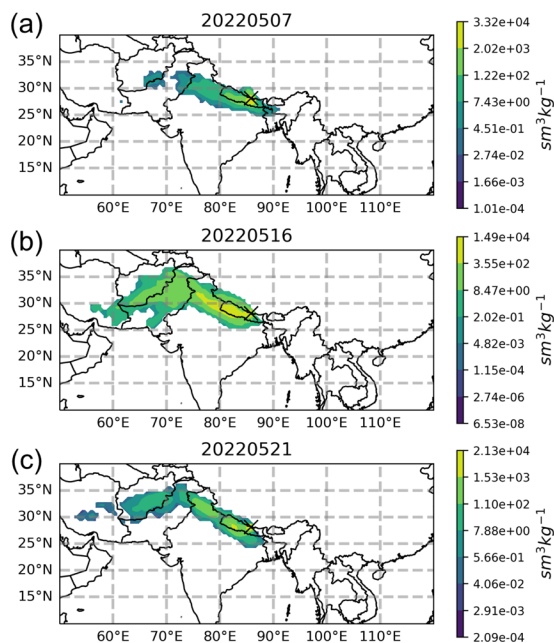


Figure 6. The spatial distribution of air backward sensitivities at QOMS between 04:00-05:00 (UTC) on 7, 16, 21 May 2022. The QOMS site is marked with a cross symbol.

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Table 3. 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

4.2 Ground-based FTIR against TROPOMI XCO measurements

In this section, we compare the ground-based EM27/SUN FTIR measurements (GB) to the TROPOMI satellite XCO measurements (SAT) at QOMS. We use the mean of GB measurements within ± 2 hours of each satellite overpass time (approximately from 12:30 to 17:30 LT), and select all the satellite measurements within a certain distance around QOMS. For each SAT-GB data pair, we apply the SAT *a priori* profile (TM5 model) as the common prior to reducing the uncertainty from different *a priori* profiles (Rodgers and Connor 2003). In addition, to get rid of the discrepancy caused by different surface altitudes of the FTIR and satellite measurements, the FTIR retrieved XCO is scaled to the same vertical range of each satellite measurement (Langerock et al. 2015). Figures 7b and 7c show the mean and std of the differences between co-located TROPOMI satellite and ground-based FTIR XCO measurements (SAT-GB) between 5 and 24 May, varying with the co-located distance criterion ranging from 10 to 105 km. The mean difference varies between -3.9 and -6.3 ppb. The mean difference enlarges with increasing distance between FTIR and TROPOMI measurements, while the std reaches the minimum (5.36 ppb) at the distance of 25 km. To ensure enough data pairs to get a robust comparison, we set the co-located distance to 25 km, resulting in 17 days with co-located FTIR and satellite measurements.

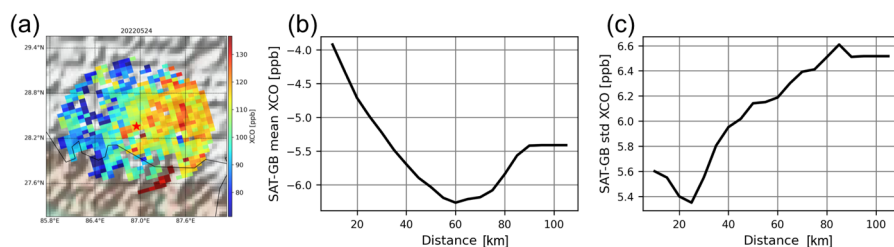


Figure 7. The TROPOMI XCO measurements within 100 km around the QOMS (red star) on 24 May 2022 (a). The mean (b) and std (c) of the differences between co-located TROPOMI satellite

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and ground-based EM27/SUN XCO measurements (SAT-GB) between 5 and 24 May varying with the co-located distance criterion.

The time series and scatter plot of the co-located FTIR and TROPOMI satellite XCO measurements at QOMS are shown in Figure 8. A good correlation between FTIR and TROPOMI XCO measurements is observed, with the R value of 0.81 ($p_value < 0.001$). The difference between FTIR and TROPOMI XCO measurements is -5.06 ± 5.36 ppb ($-4.7 \pm 5.1\%$), which is within the S5P mission requirements with a systematic error of 15% and random error of 10%. The relative bias at QOMS is also comparable with other places around the world (Sha et al. 2021; Martínez-Alonso et al. 2022). The EM27/SUN measurements at QOMS thus indicate that the TROPOMI XCO data has a good performance in this region.

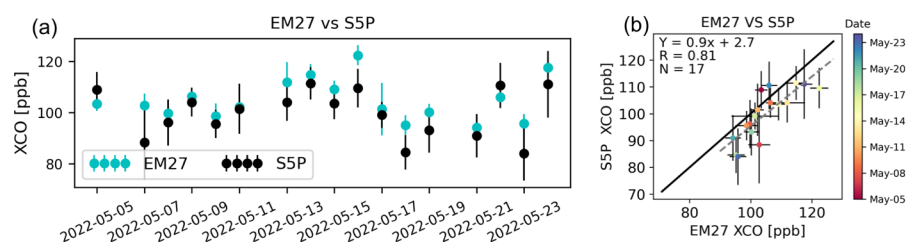
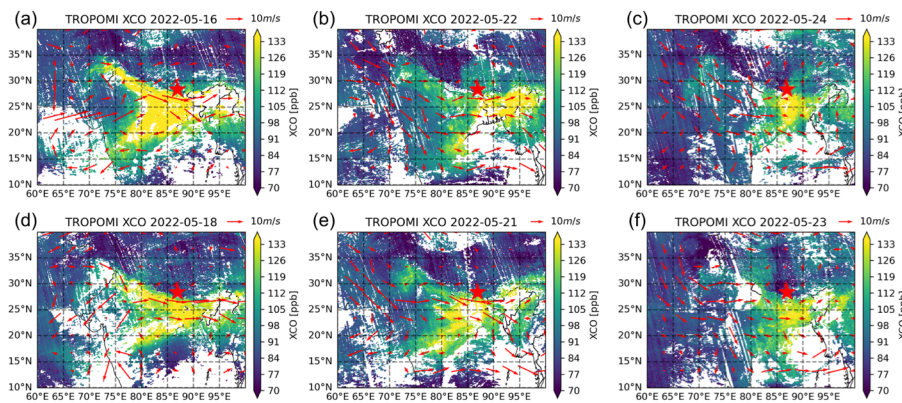


Figure 8. The co-located TROPOMI/S5P and EM27/SUN XCO measurements between 5 and 24 May 2022 (a). The errorbar of the satellite measurement is the std of all measurements within 25 km around the site, and the errorbar of the FTIR measurement is the std of all measurements within ± 2 hours within the satellite overpass time. The scatter plot of the TROPOMI/S5P and EM27/SUN XCO co-located measurements (b). The dashed line is the linear regression, R is the Pearson correlation coefficient, and N is the number of the co-located measurements. The dot is colored by the measurement date.

The high and low XCO values at QOMS are observed simultaneously from the ground-based FTIR and TROPOMI satellite measurements. Thanks to the good coverage of the TROPOMI satellite measurements, it is applied to show the spatial distributions of XCO around QOMS in a larger domain. Figure 9 shows the TROPOMI XCO measurements on 16, 22, and 24 May with relatively high XCO values, and on 18, 21, and 23 May, with relatively low XCO values around QOMS. The wind speed and wind direction are derived from the ERA5 reanalysis pressure-level data at 500 hPa (~ 5 km a.s.l.). We find that XCO at the south side of Mt. Qomolangma is much larger than that at the side edge, because of high anthropogenic emissions in Nepal and India

350 (Crippa et al. 2018). The relatively high XCO values at QOMS on 16, 22, and 24 May correspond
 351 to south winds, which bring the air mass with high CO mole fraction to QOMS. On 18 and 21 May,
 352 the wind direction was from the west to the east along the southern edge of the Himalayas
 353 mountains and did not bring air mass to QOMS so low XCO values are observed in all regions of
 354 the southern Tibetan Plateau. On 23 May, relatively high XCO values are observed at
 355 approximately 200 km west or east of QOMS, but low XCO values are observed at QOMS. Based
 356 on the TROPOMI satellite measurements and the wind data, we conclude that the day-to-day
 357 variation of XCO observed at QOMS is largely influenced by atmospheric transport, and the
 358 air mass transported from southern Asia can enhance the CO mole fractions over the Tibetan
 359 Plateau.

360



361

362 **Figure 2.** The XCO observed by the TROPOMI/S5P satellite on 16 (a), 22 (b), and 25 (c) May
 363 2022, with relatively high XCO values at QOMS (the red star), and on 18 (d), 21 (e), and 23 (f)
 364 May 2022, with relatively low XCO values at QOMS. The wind direction and wind speed are
 365 derived from the ERA5 reanalysis data at 500 hPa (~ 5 km a.s.l.).

366

367 4.3 Ground-based FTIR against OCO-2 XCO₂ measurements

368 Unlike the TROPOMI XCO measurements, very limited OCO-2 XCO₂ measurements are
 369 available due to its narrow swath. During this campaign, we only had two days, when the OCO-2
 370 satellite provided valid measurements (qflag = 0) around QOMS (Figure 10). The distances
 371 between the OCO-2 measurements and the FTIR site are about 480 km and 250 km on 8 and 24
 372 May, respectively. Note that the observation mode of OCO-2 is land glint on 8 May and land nadir
 373 on 24 May. As the OCO-2 and EM27/SUN both use GEOS-FPIT model simulations as the *a priori*

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profiles, no prior substitution correction is needed (Zhou et al. 2016). For each FTIR-satellite data pair, we correct the FTIR measurement to the same altitude of the satellite footprint. As we do not have the FTIR measurement around the OCO-2 overpass time (~15:30 CST), we use the mean of the FTIR measurements in the latest 1 hour.

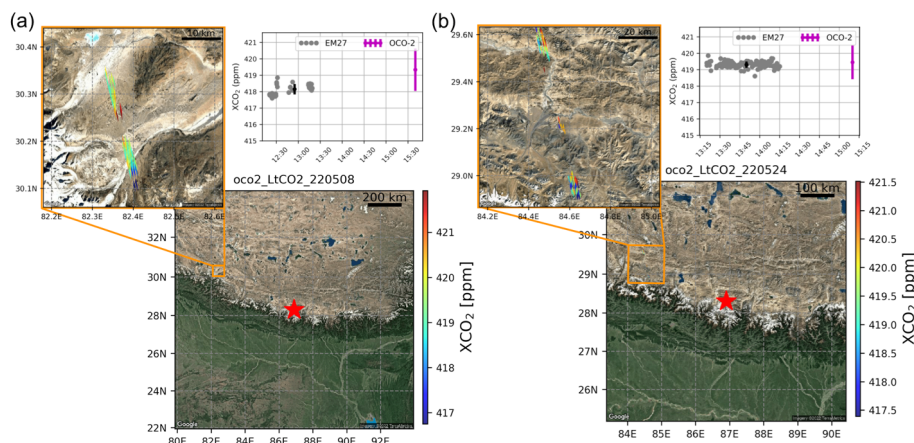


Figure 10. The XCO₂ observed by the OCO-2 satellite with glint mode on 8 May (a) and with nadir mode on 24 May (b) around QOMS (the red star on the satellite image from © Google Maps). For each day, the time series of the EM27/SUN XCO₂ individual measurements in the latest 1 hour are shown together with the mean and std of EM27/SUN measurements (black) and all co-located OCO-2 measurements (magenta) in the top right panels.

The mean and std of the differences between EM27/SUN and OCO-2 XCO₂ measurements (SAT-GB) are 1.21 ± 1.29 ppm and 0.21 ± 0.98 ppm on 8 and 24 May, respectively. The bias of OCO-2 XCO₂ measurements at QOMS is in the same order of magnitude as the bias found at global TCCON sites (Wunch et al. 2017). The reported uncertainty of the OCO-2 XCO₂ nadir measurements on 24 May is 0.65 ppm, which is slightly lower than the std of 0.98 ppm. However, the reported uncertainty of the OCO-2 XCO₂ glint measurements on 8 May is 0.57 ppm, which is much smaller than the std of 1.29 ppm.

As the footprint of OCO-2 is a bit far away from the QOMS site, we use the Copernicus Atmosphere Monitoring Service (CAMS) model simulations (Agustí-Panareda et al., 2023) in May 2020 to estimate the spatial variability of XCO₂ between the QOMS and OCO-2 footprints in this study. The mean and std of the differences in CAMS XCO₂ between the QOMS and OCO-

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2 footprint around 29.2°N, 84.5°E (~250 km) are 0.02 ppm and 0.27 ppm, respectively. The mean and std of the differences in CAMS XCO₂ between the QOMS and OCO-2 footprint around 30.2°N, 82.4°E (~480 km) are 0.10 ppm and 0.34 ppm, respectively. According to the CAMS model simulations, the XCO₂ spatial variability in this region is relatively small.

The OCO-2 measurements on these two days are both concentrated in a small region without intense human activity. It is assumed that the XCO₂ is stable in such a region. Figure 11 shows the bias of OCO-2 glint measurements (SAT-GB) varying with surface altitudes and retrieved surface albedos in the O₂-A band on 8 May 2022. It is found that the bias of OCO-2 land glint measurements is strongly related to the retrieved surface albedos (R=0.54) and footprint surface altitudes (R=0.48). Further correction of OCO-2 glint measurements in this region using the O₂-A band surface albedo/surface altitude is recommended, but more data are required. Regarding the OCO-2 land nadir measurements on 24 May, we do not find significant correlations between the bias and altitudes (R=-0.26) or surface albedos (R=0.17). Instead, the biases are randomly distributed.

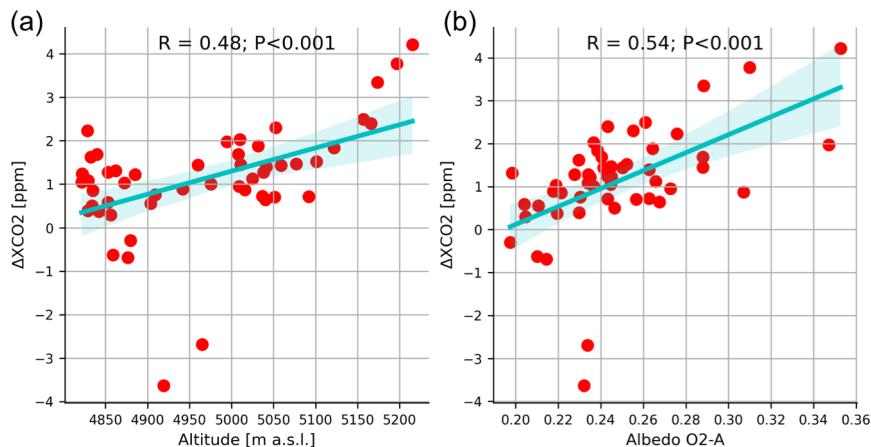


Figure 11. The bias of XCO₂ observed by OCO-2 land glint measurements ($\Delta\text{XCO}_2 = \text{SAT-GB}$) on 8 May 2022 varying with surface altitude (a) and surface albedo in O₂-A band (b).

5 Conclusions

The QTP serves as a huge carbon storage but is also sensitive to climate change. Currently, there is still a large uncertainty about the terrestrial ecosystem carbon sink in the QTP. Due to the

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tough environment, GHG measurements are scarce. In May 2022, an integrated greenhouse gases measurement campaign was carried out at QOMS within the framework of the Second Tibetan Plateau Scientific Expedition Program. In this study, we present the experiments about the *in situ* measurements near the surface and the ground-based EM27/SUN FTIR column measurements.

The following results are presented and discussed:

- 1) The *in situ* measurements near the surface at QOMS between 13 and 24 May 2022 show that the CO₂ and CH₄ mole fractions are 424.2±2.1 ppm and 1985.2±19.7 ppb, respectively. In addition, a good correlation (R=0.82) between the surface CO₂ and CH₄ mole fractions is observed.
- 2) The ground-based FTIR measurements at QOMS between 5 and 24 May show that the mean XCO₂, XCH₄, XCO, and XN₂O are 418.4±0.6 ppm, 1888.3±8.0 ppb, 106.2±8.3 ppb, and 321.6±3.2 ppb, respectively. The mean of XCO₂ at QOMS is about 0.5-3.5 ppm lower than six TCCON sites in the mid-latitude northern hemisphere during the same time period. The GHG measurements at QOMS seriously differ from the GEOS-PFIT model simulations, indicating the large uncertainty of the model simulations in this region.
- 3) The ground-based FTIR measurements at QOMS are compared to TROPOMI XCO satellite observations. The difference between FTIR and TROPOMI XCO measurements is -5.06±5.36 ppb (-4.7±5.1%), which is within the S5P mission requirements. A good correlation between FTIR and TROPOMI XCO measurements is also found, with an R of 0.81. Utilizing the good spatial coverage of TROPOMI satellite measurements together with the wind data, we find that the day-to-day variation of XCO observed at QOMS is largely affected by atmospheric transport. It is important to carry out long-term measurements to calculate the cross-regional transport in this region quantitatively.
- 4) The ground-based FTIR measurements at QOMS are also compared to OCO-2 XCO₂ observations. There were only two days with OCO-2 measurements within 500 km around QOMS (land glint mode on 8 May 2022 and land nadir mode on 24 May 2022). The mean differences between FTIR and OCO-2 XCO₂ measurements are 1.21±1.29 ppm and 0.21±0.98 ppm on 8 and 24 May, respectively. It is found that the bias of OCO-2 glint measurements on 8 May is relatively large, and it is statistically related to the retrieved surface albedos and surface altitudes. The quality of the OCO-2 XCO₂ land glint

measurements in this region should be further assessed when more ground-based measurements become available.

Appendix I

The uncertainties of the EM27/SUN CO₂, CH₄, CO and N₂O measurements at QOMS are estimated by perturbing the inputs using the GGG2020 code. In this study, we include contributions from instrumental effects (ILS), observation geometry (pointing offset), temperature profile, spectroscopy (line intensity), a priori dependence, and measurement noise (Table A1).

Table 12. List of sources, values used for the uncertainty analysis, and CO₂, CH₄, CO and N₂O column retrieval uncertainties for all measurements at QOMS site using the GGG2020 code. (ME: modulation efficiency amplitude; SNR: signal-to-noise ratio). The third column provides the partitioning of the error values between random (ran) and systematic (sys) contributions. Note that an uncertainty less than 0.01% is indicated as '-'.

Error source	Uncertainty value	sys/ran contribution [%]	CO ₂ column uncertainty (sys/ran) [%]	CH ₄ column uncertainty (sys/ran) [%]	CO column uncertainty (sys/ran) [%]	N ₂ O column uncertainty (sys/ran) [%]
Prior	2%	50/50	0.01/0.01	0.02/0.02	0.04/0.04	0.08/0.08
ILS (ME and phase error)	1% and 0.01rad	50/50	0.1/0.1	0.1/0.1	0.1/0.1	0.1/0.1
Pointing offset	0.1°	10/90	-/0.1	-/0.1	-/0.1	-/0.2
Temperature profile	2 K	50/50	0.03/0.03	0.15/0.15	0.80/0.80	0.07/0.07
Spectroscopy	2%	100/0	2.0/-	2.0/-	2.0/-	2.0/-
Measurement noise	1/SNR	0/100	-/0.1	-/0.1	-/0.3	-/1.2
Total			2.0/0.2	2.0/0.2	2.0/0.9	2.0/1.2

Data availability

The TCCON data were obtained from the TCCON Data Archive hosted by CaltechDATA at <https://tccodata.org>. The TROPOMI satellite data are available at <https://dataspace.copernicus.eu/> (registration request). OCO-2 satellite data are available at <https://ocov2.jpl.nasa.gov/science/oco-2-data-center/> (registration request). The ground-based FTIR measurements at QOMS are available upon request. The CAMS model simulations are publicly available at <https://ads.atmosphere.copernicus.eu/datasets/cams-global-ghg-reanalysis-egg4?tab=overview>.

Competing interests

The authors declare that they have no conflict of interest.

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Author contributions

MZ, YW, and MZ design the study. MZ wrote the manuscript with inputs from YW and MZ. XT and JD serve as the project leaders of the campaign. JB set up the EM27/SUN instrument. YM, WM, and ZX provided local support to collect the data. All authors have read and commented the manuscript.

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