## Interactive comment on "Observational constraints on methane emissions from Polish coal mines using a ground-based remote sensing network" by Andreas Luther et al.

Dear Brad Weir,

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Thank you very much for your review! All your points are useful and improve the manuscript a lot. We answered all questions and implemented your suggestions. Answers are written in italics, changes regarding the manuscript are written in blue italics.

## 10 Specific points

A correction after COCCON calibration? I found the correction applied after the COCCON calibration to be confusing/worrying. Isn't one goal of the COCCON calibration to prevent issues like these? Obviously this is more a question for the COCCON group than for this paper, but many of those people are coauthors, so it seems relevant. Could you please clarify here?

The centralized COCCON calibration activities as reported by Frey et al. (2019), and Alberti et al. (2021) are of utmost importance in order to achieve optimal performance of the EM27/SUN network and for the proper tying of this network as a whole to the TCCON scale. Although the EM27/SUN
spectrometer is very robust, we cannot exclude that repeated transports might slightly alter the optical alignment. The EM27/SUN instruments that were operated during the CoMet campaign were used for different measurement campaigns around the globe and therefore transported and operated in a variety of different locations after their previous centralized calibration check by the central facility. Therefore it is advisable to perform a dedicated side-by-side calibration in the framework of a campaign which
focuses on the detection of differential column signals using several distributed spectrometers and to use the resulting relative calibration factors for the interpretation of the observations. This is a recommended and common procedure applied during similar network deployments to exclude spurious gradients (Frey et al., 2015; Chen et al., 2016; Frey et al., 2019; Vogel et al., 2019; Makarova et al.,

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2020; Dietrich et al., 2021; Jones et al., 2021).

2. Independent data evaluation. I would've preferred an evaluation against indepedent data, perhaps say a comparison of prior and posterior  $CH_4$  against the aircraft data from Kostinek et al. (2021). The set-up in this paper is so close to that paper, it really is puzzling how they disagree on greater vs. lesser emissions for the June 6th case. It would be interesting to see how well they agree with each other in  $CH_4$  mixing ratio apage and if it has anything to do with the abave reaching. However, the output of the set of the s

<sup>35</sup> mixing ratio space and if it has anything to do with the above rescaling. However, the authors can't be expected to do everything, the paper is quite thorough, and the scientific evaluation in Section 4 and the Conclusions, especially the discussion in the context of Kostinek et al. (2021) is sufficient. Since this is a paper about a campaign and network, I thought it might be nice to have a few more sentences on how to resolve these discrepancies other than adding more and more data until they go away.

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Within CoMet we aimed at comparing multiple methane emission estimation approaches. Beside the ground-based teams, various airborne datasets were collected, e.g. flask samples, methane lidar measure-



Figure 1. Comparison of shaft-wise emission estimates reported in this study with estimates by Kostinek et al. (2020) for June 6, 2018. The estimated total sums for the listed shafts are  $298 \text{ kt a}^{-1}$  for E-PRTR,  $295 \text{ kt a}^{-1}$  for Kostinek et al. (2020), and  $545 \pm 73 \text{ kt a}^{-1}$  for this study.

ments, and imager observations (Krautwurst et al., 2021). Combining all these data will be challenging, which could also be a take-away message from this work. Fig. 1 directly compares the shaft-wise results for June 6 from our study to those from Kostinek et al. (2020). Note, that our estimates in Fig. 1 are 45 assembled from both, the southern (Pustelnik) and the western (Raciborz) station, with observational periods from around 8UTC to 15:30UTC with one hour gap during noon for the southern station and only roughly 2 hours of data around noon for the western station. The emission estimates by Kostinek et al. (2020) are based on two flights, one in the morning, and one in the afternoon. Since these two data 50 sets are sampled on the same day but not during the same time periods, it is possible, that variability of the mining operations contributes to the discrepancies, and it is also clear that our ground-based stations are sensitive to shafts that the aircraft data of Kostinek et al. (2020) are not, and vice versa. Overall, while our study intends to demonstrate how to measure  $CH_{4}$  emissions from mining facilities using an EM27/SUN network, it is evident that only long-term observations covering various observation

techniques can enable emission monitoring. 55

3. EDGAR version and year. The year 2017 on line 32 must be a typo because EDGAR v4.3.2 ends in 2012. I hate to be this reviewer, but that is a very old version of EDGAR. The current version, v6.0, actually includes the study year of 2018. It also includes a seasonal cycle, which could be relevant to the results in this paper. My understanding is that E-PRTR and not EDGAR is used in the study's prior. In that case, it should be straightforward to include the number cited in the text for the newest version and the study year. If EDGAR was used in the prior, I'd be happy if you just noted both numbers (2012 from

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v4.3.2 and 2018 from v6.0), no need to redo experiments. Also, it might be nice to list the exact sectoral breakdown that was used.

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Indeed, the EDGAR paragraph contains errors and is rewritten. We apologize for this negligence. Since EDGAR is not used as prior (but E-PRTR plus updates), this does not affect the emission estimates. The respective paragraph is rewritten: EDGAR v4.3.2 (Emission Database for Global Atmospheric Research) accounts emissions of 675kt CH<sub>4</sub> a<sup>-1</sup> (Janssens-Maenhout et al., 2017) for fuel exploitation emissions
70 (EDGAR abbreviation PRO) for the USCB in the year 2012. EDGAR v6.0 (Crippa et al., 2020) states 454kt CH<sub>4</sub> a<sup>-1</sup> for the sector IPCC 1B1a (fuel exploitation COAL) for the USCB in 2018.

4. Satellites. Campaigns like CoMet and networks like COCCON have another very obvious use in that they are essential in the bias correction and interpretation of retrievals from satellites. There are now several satellites in orbit that observe CH<sub>4</sub>, e.g., the GOSATs, TROPOMI, with the expectation of several more (GeoCarb, Merlin) including several from commercial partners. Could you dedicate at least a sentence or two, perhaps in addressing point 2 above, to describe what impact your results might have on those missions and vice-versa?

80 It was part of our network design to eventually compare our observations to TROPOMI and/or GOSAT data. However, the satellite data (after cloud and quality screening) were too sparse to allow for a profound comparison. A dedicated satellite validation campaign with a long term stationary network deployment could indeed contribute to improvements on both sides, the satellite and the ground-based emission estimation. We added the following sentence to the discussion: In addition to the monitoring
85 aspect, permanent networks could also validate satellite missions and the different ground-based and space-borne emission estimation approaches could be consolidated.

5. The background variability of 0.6 ppb on line 178 seems an order of magnitude too small. Could you provide more support for why this is reasonable? Looking at Table 3 from the Barkley et al. (2017) paper
90 from ACT-America, they have day-to-day variability in background CH<sub>4</sub> over North America of several ppb. I'm not sure that exactly maps to the purposes here, but assuming much of that is passing weather patterns, I'd guess a number of at least 6 ppb would be more appropriate. Maybe this gets picked up in the L-curve fitting, so it isn't of much practical importance? Either way, 0.6 ppb seems low.

- 95 Our wording was confusing in the respective paragraph. We assume 0.6ppb for the measurement noise based on our spectrometric observation. The "background variability" (we should have called it "background error") is derived from a sensitivity study for each individual case by shifting the airmass travel time between upwind and downwind location by plus-minus 30min. The resulting background error ranges between 0.3ppb and 2.2ppb. Since we calculate the background and its error directly from
- 100 the daily station data, day-by-day methane background variability does not play a role. For clarification, we rewrote the paragraph: The estimated background error ranges between 0.3 ppb and 2.2 ppb for the individual case studies, based on the differences of minimum and maximum average  $\Delta XCH_4$  calculated under consideration of  $\pm 30$  min time shifts of the simulated methane travel times. The measurement noise amounts to 0.6 ppb, calculated as the standard deviation of the averaged measurements of The Glade (E)

## **Technical points**

1. Can you give some indication of the correlation lengths/patterns of model background ( $S_{\epsilon}$ )? Correlation lengths affect the interpretation of the values on the diagonal, so this would be helpful putting those numbers in the context of other studies. Since the authors used an ensemble approach, a correlation "length" might not be 100% appropriate, but I'm sure they can come up with some number/figure to indicate how errors decorrelate horizontally, even if it is just one number or a range.

- 115 Our observation error covariance matrix  $S_{\epsilon}$  is diagonal representing the measurement error from the XCH<sub>4</sub> retrieval, the transport model error from our sensitivity studies and the background error. The manuscript states this in section 4 after equation (1). So, we did not consider observational error correlations. Following the reviewer comment, we looked into an easy way to estimate observation error correlations and combine them with the L-curve technique for regularization. We found no straight-120 forward way how to come up with reliable error correlations without substantially investing in ensemble calculations for every single case. Also, the L-curve appeared distorted for the attempts undertaken. Therefore, we propose to postpone the assessment to future studies. We added the following statement to section 4: Note that, for simplicity, we did not consider correlations in  $S_{\epsilon}$ .
- 125 2. At least two words about chemistry. Given the short time scale of the experiments in this paper, oxidation probably doesn't play much/any of a role, but it might be worthwhile to say that somewhere? We added the following sentence to the discussion: We did not consider chemical reactions as e.g. oxidation of methane by OH, since the time scales relevant for this study are too short that oxidation would have a significant influence on the emission estimations.

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3. "An" averaging kernel instead of "the". I found the use of the averaging kernel very helpful, but it might be better to call it "an" averaging kernel instead of "the" so that the reader does not confuse it with the averaging kernel of the EM27/SUN retrieval. Either that or qualifying adjectives like "the emissions averaging kernel" might help.

4. For those of us that haven't dedicated a good chunk of our life to this campaign over the past few years, it might be helpful to add the cardinal directions to the site names in the text as is done in Table 1. I did find myself going back and forth a lot trying to figure out which station was where, but got the hang of it once I finished.

This is a very helpful advise! We added the cardinal directions.

<sup>135</sup> We changed "the averaging kernel" to "the emissions averaging kernel".

## References

Alberti, C., Hase, F., Frey, M., Dubravica, D., Blumenstock, T., Dehn, A., Surawicz, G., Harig, R., Orphal, J., and the EM27/SUN-partners

- 145 team: Improved calibration procedures for the EM27/SUN spectrometers of the COllaborative Carbon Column Observing Network (COC-CON), Atmospheric Measurement Techniques Discussions, 2021, 1–48, https://doi.org/10.5194/amt-2021-395, 2021.
  - Chen, J., Viatte, C., Hedelius, J. K., Jones, T., Franklin, J. E., Parker, H., Gottlieb, E. W., Wennberg, P. O., Dubey, M. K., and Wofsy, S. C.: Differential column measurements using compact solar-tracking spectrometers, Atmos. Chem. Phys., 16, 8479–8498, https://doi.org/10.5194/acp-16-8479-2016, 2016.
- 150 Crippa, M., Solazzo, E., Huang, G., Guizzardi, D., Koffi, E., Muntean, M., Schieberle, C., Friedrich, R., and Janssens-Maenhout, G.: High resolution temporal profiles in the Emissions Database for Global Atmospheric Research, Scientific data, 7, 1–17, https://doi.org/10.1038/s41597-020-0462-2, 2020.

Dietrich, F., Chen, J., Voggenreiter, B., Aigner, P., Nachtigall, N., and Reger, B.: MUCCnet: Munich Urban Carbon Column network, Atmospheric Measurement Techniques, 14, 1111–1126, https://doi.org/10.5194/amt-14-1111-2021, 2021.

- 155 Frey, M., Hase, F., Blumenstock, T., Groß, J., Kiel, M., Mengistu Tsidu, G., Schäfer, K., Sha, M. K., and Orphal, J.: Calibration and instrumental line shape characterization of a set of portable FTIR spectrometers for detecting greenhouse gas emissions, Atmos. Meas. Tech., 8, 3047–3057, https://doi.org/10.5194/amt-8-3047-2015, 2015.
  - Frey, M., Sha, M. K., Hase, F., Kiel, M., Blumenstock, T., Harig, R., Surawicz, G., Deutscher, N. M., Shiomi, K., Franklin, J. E., Bösch, H., Chen, J., Grutter, M., Ohyama, H., Sun, Y., Butz, A., Mengistu Tsidu, G., Ene, D., Wunch, D., Cao, Z., Garcia, O., Ramonet, M.,
- 160 Vogel, F., and Orphal, J.: Building the COllaborative Carbon Column Observing Network (COCCON): long-term stability and ensemble performance of the EM27/SUN Fourier transform spectrometer, Atmos. Meas. Tech., 12, 1513–1530, https://doi.org/10.5194/amt-12-1513-2019, 2019.
- Hase, F., Frey, M., Blumenstock, T., Groß, J., Kiel, M., Kohlhepp, R., Mengistu Tsidu, G., Schäfer, K., Sha, M. K., and Orphal, J.: Application of portable FTIR spectrometers for detecting greenhouse gas emissions of the major city Berlin, Atmos. Meas. Tech., 8, 3059–3068, https://doi.org/10.5194/amt-8-3059-2015, 2015.
- Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Muntean, M., Schaaf, E., Dentener, F., Bergamaschi, P., Pagliari, V., Olivier, J. G. J., Peters, J. A. H. W., van Aardenne, J. A., Monni, S., Doering, U., and Petrescu, A. M. R.: EDGAR v4.3.2 Global Atlas of the three major Greenhouse Gas Emissions for the period 1970–2012, Earth System Science Data Discussions, 2017, 1–55, https://doi.org/10.5194/essd-2017-79, 2017.
- 170 Jones, T. S., Franklin, J. E., Chen, J., Dietrich, F., Hajny, K. D., Paetzold, J. C., Wenzel, A., Gately, C., Gottlieb, E., Parker, H., Dubey, M., Hase, F., Shepson, P. B., Mielke, L. H., and Wofsy, S. C.: Assessing urban methane emissions using column-observing portable Fourier transform infrared (FTIR) spectrometers and a novel Bayesian inversion framework, Atmospheric Chemistry and Physics, 21, 13131–13147, https://doi.org/10.5194/acp-21-13131-2021, 2021.
- Kostinek, J., Roiger, A., Eckl, M., Fiehn, A., Luther, A., Wildmann, N., Klausner, T., Fix, A., Knote, C., Stohl, A., and Butz, A.: Estimating
   Upper Silesian coal mine methane emissions from airborne in situ observations and dispersion modeling, Atmospheric Chemistry and Physics Discussions, 2020, 1–24, https://doi.org/10.5194/acp-2020-962, 2020.
- Krautwurst, S., Gerilowski, K., Borchardt, J., Wildmann, N., Galkowski, M., Swolkien, J., Marshall, J., Fiehn, A., Roiger, A., Ruhtz, T., Gerbig, C., Necki, J., Burrows, J. P., Fix, A., and Bovensmann, H.: Quantification of CH<sub>4</sub> coal mining emissions in Upper Silesia by passive airborne remote sensing observations with the MAMAP instrument during CoMet, Atmospheric Chemistry and Physics Discussions, 2021, 1–39, https://doi.org/10.5194/acp-2020-1014, 2021.
- Makarova, M. V., Alberti, C., Ionov, D. V., Hase, F., Foka, S. C., Blumenstock, T., Warneke, T., Virolainen, Y., Kostsov, V., Frey, M., Poberovskii, A. V., Timofeyev, Y. M., Paramonova, N., Volkova, K. A., Zaitsev, N. A., Biryukov, E. Y., Osipov, S. I., Makarov, B. K., Polyakov, A. V., Ivakhov, V. M., Imhasin, H. K., and Mikhailov, E. F.: Emission Monitoring Mobile Experiment (EMME): an overview and first results of the St. Petersburg megacity campaign-2019, Atmospheric Measurement Techniques Discussions, 2020, 1–45, https://doi.org/10.5194/amt-2020-87, 2020.
- Vogel, F. R., Frey, M., Staufer, J., Hase, F., Broquet, G., Xueref-Remy, I., Chevallier, F., Ciais, P., Sha, M. K., Chelin, P., Jeseck, P., Janssen, C., Té, Y., Groß, J., Blumenstock, T., Tu, Q., and Orphal, J.: XCO<sub>2</sub> in an emission hot-spot region: the COCCON Paris campaign 2015, Atmospheric Chemistry and Physics, 19, 3271–3285, https://doi.org/10.5194/acp-19-3271-2019, 2019.