Response to Referee #1

We would like to thank the reviewer #1 for taking the time to review this manuscript and provide valuable and constructive feedback that have improved the manuscript.

In this author comment all the points one-by-one raised by the reviewer are copied here and shown in bold text, along with the corresponding reply from the authors in plain text. We will go through our manuscript text and figures and try to shorten the paper where appropriate and we will submit a revised and restructured version of the manuscript when the second referee comment becomes available (for including all suggestions).

1. **The Introduction takes a very long time to get to the point, and at the end we still don't have a clear statement of the problem. Is the paper to quantify emissions from Madrid, or from three landfills, or is this the same thing? Why should we care about Madrid? What is actually new here?**

Sorry for this vaguely-referred information. We will make this clearer in the introduction. We estimate emissions from waste disposal sites as stated in the title of the paper. These three landfills are so close to the Madrid metropolitan area that they can be considered as city sources, and they are the most significant (also the unique significant) CH_4 sources in Madrid area. Meanwhile, these together disposal sites are rather strong source as compared with the inventory in Madrid. The metropolitan cities are continuously growing due to population movements, industries, etc., and thus, more and more cities incorporate landfills (and other CH_4 potential sources) into their limits/influential areas. This study might be also interesting for different big/medium cities and the method can be applied there.

2. **Section 2.1.2: give reference for the specific TROPOMI product that you are using. TROPOMI has a very low success rate (3% globally), is that an issue here? Or is Madrid sunny enough?**

(1) The specific TROPOMI products are from Lorente et al. (2021)*. We will add this to Section 2.1.2. according to the referee's comment.

(2) Yes, you are right that TROPOMI globally has a low success rate. This is more problematic at high latitudes and in winter. Southern Europe, like Madrid is pretty sunny and the weather during the field campaign was sunny as well.

The TROPOMI data with high quality (qa=1.0) used in this study are further collocated with the IASI data, covering the time period from November 2017 to October 2020. There are nearly 29,000 measurements. Although these observations are from many different days they can be used consistently for the local emission estimates, because we remove the daily varying background signal. This background removal is very important and allows for using data from many different days for achieving good coverage. The multi-year data set provides a large and consistent observational dataset (horizontal: for the whole area around Madrid; temporal: for different wind regimes) for studying the local emissions.

*Lorente, A., Borsdorff, T., Butz, A., Hasekamp, O., aan de Brugh, J., Schneider, A., Wu, L., Hase, F., Kivi, R., Wunch, D., 740 Pollard, D. F., Shiomi, K., Deutscher, N. M., Velazco, V. A., Roehl, C. M., Wennberg, P. O., Warneke, T., and Landgraf, J.: Methane retrieved from TROPOMI: improvement of the data product and validation of the first 2 years of measurements, Atmos. Meas. Tech., 14, 665–684, https://doi.org/10.5194/amt-14-665-2021, 2021.

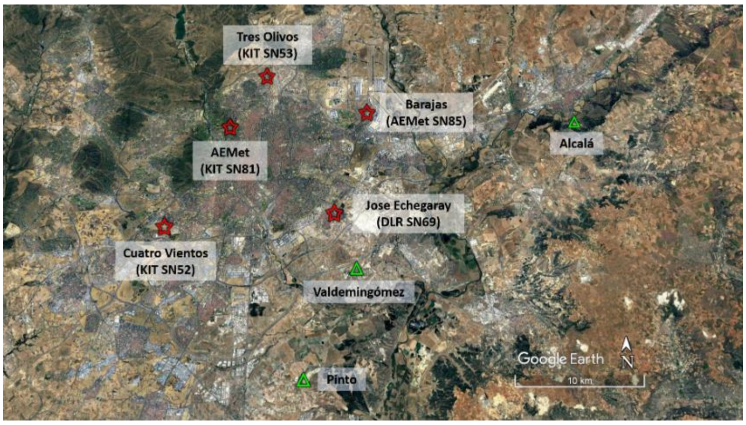
3. **Section 2.1.3: this combined IASI+TROPOMI TXCH$_4$ product is probably totally dominated by TROPOMI information in the PBL, which is what matters here. So how is it independent from TROPOMI?**

TROPOMI measures column integrated methane (XCH$_4$). XCH$_4$ is affected by tropospheric CH$_4$ concentrations but also strongly by the altitude of the tropopause (a high tropopause causes low stratospheric contributions to XCH$_4$ and thus high XCH$_4$ values, a low tropopause vice versa). One possibility to avoid this contribution that affects the study of lower tropospheric CH$_4$ emissions is to remove the background and work only with anomalies. If one assumes that the background captures all the tropospheric background and in addition the stratospheric contribution, the anomaly contains the interesting signal. However, if the background calculation misses some of the stratospheric contribution signals, the interpretation using the anomaly for investigating local emission can lead to large errors. For instance, it might be that winds from the north-east and south-west are somehow correlated with tropopause altitudes (because both wind directions and tropopause altitudes have a seasonal cycle) and if this seasonal cycle is not well resolved in the background, the wind-assigned anomaly can have an artificial signal that comes from the stratosphere.

IASI has good information about the stratospheric contribution and combining IASI with TROPOMI allows for the generation of a product (TXCH$_4$) that is largely unaffected by the stratospheric contribution. Consequently, the risk for the TXCH$_4$ anomalies to be affected by the stratospheric contributions is much lower than for XCH$_4$. We calculate the emission for XCH$_4$ and for TXCH$_4$ and get similar emission rates for both data sets. This means our background correction is working correctly already for XCH$_4$. Furthermore, we repeat the calculations for the upper tropospheric and stratospheric CH$_4$ (UTSXCH$_4$, information from the IASI data) and found that there is no emission signal. All these prove that the emission signals we observe are not by accident, instead they come from local surface emissions and that our method for background calculation together with the wind-assigned anomaly method is able to detect these signals correctly in the XCH$_4$ as well as in the TXCH$_4$ data.

4. **Figure 1 is very difficult to read.**

Thanks for the comment. The figure is redone as below:



5. **Section 2.3: this section is very confusing because it is not clear what the authors are trying to optimize. Are the 'daily plumes' for the individual landfills? Are they summed over the three landfills? Are the three landfills treated as a single plume? The cone model is surely wrong for instantaneous plumes but is reasonable for time-averaged plumes, which is what is fitted but it takes the paper a while to explain this.**

Each individual landfill is considered as an individual point source. The daily plumes from the individual landfills are super-positioned to have a total daily plume. The final results we provide are emission rates averaged for the whole three years.

6. **Line 206: I don't get the point about seeking an analogy with NO_2. The landfills don't emit NO_x, NO_x is an area source, and the decay of the NO_2 plume is by oxidation rather than dilution into background.**

Yes, you are right that no NO_x is emitted from the landfills (while the Madrid metropolitan area is a strong source of NO_2). The usage of NO_2 in this study is to check if our method is reliable. The NO_2 is exploited as a tracer for atmospheric transport, offering sufficient chemical lifetime for forming a nice plume structure, which is a simple target gas for TROPOMI. NO_2 is a suitable (approximately stable) tracer for qualitatively demonstrating the method developed for the wind-assigned anomaly. We do not correctly consider the photochemical loss of NO_2, and the demonstration is not intended to provide a high-quality quantitative analysis of the NO_2 source strength (a refined model including NO_2 decay would generate slightly reduced outer plume lobes. The lifetime of NO_x should be in the order between 6.3h during night and 29h during daytime in winter (Kenagy et al, 2018) and 5.9h in summer (Shah et al., 2020)). Our intention is simply to check the implementation of our approach and the data we feed into the simulation: if our wind data or plume dispersion modelling would be incorrect, we would not be able to reasonably reproduce the plume properly in our model runs.

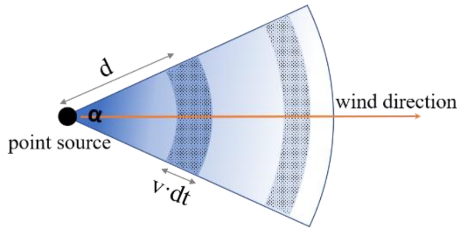
* Kenagy, H. S., Sparks, T. L., Ebben, C. J., Wooldrige, P. J., Lopez-Hilfiker, F. D., Lee, B. H., Thornton, J. A., McDuffie, E. E., Fibiger, D. L., Brown, S. S., Montzka, D. D., Weinheimer, A. J., Schroder, J. C., Campuzano-

Jost, P., Day, D. A., Jimenez, J. L., Dibb, J. E., Campos, T., Shah, V., Jaeglé, L. and Cohen, R. C.: NOx Lifetime and NOy Partitioning During WINTER, J. Geophys. Res. Atmos., 123(17), 9813–9827, doi:https://doi.org/10.1029/2018JD028736, 2018.

Shah, V., Jacob, D. J., Li, K., Silvern, R. F., Zhai, S., Liu, M., Lin, J., and Zhang, Q.: Effect of changing NOx lifetime on the seasonality and long-term trends of satellite-observed tropospheric NO2 columns over China, Atmos. Chem. Phys., 20, 1483–1495, https://doi.org/10.5194/acp-20-1483-2020, 2020.

7. Wind speed is denoted v in the text, w in Figure 2.

Many thanks to point out this mistake. We corrected it.



8. Equation (9): not clear how you get y_BG

For clarification, Eq. 9 y_{BG} should be replaced by y (the original satellite data y=y_{BG}+y_{plume}, see Eq. 7). We use y to estimate the coefficients that describe the background. Information from y is only used when the observations are not affected by the plume. In that case $\mathbf{K_{BG}}^*$ and thus $\mathbf{G_{BG}}$ are zero. On the other hand, $\mathbf{K_{BG}}^*$ (and thus $\mathbf{G_{BG}}$) is set to zero whenever y_{plume}=0. Then y_{BG}=y and Eq. 9 is correct as it. But in order to make it clearer we will write in Eq. 9 y instead of y_{BG}.

9. Section 2.3: there are many uncertainties in the procedure for inferring emissions. How can it be validated? An obvious way would be to use the independent COCCON observations to evaluate the posterior concentrations resulting from the TROPOMI inversion.

Estimated emission rates can include large uncertainties. In this study, the uncertainty of the estimated emission rates derived from TROPOMI XCH_4 ($7.4\times10^{25} \pm 6.4\times10^{24}$ molec s^{-1}) based on our method is about 9% ($6.4\times10^{24}/7.4\times10^{25}$). Please note that this is only the uncertainty due to the background uncertainty. Figure 10 shows sensitivity analysis due to wind, emission source, and opening angle, and reveals that there are other important uncertainty sources.

The referee gives a very good suggestion that COCCON is an independent data to evaluate the results. We applied this strategy and tried to estimate the emission rate from COCCON measurements on October 4 (Section 3.1), when a significant enhancement was observed by the downwind-side COCCON SN69. The estimated rate is about 3.7×10^{25} molec s^{-1}. This value is about half of that derived from TROPOMI XCH_4 or combined $TXCH_4$, which is a plausible match, because the satellite covers the complete area, whereas the COCCON plume observation primarily detects the emission from a single nearby landfill.

10. Figure 3: the agreement between TROPOMI and COCCON in that Figure strikes me as very poor, despite the authors' claim to the contrary. I'm not surprised by this in view of the known TROPOMI biases, but it undermines confidence in the results of the TROPOMI inversion. The paper goes on about the problems on Sept 25 and Oct 4 but that seems anecdotal and those two days don't seem any worse than the rest of the population in Figure 3.

We do not recognize an apparent bias between COCCON and TROPOMI (in Fig 3 we see some scatter, but the whole ensemble follows the 1:1 line quite well). Larger variations of XCH_4 in a metropolitan area containing localized sources as Madrid are to be expected (which will induce some scatter between the datasets, because the spatial resolution of the space-based sensor is much lower). Studies under background conditions have revealed very good agreement and low bias between COCCON and TROPOMI (Tu et al., 2020*). Moreover, it needs to be emphasized that our conclusions on emission strengths depend on averaged values of observed *gradients* in each dataset, so a general bias due to a calibration mismatch between satellite and ground-based would remain largely without effect.

Tu, Q., Hase, F., Blumenstock, T., Kivi, R., Heikkinen, P., Sha, M. K., Raffalski, U., Landgraf, J., Lorente, A., Borsdorff, T., Chen, H., Dietrich, F., and Chen, J.: Intercomparison of atmospheric CO2 and CH4 abundances on regional scales in boreal areas using Copernicus Atmosphere Monitoring Service (CAMS) analysis, COllaborative Carbon Column Observing Network (COCCON) spectrometers, and Sentinel-5 Precursor satellite observations, Atmos. Meas. Tech., 13, 4751–4771, https://doi.org/10.5194/amt-13-4751-2020, 2020.

11. Lines 329-330: how do we know that the 'COCCON instruments show a very good ability to detect the source'? No specific results or data from COCCON are shown.

The COCCON SN69 was located in the northwest of the landfill Valdemingómez with a quite close distance (4.5 km). When the wind came from southeast, the COCCON SN69 was located downwind of the landfill and detected a significant plume (nearly up to 100 ppb on October 4, 2018, Figure 4), whereas the other COCCON sites did not observe any enhancements. Another example is October 1, 2018 (Figure A-2): the wind direction was north to northeast and the COCCON stations were not on the downwind side of the landfills, which resulted in no enhancement at any of the COCCON stations. The observations on different days were largely depended on the wind situation. The obvious downwind enhancement observed by the COCCON instruments demonstrates that they have the ability to detect the emissions of the source, which has been demonstrated in Kille et al. (2019*) as well.

* Kille, N., Chiu, R., Frey, M., Hase, F., Sha, M. K., Blumenstock, T., Hannigan, J. W., Orphal, J., Bon, D. and Volkamer, R.: Separation of Methane Emissions From Agricultural and Natural Gas Sources in the Colorado Front Range, Geophys. Res. Lett., 46(7), 3990–3998, doi:https://doi.org/10.1029/2019GL082132, 2019.

12. Figure 8 is cryptic. What domain is shown? What are we learning from it?

We have tried to visualize as good as possible the different steps of the data treatment. Figure 7 shows the time series of the different satellite data (y from Eq. 7), their estimated background (y_{BG} from Eq. 12), and the anomaly signal due to the local emissions (y_{plume} from Eq. 13). Then Figure 8 shows the anomalies (i.e. y_{plume}) horizontally averaged for different wind directions. This data is used for the Δ-calculations. Eq.15 captures both: the horizontal averaging according to the wind directions as well as the Δ calculations. The results of these calculations are then shown in Fig. 9. These ΔCH_4 data are finally used to estimate the emission rates according to Eq. 18.

Furthermore, Figure 8 is very useful here, because it demonstrates that the CH_4 hotspots are south-east of Madrid and not in the center of the city. South-east of Madrid is where the waste disposal sites are located.

13. Table 5: I don't see the relevance of this Table to the paper.

This Table shows some results from other studies as a reference for our results. It helps to demonstrate that our results are reliable and lie in the reasonable range. The inventory only lists the active landfill cells and does not include the closed ones, which probably still emit for many years (Sánchez et al., 2019*). This is an additional argument for the relevance of the kind of work we are presenting here.

*Sánchez, C., de la Fuente, M. del M., Narros, A., del Peso, I. and Rodríguez, E.: Comparison of modeling with empirical calculation of diffuse and fugitive methane emissions in a Spanish landfill, J. Air Waste Manage. Assoc., 69(3), 362–372, doi:10.1080/10962247.2018.1541029, 2019.