Response to Referee #2

We would like to thank the reviewer #2 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 raised by the reviewer are copied here one-by-one and shown in bold text, along with the corresponding reply from the authors in plain text.

1. **The Introduction gives extensively credit on former works and is therefore also an important part, which should not be missing even it might not help the reader in understanding of the new technics.**

   Therefore, I would ask for a kind of road map at the beginning of second section “method”, before 2.1 which gives an idea, why and how the measurements and models are combined, or maybe it could be included directly at the end of introduction where the different sections are already mentioned.

Here, we add two more sentences to describe our methodology in the last paragraph of the introduction:

“Section 2 describes our methodology. We calculate the difference of the satellite data maps for two opposite wind regimes (we refer to the resulting signals as wind-assigned anomalies). A simple plume model is applied to predict the wind-assigned anomalies for a chosen position and strength of a source. The results of our study are presented and discussed in Section 3……”

2. **TROPOMI+IASI**

   2.1 You report two estimations in the abstract, which are slightly contra dictionary. The error in the estimation is higher than the error estimation from the combined retrieval, I imagine that this is more or less the effect that partial columns are constructed with more errors (Error of TROPOMI + error of IASI), but there should be another systematic error, which is reduced if the l tropospheric partial column anomaly is sub or over estimated. It would be easier for us readers, if the authors conclude on a single best estimation, and report the other option using just TROPOMI just in the results and discussion of the article.

As stated correctly by the referee, the stratospheric contribution causes larger uncertainties when using the XCH_4 data. However, it is very difficult to estimate this error. In the paper, we do not estimate the uncertainty of the stratosphere contribution. Instead, we use combined data (IASI+TROPOMI, where the stratospheric contribution can be neglected) to show that our background removal method works well and removes the stratospheric effects, also in the XCH_4 data.

2.2 The combination of IASI and TROPOMI assumes measurements of the same air mass, and therefore at the same time, but IASI comes around 10:00 and TROPOMI around 13:00, does this matter here, o IASI characterize more the background CH_4 and is therefore less critical. Maybe it would be interested in a discussion how the time difference between IASI and TROPOMI of some hours, might affect the combined retrieval.

Please also describe a bit more how much the estimation improves due the combination with IASI, I could imagine, that this advantage will increase if you use long term averages, so that the random error loses its importance in relation to improved sensitivity in the lower atmosphere.

In Schneider et al. (2021b) we found that a temporal mismatch of up to four hours is no problem. The noise error is larger than the error introduced by combining two measurements that have a temporal mismatch of four hours. The mismatch error just increases slightly the statistical error budget.

Yes, the noise error is larger in the combined product (see Schneider et al., 2021b). And this error will get smaller, when more data are used for averaging. So, if the emission rate keeps constant over a long time, the estimation of these emission rate will become the better, the longer the time series is (the more data are used for averaging). However, in practice the emission rates might not be fully constant over time.

3. Plume model

3.1 The authors realize their proper idea and do not have to cite other works, which might be partly similar ideas, but it would help some reader to understand it easier, which is similar to other approaches (up, downwind Rotating method, Gaussian's Law).

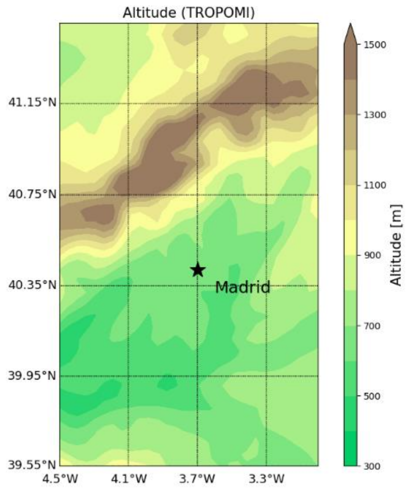
The comment is very helpful to make reader better understand of our simple model. We try to explain our model more:

Our plume model is a simplified version of the Gaussian plume model (https://link.springer.com/chapter/10.1007/978-1-4757-4465-1_7, Figure 7-1). Our simple model treats a two-dimensional concentration field (without height being involved) generated by a point source (or an array of point sources) and the gas molecules in the downwind side are evenly distributed into a sector (not a Gaussian distribution) centered along the wind direction. The opening angle of the sector is the only free model parameter, which is adjusted in order to reasonably reproduce the observe NO_2 plume.

3.2 Do I understand correctly the point, that the plume model is retrieved using the City centre of Madrid as central source, but applied to the source located at a different location (landfills). So orography should be similar or not be important. I would have been interested in the required conditions on the regional topography for this approach to be transfer also to other cities or waste deposals sites. And I also would like to know the typical mixing layer height in the area around Madrid.

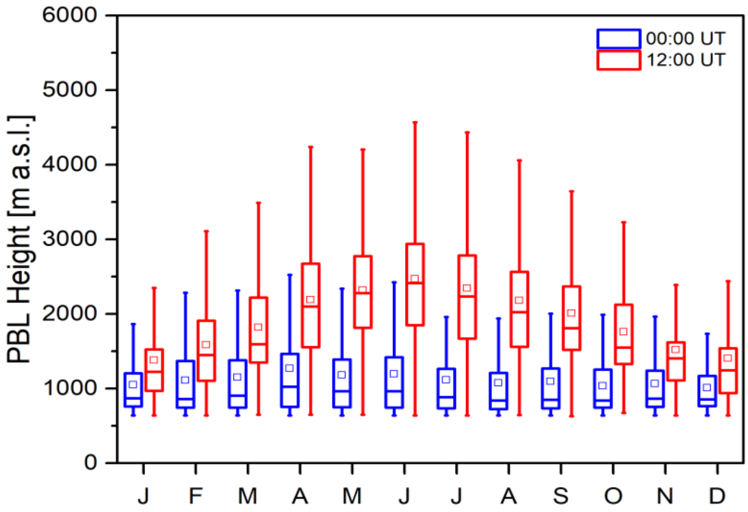
For NO_2 we use the Madrid city center as a central source for the plume model. However, we use three appropriately located point sources in case of CH_4. The a priori information about the CH_4 emissions from the landfills (Table 3) is taken from the Spanish Register of Emissions and Pollutant Sources. Each individual landfill is considered as a point source. The contributions from the individual landfills are super-positioned to generate a total daily plume.

The regional topography from Google map and the altitude derived from TROPOMI are presented as below. The mountain ranges locate along the NE-SW direction, which forces the wind flowing along this direction. We, however, do expect similar wind conditions in the city center and the landfills located nearby, as the topography is rather flat in this sub-region.



Our Spanish colleagues kindly compiled information on the Planetary Boundary Layer (PBL) height: the height of the PBL in Madrid, and its seasonal evolution, is the typical from continental areas. Figure below shows the averaged seasonal cycle of the height of PBL using meteorological radiosondes launched twice a day from the Madrid-Barajas station (WMO #08221) in the period 1981-2015. It is calculated using the Heffter method (Heffter JL. 1980. "Transport Layer Depth Calculations." Second Joint Conference on Applications of Air Pollution Meteorology, New Orleans, Louisiana). Note that the radiosonde launch station is the same used in the MEGEI-MAD campaign.

So, the noon PBL reaches typically altitudes of about 1300 m in wintertime and up to 2500 m in summertime. We discuss possible corrections due to increasing wind speed with altitude on our emission estimates further below.



3.3 **The test using NO_2 from TROPOMI results in a lower emission rate compared to estimation from the literature. The authors identify the lifetime as the reason and maybe also the value in the literature might have errors, or might not be comparable, different time ....However a validation of the wind speed estimation from a 10 m altitude would be nice.**

For CH_4 and its long lifetime the anomaly in the column should be definitively given by 1/d, as described by the equation 3.
The forward model contains the velocity and actually the quotient emission/velocity determine the concentration, or vice versa in the inversion. That is why the wind speed estimation might be so important. The sensitivity study using the wind at 10 m is very helpful and important, never the less I did not get completely get the point how you decided that 10 m is the best. I think the wind velocity you look for is the velocity you can multiply with the column anomaly to get the total CH_4 flux. Ad hoc I would take the average windspeed in the atmospheric boundary layer.

The COCCON sites at "Jose Echegaray" and "Barajas" have a distance of more or less around 8km, on 25 September 2018 (Figure 4) we see that anomalies en CH4 arrives around 1-2 hours later in Barajas than it appears in "Jose Echegaray".

So the velocity of propagation of this plume might be 8 km/2 h to 8 km/h or 1.11m/s to 2.2 m/s respectively, this fits actually very well to the assumed and modelled wind velocity in the plot below. Surely that is the intention showing the plot, but maybe it should be also explicitly be mentioned, that you can also use the 5 FTIR sites to validate the wind estimation strategy you have chosen, as here the interest in the effective flux of CH4 and it might be the best validate the velocity using the FTIR EM27 measurements of columnar CH4, so that you are independent of the vertical distribution of CH4.

EM27 measurements take typically a little bit less than a 1 minute, maybe you could do a cross correlation to retrieve the delay after interpolate data to a common 1 minute grid, and then include a point in the figure showing the wind speed or projection, but it is sufficient just to mention it.

We thank the referee for the careful consideration concerning the wind. We fully agree that the limited quality of the available wind data is a significant source of uncertainty.

According to WMO (WMO, 2018), the measurement representative of the surface winds is the wind records at 10 m a.g.l. to avoid the roughness of surrounding terrain. In this sense, the winds at 10 m are usually taken as a proxy for surface emission estimations (e.g. Viatte et al., 2017), such as in the Madrid case. In addition, we chose ERA5 wind at 10 m, because it can be directly compared to the in situ wind observations at 10m at the Cuatro Vientos Airport, which helps to estimate the uncertainty introduced by the wind data.

The wind at ~500 m a.g.l (900 hPa) would be a more appropriate choice for transport modelling if we allow for vertical mixing of the plume within the PBL. As shown in the Table 1 and **Figure 1** below, there is a significant increase of wind speed with altitude. The ERA5 wind data at 10 m and ~500 m do not differ significantly concerning the wind direction, but the wind velocity increases with altitude. The wind speed increases by 60%, i.e. using ERA5 wind information at ~500 m instead of that at 10 m would yield 60% increase in the emission rate.

Table 1: ERA5 wind at 10 m and 100 m in TROPOMI overpass days.

Wind direction range	10 m		~500 m/900 hPa	
	Number of days in total (%)	Averaged wind speed ± standard deviation (m s^{-1})	Number of days in total (%)	Averaged wind speed ± standard deviation (m s^{-1})
NE / >315° or <135°	28.4	2.3 ± 1.2	30.9	3.8 ± 2.1
SW / 135° – 315°	61.8	2.3 ± 1.4	56.7	3.6 ± 2.0

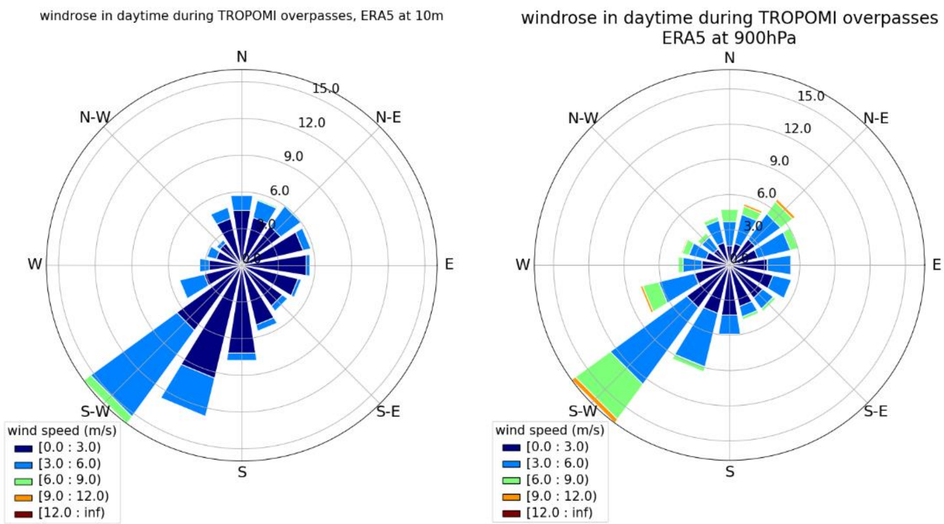


Figure 1: Wind roses for daytime (08:00 UTC – 19:00 UTC) for the ERA5 model wind at 10 m (left) and at ~500 m (900 hPa) (right).

It is really a good hint to mention that we should use the array of COCCON stations itself to validate the assumptions on the wind field. We will add this statement to our manuscript:

"These five COCCON stations can serve as an independent source of information for constraining the wind speed. For example, the distance between the Jose Echegaray and Barajas is about 10 km. The highest anomalies of XCH_4 arrived around 1.5 hours later at Barajas station than it appeared at the Jose Echegaray station on 25 September 2018, which indicates an averaged wind speed of 1.8 m/s. This value fits well to the ERA5 model wind velocity."

WMO, Guide to Instruments and Methods of Observation Volume I –Measurement of Meteorological Variables, Report No. 8, ISBN 978-92-63-10008-5, Geneva, Switzerland, 2018.

Viatte, C., Lauvaux, T., Hedelius, J. K., Parker, H., Chen, J., Jones, T., Franklin, J. E., Deng, A. J., Gaudet, B., Verhulst, K., Duren, R., Wunch, D., Roehl, C., Dubey, M. K., Wofsy, S., and Wennberg, P. O.: Methane emissions from dairies in the Los Angeles Basin, Atmos. Chem. Phys., 17, 7509–7528, https://doi.org/10.5194/acp-17-7509-2017, 2017.

4. **Page 2, Line 37:**
   4.1 "The wind-assigned plume method is also applied to the tropospheric and upper tropospheric/stratospheric column averaged CH4 mixing ratio products (in the following referred to as TXCH4 and UTSXCH4) derived from a-posteriori merged Infrared Atmospheric Sounding Interferometer (IASI) profile and TROPOMI total column data." Maybe you could split the sentence into 2 and find somehow a different description, which is easy to understand:
   For the CH4 emission estimation, the wind-assigned plume method is applied to the lower tropospheric methane /dry air column ratio (TXCH) of the combined TROPOMI Infrared Atmospheric Sounding Interferometer (IASI) Product. TXCH4 and the upper tropospheric/stratospheric column averaged CH4 mixing ratio (UTSXCH4) are derived from a-posteriori merged Infrared Atmospheric Sounding Interferometer (IASI) profile and the TROPOMI total column data.

We modify the sentence according to referee's comments:

"For the CH_4 emission estimation, the wind-assigned plume method is applied to the lower tropospheric CH_4/dry air column ratio ($TXCH_4$) of the combined TROPOMI + IASI (Infrared Atmospheric Sounding Interferometer) product. $TXCH_4$ and the upper tropospheric/stratospheric column averaged CH_4 mixing ratio ($UTSXCH_4$) are derived from a-posteriori merged IASI profile and the TROPOMI total column data."

   4.2 Does it make sense to apply the method to the UTSXCH4 Product? As mentioned earlier I would just use TXCH4 in the abstract and conclusion.

The very low emission rate derived from the $UTSXCH_4$ demonstrates that all is working well: from $TXCH_4$ we get the same as for XCH_4 (if background is carefully removed). And in $UTSXCH_4$ we see no emission signal (i.e. the background removal and the calculation of wind-assigned anomalies introduce no artificial signals).

5. **Line 40: The first sentence is a bit redundant, maybe you could drop here the sentence "Based on the NE and SW wind fields, we developed a simple plume model locating the source at three waste disposal sites east of Madrid for CH4." or move it to line 37.**

We remove this sentence according to the referee's comment.

6. **Line 44: "day and. All em…**
   Maybe you could rewrite the sentence which is also not clear." The COCCON observations indicate a weaker CH4 emission strength of around 3.7×1025 molec s-1 from local source (near to the Valdemingómez waste plant) in accordance with observations in a single day and. "Please write very clear if COCCON is used to estimate an independent "local source", if this source is part and included in the Tropomi estimation given quantitatively in the line above. Please state if TROPOMI and COCCON based estimations are contradiction, complementary and/or consistent. If you want, you might move the last sentence to line 25, as it is finally the most important finding.

The comment helps us to explain the results clearer. We change this sentence to:

"COCCON observations are investigated to estimate the local source as an independent method. The COCCON observations indicate a weaker CH_4 emission strength of around 3.7×10^{25} molec s-1 from local source (near to the Valdemingómez waste plant) in accordance with observations in a single day. That this figure is lower than the one derived from the satellite observations is a plausible result, because the analysis of the satellite data refers to a larger area, covering further emission sources in the study region, whereas the signal observed by COCCON is generated by a nearby local source."

7. **Figure 8,9:**
   Please use a fixed color scale in Fig 9 and maybe also in Fig 8. The modelled XCH_4 for total column, lower and upper troposphere seems to be exactly the same in fig 9 b,e,h. ….. Maybe when you talk about emission rates line 474 and line 477 you cloud use CH_4 and not the dry air mol fraction.
   XCH_4-> CH_4

To remove unwanted signals due to validations of ground pressure and atmospheric humidity we estimated the emission rates from total and partial column averaged dry air mixing ratios and not directly from total column amounts. The color scales in Figure 8 and 9 are chosen according to the typical dry air columns. The dry air total column is typically by a factor of 1.9 larger than the dry air tropospheric partial column. And it is typically by a factor of 2.1 larger than the dry air upper tropospheric/stratospheric partial column. These factors explain the different color scales.