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### Key Points:

- First results of long-term EM27/SUN measurements and satellite comparisons over Seoul
- EM27/SUN effective in monitoring greenhouse gases and assessing validity of satellite measurements over urban areas
- Satellites need higher resolutions and locally validated algorithms for urban monitoring

### Supporting Information:

Supporting Information may be found in the online version of this article.

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## Comparisons of Greenhouse Gas Observation Satellite Performances Over Seoul Using a Portable Ground-Based Spectrometer

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**Abstract** Satellites provide global coverage for monitoring atmospheric greenhouse gases, crucial for understanding global climate dynamics. However, their temporal and spatial resolutions fall short in detecting urban-scale variations. To enhance satellite reliability over urban areas, this study presents the first comprehensive analysis of long-term observations of column-averaged dry air mole fractions of CO<sub>2</sub>, CH<sub>4</sub>, and CO (XCO<sub>2</sub>, XCH<sub>4</sub>, XCO) using two ground-based fourier transform infrared spectrometers, EM27/SUNs, in a megacity. With over 2 years of observations, our study shows that EM27/SUN measurements can effectively capture the daily and seasonal variability of XCO<sub>2</sub>, XCH<sub>4</sub>, and XCO over Seoul, a megacity with complex topography and various emission sources. In addition, we use the advantage of having multiple greenhouse gas satellites targeting Seoul to compare with the EM27/SUNs. Our study highlights the importance of EM27/SUN observations in Seoul to identify the need for improvements in satellites to monitor greenhouse gas behaviors and emissions in urban areas.

**Plain Language Summary** This study examines how accurately satellites can monitor greenhouse gases over urban atmospheres to understand climate change. While satellites are good at covering large areas, they struggle to detect changes in cities. To improve these setbacks, this study uses ground-based instruments to measure greenhouse gases over 2 years and test satellite reliability over Seoul, a megacity with various emission sources as well as a complex terrain for observation. This study shows that the newly developed ground-based instruments, EM27/SUNs, are effective in tracking daily and seasonal changes in greenhouse gas concentrations and are useful tools in improving the validity of satellite observations in urban areas. The study suggests that using ground-based observations in addition to satellite data adapted for urban area monitoring is important for understanding greenhouse gas emissions in major cities like Seoul.

## 1. Introduction

To better manage atmospheric greenhouse gases, it is necessary to monitor and quantify emissions at all spatial scales, from global to national and urban levels. Column measurements are particularly useful for assessing greenhouse gas emissions as they are less affected by boundary layer height dynamics, surface fluxes, and vertical transport compared to in situ measurements (Dietrich et al., 2021; Wunch, Toon, et al., 2011). Satellites have been effective in large scale monitoring of column measurements of carbon compounds such as carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), and carbon monoxide (CO), including monitoring of areas where ground measurements are unable to cover. Greenhouse gas observation satellites have evolved to measure with high precision. However, despite improvements in temporal and spatial resolutions, many satellites still face setbacks due to coarse spatial coverage and sparse revisit times. In addition, although satellites show high precision in terms of global scale measurements, margins of error and bias still exist for observations over cities and point-source areas.

Ground-based remote sensing of column measurements has been of great importance for monitoring atmospheric concentrations of greenhouse gases as well as validating satellite observations. The Total Carbon Column Observing Network (TCCON), which uses Bruker IFS 125HR high-resolution spectrometers, is one of the main networks for long-term monitoring of greenhouse gases and ground validation for satellites. The TCCON sites are spread throughout various parts of the globe and retrieve precise and accurate column abundances of CO<sub>2</sub>, CH<sub>4</sub>,

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and CO amongst other gases (Wunch, Toon, et al., 2011; Wunch, Wennberg, et al., 2011). However, TCCON sites mainly exist in clean areas, lacking representation in urban and emission-heavy regions due to operational challenges and instrument portability limitations. Therefore, to fill the gap of TCCON measurements, recent studies have been measuring column-averaged concentrations of greenhouse gases using portable ground-based remote sensing Fourier transform infrared (FTIR) spectrometers. The Collaborative Carbon Column Observing Network (COCCON) consists of EM27/SUNs which are portable, low resolution FTIR spectrometers developed by Karlsruhe Institute of Technology (KIT) and BrukerOptics™ (Frey et al., 2019). EM27/SUN spectrometers are considered to be the low-cost, mobile, and easy-to-deploy supplements to TCCON spectrometers. Their performance has been extensively tested in comparison to TCCON and AirCores along with other portable FTIR spectrometers in the framework of ESA's Fiducial Reference Measurements for Greenhouse Gases (Mostafavi Pak et al., 2023; Sha et al., 2020). Using the advantage of the portability, EM27/SUNs are widely used in validating spaced-based measurements of XCO<sub>2</sub>, XCH<sub>4</sub>, and XCO (Butz et al., 2022; Frey et al., 2021; Jacobs et al., 2020; Reißmann et al., 2022; Tu et al., 2022). Validation of satellites against ground-based reference measurements is crucial as satellites broaden their global-scale observations to diverse usages such as evaluating anthropogenic greenhouse gas emissions in small-scale urban areas. Therefore, it is essential to ensure satellites accurately detect targeted gases, especially in complex urban atmospheres.

Seoul is a megacity with a population of around 10 million people and contains various anthropogenic sources of greenhouse gases within the city. Located in a basin on the west coast of the Korean peninsula, Seoul is surrounded by mountains and characterized by complex topography, including clusters of hills and the Han river running through the center of the city. The EM27/SUN site at Seoul National University is located in the southern part of Seoul in a valley next to the side of Mt. Gwanak. As measured with the Orbiting Carbon Observatory-2 satellite, South Korea has low to moderate signal-to-noise ratio in the oxygen A-band near 0.76 μm, and weak and strong CO<sub>2</sub> bands located near 1.6 and 2.0 μm, respectively (Eldering et al., 2017). The aerosol optical depth values at 500 nm in Seoul range from around 0.1 to 0.7 (Lee et al., 2021). Climatologically, heavy precipitation over Korea mainly occurs during late June through July, which accounts for more than 40% of the annual precipitation (Jung et al., 2002).

This study provides the first comprehensive analysis of long-term measurements of column-averaged dry air mole fractions of CO<sub>2</sub>, CH<sub>4</sub>, and CO (hereafter, XCO<sub>2</sub>, XCH<sub>4</sub>, XCO) in the atmosphere above a megacity–Seoul, South Korea—using two ground-based FTIR spectrometers, EM27/SUNs. The EM27/SUN measurements are performed at Seoul National University (SNU) (37.4641°N, 126.9537°E, 98 m a.s.l), and we present the variations of XCO<sub>2</sub>, XCH<sub>4</sub>, XCO observations made from May 2020 to December 2022. In addition, we use the ground-based observations to assess the reliability and validity of satellite measurements over the urban area of Seoul. Despite the complex topography of the urban area, Seoul benefits from having multiple greenhouse gas satellite observations over the city with most of them conducting target observations. We use the comparisons of five different greenhouse gas observation satellites—Orbiting Carbon Observatory-2 (OCO-2), Orbiting Carbon Observatory-3 (OCO-3), Greenhouse gases Observing SATellite (GOSAT), Greenhouse gases Observing SATellite-2 (GOSAT-2), and TROPOspheric Monitoring Instrument (TROPOMI)—to evaluate the satellite measurements of XCO<sub>2</sub>, XCH<sub>4</sub>, and XCO with that of our EM27/SUN measurements over the megacity Seoul.

## 2. Material and Methods

### 2.1. EM27/SUN

The EM27/SUN is a portable FTIR spectrometer which was developed by KIT in collaboration with BrukerOptics™. The spectrometer records direct solar spectra in the near-infrared (NIR) window and has an optical path difference of 1.8 cm which is equivalent to a spectral resolution of 0.5 cm<sup>-1</sup>. The EM27/SUN spectrometer records double-sided direct current (DC) coupled interferograms which make an average of 10 scans in 58 s. The measurements are made using a room temperature (RT) indium gallium arsenide (InGaAs) detector of a spectral coverage of about 5,500–11,000 cm<sup>-1</sup> and a DC coupled wavelength-extended RT InGaAs detector with a spectral coverage of 4,000–5,500 cm<sup>-1</sup> (Gisi et al., 2012; Hase et al., 2016). Covering the same spectral region as the TCCON and TROPOMI, the EM27/SUN measures column concentrations of CO<sub>2</sub>, CH<sub>4</sub>, CO, H<sub>2</sub>O and O<sub>2</sub> (Frey et al., 2019; Sha et al., 2020).

Two EM27/SUN spectrometers (sn142 and sn144) are located at SNU and have been measuring in mostly cloudless daytime conditions. The first EM27/SUN (sn142) started operating in May 2020 and the

second EM27/SUN (sn144) since January 2021. Both spectrometers, which are also part of COCCON, have been calibrated with the reference EM27/SUN located at KIT. The instrumental line shape (ILS) parameters for the two spectrometers at SNU were measured at the COCCON central calibration laboratory hosted at KIT (Alberti et al., 2022). The ILS parameters for sn142 and sn144 are within the range of the EM27/SUNs which are part of COCCON, and these ILS parameters are taken into account while doing the retrieval. A weatherproof data logger, the HOBO<sup>®</sup> U30-NRC Weather Station, is located next to the spectrometers to measure surface pressure and local climatological data. We use the PROFFAST retrieval algorithm Version 1 (1 April 2020) for COCCON data analysis developed by KIT with GGG2014 a priori profiles to analyze the measured spectra. The retrieval code is a least-squares fitting algorithm that adjusts atmospheric spectra by scaling the a priori trace gas profile with the ILS as the input parameter which is described in more detail in Frey et al. (2021). PROFFAST includes a preprocessing software to convert the raw interferograms to spectra with a DC correction, a phase correction scheme for double-sided interferograms, and quality control tests (Sha et al., 2020). A post-processing is also included to scale the retrievals to the TCCON measurements and apply air mass-independent and air mass-dependent corrections to generate final column-averaged dry air mole fractions of CO<sub>2</sub>, CH<sub>4</sub>, and CO (Frey et al., 2021). Our analysis covers a total of 213 clear sky days of measurements taken from 26 May 2020 to 31 December 2022, mostly during the afternoon from 12:00 to 17:00 local time. In addition, we filtered out the days with <10 measured spectra from our EM27/SUN.

## 2.2. Collocation of Satellite Observations

We compare daily median satellite measurements of XCO<sub>2</sub>, XCH<sub>4</sub>, and XCO with those of the EM27/SUNs to assess the reliability of measurements over Seoul. Five satellites are used for comparison with two EM27/SUNs measuring side-by-side at the same location in SNU. For the comparison of XCO<sub>2</sub>, we use OCO-2, OCO-3, GOSAT and GOSAT-2 observations. The OCO-2 has been making target observations of Seoul since April of 2021, while OCO-3 has been making Snapshot Area Map (SAM) observations over Seoul since June of 2020. Moreover, GOSAT-2 has also been making target observations of Seoul since November of 2020. As for the comparisons of XCH<sub>4</sub>, we use TROPOMI, GOSAT, and GOSAT-2 observations. TROPOMI provides both the standard and bias-corrected XCH<sub>4</sub> data, and we use the bias-corrected XCH<sub>4</sub> products for the analysis in our study. The bias correction, also performed for the OCO-2 and GOSAT retrievals, is based on the retrieved surface albedo to improve the accuracy and fit of the satellite products (Hasekamp et al., 2021). Lastly, for XCO, we compare the observations from TROPOMI and GOSAT-2 with our ground-based measurement. In the case of TROPOMI, CO is provided as total column density data. Therefore, we use the method from Sha et al. (2021) to calculate the XCO values from the ratio of total column CO divided by the total column of dry air. The description of satellite data products used in this study are detailed in Table S1 of Supporting Information S1.

A sensitivity test was made to determine the threshold of satellite measurement collocations to use for comparison with the EM27/SUNs. Satellite measurements that were made within a 0.1-degree buffer around the EM27/SUN measurement location were considered for analysis, and same day EM27/SUN spectral observations made during  $\pm 30$  min of the satellite overpass were taken for comparison with the satellite data (Figure S1 in Supporting Information S1). On days when satellite overpasses coincide with the measurements from both EM27/SUNs, we average the data from the two ground-based FTIR instruments. For GOSAT, we gave a larger buffer of 0.5° to obtain more satellite samples. The same collocation of  $\pm 30$  min of the EM27/SUN measurements were used to match the observations made during the GOSAT satellite overpass.

## 3. Results

### 3.1. Seoul EM27/SUN Measurements

Two EM27/SUN spectrometers which are operated side-by-side at the same location of the SNU site show good agreement for all three measurements of XCO<sub>2</sub>, XCH<sub>4</sub>, and XCO ( $r^2 = 0.99$ ,  $r^2 = 1.00$ , and  $r^2 = 0.99$ , respectively) (Figure S2 in Supporting Information S1). Both the daily and monthly measurements of the EM27/SUN XCO<sub>2</sub> capture a clear seasonal cycle with high seasonal median in the spring ( $417.14 \pm 1.43$  ppm) and winter ( $416.29 \pm 2.05$  ppm) months, and low seasonal median in the summer ( $414.70 \pm 2.76$  ppm) and autumn ( $414.31 \pm 2.79$  ppm) months (Figure S3 in Supporting Information S1). We observe the monthly and seasonal variations by detrending the EM27/SUN data of two complete years of 2021 and 2022 and calculating the

monthly medians. The maximum monthly median concentrations are observed to be in April ( $418.13 \pm 1.43$  ppm) and minimum monthly median concentrations in September ( $410.94 \pm 2.42$  ppm) (Figure S4 in Supporting Information S1). Seoul XCO<sub>2</sub> measurements reflect the typical behavior of XCO<sub>2</sub> patterns seen at Northern Hemispheric sites where the CO<sub>2</sub> concentrations show a minimum in the summer due to the drawdown of CO<sub>2</sub> during the growing season of the terrestrial biosphere, and an increase in the winter and spring where the influence of vegetation is small (Graven et al., 2013). For XCH<sub>4</sub>, there is an opposing pattern compared to what is seen in the seasonal patterns of XCO<sub>2</sub>. The lowest seasonal median is found in spring ( $1,888.38 \pm 11.39$  ppb) and highest seasonal median is found in autumn ( $1,912.95 \pm 9.49$  ppb), with XCH<sub>4</sub> concentrations showing a peak in concentration in June ( $1,914.55 \pm 19.16$  ppb) and September ( $1,915.60 \pm 6.33$  ppb). Seoul XCH<sub>4</sub> measurements show similar patterns with XCH<sub>4</sub> measurements at Xianghe and Thessaloniki, showing low values until spring, rising during the summer, and reaching a maximum in autumn (Mermigkas et al., 2021; Yang et al., 2020). In the case of XCO, the daily pattern is variable and does not show a clear seasonal pattern. The seasonal median in order from the highest to lowest is spring ( $114.62 \pm 11.34$  ppb), winter ( $111.20 \pm 14.56$  ppb), summer ( $101.88 \pm 20.27$  ppb), and autumn ( $97.17 \pm 13.41$  ppb). The maximum monthly median is found in June ( $120.62 \pm 19.49$  ppb), and the minimum monthly median is found in October ( $93.12 \pm 9.99$  ppb). A similar pattern is also shown in FTIR measurements located in Karlsruhe, Pasadena, and Paris, where the XCO values are found to be the highest in spring and lowest in summer and autumn (Che et al., 2022). Despite different seasonal patterns, all three measurements of XCO<sub>2</sub>, XCH<sub>4</sub>, and XCO in Seoul show agreeing patterns of peaks on high concentration days. This shows the effects of anthropogenic emissions from surrounding cities or long-range atmospheric transport to Seoul from different regions.

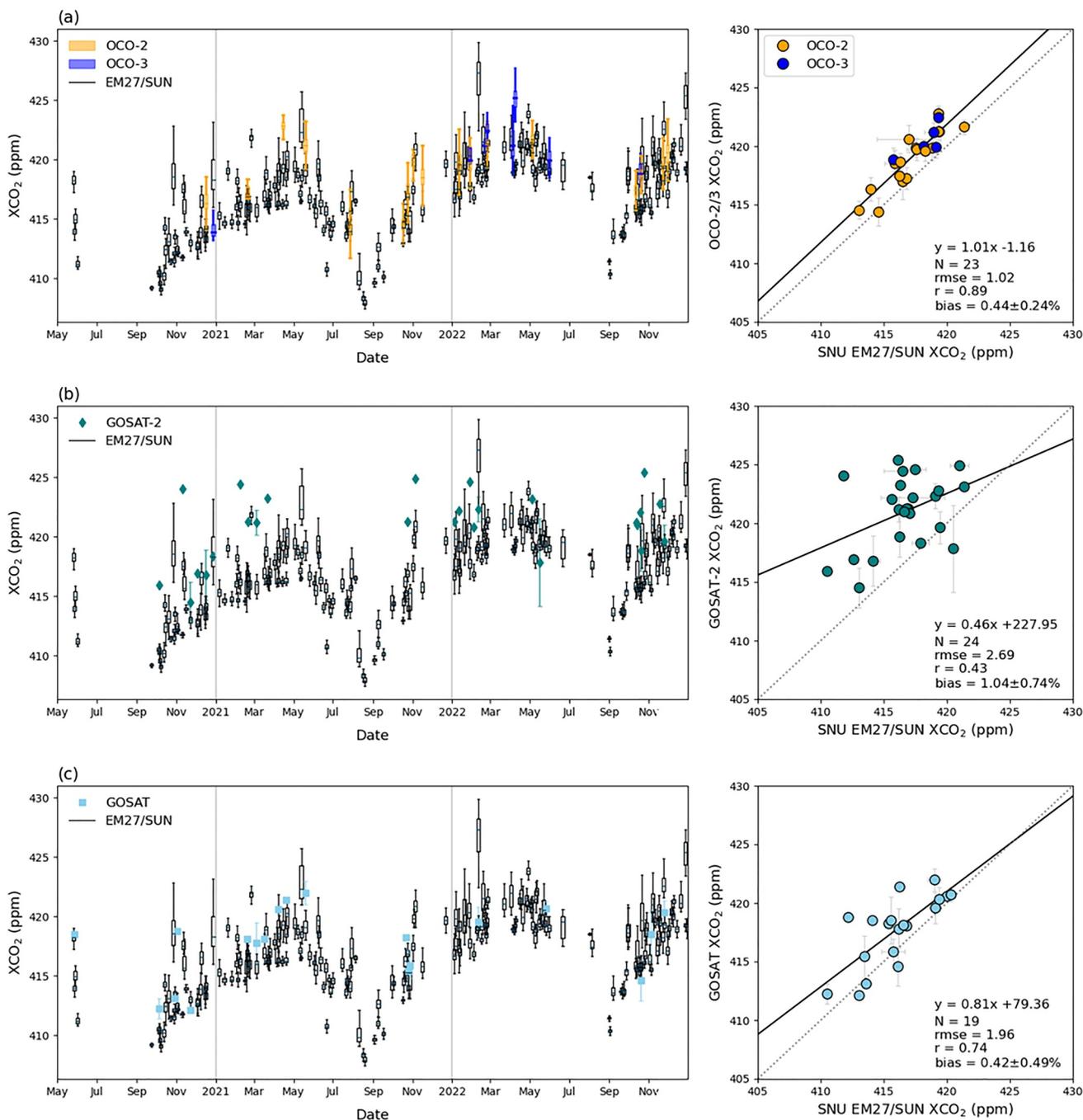
## 3.2. Comparisons of EM27/SUN and Satellite Observations

### 3.2.1. XCO<sub>2</sub>

Figure 1 shows the comparisons between EM27/SUN and satellite measurements. For XCO<sub>2</sub>, a total of 17 OCO-2 and 6 OCO-3 measurements with “good” quality flags over Seoul were used for comparison with the EM27/SUNs, while 24 and 19 “good” quality measurements of GOSAT-2 and GOSAT, respectively, were used for comparison in the analysis of the study period. The OCO-2 and OCO-3 observations showed agreement in pattern with that of the EM27/SUNs with a correlation coefficient ( $r$ ) of 0.89 and root mean square error (RMSE) of 1.02 ppm. The OCO-2/3 observations well-capture the seasonal variability of Seoul with high values in the spring and low values in the late summer (Figure 1a). The EM27/SUN measurements used for comparison with the satellites have a median value of  $417.59 \pm 1.98$  ppm, while OCO-2 and OCO-3 satellites have a median of  $419.84 \pm 2.24$  ppm. Compared to the EM27/SUN, the OCO-2 and OCO-3 satellites show a bias of  $0.44\% \pm 0.24\%$ . Our results are similar to the comparisons of OCO-2 XCO<sub>2</sub> and OCO-3 XCO<sub>2</sub> retrievals to TCCON stations which show an RMS difference of less than 1.5 ppm and an RMSE of  $\cong 1$  ppm, respectively (O'Dell et al., 2018; Taylor et al., 2020; Wang et al., 2017; Wunch et al., 2017).

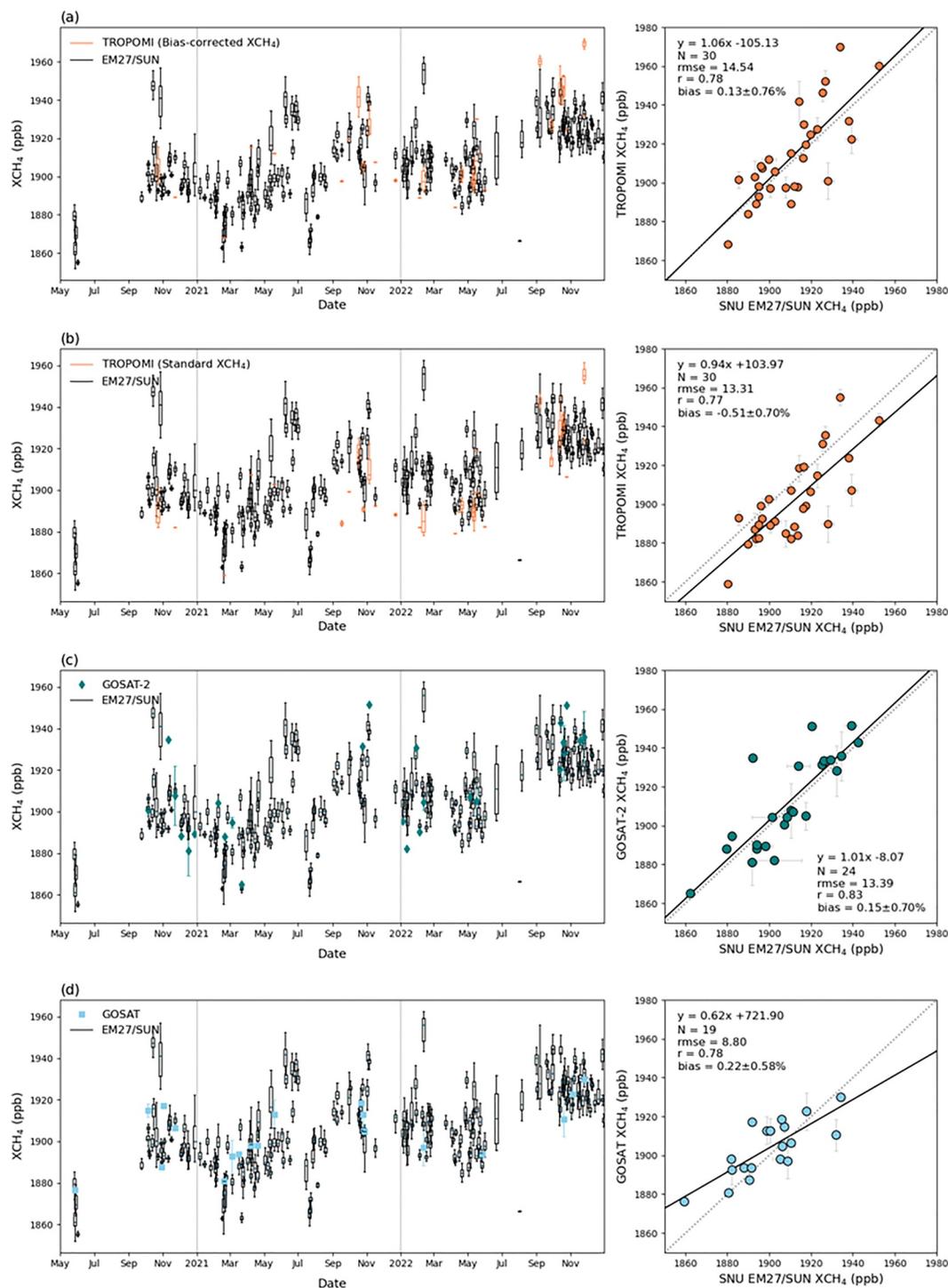
The comparison of GOSAT-2 daily observations with that of the EM27/SUN shows lower agreement ( $r = 0.43$ , RMSE = 2.69 ppm) and a higher bias of  $1.04\% \pm 0.74\%$  than that of the OCO-2/3 satellites (Figure 1b). The calculated bias of GOSAT-2 XCO<sub>2</sub> compared to EM27/SUN measurements is within the bias calculated from GOSAT-2 compared to TCCON measurements which corresponds to  $<1.7\%$  (Buchwitz et al., 2022). GOSAT-2 measurements compared with the ground-based observations have a median of  $421.26 \pm 2.97$  ppm, while the collocated EM27/SUN measurements have a median value of  $416.70 \pm 2.73$  ppm. The difference in results is most likely attributed to the low spatial resolution of the GOSAT-2 data. The OCO-2 and OCO-3 have high spatial resolutions of  $\sim 3$  and  $\sim 4$  km<sup>2</sup>, respectively, while GOSAT-2 has a lower spatial resolution of  $\sim 10$  km<sup>2</sup>. Moreover, GOSAT-2 V02.00 data products are fairly new and have not been bias-corrected, to which the bigger difference between the EM27/SUN and GOSAT-2 observations could also be attributed.

We also compared GOSAT XCO<sub>2</sub> measurements with EM27/SUN XCO<sub>2</sub> measurements. GOSAT measurements showed agreeing patterns with the ground-based observations, capturing the seasonal trends over Seoul better than the GOSAT-2 measurements (Figure 1c). The EM27/SUN measurements used for the GOSAT comparison have a median value of  $416.14 \pm 2.67$  ppm, while the collocated GOSAT measurements have a median of  $418.26 \pm 2.93$  ppm. GOSAT and EM27/SUN XCO<sub>2</sub> measurements show a correlation of 0.74 and a bias of  $0.42\% \pm 0.49\%$ . The results of the comparison of GOSAT and EM27/SUN agree well with the results of the



**Figure 1.** Satellite comparisons of (a) OCO-2, OCO-3, (b) GOSAT-2, and (c) GOSAT XCO<sub>2</sub> with EM27/SUN XCO<sub>2</sub>. Satellite soundings of OCO-2, OCO-3, and GOSAT-2 within the 0.1-degree buffer around the measurement site were used for comparison. For GOSAT, satellite soundings within a 0.5-degree buffer around the measurement site were used. EM27/SUN measurements made within  $\pm 30$  min of the satellite overpass were used for comparison. Daily median values of the EM27/SUN and satellite measurements were used for comparison.

comparison between OCO-2 and EM27/SUN, showing a similar bias but a larger RMSE for the GOSAT and EM27/SUN comparison. Finally, the average difference of GOSAT XCO<sub>2</sub> measurement compared to Seoul EM27/SUN measurements is  $1.75 \pm 2.02$  ppm.



**Figure 2.** Satellite comparisons of (a) TROPOMI bias-corrected, (b) TROPOMI standard, (c) GOSAT-2, and (d) GOSAT XCH<sub>4</sub> with EM27/SUN XCH<sub>4</sub>.

### 3.2.2. XCH<sub>4</sub>

The comparison between the bias-corrected TROPOMI XCH<sub>4</sub> measurements and EM27/SUN XCH<sub>4</sub> measurements are shown in Figure 2a. Despite the daily coverage of TROPOMI, many days of satellite XCH<sub>4</sub> measurements are lost when the quality filtering is applied and only a total of 30 days of measurements are used for

analysis. The TROPOMI bias-corrected XCH<sub>4</sub> measurements show high agreement with the EM27/SUN observations ( $r = 0.78$ , RMSE = 14.54 ppb). TROPOMI bias-corrected XCH<sub>4</sub> used for comparison has a median value of  $1,907.98 \pm 23.19$  ppb, whereas the collocated EM27/SUN XCH<sub>4</sub> has a median of  $1,911.21 \pm 17.11$  ppb. Compared to the EM27/SUN, the bias-corrected TROPOMI XCH<sub>4</sub> shows a very small bias of  $0.13\% \pm 0.76\%$  over Seoul, which is well within the bias requirement of 1.5% for TROPOMI XCH<sub>4</sub>. The comparison of the standard TROPOMI XCH<sub>4</sub> data product compared to the Seoul EM27/SUN observations show a lower correlation ( $r = 0.77$ , RMSE = 13.31) and a larger bias ( $-0.51\% \pm 0.70\%$ ) than that of the bias-corrected XCH<sub>4</sub> measurements (Figure 2b), indicating that the bias-correction of TROPOMI XCH<sub>4</sub> products over Seoul improves the retrieval results.

GOSAT-2 XCH<sub>4</sub> values showed good correlating patterns with the EM27/SUN XCH<sub>4</sub> values ( $r = 0.83$ , RMSE = 13.39 ppb) (Figure 2c). Although some days have large variations of GOSAT-2 XCH<sub>4</sub> concentrations, most of the satellite observations fall close within the range of the EM27/SUN XCH<sub>4</sub> concentrations. As a result, even with a coarser spatial resolution compared to TROPOMI, GOSAT-2 XCH<sub>4</sub> shows a slight bias of  $0.15\% \pm 0.70\%$  compared to that of the EM27/SUN. The calculated GOSAT-2 XCH<sub>4</sub> bias compared to the Seoul EM27/SUN is well within the bias requirement of GOSAT-2 XCH<sub>4</sub> products which has been validated with TCCON XCH<sub>4</sub> measurements to be less than 1.3% (Buchwitz et al., 2022). The collocated XCH<sub>4</sub> of GOSAT-2 and EM27/SUN show almost identical median values of  $1,906.06 \pm 24.03$  ppb and  $1,909.46 \pm 19.84$  ppb, respectively.

Of the three satellites, GOSAT XCH<sub>4</sub> shows the least agreement with the daily EM27/SUN XCH<sub>4</sub> observations. However, the satellite measurements still show good correlation with those of the ground-based EM27/SUN measurements ( $r = 0.78$ , RMSE = 8.80 ppb, bias =  $0.22\% \pm 0.58\%$ ) (Figure 2d). The EM27/SUN XCH<sub>4</sub> used for satellite comparison has a median of  $1,900.46 \pm 17.59$  ppb, while GOSAT XCH<sub>4</sub> has a median of  $1,904.79 \pm 14.04$  ppb. The average difference of GOSAT XCH<sub>4</sub> and EM27/SUN XCH<sub>4</sub> observations is calculated to be  $4.10 \pm 11.03$  ppb. This is within the range of GOSAT XCH<sub>4</sub> validation results with the TCCON site at Saga, Japan where the average difference between GOSAT and the ground-based high-resolution Fourier transform spectrometer measurements was  $-7.6 \pm 13.7$  ppb (Ohyama et al., 2015). Overall, we find that there is a very good agreement between the Seoul EM27/SUN and all three XCH<sub>4</sub> observing satellites used in our study.

### 3.2.3. XCO

We compared the TROPOMI XCO measurements to the EM27/SUN XCO measurements. In the case of TROPOMI XCO, daily observations are made in the Seoul area which allow for a large number of good quality samples to compare with our EM27/SUN measurements (Figure 3a). The comparison of TROPOMI XCO and EM27/SUN XCO shows satisfactory results with good agreement and the satellite capturing daily patterns similar to the ground observations ( $r = 0.92$ , RMSE = 6.61 ppb). The median values also show high agreement with the collocated TROPOMI XCO having a median of  $1,16.63 \pm 16.89$  ppb and the EM27/SUN XCO having a median of  $1,08.42 \pm 15.54$  ppb. Moreover, the calculated bias of TROPOMI XCO compared to the EM27/SUN XCO data is  $7.19\% \pm 5.86\%$ , which is well within the TROPOMI mission requirements for a bias of 15%. The bias of Seoul EM27/SUN and TROPOMI measurements is lower compared to the TROPOMI validations against TCCON stations which show a bias of  $9.14\% \pm 3.33\%$  (Sha et al., 2021), but higher than the bias calculated to be  $2.05\% \pm 7.82\%$  at Xianghe (Yang et al., 2020) and  $3.06\% \pm 5.56\%$  at Thessaloniki (Mermigkas et al., 2021). Satellite XCO observations from TROPOMI are even able to capture the days when there are peaks of XCO concentrations over Seoul which have also been measured from the ground-based measurements.

The GOSAT-2 XCO observations show a high correlation to the EM27/SUN measurements ( $r = 0.95$ , RMSE = 4.43 ppb) (Figure 3b). GOSAT-2 XCO measurements used for comparison have a median value of  $1,19.51 \pm 13.29$  ppb, while the collocated EM27/SUN XCO measurements have a median of  $1,04.05 \pm 12.66$  ppb. Although GOSAT XCO shows a high correlation to the ground observations, the satellite-observed XCO concentrations from GOSAT-2 compared to the EM27/SUN XCO concentrations have a high bias of  $15.09\% \pm 4.34\%$ . In addition, GOSAT-2 observations were not as effective in detecting spikes in urban XCO concentrations as with the TROPOMI XCO observations.

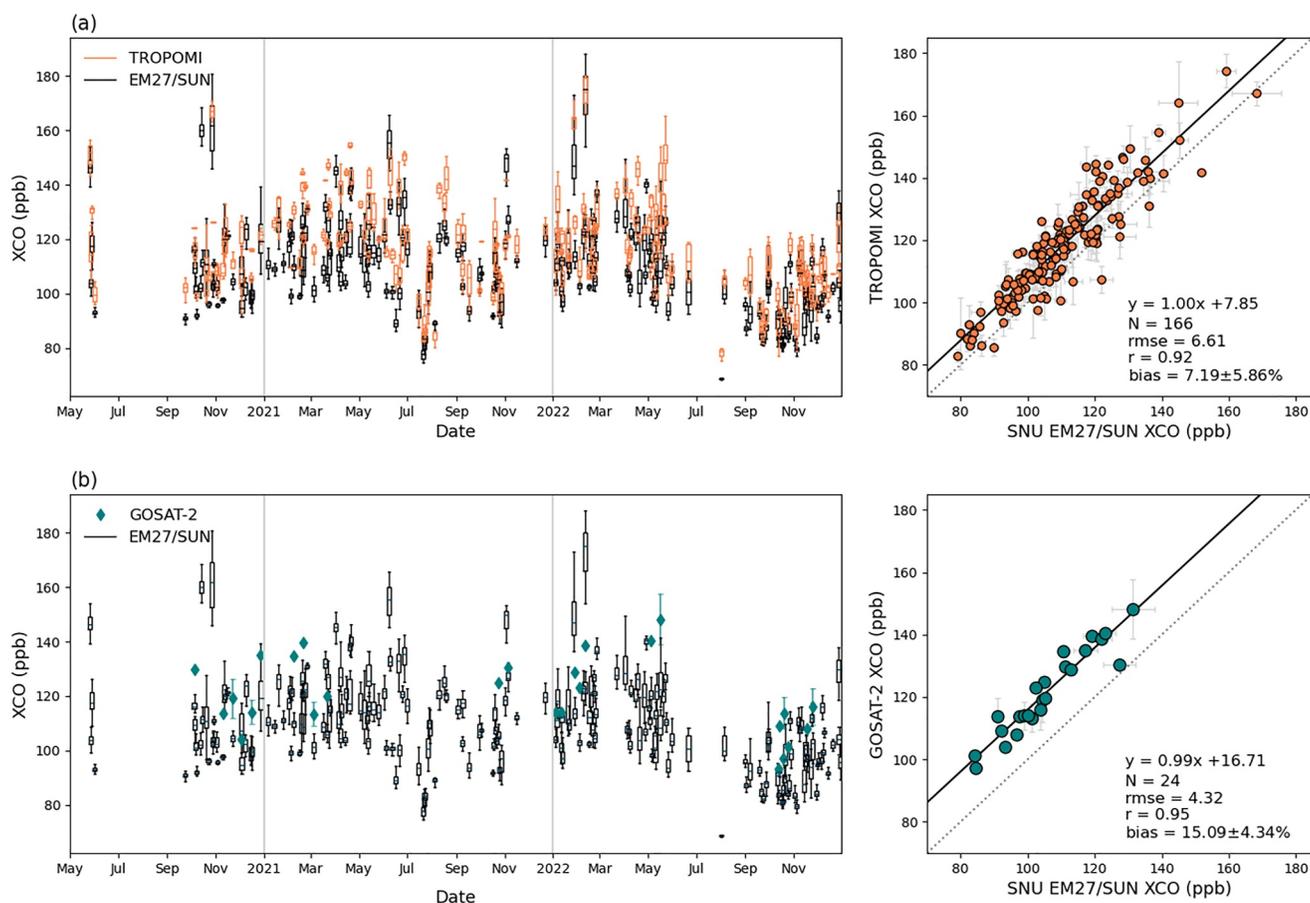


Figure 3. Satellite comparisons of (a) TROPOMI XCO and (b) GOSAT-2 XCO with EM27/SUN XCO.

#### 4. Discussion

Two EM27/SUNs, the first to be operated in South Korea, are located at the SNU site for the purpose of measuring variations of XCO<sub>2</sub>, XCH<sub>4</sub>, and XCO. We find that the long-term measurements of greenhouse gases from the two EM27/SUNs show good agreement as well as clear seasonal patterns. Aside from GOSAT-2, XCO<sub>2</sub> measurements from the OCO-2, OCO-3, and GOSAT are in good agreement with the XCO<sub>2</sub> measurements of the EM27/SUN, albeit with higher bias compared to the EM27/SUN. Seoul is a megacity with widespread anthropogenic emissions which could have affected the satellite observations that have a much wider spatial coverage than ground-based measurements, resulting in high bias. Moreover, the OCO-2 and OCO-3 XCO<sub>2</sub> concentrations show better agreement with that of the EM27/SUN than the GOSAT-2 XCO<sub>2</sub> concentrations. On the other hand, GOSAT performed quite well with the comparisons to the EM27/SUN, showing a similar bias compared to the OCO-2/3 measurements but with a higher RMSE value. Larger footprints of GOSAT and GOSAT-2 may hinder accurate CO<sub>2</sub> representation over Seoul's complex topography. Satellites with high spatial resolutions that pinpoint source areas are needed in the future to characterize the individual emission sources on a facility or area scale.

For XCH<sub>4</sub> measurements, TROPOMI, GOSAT-2, and GOSAT XCH<sub>4</sub> concentrations show a small bias compared to the EM27/SUN XCH<sub>4</sub> concentrations. GOSAT-2 XCH<sub>4</sub> has the best fit with the Seoul XCH<sub>4</sub> measurements, showing a slight bias of 0.15%. For TROPOMI XCH<sub>4</sub>, much of the data were lost after passing the quality filtering of data. Improved algorithms are necessary for retrievals of atmospheric CH<sub>4</sub> concentrations from satellites over urban areas. Especially, better filtering of clouds is needed as XCH<sub>4</sub> is much more sensitive to clouds compared to XCO retrievals. Furthermore, in the case of Seoul, CH<sub>4</sub> concentrations do not have much variation as there are not many considerable sources of methane within the city, excluding some large methane emission areas such as wastewater treatment facilities (Park et al., 2022). Although other hotspot

areas exist within the city, they are not large enough to be captured by satellites with resolutions like that of TROPOMI, GOSAT, and GOSAT-2.

In the case of XCO concentrations, TROPOMI XCO and GOSAT-2 XCO concentrations show good agreement with EM27/SUN XCO concentrations. In particular, TROPOMI XCO showed excellent agreement with the Seoul EM27/SUN measurements, having a bias of 7.19% which is well within the TROPOMI bias requirements. Especially, the daily frequency of TROPOMI observations in addition to the high spatial resolution allow the satellite to accurately capture the atmospheric variations of XCO concentrations. This also enables the satellite to detect days with high CO concentrations which have also been observed from the ground measurements. GOSAT-2 XCO measurements are also in agreement to the EM27/SUN XCO measurements with a high level of correlation, but in general, show a higher bias compared to the TROPOMI XCO measurements. However, GOSAT-2 data products are fairly new and more retrieval updates as well as bias corrections can lead to better agreement with ground-based observations. Several validation studies and their results have shown that updates and major improvements in retrieval algorithms lead to improvements in satellite and ground-based measurements as well as decrease in satellite biases (Lindqvist et al., 2015).

In addition to the systemic biases, measurement biases of satellites which are mostly dependent on aerosol content and the amount of sunlight of the observation sites also exist. Urban areas tend to have high aerosol content, which lead to changes in the light path and can result in higher measurement bias. Further studies should explore aerosol effects on satellite biases compared to ground measurements. Moreover, our study utilized an earlier version of the preprocessing software, PROFFAST, due to ongoing updates in the more recent Version 2. The data of this study was processed with the version that has been in use for a longer period of time, but future satellite comparisons should be made with updated software. Finally, this study has limitations as it did not include the application of averaging kernels and different a priori. Future studies can use the EM27/SUN data processed with the updated software along with the application of averaging kernels and considerations of different a priori to compare with satellite observations.

## 5. Conclusions

Our study demonstrates the effectiveness of EM27/SUNs in monitoring greenhouse gases in the megacity Seoul where the amount of satellite data is sparse due to long revisit times or cloud contamination. While satellite measurements are generally good, improvements are necessary to accurately capture local greenhouse gas concentrations. This entails deploying more satellites with improved temporal and spatial resolutions to target local emissions, alongside advanced retrieval algorithms for evaluating local emission sources in urban areas. In the case of Seoul, geostationary satellites for measuring greenhouse gases will be the way forward for obtaining accurate information in monitoring urban areas in addition to utilizing satellites with high temporal and spatial resolutions for detecting hotspots and leaks. In addition to performing as low-cost alternatives of TCCON for ground validation to improve satellite accuracy, EM27/SUNs can contribute to greenhouse gas monitoring as they can be deployed in various emission source locations or difficult-to-reach areas and provide modeling input data for estimating emission fluxes. The measurement site at Seoul is an important site contributing to COCCON as a megacity site in a high emission source region. The site provides valuable reference measurements for validation of current and future satellite missions with greenhouse gas measuring capabilities over South Korea. A combination of multiple, long-term EM27/SUN measurements with high precision satellites accommodating high temporal and spatial coverage for urban areas will be of great synergy for greenhouse gas emission monitoring as well as reaching emission reduction targets.

## Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

## Data Availability Statement

The OCO-2 and OCO-3 data were produced by the OCO-2 and OCO-3 project at the Jet Propulsion Laboratory, California Institute of Technology, and obtained from the OCO-2 and OCO-3 data archive maintained at the NASA Goddard Earth Science Data and Information Services Center (GES DISC) (OCO-2/OCO-3 Science Team, Chatterjee & Payne, 2022; OCO-2/OCO-3 Science Team, Payne & Chatterjee, 2022). Copernicus

Sentinel-5P TROPOMI data processed by ESA was retrieved through GES DISC (Copernicus Sentinel-5P (processed by ESA), 2021a, 2021b). GOSAT V02.97 and V02.98 and GOSAT-2 Ver.02.00 data products have been provided by JAXA and NIES and are openly available via GOSAT Data Archive Service (<https://data2.gosat.nies.go.jp/>) and GOSAT-2 Product Archive (<https://prdct.gosat-2.nies.go.jp/>), respectively.

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