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Interactive comment

# *Interactive comment on* "Munich permanent urban greenhouse gas column observing network" *by* Florian Dietrich et al.

# Florian Dietrich et al.

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Received and published: 2 December 2020

# 1 Responses to the comments of reviewer 1

We would like to thank David Griffith for thoroughly reading our paper and providing very helpful and insightful comments. Below, please find our responses to his comments.

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### 2 Summary

**Reviewer:** This paper describes a measurement system and early results from a set of portable, automated ground-based solar infrared spectrometers based on the commercially available Bruker EM27-Sun. Each spectrometer is housed in an autonomous housing allowing weather-proof operation and full automation. Five such spectrometers are deployed around Munich, Germany to deduce city-scale emissions of CO<sub>2</sub>, CH<sub>4</sub> and CO from the upwind-downwind differences of total column amounts of these gases. The paper provides full technical details, building on an earlier 2016 publication. It represents a substantial instrumental development which would be of interest to anyone concerned with quantifying extended-source emissions such as cities or large point sources. It would require substantial effort and funding for others to duplicate the work, so I am pleased to see that the authors agree to make all technical plans, drawings and code available. It does leave me curious whether the authors have any plan to licence the system to a commercial provider - weather protection and automation for solar trackers and the EM27 used here and in TCCON and COCCON networks are not commercially available, but every installation needs one. The paper is clearly suitable for publication in AMT, after minor revisions and extensions and a few technical corrections.

**Response:** Thank you very much for appreciating our work and supporting its publication in AMT. Thank you also for suggesting the commercialization of our devices. We are currently looking into the possibilities to make our systems commercially available.

#### 3 General comments

**Reviewer:** Section 5.2 describes essential side by side comparisons (they are not "calibrations" per se) amongst the 5 instruments and TCCON FTS at Karlsruhe. These

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comparisons are of critical importance in evaluating small differences between upwind and downwind measurements, since any instrument bias would be interpreted as a gradient if not corrected. Yet no details of the comparisons are presented, and the reader has no idea of the uncertainty in the bias correction factors. It is essential to present the numerical details from all 6-monthly comparisons between instruments (and occasional TCCON comparisons). This could be done as a table of regression factors for each instrument pair and date. Only then can the reader assess the statistics of these comparisons – their magnitude, stability and reproducibility. The quantitative uncertainty will be essential for later modelling of the measured gradients in any Bayesian inversion scheme.

**Response:** Thank you for pointing out the missing quantitative comparison. We added two new tables (Table B1 and Table B2), which include the calibration factors for both the comparisons to the Karlsruhe instrument, which is calibrated to the TCCON standard, and the comparisons amongst our five instruments.

**Reviewer:** In section 5.3 and Figure 13 upwind-downwind data are compared, and at L302 the changes during Covid lockdown in 2020 are compared. However without the quantitative intercomparison data requested for section 5.2, it is impossible to assess the meaningfulness of these differences. How are the error bars on the  $CO_2$  enhancements calculated (refer to 5.2 discussion)? I also do not agree that the data show a correlated drop in  $CO_2$  enhancement with traffic congestion from weeks 4 to 12.  $CO_2$  enhancement drop 4-5 weeks earlier, meanwhile the season is changing from winter to spring and presumably  $CO_2$  sources other than traffic also change in this time, such as home, industrial and commercial heating. The interpretation is too simplistic. Thus I do not agree with the statement "These results prove that our network can detect changes in the urban emissions" – see also the conclusion around L 335. Based on the detail currently presented this conclusion is not valid. However this is not to say it is not possible. The data should be extremely valuable for such interpretations when

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combined with a city-scale regional model such as described as being under development. I agree that this level of modelling and interpretation is outside the scope of this paper.

**Response:** Thank you very much for the valuable insights. After adding the requested information in section 5.2 regarding the accuracy of our measurements, it becomes clearer that the enhancements in Figure 13 represent a real signal. Furthermore, we clarified how the error bars in Figure 13 are generated: "The error bars show the  $1\sigma$  standard deviation of all enhancements within the respective two-week period."

In addition, we refined our statement regarding the correlation of our measurements to the traffic data. The new formulation is: "The plot demonstrates that the lockdown had a significant impact on traffic flow. The CO<sub>2</sub> enhancements show a similar pattern throughout the first half of the year 2020. Based on the regression plot, there seems to be a correlation between the reduced traffic volume and the lower CO<sub>2</sub> enhancements ( $R^2$ =0.63). Both curves first decrease and then increase again after the strict restrictions were gradually loosened." We also added a regression plot (Figure 13, right) showing the correlation between the CO<sub>2</sub> enhancements and the traffic congestion data ( $R^2$  = 0.63), to provide evidence for our statement.

In the conclusion, we changed the sentence to: "The results show a *possible* correlation between the  $CO_2$  column concentration gradients and the traffic emissions, both of which *appear to be drastically affected* by the lockdown."

**Reviewer:** Finally, I count the term "world's first" 5 times in the manuscript – this is excessive. I suggest it is OK and sufficient to point this out once-only in the abstract, conclusion, and introduction.

**Response:** Thanks for pointing this out. We deleted three of the five occurrences of "world's first".

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# 4 Technical corrections

1. L15: Suggest rewording "as well as concentration gradients between sites upwind and downwind of the city."

Response: Thanks, we changed it according to your suggestion.

2. L59: TCCON measurements are made at a resolution of 0.02 cm<sup>-1</sup>

**Response:** Sorry for the confusion. You are right, the resolution of TCCON measurements is  $0.02 \text{ cm}^{-1}$ . We changed it in the paper accordingly.

3. L92: "Reed sensors in the inner cover COUNT these signals..."

**Response:** We changed "are counting" to "count".

4. L93: "Reed sensors INDICATE the absolute position." There are several more cases of present perfect tense where simple present is normal English usage – a copy editor should pick these up.

**Response:** Thanks for the hint. We changed "are indicating" to "indicate". Furthermore, we tried to change all occurrences of present progressive to simple present.

5. L119: As described in the current text, the system could not operate for several hours around noon in the southern hemisphere unless the whole instrument is rotated 180° to point north, with software able to handle this switch. Presumably this is the case - I suggest the text be clarified to make this clear. Further, why is the range (30-300°) not symmetric around North?

**Response:** Thanks for pointing this out. The range is not symmetric around North as the first mirror of the solar tracker is not centered at the rotation axis. Therefore, there is a slightly asymmetric behaviour of the morning and evening azimuth. We changed the text to: "That is why we designed our new cover so

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that it can measure solar elevation angles up to about  $80^{\circ}$  and azimuth angles between  $30^{\circ}$  and  $300^{\circ}$  for setups at the northern hemisphere. The asymmetric azimuth angle range is due to the non-centered first mirror of the solar tracker. If the system is used in the southern hemisphere, it must be rotated by  $180^{\circ}$  and a setting must be changed in the software. These solar angles cover most places in the world."

6. L138: Unclear wording ("cannot" should be "can not"), I suggest either that small change or "They can control temperature to a constant level... as well as condense (not condensate) water vapour..."

**Response:** We changed to sentence according to your suggestions to: "They can control temperature to a constant level of 25 °C under normal weather conditions in Uganda as well as condense water vapour to reduce the relative humidity inside the system."

7. L178: ...with two INDEPENDENT software COMPONENTS, OPUS and Camtracker, to control....

**Response:** We changed the formulation according to your suggestion.

8. L207: Very little detail of the spectrum retrieval with GFIT is given. This is OK if it follows the Wunch and Hedelius references exactly, but any variations from those procedures should be described because they will impact on accuracy and precision. In particular, when is the analysis done? – vertical pressure-temperature-humidity profiles only become available after a few days, but the text sort-of implies the fitting is done the same day in a pipeline process.

**Response:** Yes, we follow the GFIT retrieval as described in the references. We added a few additional information and modified the sentence about when we start the retrieval algorithm emphasizing that we have to wait for the vertical pressure profiles as you pointed out: "After about five days, when the a priori vertical pressure profiles from NCEP (National Centers for Environmental Prediction)

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are available, the retrieval algorithm converts the information from the interferograms into concentrations. The retrieval algorithm used is GGG2014 (Wunch et al., 2015), which is also used to retrieve all the TCCON data. We applied the standard TCCON parameters, including the air mass independent correction factors (AICFs). The spectral windows for retrieving diverse gas species are slightly modified according to the EGI setup (Hedelius et al., 2015)."

9. L219: "respectively" is not needed here, remove.

Response: We deleted "respectively".

10. L230: "pure emissions" is not quite the right wording, I suggest ".. this setup cannot be used to determine the emissions of the central city of Munich separate from its outer surrounds".

**Response:** Thanks, we changed the sentence according to your suggestion.

11. Fig 8 L2: The urban area itself is largely contained (provide a %?) within the green inner dotted circle in the centre.

**Response:** In Figure 8, we introduced the boundary of the urban area (black line). Furthermore, we changed the sentence to: "The urban area itself (indicated by the black border) is largely contained within the inner green dashed circle in the center, [...]"

12. L236: OCO-2 and OCO-3

Response: Thanks for pointing this out. We changed it to "OCO-2 and OCO-3"

13. L254: with the parameters a to d to be fitted.

**Response:** We changed the sentence to: "with the parameters a to d to be fitted".

14. L270: See general comments, this section should be expanded to include actual regression coefficient and statistics.

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**Response:** Thank you. See response to general comments. We added two new tables (Table B1 and B2) that includes scaling factors and regression coefficients.

15. L292: Can you provide the actual starting dates and numbers of measurements, rather than "a little bit later" and "a little bit less".

**Response:** We added a new table (Table 1) that includes the dates when the respective instruments started to measure in our network as well as the measured data points taken so far.

16. L302: See general comments.

**Response:** See response to general comments. We changed our interpretation according to your advice.

With best regards, Florian Dietrich on behalf of all co-authors

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Fig. 1. Map of the greater Munich area together with the two different sensor network setups that have been implemented.

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# *Interactive comment on* "Munich permanent urban greenhouse gas column observing network" *by* Florian Dietrich et al.

# Florian Dietrich et al.

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# 1 Responses to the comments of reviewer 2

We would like to thank reviewer 2 for reading our paper in detail and giving helpful comments. Below please find our answers:

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### 2 General comments

**Reviewer:** In this study, Dietrich et al. report on a novel permanent urban greenhouse gas monitoring network using EM27/SUN spectrometers inside an automated enclosure system in Munich. They carefully describe the technical innovations from a previous design as well as results from a successful testing campaign and longterm operations. It is clear that the presented systems are a significant improvement and hold the potential to facilitate such measurements in many cities and regions in the future. The paper is clearly structured and very well written and it fits perfectly into the scope of AMT. Although the technical aspects are overall excellent, there is unfortunately a major point of concern that should be addressed before publication. The authors have made very strong statements that the manuscript itself does not address. For example, the claim that the presented systems and approach allows to determine urban greenhouse gas emissions "in any city worldwide". More instances of such sweeping statements are given in the specific comments section. I recommend that the authors revisit these statement and provide additional data and explanations to support them. On the other hand the author could also choose to let the fully supported and very impressive results, e.g. increased data availability, continuous operations during COVID-lockdown, tracking of XCO<sub>2</sub> enhancement changes speak for themselves.

**Response:** Thank you very much for your helpful and constructive feedback and your appreciation of our technical achievements. Regarding your critics, we agree with you that some of our statements regarding the emission assessment are too strong and not always supported by data or references. Therefore, we modified these statements throughout the text. Please see our comments in the specific comments section.

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# 3 Specific comments

1. L1 – Consider removing "the"

Response: We deleted "the"

2. L8 – This study does not establish that this technique by itself allows to quantify emissions. For example, how well can annual emissions be estimated when observations have a clear-sky (and maybe seasonal) bias.

**Response:** Thank you for pointing this out. In our opinion, the properties of column measurements such as insensitivity to vertical redistribution of tracer masses and surface fluxes upwind the city, are a very important prerequisite to quantify emissions. In addition, we have the column measurements conducted upwind and downwind of the city, and the possible biases are canceled out by looking at the gradients. We slightly modified the formulation to "These column measurements and column concentration differences are relatively insensitive to vertical redistribution of tracer masses and surface fluxes upwind of the city, making them a suitable input for an inversion framework and, therefore, a well-suited candidate for the quantification of GHG emissions."

3. L21 – Although it is an impressive measurement system for total column CO<sub>2</sub> and CH<sub>4</sub>, it seems far from proven that this technique and system as a "new standard for determining GHG emissions", given the complexity and challenges in urban environments.

**Response:** You are right, this statement is probably a bit excessive. Therefore, we changed the sentence to: "In summary, our achievements in automating column measurements of GHGs will allow researchers all over the world to establish this approach for long-term greenhouse gas monitoring in urban areas."

4. L27 – Gurney et al. did not claim that urban areas contribute more than 70% of GHG emissions, but rather that "Cities account for more than 70% of global fossil

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fuel emissions". There are other non-urban and non-fossil fuel sources that contribute significantly to global GHG emissions, like land-use and land-use change (CO<sub>2</sub>), agriculture (CH<sub>4</sub>, N<sub>2</sub>O), etc. Please correct this statement or provide a reference for your claim.

**Response:** Thank you for pointing out this mistake. We changed it to "70% of global fossil fuel  $CO_2$  emissions [...]"

5. L59 – Do all TCCON stations use this very high spectral resolution in their operations?

**Response:** You are right, the resolution of TCCON measurements is lower (0.02  $\text{cm}^{-1}$ ). We changed it in the paper accordingly.

 L64 – How can you be sure that you will be able to assess the effectiveness of mitigation strategies? Could the atmospheric modelling framework not be insufficient to achieve this, if for example urban heat island effects are not correctly modelled. Furthermore, are the planned emission reductions in Munich large enough to significantly alter XCO<sub>2</sub>, XCH<sub>4</sub>, XN<sub>2</sub>O and other greenhouse gases.

**Response:** Thank you for this note. We modified the language of this sentence a bit: "The combination of our sensor network with a suitable modeling framework will build the basis for monitoring urban GHG emissions over years, identifying unknown emission sources, validating satellite-based GHG measurements as well as assessing the effectiveness of the current mitigation strategies." The details of the modeling framework will be part of a follow up paper. Up to our knowledge so far, the challenges you mentioned can be solved.

Furthermore, you are right: the absolute values of the column averaged GHG concentrations will not alter significantly based on the planned emission reductions. However, the emission information in our approach is included in the concentration gradients. To date, we can see a clear concentration gradient, with an estimated bottom-up  $CO_2$  emissions of about 5.9 t per Munich citizen and year.

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As the reduction goals aim to reduce these emissions by about 50% to 3 t per citizen and year until 2030, we are certain that our instruments will sense the changes in concentration gradients.

7. L147- Consider rephrasing for readability

**Response:** We rephrased the sentence: "For controlling and automating the enclosure system, we developed two independent software: ECon and Pyra. The purpose of Econ is to control all safety and enclosure features that are monitored by the PLC, whereas Pyra is used to control the spectrometer and to automatically perform the measurements. Pyra also includes a user interface (UI) where the operator can set all parameters and observe the current state of the system."

8. L177 - See L147

**Response:** Thank you. We reformulated the sentence: "Since the measurements are based on the spectral analysis of the sun, we have named the program Pyra, which is a combination of the programming language Python and the name of the Egyptian sun god Ra."

9. L219 - consider removing "respectively"

Response: We deleted "recpectively".

10. L230 - What is meant by "pure emissions"? Does this refer to net emissions of the city of Munich?

**Response:** We changed the sentence to "[...] this setup cannot be used to determine the emissions of the central city of Munich separately from its outer surroundings.".

11. L236 – "OCO-2" is repeated here

**Response:** Thanks for pointing this out. We changed the second occurrence of "OCO-2" to "OCO-3".

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12. L252 (eq1) - Why was such a simplified fitting approach taken here, when more suitable and well-established methods are widely used to determine seasonal variations and trends in atmospheric CO<sub>2</sub> records? For example, as described in Nakazawa et al. 1997 and references therein (https://doi.org/10.1002/(SICI)1099-095X(199705)8:3<197::AID-ENV248>3.0.CO;2-C).

**Response:** Thank you for suggesting us to use a more sophisticated fitting approach. We agree that our simple method cannot be used for a detailed and quantitative investigation of interannual variability in the  $CO_2$  trend. For such purposes, a method as described in Nakazawa et al. (1997) would be necessary. However, we use the fitted curve just as a qualitative comparison and visualization in the plot. The obtained fitting parameters are not used in any further analysis. Therefore, we think that the simple fitting approach is sufficient for this case.

13. L257 - The word "hotspot" seems not to be optimal to describe data density

**Response:** We changed it to "These high density data clusters represent our campaigns [...]"

14. L264 - It would be worthwhile to explain if this refers to 52% of all days since automation or all sunny/suitable days since automation, in any case a very impressive result.

**Response:** Thank you for this comment. The two ratios refer to all days not only to suited days. As a result, we measured on average at least one hour every second day since the automation started. We tried to make our statement clearer by adding the following sentence: "In this calculation all days are taken into account, regardless of whether the measuring conditions were good or bad."

15. L281 - Adding the pollution rose plot for CO<sub>2</sub> enhancement of the station inside

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the city could also be very interesting here to learn about the source distribution inside the city limits.

**Response:** Thank you for your suggestion. We added the concentration plot for the center station in the figure.

16. L293 – How much less data is available for southern station

**Response:** We added a new table (Table 1) that includes the dates when the respective instruments started to measure in our network as well as the measured data points taken so far.

17. L296 - This study does NOT show the drastic impact on GHG emissions, but mere a decrease in local GHG enhancements. There are many other possible reasons for changes in GHG concentrations other then emission changes. It is reasonable to assume here that the concentration enhancement change is due to emission changes, but this should be stated carefully and other potential sources of uncertainty have to be included when referring to emissions.

**Response:** Thanks for your suggestion. We attenuated our statements regarding our ability to determine emissions throughout the whole document. In this section, we changed the headline to "Influences of the COVID-19 lockdown *on urban concentration gradients*". Furthermore, we changed L296 to: "[...] showing the influence of such a drastic event on the *urban GHG gradients* of a city like Munich."

18. L302 - Please provide the R2 for this relationship. Also, looking at figure 14 it seems clear that CO<sub>2</sub> enhancements decreased strongly in week 6 and 8 already, well before the lockdown period, while congestion was above 25%, i.e. fairly normal. A scatter plot of the two quantities could be a useful addition in the supplemental information of this paper.

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**Response:** Thanks for this valuable suggestion. We modified our statement regarding the correlation of our measurements to the traffic data and added the R2 value. The new formulation is: "The plot demonstrates that the lockdown had a significant impact on traffic flow. The CO<sub>2</sub> enhancements show a similar pattern throughout the first half of the year 2020. Based on the regression plot, there seems to be a correlation between the reduced traffic volume and the lower CO<sub>2</sub> enhancements (R<sup>2</sup>=0.63). Both curves first decrease and then increase again after the strict restrictions were gradually loosened." As per your suggestion, a scatter plot was introduced to Figure 13.

19. L304 - See comment L296, L302, this study does not establish a decrease in emissions within Munich. Further modelling (including biospheric CO<sub>2</sub>) and assessment of uncertainties seems necessary before the authors should claim that they have proven that their system is sufficient to track emission changes. The authors later refer these uncertainties, so they seem aware of this problem, so why make such a strong claim here? Being able to reliably track XCO<sub>2</sub> enhancement changes during COVID lockdown with an automated system is already an excellent achievement in itself.

**Response:** Thank you. We have changed the statement (see response to comment of L302)

L327 - This statement completely ignores the potentially large impact on CO<sub>2</sub> concentrations by the urban biosphere, that has been found to be an important CO<sub>2</sub> sink (and sometimes source) in urban areas, for example, Miller et al. 2020 (PNAS, https://doi.org/10.1073/pnas.2005253117).

**Response:** Yes, our statement is too simplistic here. We changed the sentence to "For that, the concentration gradients between the downwind and upwind stations are decisive, as they represent the anthropogenic emissions superimposed with biological processes."

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21. L335: No data set of traffic emissions was presented in this paper. I agree that the seen decrease in congestion makes emission reductions extremely likely, but this should be stated carefully. Also the decrease does not seem to be concurrent.

**Response:** We modified our sentence to: "The results show a *possible* correlation between the  $CO_2$  column concentration gradients and the *traffic amount*, both of which appear to be drastically affected by the lockdown."

22. L342: It is unclear how this study has established that column measurements can be used in "any city worldwide". It seems apparent that the concentration gradients in the total column for smaller cities might be too small to detect reliably or the CO<sub>2</sub> emission signal might be masked due to biospheric uptake in cities in the tropics. What about cities with very strong aerosol loads, like Beijing, would the EM27SUN be able to penetrate dense smog?

**Response:** Thank you for your comment. As mentioned before, we do not claim anymore that we can measure emissions but concentration gradients. We changed it in the paper accordingly. Based on that, we think that our approach can be used in many cities worldwide. We changed "any city" to "over a wide range of latitudes". The statement we want to make is that we developed the sensor system that is necessary to establish a permanent ground-based remote sensing network using EM27/SUN independent on the location.

Furthermore, there exist FTS sites in large Chinese cities such as Beijing (Bi et al., 2018: https://doi.org/10.1007/s13351-018-7118-6) and Hefei (Wang et al., 2017: http://dx.doi.org/10.5194/amt-10-2627-2017).

With best regards,

Florian Dietrich on behalf of all co-authors

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Discussion paper

**Fig. 1.** Concentration enhancements over the background for each of the five stations displayed as a polar histogram.



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**Fig. 2.** Correlations between the CO\$\_2\$ enhancements over the background measured at our inner city station in Munich, and the traffic amount represented by the congestion rate (time series + regression plot)

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# Munich permanent urban greenhouse gas column observing network

Florian Dietrich<sup>1</sup>, Jia Chen<sup>1</sup>, Benno Voggenreiter<sup>1</sup>, Patrick Aigner<sup>1</sup>, Nico Nachtigall<sup>1</sup>, and Björn Reger<sup>1</sup> <sup>1</sup>Environmental Sensing and Modeling, Technical University of Munich (TUM), Munich, Germany **Correspondence:** Florian Dietrich (flo.dietrich@tum.de) and Jia Chen (jia.chen@tum.de)

#### Abstract.

5

In order to mitigate climate change, it is crucial to understand the urban greenhouse gas (GHG) emissions precisely, as more than two third thirds of the anthropogenic GHG emissions worldwide originate from cities. Nowadays, urban emission estimates are mainly based on bottom-up calculation approaches with high uncertainties. A reliable and long-term top-down measurement approach could reduce the uncertainty of these emission inventories significantly.

We present the world's first urban sensor network that is permanently measuring GHGs, based on the principle of differential column measurements (DCM) starting in summer 2019. These column measurements and column concentration differences are relatively insensitive to vertical redistribution of tracer masses and surface fluxes upwind of the city. Therefore, they are, making them a suitable input for an inversion framework and, therefore, a well-suited to quantify candidate for the quantification of GHG emissions.

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However, setting up such a stationary sensor network requires an automated measurement principle. We developed our own fully automated enclosure systems for measuring  $CO_2$ ,  $CH_4$  and CO column-averaged concentrations with a solar-tracking Fourier Transform spectrometer (EM27/SUN) in a fully automated and long-term manner. This includes also includes a software that starts and stops the measurements autonomously and can be used independently from the enclosure system.

- 15 Furthermore, we demonstrate the novel applications of such a sensor network by presenting the measurement results of our five sensor systems that are deployed in and around Munich. These results include the seasonal cycle of  $CO_2$  since 2015, as well as concentration gradient measurements gradients between sites upwind and downwind of the city. Thanks to the automation, we were also able to continue the taking measurements during the COVID-19 lockdown in spring 2020. By correlating the CO<sub>2</sub> column concentration gradients to the traffic amount, we demonstrate that our network is well capable to detect easily
- 20 capable of detecting variations in urban emissions.

The measurements from our unique sensor network will be combined with an inverse modeling framework that we are currently developing, in order to monitor urban GHG emissions over years, identify unknown emission sources and assess how effective the current mitigation strategies are. In summary, our achievements in automating column measurements of GHGs will allow researchers all over the world to establish this novel measurement approach as a new standard for determining GHG

emissions approach for long-term greenhouse gas monitoring in urban areas. 25

#### 1 Introduction

#### 1 Introduction

Climate change is one of the defining issue issues of our time, which and one that affects the entire planet. For an effective reduction of the To reduce greenhouse gas (GHG) emissions effectively, accurate and continuous monitoring systems for local and regional scale emissions are a prerequisite.

30

Especially for urban areas, which contribute to more than 70 % of GHG global fossil fuel CO<sub>2</sub> emissions (Gurney et al., 2015) and are therefore hotspots, there is a lack of accurate assessments of emissions shortage of accurate emissions assessments. The city emission inventories often underestimate the emissions as there exist emissions due to unknown emission sources that are not yet included in the inventories (Chen et al., 2020; Plant et al., 2019; McKain et al., 2015).

- 35 In the recent years, several city networks were established to improve the emission monitoring. These include networks using in-situ high precision instruments (McKain et al., 2015; Bréon et al., 2015; Xueref-Remy et al., 2018; Lamb et al., 2016) and low-cost sensor networks deploying NDIR sensors (Kim et al., 2018; Shusterman et al., 2016). In addition, eddy covariance flux tower measurements are used for directly inferring city fluxes (Feigenwinter et al., 2012; Helfter et al., 2011). However, all these approaches involve some challenges for measuring urban emission fluxes, such as high sensitivity to the boundary
- layer height dynamics, large variations due to mesoscale transport phenomena or the fact that they can only capture the fluxes 40 of a rather small area.

Column measurements have proven to be a powerful tool for assessing GHG emissions from cities and local sources, because they are relatively insensitive to the dynamics of the boundary layer height and to surface fluxes upwind of the city if a differential approach is used (Chen et al., 2016). Therefore, this method has recently been widely deployed for emission

- studies of cities and local sources using mass balance or other modeling techniques. In St. Petersburg, Makarova et al. (2020) 45 deployed 2 compact solar-tracking Fourier-transform infrared (FTIR) spectrometers (EM27/SUN) and a mass balance approach to study the emissions from the fourth largest European city. The EM27/SUN spectrometer has been developed by KIT in collaboration with Bruker and is has been commercially available since 2014 (Gisi et al., 2011, 2012; Hase et al., 2016). Hase et al. (2015) and Zhao et al. (2019) used the measurements of 5 EM27/SUNs to measure the Berlin city emissions of CO<sub>2</sub>
- 50 and  $CH_4$  in Berlin. With a similar sensor configuration, Vogel et al. (2019) studied the Paris metropolitan area and applied the CHIMERE-CAMS model to show that the measured concentration enhancements are mainly due to fossil fuel emissions. Jones et al. (2018) combined the Indianapolis city measurements measurements from Indianapolis (5 EM27/SUNs) with an adapted inverse modeling technique to determine the urban GHG emissions.
- Besides these urban studies, column measurements are also used to investigate local sources: Chen et al. (2016) and Viatte et al. (2017) determined the source strength of dairy farms in Chino, California. By combining column measurements with 55 a computational fluid dynamics (CFD) model, Toja-Silva et al. (2017) verified the emission inventory of the largest gas fired gas-fired power plant in Munich. With mobile setups, Butz et al. (2017) studied emissions from the volcano Mt. Etna, Luther et al. (2019) quantified the coal mine emissions from upper Silesia and Klappenbach et al. (2015) utilized column measurements on a research vessel for satellite validations above the ocean. However, these studies are all based on the campaign mode and not
- 60 suited for monitoring the urban emissions permanently. Only TCCON (Total Carbon Column Observing Network, Wunch et al.

(2011)) and COCCON (Collaborative Carbon Column Observing Network, Frey et al. (2019); Sha et al. (2019)) are measuring the global GHG column concentrations permanently. For this purpose, TCCON uses IFS 125HR spectrometers (resolution:  $0.00090.02 \text{ cm}^{-1}$ ), while COCCON uses calibrated EM27/SUN spectrometers (resolution:  $0.5 \text{ cm}^{-1}$ ). Both networks focus, however, However, both networks focus on detecting GHG background concentrations and are not mainly primarily designed

#### 65 to study urban emissions.

In this paper, we present the world's first permanent urban GHG network in Munich that is based on the differential column measurements (DCM) principle . The network is located in Munich and consists of 5 fully automated FTIR spectrometers. By combining the The combination of our sensor network with a modelling framework , we will be able to monitor suitable modeling framework will build the basis for monitoring urban GHG emissions over years, identify identifying unknown

70 emission sources, validate validating satellite-based GHG measurements as well as to assess how effective the assessing the effectiveness of the current mitigation strategies are.

#### 2 Measurement principle

As a measurement principle, the DCM method is used (Chen et al., 2016). DCM is an effective approach to determine for determining the emissions of large area sources using just a small number of stationary ground-based instruments. The basic

- 75 principle of DCM is illustrated in Fig. 1. The column-averaged concentrations of a gas in the atmosphere are measured upwind and downwind of an emission source, utilizing ground-based FTIR spectrometers that use the sun as light source. The concentration enhancements between the two stations are caused by the urban emissions. Chen et al. (2016) have shown that the differences between the upwind and downwind column concentrations are relatively insensitive to the boundary layer height and upstream influences. Therefore, DCM in combination with a wind-driven atmospheric transport model can be used to
- 80 determine emissions.

#### 3 Measurement system

In order to use the DCM principle for long-term monitoring of the urban GHG emissions, a fully automated measurement system is needed. Therefore For this, we developed an electronically controlled enclosure system including that includes the related software.

#### 85 3.1 Hardware

The enclosure system protects the spectrometer inside against harsh weather conditions and other harmful events, such as power or sensor failures. Furthermore, it enables a communication between the devices inside the system and allows the host to remote control the measurements over the internet. At suited measurement Under suitable measuring conditions, such as sunny weather and valid sun elevations, the system automatically starts the measurement process. During the day, the measurements are checked regularly by the enclosure software to detect and solve malfunctions autonomously. When the measurement conditions

90 checked regularly by the enclosure software to detect and solve malfunctions autonomously. When the measurement conditions



Figure 1. Basic principle of the differential column measurements: with the help of an upwind and at least one downwind station, the column-averaged GHG concentrations are measured. The differences between the two stations are representative for the emissions generated in the city.

are not suited anymore measuring conditions are no longer suitable, the system stops the taking measurements and closes the cover to secure the spectrometer. An operator is informed about any unexpected behavior via emailnotification by email.

#### 3.1.1 Standard edition

- The described enclosure is based on our first prototype system presented in Heinle and Chen (2018), which is has been con-95 tinuously running on the rooftop of the Technical University of Munich (TUM) in the inner city of Munich Munich's inner city since 2016. This system was developed to semi-automate the measurement process using an EM27/SUN spectrometer throughout over the years. For the permanent urban GHG network, we improved this system to make it more reliable, easier to transport and fully autonomous.
- Our new enclosure system is based on a lightweight yet robust aluminum housing (Zarges K470 box, waterproof according to IP54) that we modified for our purposes. The CAD model of this system is shown in Fig. 2. A rotating cover at the top of the housing allows the sunrays to hit the mirrors of the solar tracker at arbitrary azimuth and elevation angles. Every 10 degrees, a magnet is fixed in the outer cover (see Fig. 3). Reed sensors in the inner cover are counting count these signals so that the relative position of the cover can be computed. Before opening the cover the cover is opened and after every full rotation, two additional Reed sensors are indicating indicate the absolute zero position. The target position of the cover is computed automatically depending on the coordinates of the site and the time. Optical rain and direct solar radiation sensors indicate
  - whether the current environmental conditions are suited for measurements.

Signal lamps, push buttons and an emergency stop button can be used to control the basic functions of the enclosure directly at the site. Full control can only be achieved by remote access to the enclosure computer, which is an industrial embedded box

PC. In addition to the remote access, the computer is also responsible for controlling the spectrometer and the solar tracker and 110 to store for storing the interferograms before they are transferred to our retrieval cloud via the internet.

The enclosure system itself is controlled by a Siemens S7-1200 PLC (programmable logic controller) and not by the enclosure computer that runs with on a Microsoft Windows operating system. This approach ensures that the safety features, such as rain or power failure detection, cover motor control, temperature control, etc. are separated from the Windows operating system, making the enclosure less error-prone and more fail-safe.

- All the additional electronics are placed in the rear part of the enclosure systems and are shown in Fig. 4 more in detail. Besides the PLC, we installed an LTE router, a heater, the motor driver, two circuit breakers, surge protection devices and an RCCB (residual current circuit breaker). In addition, new relays were added to the system to be able to reset all error-prone devices such as the computer, the router or the router or PLC remotely. In order to make the system as lightweight as possible, we replaced the large and heavy thermo-electrical cooler (TEC) by a cooling fan and a heater, and replaced the lead-acid
- 120 battery of the UPS (uninterruptible power supply) by a capacitor-based energy storage. All the devices inside the system are communicating over communicate via the two standard protocols TCP/IP and USB.

A photo of one of the four newly developed enclosure systems for the Munich network can be seen in Fig. 5. It shows the measurement setup at our southern site on top of a flat rooftop.

#### 3.1.2 Universal editions

- 125 Our enclosure system was originally developed to measure the GHG concentrations in Munich at a latitude of 48.15° N. Therefore, the rotating cover that protects the solar tracker from bad weather was designed to enable measurements for all possible solar angles at such a latitude. If However, if the enclosure system is , however, used somewhere else in the world, these limitations need to be considered. That is why we designed our new cover so that it can measure solar elevation angles up to about 80° and azimuth angles between 30° and 300, which covers most of the places worldwide° for setups at the northern
- 130 hemisphere. The asymmetric azimuth angle range is due to the non-centered first mirror of the solar tracker. If the system is used in the southern hemisphere, it must be rotated by 180° and a setting must be changed in the software. These solar angles cover most places in the world. Furthermore, we adapted some features to overcome challenges such as extreme temperatures as well as and high relative humidity. We developed two of these special editions and tested them both at very low and high latitudes; one in Uganda next to the equator and one in Finland next to the polar circle.
- As part of the NERC MOYA project, the University of Leicester is has been using our enclosure system to measure  $CH_4$  emissions from the wetlands north of Jinja, Uganda (latitude:  $0.4^{\circ}$  N) since the beginning of 2020 (Boesch et al., 2018; Humpage et al., 2019). Besides significant Quite apart from the significantly higher temperatures and relative humidity in comparison to than in Munich, the very high solar elevation angles (up to 90°) are challenging. These high angles are a problem for both the cover of the enclosure as it is blocking blocks the sun in such cases and for the solar tracker of the spectrometer. The
- 140 solar tracker of the EM27/SUN can only measure up to elevation angles of about 85°. For At higher elevations, the control algorithm is not stableanymoreno longer stable. Therefore, both the spectrometer and the cover cannot work properly at such high elevation angles.



Figure 2. Side view of the enclosure system (CAD model)

To overcome this challenge, we tilted the whole enclosure system by a few degrees to simulate that the instrument is the instrument being located at a site with a higher latitude than it actually is. This is done using two state-of-the-art car jacks

(see Fig. 6), which can elevate the side of the enclosure that points towards the equator up to  $15^{\circ}$ . In this way As a result, the 145 very low elevation angles cannot be measured anymore can no longer be measured, as the sun is then blocked by the lid of the enclosure, which is however although this is not an issue. This is because the air mass dependency of the slant column cannot be reliably handled by the GFIT retrieval algorithm at these high solar zenith angles (Wunch et al. 2011). Using this unique approach, both the solar tracker and the rotating cover work properly at high elevation angles, which makes this approach 150 suited for locations at low latitudes.

Since the temperature and relative humidity are much higher than in central Europe, the normally used fan and heater normally used are replaced by two 150 W theromoelectrical coolers. They cannot only control the temperature to can keep the temperature at a constant level of 25 °C under normal weather conditions in Ugandabut also condensate water vapour, as well as condense water vapor to reduce the relative humidity inside the system.

- 155 Our enclosure is, however, not only suited to work not only at very low latitudes but also at high ones. To test the system under such conditions, we built another enclosure system for the COCCON site next to the TCCON station in Sodankylä at a latitude of 67.4° N (Tu et al. 2020). Therethe system is continuously measuring, the system has been measuring continuously since 2018, which shows that our system cannot can not only withstand cold winters but is also suited suitable to measure a large azimuth angle range.
- 160 Overall, we developed a system that is universally applicable and can be used for a wide latitude range to enable groundbased GHG measurements worldwide with minimum effort and maximum measurement data.



**Figure 3.** CAD model of the newly designed cover (outer + inner one) with a small opening and a steeper slope compared to the first version in Heinle and Chen (2018). With the help of the reed sensors 1+2, the relative position of the cover is calculated (in  $10^{\circ}$  steps). The second sensor is indicating indicates the direction. The reed sensors 3+4 are used to determine the absolute zero position each time before the cover is opening opens.

#### 3.2 Software

For controlling and automating the enclosure system, we developed two independent software were developed: one programs: ECon and Pyra. The purpose of Econ is to control all safety and enclosure features that are monitored by the PLC(ECon) and the other one is, whereas Pyra is used to control the spectrometer and automatically perform the measurements (Pyra). Latter one take measurements automatically. Pyra also includes a user interface (UI) where at which the operator can set all parameters and observe the current state of the system.

#### 3.2.1 Enclosure control (ECon)

The enclosure control software ECon was already a part of the first enclosure version (Heinle and Chen, 2018). There, a microcontroller program is used to control the enclosure features such as opening and closing the rotating cover, analyzing the rain sensor data, powering the spectrometer and monitoring the UPS. For the new version, we separated these safety operations from the measurement-related software that is running on a Windows computer to make these features fail-safe. As the microcontroller is in the new version replaced by a PLC in the new version, the ECon software needed to be renewed as well.





ECon is structured as a sequence control that loops through the main program, which is grouped into several functions, over and over again. These functions include<del>for example, for example,</del> the detection of any alarm caused by the UPS, encoder or power failures, the request of the current solar azimuth angle and the control of the cover motor and other outputs such as relays or signal lamps.

The most safety-relevant function is thereby the control of the cover motor. The program is structured in such a way that 180 closing the cover is prioritized in any condition. Even in <u>case the event</u> of a Reed sensor failure, the program will make sure that the cover closes correctly by evaluating the sensor signals, which are implemented redundantly.

Furthermore, ECon monitors whether the ethernet connections to the computer, spectrometer and internet are working properly. In case that work properly. If any malfunction is detected, the program automatically restarts either the spectrometer, computer or the routerrouter, depending on the kind of failureby shortly, by briefly interrupting the power supply of to the respective device using relays. This approach ensures a minimum requirement for human interactions in case of malfunctions

if malfunctions occur, which is particularly beneficial for operating very remote sites.

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**Figure 5.** Image of the new enclosure system on the roof of a school at our southern site Taufkirchen. The systems includes, inter alia, the newly designed rotating cover, the lightweight aluminum case, the solar radiation sensor and a surveillance camera attached to a post.



**Figure 6.** Setup of the tropical version of our enclosure in Jinja, Uganda (Latitude:  $0.4^{\circ}$  N). With the help of car jacks the whole system is tilted in order to enable measurements at high elevation levels close to 90°. Furthermore, the system is equipped with two 150 W thermoelectrical coolers (attached at the two sides of the system) that keep the temperature inside the enclosure constant at 25 °C. Photo by Neil Humpage, University of Leicester.

To keep the temperature within a predefined range, ECon also controls the temperature inside the enclosure by either powering the heater or the fan, depending on the actual and the given nominal temperature.

#### **3.2.2** Automation software (Pyra)

- 190 In order to control the measurements of the spectrometers automatically, it was necessary to develop a software that covers all the tasks that a human operator normally does to perform the measurements. We decided to use Python as a programming language to develop both the automation software and a user interface that allows an operator to set all necessary parameters and observe the current state of the system. The program is running runs all the time on each enclosure computer and serves as a juncture between the spectrometer, the enclosure system and the operator. As operator. Since the measurements are based on the spectral analysis of the sun, the program is called we have named the program Pyra, which is a combination of the
- 195

programming language Python and the name of the Egyptian sun god Ra.

The manufacturer Bruker provides the EM27/SUN spectrometers with the two software independent software components OPUS and CamTracker, to control both the spectrometer itself and the camera-based solar tracker that is attached to the spectrometer. Pyra does not replace these two software elements but provides the possibility to start, stop and control them au-

200 tomatically. Besides these necessary tasks, Pyra also monitors the operating system and the spectrometer to detect malfunctions such as insufficient disk space or non-working connections. Furthermore, it evaluates whether the environmental conditions are suited for measurements and logs each event to a file.

There are Pyra has four different operating modes of Pyra. The: the manual one, where in which the operator can start and stop the measurements with just one click, two semi-automated modes, where Pyra is starting and stopping in which Pyra starts

205 and stops the measurements based on either a defined time or the solar zenith angle (SZA) range and the fully-automated mode. In the latter, Pyra is evaluating evaluates the direct solar radiation sensor data and combines it with the online calculated SZA information SZA information calculated online to start and stop the measurements whenever the environmental conditions are suitedsuitable.

A more detailed description about the features of the Pyra can be found in Appendix A.

210 Although - Pyra was developed to automate the process of EM27/SUN spectrometers that are operated in our enclosure system, it can also be used without this system or in a different shelter. In this case, only the fully-automated mode is not working anymore fully automated mode no longer works, as the information of from the direct solar radiation sensor is not available. However, all the other modes are workingwork, which leads to less human effort and more reliable measurements.

All the aforementioned features of Pyra are combined in a common user interface (see Fig. 7). It is a clear and handy interface 215 that allows any operator to do-make all the necessary settings for performing automated measurements using EM27/SUN spectrometers. There are in total total, there are three Pyra tabs (Measurement, Configuration and Log) and one ECon tab, which we also included in this user interface. The ECon tab allows us to control the PLC that operates the enclosure system (details for details, see section 3.2.1). Thereby, the program itself is not running does not run on the enclosure computer but on the PLC, which makes the safety-related features fail-safe. As the PLC does not provide an-its own graphical user interface, we

decided to include these functions, such as controlling the cover motor, heater, fan, relays, etc. in the Pyra UI as well. For that, 220

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leasurement C	onfiguration Log ECor		
Main Info Measureme	nt VBDSD automation	Start Automation	Stop Automation
Status: Mode: Start Time (UT	Active Monitoring -0 13:02	Start Measurement	Stop Measurement
Live Monitor		Static Monitor	
Overview Error present: Programs	No	System Info Storage Path: Available Storage Space	N\MUNICH\ <y><m><d>\</d></m></y>
BktTimeSync: Camtracker: OPUS:	Not active Is active Is active	Measurement Files	MUC_86.mtx
Network EM27:	Connection enable	Experiment:	dual_munichcampaign.xpm
Sun Status: Elevation Mot	good or Offset: 4.271238°	Set Measurement Time 11/07/2020 12:00:00 AM	M Cancel

Figure 7. User interface of for the control software Pyra. In total there are four different tabs (*Measurement, Configuration, Log and Enclosure control*) that can be selected. In this image, the *Measurement* tab is shown.

the Python library *snap7* is used, which allows makes it possible to communicate with a Siemens S7 PLC using an ethernet connection.

#### 3.2.3 Automated retrieval process

For a fully-automated fully automated greenhouse gas observation network, not only the measurements need to be autonomous but also the data processing. Therefore, we automated the data processing chain as well.

Each enclosure computer automatically uploads at At the end of a measurement day, each enclosure computer automatically uploads all the interferograms and the weather data over weather data via an SSH connection to our Linux cloud server at the Leibniz Supercomputing Center in Garching. After the data of a full day from a station is completely transferredabout five days, when the a priori vertical pressure profiles from NCEP (National Centers for Environmental Prediction) are available, the
retrieval algorithm that is based on the software GFIT (Wunch et al., 2011; Hedelius et al., 2016) is retrieving the concentration data converts the information from the interferograms and stores all values in an ASCII file. This file contains the concentrations as well as the wind information throughout the dayinto concentrations. The retrieval algorithm used is GGG2014 (Wunch et al., 2015), which is also used to retrieve all the TCCON data. We applied the standard TCCON parameters, including the air mass independent correction factors (AICFs). The spectral windows for retrieving diverse gas species are slightly modified according to the EGI setup ?.

12

#### 4 Network setup

We tested the automated network consisting of five spectrometers in a measurement campaign in August 2018 (Dietrich et al., 2019), before the permanent network was installed in September 2019. In addition, our first enclosure system is has been permanently measuring on the university rooftop since 2016 permanently. 2016.

#### 240 4.1 Test campaign - Munich August 2018

After building in total a total of five enclosure systems, we established the world's first fully-automated first fully automated GHG sensor network based on the differential column measurements (DCM) principle (Chen et al., 2016) in a one-month measurement campaign in Munich.

- For testing To test our enclosure systems and the network configuration, we borrowed spectrometers from the Karlsruhe Institute of Technology (KIT) and the German Aerospace Center (DLR). Besides In addition to our long-term operating station in the inner city, we set up a system in each compass direction , respectively (see red shaded enclosure systems in Fig. 8). The distance A distance of approximately 20 km was selected between the downtown station and the outer stations was chosen as approximately 20 each each outer station, to ensure that the outer stations are not directly affected by the city emissions if they are located upwind.
- Thanks to the automation, we were able to measure take measurements on each of the 25 sunny August days mostly from the very early morning to the days in August, both weekdays and weekends, mostly from very early in the morning to late evening (approx. 07:00 to 20:00 local time)both on weekdays and weekend days. Thereby, the human interactions were reduced 7 am to 8 pm). This kept human interactions to a minimumand were mostly restricted , and restricted them mostly to setting up and disassembling the enclosure systems on the rooftops that we used as measurement sites. Therefore, this campaign was characterized by a very small effort as well as a very high data amountvolume. These results are the desired outcomes of
- such campaigns and are the foundation to use such a setup also for also using this kind of setup for a permanent urban GHG observation network.

#### 4.2 Permanent Munich GHG network setup

Although the configuration of the outer stations in the August 2018 campaign was well suited to capture for capturing the background concentrations, this setup cannot be used to determine the <u>pure</u> emissions of the <u>central</u> city of Munich <u>separately</u> from its outer surroundings. Instead, the greater Munich area emissions are captured as well. As our focus <u>lies in the city</u> emissions is emissions from the city itself, we decided to go closer to the city boundaries for our permanent sensor network. The distance between the downtown station and each outer station was halved to 10 km (see green enclosure systems in Fig. 8). Thus, the outer stations are located approximately at the city boundaries of Munich. The second benefit of such a dense sensor

setup is that it can be better used for validating concentration gradients measured by satellites. Due to the unique dataset of our sensor network, the NASA satellites OCO-2 and  $\frac{\text{OCO-2 are OCO-3 have been}}{\text{OCO-2 are OCO-3 have been}}$  measuring CO<sub>2</sub> concentrations over Munich in the target mode since spring 2020. The area OCO-2 can cover over Munich in this mode over Munich is approximately 21 km



**Figure 8.** Map of the greater Munich area together with the two different sensor network setups that have been implemented. The urban area itself (indicated by the black border) is largely contained within the highlighted shape inner green dashed circle in the center, which represents the current setup of the stationary network. The light red shaded sensor systems, together with the center station, represent the setup during our 2018 summer campaign2018; the four systems on the green circle together with the center station represent the current setup of the stationary network. Both setups are characterized by having a center station and a station in each compass direction to measure the inflow and outflow of GHG concentrations at under arbitrary wind conditions. Map data are from © Google Maps

 $\times$  13 km. As the satellite trajectory is not exactly aligned <u>in on</u> the north-south axis, the distance of 10 km between the inner and outer stations is optimal <u>in order to capture</u> for capturing the urban concentration gradients.

270 In addition to the relocation, the enclosure systems were slightly improved based on the experiences from the August 2018 campaign. This includes especially adding In particular, this includes the addition of a direct solar radiation sensor in order to start and stop the measurements depending on the actual weather conditions. Furthermore, we replaced the three borrowed spectrometers by with our own ones so that all five instruments can measure long-term.

All in all, we were able to set up the first permanent urban column concentration network for GHGs in September 2019 275 using our own five spectrometers. Starting from Since this date, we are not only measuring have been measuring not only the absolute GHG concentration trend of Munich but also the city gradients, which will be used to determine the urban GHG emissions of in Munich over the course of the years and years, as well as to find unknown emission sources.

#### 5 Results

Since 2015, we are have been continuously measuring the GHG concentrations in Munich with at least one instrument. Over time, the data amount is increasing significantly as we amount of data has increased as we have improved our automation and used more and more instruments.

#### 5.1 Seasonal Cyclecycle

In Fig. 9, we show the measurement curve of our downtown station over the first five years of measurements. In order to display the seasonal cycle, a sinusoidal function of the form

285 
$$c_{CO_2}(t) = a \cdot (sin(\frac{2\pi(t-b)}{365})) + ct + d$$
 (1)

with the parameters a to d is to be fitted. One can see clearly clearly see the globally rising trend in  $CO_2$  (about 2.4 ppm per year), as well as the seasonal cycle for the five years over the five-year peroid.

Although the whole time period between entire period from fall 2015 and to summer 2020 is covered, there are times with a much larger amount of data compared to the rest. These data hot spots are representing some times within this range yield a much greater volume of data than others. These high-density data clusters represent our campaigns in summer 2017 and 2018, as well as our Oktoberfest campaign in 2018. A further hot spot can be detected in fall 2016where, when the first version of our enclosure system (Heinle and Chen, 2018) has been was established and intensively tested in the semi-automated mode. Since summer 2019the fully-automated enclosure system is, the fully automated enclosure system has been measuring whenever the weather conditions are suited suitable, which results in a very high and dense data amountvolume.

In total, we have measured on 498 days throughout the last five years. Thereby Of these, only days with continuous measurements of at least 1 hour are taken into account. The ratio of measurement days compared to non-measurement days is about 17 % for the time period before summer 2019. After establishing the full automation Once full automation was established, this ratio increased to about 52 %, which shows the great benefit of our fully-automated sensor network approach. In this calculation all days are taken into account, regardless of whether the measuring conditions were good or bad.

#### 300 5.2 Side-by-side calibration and urban gradient comparison

The results in the previous section show that our automation is working works and that we are able to gather a lot of GHG measurement data. The final goal of our network is, however, the quantification of the to quantify the urban emissions. For that, the gradients between the single stations need to be analyzed. As the concentration enhancements of column averaged dry air mole fractions are quite small for an urban emission source, it is absolutely necessary to calibrate the instruments

305 regularly. Besides calibrating the absolute concentrations values next to a TCCON station from time to timeIn addition to the calibration of absolute concentration values during measurements next to the TCCON station in Karlsruhe, the relative comparison between the single instrument is even more decisive. Therefore, we calibrate all instruments regularly twice a year and additionally at the beginning and end of each campaign. The setup of such a with respect to our defined standard represented by the instrument ma61. Fig. 10 shows the setup of this kind of side-by-side calibration measurement dayis shown

310 in Fig. 10 measurement day, where five automated sensor systems are measuring measure next to each other on top of our university roof.

For each instrument and gas species a constant calibration factor is determined, a constant correction factor f (see Table B1) is determined to convert the raw concentration value  $c_{raw}$  to the corrected concentration value  $c_{corr}$  using linear regression with  $R^2 > 0.9$ . As :

$$\begin{array}{ll} 315 \quad c_{\rm corr} = \frac{c_{\rm raw}}{f} \\ & \\ & \\ \end{array} \tag{2}$$

As an absolute reference value<del>we take the concentration value of the</del>, we will use the instrument that was <del>last</del> calibrated at a TCCON station - most recently. Currently, the correction values determined by Frey et al. (2019) before shipping the instruments from Bruker to Munich are used (see Table B2).

Fig. 11 shows the  $CH_4$  gradients of a standard measurement day on a Saturday during Oktoberfest 2019. It indicates that our sensor network can detect the differences in  $CH_4$  concentrations well, which allows us to determine the urban emissions using these measurements as an input. Furthermore, one can see that our automated network allows us to measure not only on weekdays but also on weekends from early morning to evening, without the need for human resources.

In Fig. 12, we show the  $CO_2$  concentration enhancements above the background concentration for the four outer city stations depending on the wind direction. For this purpose, we use an ultrasonic wind sensor (Gill WindObserver II) on a roof in the

- 325 inner city of Munich (48.148° N, 11.573° E, 24 m agl.). To determine the background concentration, we use the data of all from all of our measurement stations and determine the lowest measurement point for each time step. Afterwards, a moving average with a window size of 4 h is used to smooth the curve as we assume that the background concentration must not change rapidly. For each station, a polar histogram shows where and how often the concentration enhancements come from originate and how frequent they are. In contrast to a standard wind rose, the different colors indicate the strength of the concentration
- 330 enhancement; yellow means low and red high enhancement. The wind speed is displayed by the distance of the respective cell to the center point of each circle.

One can see clearly that for all 4 stations four stations, the enhancements are higher towards the city. For the eastern station for example, for example, the highest enhancements, indicated by the reddish color, are located in the west. These results indicate that the gas molecules are mainly generated in the city and that our network is able to detect and quantify such urban emitters.

335 Due to technical issues, the southern station started its measurements a bit later than the other four stations not all stations started their measurements at the same time. Therefore, the data amount is a little bit less. This is, however, not decisive as north wind in Munich is quite rarevolume collected at the southern station, which started no earlier than May 2020, is much smaller. An overview of when each station started its measurements in the permanent network, including the data collected so far, is shown in Table 1.

Instrument	Location	Start date	Datapoints
<u>ma61</u>	Center	September 2015	1,550k
<u>mb86</u>	East	August 2018	<u>850k</u>
mc15	West	September 2019	<u>310k</u>
<u>md16</u>	North	December 2019	<u>270k</u>
me17	South	<u>May 2020</u>	<u>16k</u>

Table 1. Start of operation including the number of measurements taken by each station so far (until August 12, 2020)

#### 340 5.3 Influences of the COVID-19 lockdown on urban concentration gradients

Thanks to the automation, we measured-took measurements throughout the COVID-19 lockdown in spring 2020, which results resulted in a unique dataset showing the influences influence of such a drastic event to the GHG emissions on the urban GHG gradients of a city like Munich. Fig. 13 displays the gradients between the measurements of the inner city station and the background concentrations (cf. section 5.2).

All concentration enhancements gradients are clustered into biweekly bins. In Fig. 13, the median of these bins is displayed as 345 the blue curve. The error bars indicate the  $1\sigma$  standard deviation of these biweekly distributions. In addition, the Munich traffic amount-volume of traffic in Munich is displayed in red using the congestion rate that is provided by TomTom International BV. Furthermore, the COVID-19 lockdown period is shown as the grey shaded area.

The plot demonstrates that the lockdown had a large impact on the traffic amount and that there is a good correlation with 350 the significant impact on traffic flow. The  $CO_2$  enhancements show a similar pattern throughout the first half of the year 2020. Based on the regression plot, there seems to be a correlation between the reduced traffic volume and the lower  $CO_2$ concentration enhancements enhancements ( $R^2$ =0.63). Both curves decrease drastically at the beginning of the lockdown and increases first decrease and then increase again after the strict restrictions were loosened bit by bit. These results prove that our network can detect changes in the urban emissions well. gradually loosened.

355 Nevertheless However, our statistical approach, which uses about 100k measurement points, shows large variations in the  $CO_2$  enhancements for the single bins. Such high variations are, however, not concerning as the approach is not considering does not take into account wind speed and direction, for example. Furthermore, the assumption of homogeneously distributed emissions sources does not reflect the truth and photosynthetic effects are not considered. Therefore, it can only serve as a first indication of how the emissions were reduced during the lockdown period. For the future, we will apply more sophisticated modelling modeling approaches to quantify the emissions. 360

#### Conclusion 6

We present the world's first permanent urban GHG column network consisting of 5-five compact solar-tracking spectrometer systems distributed in and around Munich. We developed the hardware and software to establish such this kind of a fully



Figure 9. Daily mean values of the  $CO_2$  measurements from the downtown station in Munich. The concentrations are following follow the globally rising trend. Furthermore, the seasonal cycle with lower concentrations in summer and higher concentrations in winter is clearly visible for the five-year period shownfive years.



Figure 10. Calibration measurements of all our five sensor systems on top-the roof of our institute's building. One can see four slightly different versions of our enclosure systems.

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automated GHG sensor network for quantifying large area emission sources, such as cities. Both the enclosure system and the related Python program for automating the measurement process can be used by the community to build up similar sensor networks in cities worldwide. Also, COCCON would significantly benefit from such an benefit greatly from this kind of automated system, as the current approach of operating EM27/SUN spectrometers in this network still requires manpower



**Figure 11.**  $CH_4$  measurement values (5-minute average) of from all five stations during our Oktoberfest 2019 campaign on September 21, 2019. The concentration gradients between the single stations are clearly visible, which indicates the presence of strong  $CH_4$  sources in the city.



on site for starting up measurements and for protecting to protect the spectrometer from adverse meteorological conditions. Permanent and long-term observations will help to improve the understanding of the global carbon cycle.



**Figure 13.** Correlations between the CO<sub>2</sub> enhancements over the background measured by at our inner city station in Munich, and the traffic amount represented by the congestion rate (left: time series, right: regression plot). The time period includes the COVID-19 lockdown in spring 2020. We show the median trend of all column concentration gradients clustered into biweekly bins. The error bars indicate show the  $1\sigma$  standard deviation of these all enhancements within the respective two-week period. Traffic data are from ©2020 TomTom International BV.

With our sensor systems, we did-carried out several test campaigns between 2016 and 2019 and finally set up the world's first-permanent urban GHG sensor network based on the differential column methodology in fall 2019. The results show the advantages of such an automated network based on the differential column methodology in fall 2019. The results show the advantages of such an automated network based on the differential column methodology in fall 2019. The results show the advantages of such an automated network based on the differential column methodology in fall 2019. The results show the advantages of such an automated network based on the differential column methodology in fall 2019. The results show the advantages of such an automated network based on the differential column methodology in fall 2019. The results show the advantages of such an automated network based on the differential column methodology in fall 2019. The results show the advantages of such an automated network based on the differential column methodology in fall 2019. The results show the advantages of such an automated network based on the differential column methodology in fall 2019. The results show the advantages of such an automated network based on the differential column methodology in fall 2019. The results show the day of the week or the season, we show in this study this study shows that our network can effectively detect both the globally rising trend of CO<sub>2</sub> concentrations and the seasonal cyclevery well.

The final goal of such a this kind of network is the quantification of urban GHG emissions. For that, the concentration gradients between the downwind and upwind stations are decisiveas they are representing the amount of pollutants that are generated by the emission sources in between, as they represent the anthropogenic emissions superimposed with biological processes. Our results demonstrate indicate that these gradients can be captured clearly with our sensor setup. Additional analyses, including wind informationdemonstrate, shows that the city is causing these emissions causes these enhancements.

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Furthermore, the network can be used to validate GHG satellites in a unique way, as not only absolute values but also concentration gradients can be compared. Since spring 2020, the NASA OCO-2 and OCO-3 satellites (Crisp, 2016; Eldering et al., 2018) have been measuring urban  $CO_2$  concentration gradients of in Munich using the spatially highly resolved target mode in a recurring pattern to compare the satellite measurements with our ground-based ones.

385 Thanks to the With the benefit of full automation, we were also able to measure the concentration gradients also concentration gradients during the COVID-19 lockdown period in spring 2020. The results show a elear possible correlation between the CO<sub>2</sub> column concentration gradients and the traffic emissions, which were both drastically influences amount, both of which appear to be drastically affected by the lockdown.

In order to quantify the Munich GHG emissions, we are currently developing an atmospheric transport model based on 390 Bayesian inversion. Such a This kind of modeling framework will help us in the future to quantify the GHG emissions of Munich-quantify Munich's GHG emissions in the future, and find correlations between parameters such as time of the day, season, weather conditions, etc. Furthermore, we will use our rich dataset to detect and quantify unknown GHG emission sources.

In sumsummary, this study provides the framework for establishing a permanent GHG sensor network to determine urban emissions concentration gradients using column measurements in any city worldwideover a wide range of latitudes. The characteristics of the presented approachhardware presented here, such as high precisionreliability, ease of use and low operating costs, form the basis for it to become a new standard for monitoring urban GHG emissions concentrations. *Code and data availability.* The Python software Pyra as well as the technical drawings, schematics and component list for the enclosure system can be provided by the authors upon request.

#### 400 Appendix A: Pyra - software features

To control the spectrometer program OPUS, we use the Microsoft Windows technology *dynamic data exchange* (DDE), which is also supported by OPUS. It is a protocol to exchange data based on the client-server model and allows us to send requests, such as starting a measurement or loading a specific setting file, to OPUS. With the help of DDE, combined with an MTX macro file for OPUS, Pyra can start recurring measurements of the spectrometer. The necessary settings are stored in an XPM experiment file and are loaded into the program in the same way.

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The communication <u>Communication</u> with the solar tracker program CamTracker is <u>done in a simplerway as the settings of</u> this programdo not simpler, as this program's <u>settings no longer</u> need to be changed after the initialization<del>anymore</del>. Therefore, we asked the manufacturer Bruker to implement an <u>auto start autostart</u> option for the tracker. Whenever CamTracker is called with this option, the solar tracker automatically aligns its two mirrors to with the calculated live position of the sun and enables

410 the tracking of the sun. After terminating the program Once the program has been terminated, the tracker automatically moves back to its parking position.

In order to detect malfunctions, Pyra is equipped with several live monitoring functions. It monitors every 0.2 s whether the two programs OPUS and CamTracker are still running correctly. If this is notthe case anymore they are not, it automatically restarts the non-working program to proceed with the measurements. Furthermore, the log files of CamTracker are read con-

415 tinuously, which allows us for example to automatically detect to detect automatically if the solar tracker is not tracking does not track the sun correctly anymore, for example. Such a behaviour behavior is quite common as the solar tracker is using uses a camera-based approach to follow the sun in over the course of the day. In ease of cloudy conditions, the algorithm sometimes mistakenly detects objects other than the sun, resulting in an incorrect tracking. In such a case, the tracking is restarted using the calculated position of the sun at the given coordinates and time. In addition to trying to solve the error automatically, Pyra

420 is also sending also sends an error notification email to an operator, whose email address can be defined in the settings.

#### Appendix B: EM27/SUN calibration factors

*Author contributions.* FD and JC conceived the study and developed the concept, FD led the hardware and software development as well as the setup of the sensor network. FD, BV and BR built the enclosure systems. PA, BV and FD developed the software Pyra. FD and JC have performed the measurements. FD, JC and NN analyzed the measurement data. FD and JC wrote the manuscript.

<sup>425</sup> Competing interests. The authors declare that they have no conflict of interest.

Table B1. Scaling factors of the side-by	r-side measurements with reference to our	standard instrument ma61 for CO <sub>2</sub>	and $CH_4$
			~ ~ ~ ~ ~ ~ ~ ~

#~~~	Date	Species	ma61	mb86	<u>mc15</u>	<u>md16</u>	mel7
1~~	Aug 2018	$\underbrace{\mathrm{CO}}_{2}(\mathbf{R}^{2})$	1.(1.00)	0.99998 (0.99)	~	~	~
		$\underline{CH_4}(\underline{R}^2)$	1(1.00)	0.99966 (0.99)	$\tilde{\sim}$	≂	≂.
2	2 Eab 2010	$\underbrace{CO_2(\mathbb{R}^2)}$	1(1.00)	0.99960 (0.99)	$\tilde{\sim}$	≂	≂.
		$\underline{CH_4}(\underline{R}^2)$	1(1.00)	0.99996 (0.99)	$\tilde{\sim}$	≂	≂.
3	Sept 2019	$\underbrace{CO_2(\mathbb{R}^2)}$	1(1.00)	$\tilde{\sim}$	0.99922 (0.96)	≂	≂.
		$\underbrace{CH_4}(\underline{R}^2)$	1(1.00)	$\tilde{\sim}$	0.99946 (0.99)	≂	≂
4	4 <u>Dec 2019</u>	$\underline{CO_2(\mathbb{R}^2)}$	1(1.00)	0.99995 (0.98)	$\tilde{\sim}$	1.00034 (0.97)	≂.
≁~		$\underbrace{CH_4}(\underline{R}^2)$	1(1.00)	0.99999 (0.86)	$\tilde{\sim}$	1.00041 (0.90)	≂
5	<u>Nov 2020</u>	$\underline{CO_2(\mathbf{R}^2)}$	1.(1.00)	$\overline{\sim}$	$\bar{\sim}$	$\tilde{\sim}$	0.99989 (0.98)
		$\underbrace{CH_4}(\underline{R}^2)$	1.(1.00)	$\overline{\sim}$	$\tilde{\sim}$	$\tilde{\sim}$	1.00175 (0.99)

 Table B2. Scaling factors according to Frey et al. (2019) of our five EM27/SUN instruments with respect to the reference EM27/SUN (S/N 037) at KIT.

Instrument	<u>S/N</u>	Date	$\underbrace{CO_2}$	<u>CH4</u>	<u>CO</u>
<u>ma61</u>	<u>61</u>	20170713	0.9993	0.9996	1.0000
<u>mb86</u>	<u>86</u>	20180214	0.9986	1.0002	0.9975
mc15	115	20190725	0.9998	1.0005	1.0272
<u>md16</u>	116	20191014	0.9998	0.9996	1.0055
me17	117	20191031	1.0015	1.0004	1.0058

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