Below, first our response to Anonymous Referee #1, followed by our response to Anonymous Referee #2.

Response to the review of Anonymous Referee #1

Dear referee, thank you very much for your very thorough and helpful review of our paper!
We will discuss below our changes in the paper and/or our thoughts about the points that you mention in your review in blue. We wrote our comments in italic and the improved text within quotes.

1. Missing emissions
a) Please provide motivation for increasing GFAS emissions by 5 or 10 times. For example, are there reasons to believe these are the bounds on fire emission uncertainty?
Add in to methods on page 5 or work into the analysis on page 8.

The exact uncertainty in the fire emissions from the database is not known. The papers of Mota and Wooster, (2018) Cusworth et al. (2018) and Huijnen et al. (2016) indicated all that there very likely is a (large) underestimation of the fire emission databases that are currently existent. {Mota:2018uj} mentions a 64% underestimation of particulate matter (PM) emitted in GFEDv4.1. Cusworth mentions that compared to the GFED+agriculture emissions, GFEDv4.1 PM emissions are a factor 3 too low.
For CO we could not find an estimate of the underestimation, but through personal contact with Dr. Pankaj Sadavarte (now SRON, previously at Indian Institute of Technology Bombay, IITB) we learned that they use a regional emission inventory at IITB in which the emissions of CO were a factor 2.7 times the GFAS estimate of CO emissions in November and the emissions might even be higher.

*To make this more clear, we added in the text, section 2.3: "*However, there are strong signs that fire emission inventory datasets, such as GFAS and GFED, do not capture all of the biomass burning emissions (Mota and Wooster, 2018; Cusworth et al., 2018; Huijnen et al., 2016). Also, the conversion of fire occurrence to CO emissions depends on factors such as combustion efficiency, biome type, and soil characteristics, which adds uncertainty to the emission estimates (Werf van der et al., 2010). For PM, it is estimated that the GFAS emissions are underestimated approximately a factor 2-6 (Mota and Wooster, 2018; Cusworth et al., 2018; Huijnen et al., 2016). For CO, less information is available, but similar underestimates are expected."

b) Low emissions are not the only way an inventory can be wrong. With small fires, such as agricultural fires, the issue is that fires could be completely missed (as discussed on page 12, lines 8-12), meaning that increasing emissions will not help (i.e. 10 x 0 = 0). This might be what is occurring for GFAS in this study, especially for 11-14 November. TROPOMI shows many more CO hotspots in NW India between 75 E and 80 E (Fig. 5c), compared to the 5 _ GFAS plot (Fig. 5e). In comparison, NW India between 70 E and 75 E seems better captured by the modeled hot-spots. This discussion is currently missing in the manuscript. Potential missing emissions could be further analyzed by spatially comparing the 5 _ GFAS to the 1 _ GFAS simulation to determine how much the emission increase resulted in CO column increase in the underrepresented area. A difference plot to TROPOMI could help determine the location of missing emissions. Also, a comparison to an inventory like GFED 4s, which includes an algorithm to capture small fires, could be used to analyze the spatial uncertainties in the emissions. As a result, fire emissions required in the underrepresented area could be estimated and assessed for plausibility.
Yes, it is true that some fires might be missed. We have been looking into this and have done simulations based on fire radiative power in the Suomi-NPP Visible Infrared Imaging Radiometer Suite (VIIRS) which has a higher resolution and better sensitivity to fires. Furthermore, simulations have been performed based on a land use map, as an alternative method to locate crop stubble burning in November. Unfortunately, the GFED 4s data is not yet available for this recent time

period. The VIIRS and land-used based simulations, indicated that although it is quite likely that some small burning areas are missed in the GFAS data, the resolution of the satellite and model are relatively low for this purpose and the spreading over the IGP area went relatively quick, making the exact location of the emissions not very important. So, on this resolution, increasing the emissions at the GFAS locations is probably sufficient to account for missing emission locations.
*We added in the conclusions: "*Fires missed by GFAS observations might explain part of this increase.*" We added in section 4.1: "*Or a substantial amount of fires is missed in GFAS in this period."

c) Is there reason to believe MACCity is too low by 20%?
No, but there is quite a large uncertainty in the data, estimated at 50%-200% as mentioned in the paper. So, 20% too low is still well within the uncertainties.

2. Selection of the two date periods
a) More motivation is needed for selecting the date periods. The manuscript mentions "based on patterns seen in TROPOMI" (page 7, line 1), and suggests this is presented in Section 4.3, which is not the case. The ground-based timeseries (Fig. 7) implies the cut-off could be Nov. 11-14, but also Nov. 11-16.
*Section 4.3 is only mentioned to refer to the weather conditions. We agree that based on the weather conditions only, the split could have been on November 15 or 16 also. By choosing two periods of four days, both periods have sufficient data to compare. We clarified this as follows: "*We also averaged over several days of data, concentrating on two periods: 11-14 November 2017, and 15-19 November 2017, in order to obtain a gap free image of Northern India (18 November no TROPOMI data was available). We selected these two periods based on the patterns seen in TROPOMI data, the equal number of days per period, and the weather conditions (see 4.3)."

b) Clarify why TROPOMI data before November 11 was not used (was it not available?).
On 9 November the first radiance measurements were done. The near nominal temperature was first reached on November 11 and so considered as the first day with reliable data. We added in the text: "The instrument reached near-nominal temperatures on 11 November, which is considered the first day of reliable data from TROPOMI (Borsdorff et al., 2018a)"

c) Section 3.4 discusses that ground stations show the polluted period begins on November 3rd. Why are the ground-based timeseries only shown from November 11th onwards? It would be valuable to plot all of November in order to see the increase from the 3rd of November onwards. Alternatively, the investigation of ground station data could be informed by TROPOMI and only the same time periods investigated.
We chose to show the ground-based time series from 11 November onwards to make the data periods more consistent, because for these days also TROPOMI data is available. It is however important to consider that we saw in the longer timeseries a build-up of CO levels from October on. This can be seen in the data series of 2017 in Fig. 4 as well. We changed the first sentence of this section to clarify this: "CO concentrations at the ground-level measurement stations that are used are generally increasing until 14 November, compared to earlier measurements. This is in accordance with the total column CO levels seen in the 2017 timeseries from CAMS."

3. The paper would benefit from a little more direct discussion on why investigating CO is important - e.g. in addition to a CO health threshold, co-emitted species are also relevant for human health; CO can be measured from space so using this to analyze transport of CO can determine the impact of transported pollution. This would be best built into the Introduction, Discussion and Conclusion. Also, discussing the potential impact on many people in this region would help motivate why a study of this specific region is important. Finally, while meteorology and stagnant conditions seem to drive this pollution event, there would not be as high pollution in the region without the high anthropogenic emissions. The authors could be a little more direct in highlighting this (e.g. page 15, line 12). Quantification of the anthropogenic contribution could be added

to the Abstract, Discussion and Conclusion.
We added to the Introduction: "The total population in the IGP region (including parts of Pakistan, Bangladesh and Nepal) exceeds 400 million and is growing. This means that the bad air quality is effecting hundreds of millions of people for a large part of the year. This makes it very important to investigate the origin and transport of pollution in the area. Due to its lifetime of several weeks, CO can also be used as a proxy of other, co-emitted, anthropogenic pollution."
We added to the Discussion: "This means that not only the people living in Delhi, but also a large part of the hundreds of millions of people inhabiting the rest of the IGP are affected by the bad air quality."
We added to the Conclusion: "This demonstrates the high importance of investigating the sources and the transport of pollution as hundreds of millions of people are living in the IGP and their health is likely affected by the bad air quality. CO is very suitable for investigating air pollution, not only because of the negative health impact of CO itself, but also as tracer to track the dispersion of other pollutants, due to its life-time of several weeks."

4. Expand the discussion of India versus China
a) Using the CAMS reanalysis timeseries of 2012-2017 as November average CO over the IGP and comparing with NE China could add more support to the argument on page 7 that India is now more polluted than Chna. Expanding this discussion would help make the manuscript less focused on a specific area.
Thanks for the suggestion. We have also thought about expanding the discussion to include China, but decided to keep the focus on India. We think that when extending this discussion more to China, we also should include more discussion on the sources of CO emission in China and the meteorological situation in China. This would distract too much from the message we want to convey with this paper: the high pollution levels in India and the sources thereof (and thus the possible solutions to decrease these high pollution levels). In China, already quite some effort has been and will be spend to decrease pollution levels.

b) Page 8, line 1: Please explain that if NCAP does not have emission reduction targets, what does it aim to do?
NCAP is more focused on making an inventory of how the air quality is at the moment and making plans on how to best reduce the emission. We added the following to the text (last part of the sentence):
"NCAP does not yet include strict targets for emission reductions and rather focusses on setting up an effective ambient air quality monitoring network and making plans for prevention, control and abatement of air pollution."

5. Emission resolution
How are the 0.5 _ 0.5 degree MACCity emissions interpolated to the 10 km by 10 km WRF grid? Are they downscaled? Is the result mass conserving?
The emissions are bilinearly interpolated. The result is not fully mass conserving, but the resulting differences are very small and much smaller than the error of the emission uncertainty.

6. Introduction:
a) Page 1, line 21: What is the temporal resolution of the "high concentrations", i.e. is 10mg/m3 per 8hr, 24 hr? Also describe temporal resolution for 400 mg/m3 on page 2, line 8 so we can compare to the health standard more accurately.
We added the time resolution: "carbon monoxide with values of up to 10 mg/m^3 several days in a row". And: "*reaching 15 minute average values up to 400 mg/m^3*".

b) In general, the motivation for using both mg/m3 and ppb in the manuscript is unclear. I suggest to make it clearer on page 2, line 8 why the conversion to ppb is described, and to only use ppb from there onwards (e.g. Page 11 swaps back to using mg/m3 even though it talks about plots that are displayed in ppb).

Thanks for noticing. The mg/m^3 was intended to make it easier to compare to the standards. But we decided indeed to look further at ppb levels, to make the CO levels less dependent on orography and to make total columns easier to compare with ground-level CO levels. The mg/m^3 on page 11 was meant to be ppb already. We will change this in the text and added: "… we will use ppb from here on".

c) Page 2, paragraph starting line 10: Why is GFAS uncertainty brought up in this paragraph, i.e. why is it relevant for this study? I suggest to include an introduction of the fire emission sensitivity tests (_ 5, _ 10) in the paragraph on page 3.
*We moved the following sentence from the introduction to the paragraph explaining the WRF inputs and the 1,5,10 sensitivity test: "*However, there are strong signs that these datasets do not capture all of the biomass burning emissions (Mota and Wooster, 2018; Cusworth et al., 2018; Huijnen et al., 2016). Also, the conversion of fire occurrence to CO emissions depends on factors such as combustion efficiency, biome type, and soil characteristics, which adds uncertainty to the emission estimates (Werf van der et al., 2010).".

d) Page 2, lines 27-29: Clarify "this period". Did TROPOMI (and CAMS) observe high mixing ratios for November? All days or certain days?
*We changed the previous sentence to: "*TROPOMI observed very high column mixing ratios over the Northern part of India from November 11 - 19".

7. Section 2:
a) More information is needed about the TROPOMI a priori. For example, does it come from a model climatology? Is it spatially and temporally varying?
When the right regularization is used, as is done for the TROPOMI data, the TROPOMI CO a priori is not relevant in the retrieved data. We explain this now more extensive:
"… According to Borsdorff et al. (2014):
$C_{retr} = C_{prior} + AK(C_{true} - C_{prior}) + e_x$ (1)
where ex represents the error on the retrieved trace gas profile. The Equation simplifies to Eq. 2:
$C_{retr} = AK(C_{true}) + e_x$ (2)
when the effective null space contribution of the a priori profile is eliminated, which is the true for the chosen regularisation parameter for the TROPOMI CO data, as is explained in Borsdorff et al. (2014)

b) Are land-only retrievals used or ocean as well? If ocean retrievals are used, explain retrievals over ocean (e.g. must have clouds).
Both land and ocean retrievals are used, ocean retrievals indeed require clouds. We explain already all the filtering we apply on the data in the TROPOMI subsection of Data and methods. We added the following sentence to make it more clear that also some data over sea was included: " This retrieval method allowed to include some measurements over sea with low-level clouds (Borsdorff et al., 2018c)."

c) Page 4, line 14: Biases in the KNMI, 2018 report are relative to MOPITT and IASI which are assimilated into CAMS, so are not independent measurements. Please remove "satellite" from this sentence, leaving the bias relative to TCCON.
Good point. We removed the word "satellite".

d) Define vertical resolution of CAMS rather than "various pressure levels" (page 4, line 16).
The pressure levels are the following: "1000 950 925 900 850 800 700 600 500 400 300 250 200 150 100 70 50 30 20 10 7 5 3 2 1"
*Describing them all would not add much useful information, compared to the amount of text needed. We changed "various pressure levels" to "*25 pressure levels*". In the link following the text, the pressure levels can be found.*

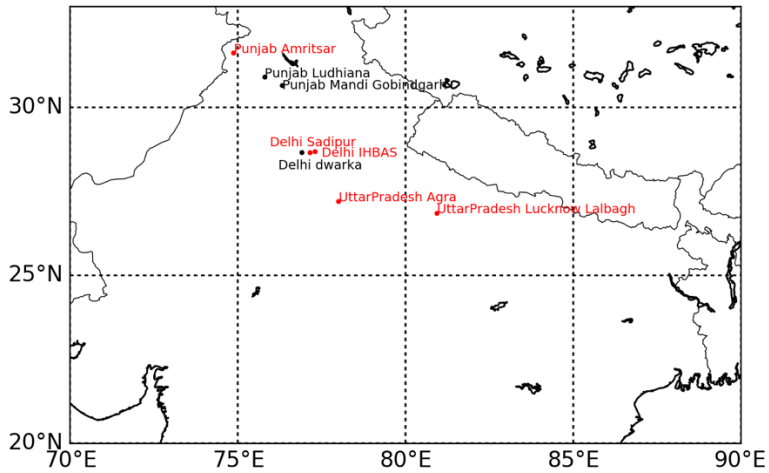
e) Define time period of CAMS data used. I interpret that 2012-2017 is used to look at October-December interannual variability, and an earlier version of CAMS was used for November 2017 boundary conditions in WRF.
We used 2012-2017 CAMS total column data for the interannual variability and the CAMS data for the pressure levels for October and November 2017 for the WRF boundary conditions.
We added October and November in the description of the WRF data: "The boundary conditions for CO came from the CAMS CO October and November data on pressure levels, interpolated to the WRF model levels." *We added the following sentence to the CAMS subsection*: "September-January total column CAMS data for the years 2012-2017 is used in section 3.1."

f) Does WRF nudge completely towards the meteorology, or does it nudge as a percentage?
Only partly: WRF nudges the interior meteorology towards the values prescribed at the lateral boundary conditions in a "relaxation zone" of, in our case, 5 cells outside the WRF grid to prevent very sharp changes in the meteorology.

g) The WRF set-up is unclear. What is the temporal period of WRF modeling? Is there a spin-up period? Are there some WRF with tracers simulations and some WRF-chem simulations? I did not realize until page 13, line 34 that there was a MACCity-only simulation. Is OH oxidation of CO performed in the sensitvity run (a major sink) subtracted from the total CO in all simulations? It would be helpful to define the base-run that everything else is compared with. Perhaps a table recording all the simulations performed would also be helpful.
*We run our model in all cases for two months: October and November 2017. October 1 to November 11 can thus be considered as spin-up period. We did not do a full WRF-Chem simulation, we only use its tracer function. OH oxidization is only taken into account in the sensitivity run, not in the other simulations. Due to the outflow out of the domain, part of the CO is already decreasing. We made this more clear in the text now: "*Different CO emission inventories are available for Southern Asia. As in CAMS, we used MACCity anthropogenic CO emissions for the year 2017 at a resolution of 0.5°x0.5°(Lamarque et al., 2010). We implemented nine different CO tracers representing the MACCity emission categories in WRF-Chem (see Table 1). The MACCity database estimates worldwide monthly emission strengths for these emission categories. An additional tracer was used to account for CO transported from the CAMS derived boundary conditions: we refer to this CO tracer as background in this paper (Table 1). For biomass burning emissions, we used GFAS data with a resolution of 0.1°x0.1°(available for download from: http://apps.ecmwf.int/datasets/data/cams-gfas/). However, there are strong signs that fire emission inventory datasets, such as GFAS and GFED, do not capture all of the biomass burning emissions (Mota and Wooster, 2018; Cusworth et al., 2018; Huijnen et al., 2016). Also, the conversion of fire occurrence to CO emissions depends on factors such as combustion efficiency, biome type, and soil characteristics, which adds uncertainty to the emission estimates (Werf van der et al., 2010). For PM, it is estimated that the GFAS emissions are underestimated approximately a factor 2-6 (Mota and Wooster, 2018; Cusworth et al., 2018; Huijnen et al., 2016). For CO, less information is available, but similar underestimates are expected.
In our base setup, we ran the model for the period 1 October - 20 November, including the tracers listed in Table 1, including the original GFAS emissions and the background. In this paper, results are shown for November 11-20, October is considered as a spin-up period. Besides the base run, we did some extra simulations including 0 (referred to as: MACCity), 1 (base setup), 5 (+5GFAS) and 10 (+10GFAS) times the original GFAS emissions, based on the estimated underestimation of fire PM emissions."

h) Figure 2: Names which are combinations of region and station are a little confusing. The acronyms are not used further in the manuscript, so I suggest writing the whole name on the map and coloring/shading the regions underneath, or adding boxes around the three regions investigated. Add latitudes and longitudes to the map.

We now included latitudes and longitudes to the map and wrote the full station locations name instead of acronyms. There are no further regions than the station locations indicated with the dots.

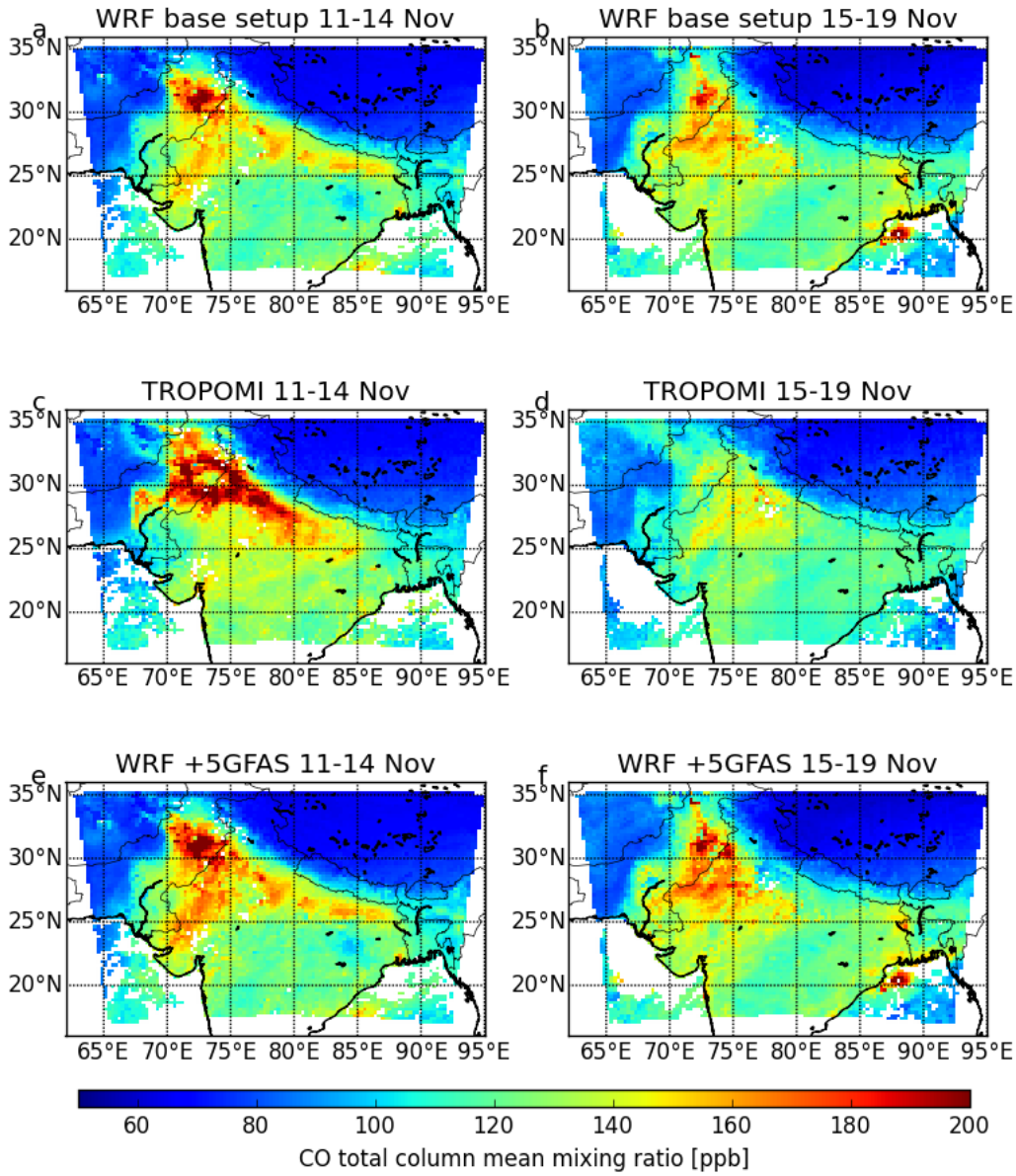


i) Why is the OH climatology scaled by 0.92?
The OH field has been optimized using methylchloroform data, leading to a scaling factor of 0.92 (Krol et al., 2013).
We changed the text to: "and the corresponding OH climatology, based on Spivakovsky et al. (2000), scaled by 0.92 (Huijnen et al., 2016, 2010; Krol et al., 2013)"

j) Page 7, line 2: Expand how the amount of data per day was used to select periods -
e.g. how many data points are in each period, were they chosen to be similar?
*Yes, both periods contain 4 days of data this way. We changed the text to: "*We also averaged over several days of data, concentrating on two periods: 11-14 November 2017, and 15-19 November 2017, in order to obtain a gap free image of Northern India (18 November no TROPOMI data was available). We selected these two periods spanning an equal number of days, based on the patterns seen in TROPOMI data and the changing weather conditions (see 4.3)"

8. Section 3:
a) While CAMS has been compared to TROPOMI globally (page 3, line 22) bias and correlation between TROPOMI and CAMS for the IGP region is needed to ensure CAMS can be used to probe the relationship between 2017 and earlier years. A sentence or two on this subject would be a valuable addition.
We added the following sentence: "For the India region, a 2.9% difference was found with CAMS with a standard deviation of 6% and a Pearson correlation coefficient of 0.9 (Borsdorff et al., 2018a)"

b) Figure 5: Column and row headings in this figure would make it faster and easier for a reader to compare the plots.
We added the headers now per subfigure.



c) I suggest that section 3.3. does not need to be separate to section 3.2, because it is still comparing WRF and TROPOMI.

*We prefer to keep these sections separated to highlight the findings presented here. As a separate section, it appears clearer to the reader that this is an important message of our paper. We now divided 3.2 in two subsections, 3.2.1: "*Agreement between WRF and TROPOMI*", and 3.2.2 is what was section 3.3 before.*

d) Figure 7 Caption: Plots are in ppb not mg/m3.

Changed the caption.

9. Section 4:

a) Page 11 lines 1 to 9: This text can be removed because the methodology section (page 6) mentions that stations close to sources are not used in the analysis.

*We wanted to make clear that in the excluded stations, some high CO mixing ratios were found and that we are not sure what the cause thereof is. We now removed the lines 1-9 in this section and added a more general statement to the next subsection on source attribution: "*Some ground stations were excluded because of the proximity to large CO sources, leading to very high observed CO concentrations. Although these measured enhancements are likely traffic related, it is possible that some of the enhancements are caused by local biomass burning. Overall, however, our results point to a relatively small role of biomass burning in the enhanced CO concentrations over the largest part of the IGP, as shown in Fig. 8 for inner-city stations. A similar contribution of biomass burning is found out of the city (not shown)."

b) The authors might want to discuss the episodic nature of fire emissions versus the consistent anthropogenic emissions. Also, while a source might contribute a small amount to the total amount of CO, it might be a more substantial contributor for anomalous CO. Finally, in Table 2, why wasn't the period split into 11-14 November and 15-19 November? It is unclear what showing October 1 to November 19 in Table 2 adds to the manuscript, when the focus has been on the two periods.

We agree that the episodic nature of fire CO emissions can be underlined more in the paper. In Table 2, we deliberately show not only the average but also maximum values to represent the CO variability.
We added the following sentence to the start of the section: "The CO emission contribution from fires has certainly a more episodic nature than the anthropogenic sources in MACCity. In this section, we do not only include the average contribution, but also consider the maximum contribution of fire emissions to investigate short-term variations in the contribution of fires to CO."
In our opinion splitting this period into 11-14 and 15-19 November does not add extra information in the discussion of the contribution of the fire emissions, which is similar in both periods.
We included the longer period, because the fire emissions before November 11 have been higher, and we wanted to give a fair comparison including also the higher contribution. We included now the following sentence: "The steadily increase in CO levels started before November 11; to make sure we did not miss part of the biomass burning contribution, we included in Table 2 also a longer period of the post-monsoon season: 1 October - 19 November."

c) Page 13, line 30: Show the plots of outflow in WRF upper layers to support the argument. A map of the mean meteorological conditions in each period overlaid on the WRF maps of CO may be helpful to visualize the differences between conditions. Alternatively, a Hovmöller diagram of the WRF results may help visualize this outflow.

d) Page 14, lines 7-10: Reword to something such as "In our study, 2017 meteorological conditions are more favorable towards pollution accumulation than 2015 conditions." Also, what data exactly supports this claim? Is it the CAMS concentrations? Or is it meteorological data that isn't shown?
The main point is not that 2017 is significantly different from 2015, which we can't prove by comparing the available data, but that both 2015 and 2017 show the build-up of air pollution. From the data that we have, 2017 seems slightly worse than 2015. We changed the text as follows: "From the meteorological and observational data we conclude that 2017 shows a similar build of pollution as in the 2015 study, however, circumstances for CO accumulations were slightly stronger in November 2017. In our study, as in Verma et al. (2017), stagnant weather conditions are an important cause of the high pollution levels."

e) Page 14, Section 4.3: Does the inclusion of OH oxidation fix the overestimation for 15-19 Nov in the non-IGP region? I suggest to add another bar to Fig. 6 for the simulation that includes OH oxidation. Section 4.2 would flow better if it was moved to the end of section 3.2.

No, the inclusion of OH oxidation does not fix the overestimation. We thought about adding an extra bar to Figure 6, but we think it will complicate the Figure and discussion more than necessary. We also would like to keep the section in the discussion and not in the results.

10. Section 5:
a) Page 15, line 22: Mention the quantitative amount that previous studies have found GFAS to be underestimating fire emissions of CO.
We added these values at the end of the sentence now: "The difference could be accounted for by increasing the GFAS emissions by 500%-1000%, a rather large increase compared to the 200%-600% increase found in the last named studies"

b) Page 15, line 29: Are there any leads on geoengineering meteorology? Also, geoengineering meteorology could have unforeseen consequences for the local or downwind regions, while reducing emissions is likely good overall.
Yes, we agree with this and would definitely not support geo-engineering. As we are no expert on this field, we think it is best to leave the geo-engineering part out in total. We changed the sentence now to: "Meteorology is found to be key driver of the extreme pollution episodes, however, in conjunction with strong CO emissions."

Technical corrections:
I suggest to use consistent date formats throughout the manuscript. For example on the first page there are already several formats being used:"13 October 2017" (page 1, line 1); "11 and 19 November 2017" (page 1, line 6); "11-14 November" (page 1, line 11); "15th of November" (page 1, line 11). The changing of formats can be a bit discontinuous for the reader.
We changed the dates now to be consistent as "11-14 November"

Page 1, Line 1: 2017, measures -> 2017, has measured
Done
Page 1, line 11: The meteorological situation -> Meteorological conditions... were most likely
Done
Page 1, lines 13-14: "...emphasizing the important role of atmospheric dynamics." seems like an unfinished sentence. Expand to explain the role specifically.
We split the sentence and added some extra explanation: "This emphasizes the important role of atmospheric dynamics in determining the air quality conditions at ground-level and in the total column."
Page 2, lines 4-5: Flip the sentence, "Nine out of ten most polluted cities are in the IGP according to the 2018 WHO..." etc.
Done
Page 2, line 4: Define WHO acronym.
Done
Page 2, line 12: Sentence beginning "In addition" is suggested to move to end of paragraph.
Done
Page 2, line 19: practices, these other
Done
Page 2, line 21: extend -> extent
Done
Page 2, line 25: measurement instrument -> measurements
Done
Page 2, line 26: the orbits of scientific -> the scientific
Done
Page 2, line 30: combining -> comparing
Done

Page 2, line 32 to page 3, line 2: Unusual to stop the list of four objectives in the middle with a full stop after objective (2). Suggest to re-write.
*We rewrote the sentences so all objectives are now in one sentence. While doing this we identified an extra objective: "*We assess this according to our five objectives: (1) whether TROPOMI is in accordance with ground-based measurements and (2) how well WRF is able to reproduce these data; how the pollution is dispersing over India (3), the role of meteorology in the accumulation and transport of CO (4), and shedding more light on sources contributing most to the high pollution over the Indo-Gangetic Plain (IGP) of India (5) in support of future pollution mitigation efforts."
Page 3, line 6: section also the role of met... -> section the role of meteorological conditions is also...
Done
Page 3, line 15: Define SICOR acronym.
Done
Page 4, line 6: global resolution -> global horizontal resolution
Done
Page 4, line 20: Remove "ARW" if not used again.
Done
Page 4, line 24: Remove "MYJ" if not used again.
Done
Page 4, line 24: Should "Eta" be capitalized?
No, it usually is written with all small letters in this context
Page 4, line 27: Add citation after RRTM and add ", respectively" to the end of the sentence.
We removed RRTM and placed the citation at that place. We added respectively to the end of the sentence.
Page 6, line 17: As outlined in section 2.1, the...
Done
Page 7, line 6: ...regions (Fig. 2 red and black labels are inner and outer city, respectively).
"regions (Fig. 2 red and black labels indicate inner and outer city, respectively)
Page 7, lines 12-13: ...China was the most polluted...
Done
Page 8, lines 3-4: To determine how unique these high CO values were, we analysed the last four years of CAMS data.
Changed to: "To determine how unique these high CO values were during this time of the year over the IGP, we analysed the last four years of CAMS data."
Page 8, line 8: However, ->These
Done
Page 8, Line 25: Reword the last sentence to say something such as "Atmospheric chemistry is not sufficient to explain differences in CO between the two time periods."
We would like to keep it clear that we did not include the atmospheric chemistry in our WRF base setup. We changed the sentence to: "The simulations in WRF were not including the atmospheric chemistry, but this is probably only playing a minor role (see section 4.3)."
Page 9, line 4: then -> than
Done
Page 11, Figure 8 caption: (b.) total column mixing ratio.
Done
Page 13, line 21: According to Fig. 8, the wind speeds clearly...
Changed to: "As can be seen in Fig. 8, the wind speeds clearly…"

Response to the review of Anonymous Referee #2

Thank you, referee #2 for your review! We improved our paper with your helpful suggestions, as indicated below in blue. Our comments are written in italic, and the improved text is within quotes.

The authors suggest that meteorology is a dominant explanation for the CO episodes. What I found lacking in the argument provided here was in explaining the genesis/formation of the episode. Was there a sudden shift in meteorology (e.g., reduction in wind speeds and PBLH) that coincided with the initiation of the pollution episode? Here, I concur with what Reviewer #1 has suggested, namely regarding a more detailed explanation of why the specific time periods included here were chosen for analysis. If it is possible, showing a longer time period that illustrates the formation of the episode could be helpful.

As can be seen in Fig. 4, total column CO levels over the IGP are rising from the first of October till the 13th of November. For TROPOMI, there are only data available from 11-17 and at 19 November. To have equal amounts of data in two periods of higher and lower CO mixing ratios we chose to have the first 4 days of TROPOMI data in the period of high CO levels and 4 days of lower CO levels 15,16,17, and 19 November. The meteorology also clearly changes around the 15th of November. As can be seen in Figure 9, the wind speed is largely increased from 16 November on, the surface pressure starts to increase from 13 November on, the boundary layer height is high on November 14 and from 18 November on. We described the choice of our periods clearer in the text now: "We averaged over several days of data, concentrating on two periods: 11-14 November 2017, and 15-19 November 2017, in order to obtain a gap free image of Northern India (18 November no TROPOMI data was available). We selected these two periods spanning equal number of days based on the patterns seen in TROPOMI data and the weather conditions (see 4.3)."

To strengthen your argument, you might also appeal more directly to the existing emissions inventories. For example, it would be useful to quantify in a table or figure the share of primary emissions (for this season) across the IGP that are attributable to the various sectors and to crop burning.

We already included Figure 8 that shows the share of the primary emissions over the IGP and in Table 2 we focused on the share of fire emissions therein. In the text: "According to the tracer simulations in our model that are based on MACCity and GFAS emissions, more than 50% of the CO near ground-level in November was caused by residential and commercial combustion. Other main sources are industrial combustion and traffic."

You might consider distinguishing here between fire- and non-fire periods, perhaps with a sensitivity case that allows for the possibility you alluded to that fire CO emissions are underestimated.

The problem with distinguishing between "fire- and non-fire" periods is that we have a limited amount of data available (TROPOMI: 11-19 November) and in the fire data that we have, there are probably lacking fire CO sources (risking to mark fire days as non-fire days).

Other Minor suggestions:

* Units: the manuscript switches frequently between using mass concentration (µg/m3, mg/m3) units and mixing ratio units (ppb) for CO. While I recognize that there are good reasons for using these alternative unit systems, it would be helpful to orient readers by providing an approximate conversion factor and when possible crafting the core narrative of the paper around one unit system).

We stick with the ppb now, except from the introduction on guidelines for CO on ground-level, where we include mg/m3 with a conversion factor.

* Health-based standards: CO is subject to national ambient air quality standards in India. It would be helpful to compare the observed concentrations to Indian guidelines and other relevant values (ie., Chinese, US, EU). I believe that Indian standards may be more stringent than most other countries for CO – unlike Indian standards for PM2.5, which tend to be quite permissive.

This is quite interesting. We weren't aware that this is the case. We looked up the standards to find out that the Indian standards indeed are much more stringent:

The Indian standard is 2 mg/m^3 for 8 hours, 4 mg/m^3 for 1 hour,
China: 10 mg/m^3/hour, 4 mg/m^3 for 1 hour.
EU: 10 mg/m^3 for 8 hours.
US: 10 mg/m^3 for 8 hours, 40 mg/m^3 for 1 hour.
WHO: 10 mg/m^3 for 8 hours, 30 mg/m^3 for 1 hour.
Based on:
https://acm.eionet.europa.eu/reports/docs/ETCACM_TP_2016_10_AAQstandards.pdf

On the other hand, the standard is of course sometimes just a number, if there is no consequence of exceeding the standards (as far as we know, there is not yet any kind of penalty associated with violating these standards in India), a strict standard is not any better than a loose standard.

*We therefor don't think that adding the discussion of the standards in this paper will add something useful, but we added the Indian standards in the introduction: "*At several ground-based measurement stations in the IGP maintained by the Central Pollution Control Board (CPCB, http://cpcb.nic.in/), carbon monoxide (CO) levels amply exceeded the world health organization guidelines (100 mg/m^3 for 15 minutes, 10 mg/m^3 for 8 hour) and the more stringent Indian standards for CO (2 mg/m^3 for 8 hours, 4 mg/m^3 for 1 hour) during several days in November 2017 reaching 15 minute average values up to 400 mg/m^3"

* Section 4.2: the role of adverse meteorology in driving North Indian pollution episodes is widely appreciated in the literature. I would suggest looking at (and citing when appropriate) the following articles:
* Guttikunda and Gurjar, "Role of meteorology in seasonality of air pollution in megacity Delhi, India": doi: 10.1007/s10661-011-2182-8
* Gani et al., "Submicron aerosol composition in the world's most polluted megacity: The Delhi Aerosol Supersite campaign", doi: 10.5194/acp-2018-1066
* Tiwari et al., "Aerosol optical properties and their relationship with meteorological parameters during wintertime in Delhi, India", doi:10.1016/j.atmosres.2014.10.003
* Tiwari et al., "Variability in atmospheric particulates and meteorological effects on their mass concentrations over Delhi, India", doi: 10.1016/j.atmosres.2014.03.027

Thank you for referring to these papers. We added a bit of extra discussion based on these papers and referred to them now:

Introduction: "In addition, post-monsoon meteorological conditions can lead to an accumulation of pollutants in Northern India (Liu et al., 2018; Gani et al., 2018; Tiwari et al., 2015, 2014; Guttikunda and Gurjar, 2011)."

Meteorological conditions: "Several studies have been performed studying this relation for Delhi based on PM (e.g. Gani et al., 2018; Tiwari et al., 2015;Guttikunda and Gurjar, 2011). Guttikunda and Gurjar (2011) for example, found that: "… irrespective of constant emissions over each month, the estimated tracer concentrations are invariably 40% to 80% higher in the winter months (November, December, and January) and 10% to 60% lower in the summer months (May, June, and July), when compared to annual average for that year."

* From the various comparisons made with conditions in China, it is evident that the authors are somewhat surprised by the severity of the pollution problem in North India. Yet the magnitude of the PM2.5 problem in North India is becoming quite well documented, especially in literature focused on remotely sensed PM2.5 levels. The evidence is now quite clear that India has overtaken China for population-weighted PM2.5 levels, while other large countries (Bangladesh, Pakistan, Nigeria, Egypt) can be even more polluted. In my view, the focus on China's pollution in the popular imagination – even when nowhere near the highest in the world – arises in part because of the rapid improvement in the availability of ground-based data there. See for example Shaddick et al, ES&T 2018: https://pubs.acs.org/doi/pdf/10.1021/acs.est.8b02864

Yes, during the writing of this paper, we were also more and more convinced of that. But China still has a worse reputation than India indeed, that is why we also try to emphasize the problem in India in our paper.

* A discussion of data quality and measurement uncertainty for the in-situ CPCB pollution measurements would be appropriate. Do the surface measurements appear to be generally well calibrated and reliable?

We could not get any information on the data quality unfortunately

What caused the extreme CO concentrations during the 2017 high pollution episode in India?

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Abstract. The TROPOspheric Monitoring Instrument (TROPOMI), launched 13 October 2017, ~~measures~~ has measured carbon monoxide (CO) concentrations in the Earth's atmosphere since early November 2017. In the first measurements, TROPOMI was able to measure CO concentrations of the high pollution event in India of November 2017. In this paper we studied the extent of the pollution in India, comparing the TROPOMI CO with modelled data from the Weather Research and Forecast model (WRF) to identify the most important sources contributing to the high pollution, both at ground-level and in the total column. We investigated the period ~~between~~ 11 ~~and~~ - 19 November 2017. We found that residential and commercial combustion was a much more important source of CO pollution than the post-monsoon crop burning during this period, which is in contrast to what media suggested and some studies on aerosol emissions found. Also, the high pollution was not limited to Delhi and its direct neighbourhood but the accumulation of pollution extended over the whole Indo-Gangetic Plain (IGP) due to the unfavourable weather conditions in combination with extensive emissions. From the TROPOMI data and WRF simulations, we observed a build-up of CO during 11-14 November and a decline in CO after ~~the~~ 15 th ~~of~~ November. The meteorological ~~situation~~conditions, characterized by low wind speeds and shallow atmospheric boundary layers, ~~was~~ were most likely the primary explanation for the temporal accumulation and subsequent dispersion of regionally emitted CO in the atmosphere~~, emphasizing~~ . This emphasizes the important role of atmospheric dynamics in determining the air quality conditions at ground-level and in the total column. Due to its rapidly growing population and economy, India is expected to encounter similar pollution events more often in future post-monsoon and winter seasons unless significant policy measures are taken to reduce residential and commercial emissions.

1 Introduction

During November 2017, India encountered an extreme pollution episode. Various ground-level measurement stations reported Air Quality Index (AQI, http://aqicn.org/) values of 999, i.e. far above the standard scale that is limited to 500. These high AQIs were caused by high concentrations of several pollutants, but most importantly particulate matter (PM_{10} and $PM_{2.5}$)

with reported values of >700 μg/m^3 $PM_{2.5}$, and carbon monoxide with values of up to 10 mg/m^3 several days in a row. Most of this pollution was found over the Indian part of the Indo-Gangetic Plain (hereafter called IGP), a highly populated region in the North of India near the Himalayas, including the cities of Delhi, Agra, Kanpur, Lucknow, Patna and Kolkata. Heavy air pollution is an annual recurring problem in this region, especially during the post-monsoon and winter months (Cusworth et al., 2018; Vadrevu et al., 2011; Girach and Nair, 2014). ~~In the~~ Nine out of ten most polluted cities were located in the IGP, according to the 2018 ~~WHO~~ World Health Organisation (WHO) list of most polluted cities in the world which is based on fine particulate matter ($PM_{2.5}$) ~~, based on~~ from the year 2016 ~~data, nine out of ten cities were located in the IGP~~ (Bhattacharya, 2016; WHO, 2018). At several ground-based measurement stations in the IGP maintained by the Central Pollution Control Board (CPCB, http://cpcb.nic.in/), carbon monoxide (CO) levels amply exceeded the world health organization guidelines (100 mg/m^3 for 15 minutes, 10 mg/m^3 for 8 hour) and the more stringent Indian standards for CO (2 mg/m^3 for 8 hours, 4 mg/m^3 for 1 hour) during several days in November 2017 reaching 15 minute average values up to 400 mg/m^3 (1 mg/m^3 is roughly equal to 870 ppb at ground level~~).~~ , we will use ppb from here on). The total population in the IGP region (including parts of Pakistan, Bangladesh and Nepal) exceeds 400 million and is growing. This means that the bad air quality is effecting hundreds of millions of people for a large part of the year. This makes it very important to investigate the origin and transport of pollution in the area. Due to its lifetime of several weeks, CO can also be used as a proxy of other, co-emitted, anthropogenic pollution.

Several explanations have been proposed for the high pollution levels in this period, but the exact cause is still unclear. Agriculture is very important in the IGP, with the post-monsoon burning of crop residues taking place in October and November. ~~In addition, post-monsoon meteorological conditions can lead to an accumulation of pollutants in Northern India (Liu et al., 2018).~~ CO emissions from fires during this period are estimated, for example, by the Global Fire Assimilation System (GFAS) and the Global Fire Emission Database (GFED). ~~However, there are strong signs that these datasets do not capture all of the biomass burning emissions (Mota and Wooster, 2018; Cusworth et al., 2018; Huijnen et al., 2016). Also, the conversion of fire occurrence to CO emissions depends on factors such as combustion efficiency, biome type, and soil characteristics, which adds uncertainty to the emission estimates (Werf van der et al., 2010).~~ Other anthropogenic sources of CO, e.g., traffic and heating systems are very high in the highly populated Indo-Gangetic plains, especially during the colder post-monsoon and winter months. Thus, apart from fire emissions from post-harvest burning practices, ~~also~~ these other anthropogenic sources might be an important factor explaining the high CO pollution. In addition, post-monsoon meteorological conditions can lead to an accumulation of pollutants in Northern India (Liu et al., 2018; Gani et al., 2018; Tiwari et al., 2015, 2014; Guttikunda and Gurjar, 2011).

Satellite data can play an important role to obtain more insight in the origin and ~~extend~~ extent of pollution, providing information on the distribution of pollutants over large regions on a daily basis. In October 2017 the TROPOspheric Monitoring Instrument (TROPOMI) was launched, measuring various trace gases, including CO, with unprecedented high spatial and temporal resolution (Landgraf et al., 2016). TROPOMI was still in its commissioning phase in November and algorithm tests and calibrations were ongoing. Fortunately, the first calibration results were positive and proved the high quality of the ~~measurement instrument~~ measurements and high signal to noise ratio (Borsdorff et al., 2018a, b), confirming the usefulness

of the ~~orbits of~~ scientific data that were collected. TROPOMI observed very high column mixing ratios over the Northern part of India from 11-19 November, in accordance with the ground-based data. The Copernicus Atmosphere Monitoring Service (CAMS, see section 2.2) data showed similar enhancements in CO columns during this period, further corroborating the TROPOMI retrieved CO variations (Borsdorff et al., 2018a).

In this study, we examine the high CO pollution episode of November 2017, focussing on 11-19 November, in more detail. We do this by ~~combining~~ comparing the daily CO observations over India from TROPOMI with ground-based measurements and simulated CO mixing ratios from the Weather Research and Forecast (WRF) model. We assess this according to our ~~four~~ five objectives: (1) whether TROPOMI is in accordance with ground-based measurements and (2) how well WRF is able to reproduce these data~~. We investigate the dispersion of pollution over India to~~ ; how the pollution is dispersing over India (3)~~shed~~ , the role of meteorology in the accumulation and transport of CO (4), and shedding more light on sources contributing most to the high pollution over the Indo-Gangetic Plain (IGP) of India (5) in support of future pollution mitigation efforts. ~~With WRF we (4) also study the role of meteorology in the accumulation and spreading of CO.~~

The data and methods section describes the datasets that are used and the setup of the WRF model. In the results section, CO levels measured by TROPOMI over Southeast Asia and by ground-level pollution measurement stations are compared with WRF data. The model is used also to attribute the high total column average mixing ratios over India to specific emission categories as presented in section 4. In this section ~~also~~ the role of meteorological conditions is also discussed as well as the results of sensitivity tests on CO chemistry in the model.

2 Data and methods

2.1 TROPOMI

TROPOMI has a shortwave infrared spectrometer module, from which the total column average mixing ratio (XCO) is retrieved using the measured radiance around 2.3 μm. Due to its high spatial and temporal resolution, TROPOMI is able to observe global CO vertical columns on a daily basis (Landgraf et al., 2016).

We used data from 14 orbits of TROPOMI retrieved ~~between~~ 11 ~~and~~ - 19 November 2017 that covered the Northern part of India. The instrument reached near-nominal temperatures on 11 November, which is considered the first day of reliable data from TROPOMI (Borsdorff et al., 2018a). As in the study of Borsdorff et al. (2018a) on the first TROPOMI CO results, we used XCO values that were retrieved using the operational algorithm SICOR (Shortwave Infrared CO Retrieval, Landgraf et al., 2016). TROPOMI data were filtered for clear sky observations, and cloudy sky observations with a cloud top height < 5000 m and an aerosol optical thickness >0.5. Borsdorff et al. (2018c) found that including low-level cloud data increased the amount of available measurements, while hardly affecting the ability to measure relatively small scale sources by applying the SICOR algorithm to data from the SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY). This retrieval method allowed to include some measurements over sea with low-level clouds (Borsdorff et al., 2018c).

We removed the two most westward pixels of every swath, which suffer from a not yet resolved performance issue ~~(Borsdorff et al. , 2018a).~~ (Borsdorff et al., 2018a). The first validation study showed that the TROPOMI data is in good agreement with

CAMS data with a global mean difference of +3.2% and a Pearson correlation coefficient of 0.97 (Borsdorff et al., 2018b). For the India region, a 2.9% difference was found with CAMS with a standard deviation of 6% and a Pearson correlation coefficient of 0.9 (Borsdorff et al., 2018a). Moreover, only a small mean bias of 6 ppb, with a standard deviation of 3.9 and 2.4 ppb for respectively clear and cloudy skies has been found compared to ground-based total column measurements of TCCON (Total Carbon Column Observing Network). The signal-to-noise ratio of TROPOMI is high compared to previous satellite instruments retrieving CO (Borsdorff et al., 2018a).

The TROPOMI averaging kernel (AK) provides information on the vertical sensitivity of the satellite instrument for each single retrieved CO column (Borsdorff et al., 2014). The relationship between the reported CO vertical profile (C_{retr}) and the true CO profile (C_{true}) is given by Eq. ~~1.~~ 1. In this equation C_{retr} is the retrieved CO profile and C_{prior} the a priori CO profile. According to Borsdorff et al. (2014):

$$C_{retr}\boldsymbol{C_{retr}} = C_{prior}\boldsymbol{C_{prior}} + \boldsymbol{AK}(C_{true} - C_{prior}\boldsymbol{C_{true}} - \boldsymbol{C_{prior}}) + \boldsymbol{e}_x \quad (1)$$

where e_x represents the error on the retrieved trace gas profile. The Equation simplifies to Eq. 2:

$$\boldsymbol{C_{retr}} = \boldsymbol{AK}(\boldsymbol{C_{true}}) + \boldsymbol{e}_x \quad (2)$$

when the effective null space contribution of the a priori profile is eliminated, which is the true for the chosen regularisation parameter for the TROPOMI CO data, as is explained in Borsdorff et al. (2014). In this study, we compare the CO columns from TROPOMI, derived from C_{retr}, with the modelled columns from WRF. To make a fair comparison between the TROPOMI CO columns and the modeled CO columns, the AK has been applied in the same way to the modeled CO vertical profile (Eq. ~~1~~2), by replacing C_{true} with the modelled profiles and ignoring the error.

2.2 CAMS

The Copernicus Atmosphere Monitoring Service (CAMS, https://atmosphere.copernicus.eu) provides data on air quality in 6-hourly time intervals at a global horizontal resolution of 0.25°x0.25°. The CAMS CO reanalysis product is derived from the output of a 4-dimensional variational (4D-Var) data assimilation system, based on ECMWF (European Centre for Medium range Weather Forecast) numerical weather prediction reanalysis data. It uses MACCity anthropogenic emissions, which combines information from the European Union MACC (Monitoring Atmospheric Composition and Climate) and CityZen (megacity Zoom for the Environment emission database, Granier et al., 2011) inventories. For biomass burning the GFAS fire emission inventory is used, which is based on MODIS fire counts and is provided at a 0.1°x0.1°resolution. The CAMS model is constrained by CO satellite observations from the Measurements of Pollution in the Troposphere (MOPITT) and the Infrared Atmospheric Sounding Interferometer (IASI) satellite instruments. Constraining the model with satellite observations provides a relatively good estimate of the actual XCO over the globe. Biases are found to be within $\pm$10% with respect to ~~satellite and~~ TCCON observations according to the latest validation report (KNMI, 2018), with data delivery lagging behind real-time by about one week. In this research we used the CAMS CO reanalysis products at ~~various~~ 25 pressure levels and the total column product (http://apps.ecmwf.int/datasets/data/cams-nrealtime/levtype=sfc/). September-January total column CAMS data for the years 2012-2017 is used in section 3.1.

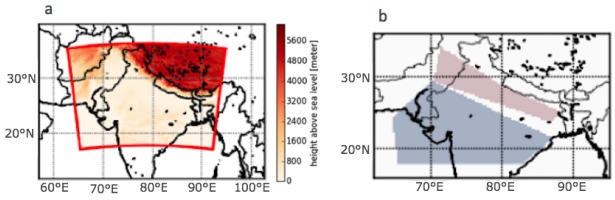

Figure 1. (a) WRF domain over India, the colours depict the height above sea level (m), showing the Himalaya mountain range. (b) Within the WRF domain: the area for averaging over the IGP (pink) and for averaging over Non-IGP India (blue).

2.3 WRF

To model XCO and ground concentrations at high spatial resolution we used WRF version 3.8.1 (http://www.wrf-model.org/) with the Advanced Research WRF core ~~(ARW)~~. WRF is a numerical non-hydrostatic model developed at the National Centers for Environmental Prediction (NCEP). It has several choices of physical parameterizations, allowing application of the model to a large range of spatial scales (Grell et al., 2005). Our model domain of 2900 km by 2010 km is over the northern part of India and parts of Pakistan, Nepal, China and Bangladesh, including parts of the Himalaya mountain range (see Fig. 1a). Our model employed a 10x10 km^2 resolution and 29 vertical eta levels, and used the Mellor-Yamada-Janjic ~~(MYJ)~~ planetary boundary scheme (Janjic, 1994), the Unified Noah land surface model for surface physics (Ek et al., 2003; Tewari et al., 2004), and the Dudhia scheme (Dudhia, 1989) and the Rapid Radiative Transfer Method ~~(RRTM)~~ (Mlawer et al., 1997) for short-wave and long-wave radiation ~~(Mlawer et al., 1997)~~respectively. Cloud physics are solved with the Grell-Freitas cumulus physics ensemble scheme (Grell and Freitas, 2014).

Our boundary and input meteorological conditions, on 6-hourly basis, were based on ECMWF reanalysis data, similar to the CAMS model. WRF calculates its own meteorology in between these 6-hourly time steps and nudges towards the meteorological boundary conditions every 6 hours. The boundary conditions for CO ~~were~~ came from the CAMS CO October and November data on pressure levels, interpolated to the WRF model levels.

Different CO emission inventories are available for Southern Asia. As in CAMS, we used MACCity anthropogenic CO emissions for the year 2017 at a resolution of 0.5°x0.5°(Lamarque et al., 2010). ~~WRF-Chem simulations are performed for eight~~ We implemented nine different CO tracers representing the MACCity emission categories ~~and one representing GFAS biomass burning~~ in WRF-Chem (see Table 1). The MACCity database estimates worldwide monthly emission strengths for ~~nine different~~ these emission categories. An additional tracer was used to account for CO transported from the CAMS derived boundary conditions: we refer to this CO tracer as background in this paper (Table 1).

For biomass burning emissions, we used GFAS data with a resolution of 0.1°x0.1°(available for download from: http://apps.ecmwf.int/dat
gfas/). ~~An additional tracer was used to account for CO transported from the CAMS derived boundary conditions: we refer to this CO tracer as background in this paper.~~ However, there are strong signs that fire emission inventory datasets, such as GFAS and GFED, do not capture all of the biomass burning emissions (Mota and Wooster, 2018; Cusworth et al., 2018; Huijnen et al., 2016). Also, the conversion of fire occurrence to CO emissions depends on factors such as combustion efficiency, biome type, and

Table 1. Overview of the different tracers implemented in WRF.

Tracer (short name)	Explanation	Source
en_prod	Energy production and distribution	MACCity
res_com	Residential and commercial combustion	MACCity
agr_waste	Agricultural waste burning	MACCity
ind_proc	Industrial processes and combustion	MACCity
agr_prod	Agricultural production	MACCity
solv_proc	Solvent production	MACCity
land_transp	Land transport	MACCity
mar_transp	Maritime transport	MACCity
waste_treat	Waste treatment and disposal	MACCity
COgfas	Biomass burning	GFAS
COobg	Boundary condition, referred to as *background*	CAMS

soil characteristics, which adds uncertainty to the emission estimates (Werf van der et al., 2010). For PM, it is estimated that the GFAS emissions are underestimated approximately a factor 2-6 (Mota and Wooster, 2018; Cusworth et al., 2018; Huijnen et al., 2016). For CO, less information is available but since its emissions are linked to that of other compounds, similar underestimates are expected.

In our base setup, we ran the model for the period 1 October - 20 November, including the tracers listed in Table 1, so including the original GFAS emissions and the background. In this paper, results are shown for 11-20 November, October is considered as a spin-up period. Besides the base run, we did some extra simulations including 0 (referred to as: MACCity), 1 (base setup), 5 (+5GFAS) and 10 (+10GFAS) times the original GFAS emissions, based on the estimated underestimation of fire PM emissions.

Deposition and chemical production from Volatile Organic Compounds (VOCs) are not included in our base setup. The deposition process is slow compared to transport of CO out of the model domain, and direct CO sources over the highly populated (IGP) of Northern India are much larger than the indirect source from VOC oxidation.

However, in a sensitivity simulation (see Section 4.3) we accounted for the chemical reaction between the Hydroxyl radical (OH) and CO using the JPL recommended temperature and pressure dependent reaction rate (Burkholder et al., 2015). Carbon monoxide production from the oxidation of methane and other VOCs are included in this simulation as well. In this chemistry simulation, we used the CO production from the TM5-4DVAR system (Krol et al., 2013) and the corresponding OH climatology, based on Spivakovsky et al. (2000)~~and~~ , scaled by 0.92 (Huijnen et al., 2016, 2010; Krol et al., 2013).

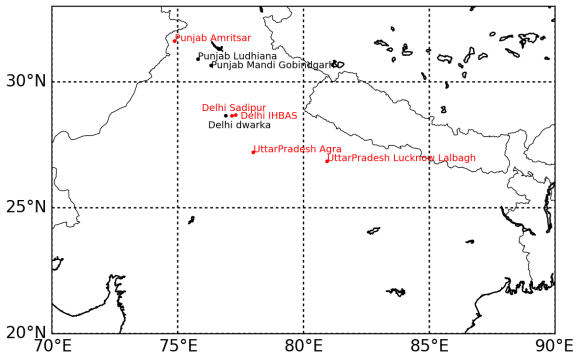

Figure 2. Locations of ground-level measurement stations, red dots and labels are inner city stations, black dots and labels outer city locations.~~PUN = Punjab: AMR = Amritsar, LUD = Ludhiana, MAN = Mandi Gobindargh; DI = Delhi: SADI = Sadipur, IBS = IHBAS, DWAR = Dwarka; UP = Uttar Pradesh: AGRA = Agra, LULA = Lucknow Lalbagh~~

2.4 Ground-level measurements

The central pollution control board (CPCB) of India measures the air quality at several stations in India (http://cpcb.nic.in/automatic-monitoring-data/). All the samples are taken at ground-level and are made available as fifteen minute averages. We only used stations here with CO measurements available ~~between the~~ for 15 th ~~of October and the~~ October - 20 th ~~of~~ November. To obtain measurements representative of the urban background, we excluded stations near large roads showing large CO enhancements. This selection is needed for a meaningful comparison to WRF simulations at 10x10 km^2 using MACCity emissions at only 0.5°x0.5°resolution. In Fig. 2 all stations used for comparison with WRF are listed.

2.5 Comparing WRF with TROPOMI and ground-level measurements

As outlined ~~before (~~in section 2.1~~)~~, the averaging kernel was applied to the WRF data using Eq. ~~1.~~ 2. Both WRF and TROPOMI data were averaged on a 0.25°x0.25°grid to make the comparison less sensitive to local outliers in the data. We ~~also~~ averaged over several days of data, concentrating on two periods: 11-14 November 2017, and 15-19 November 2017, in order to obtain a gap free image of Northern India (18 November no TROPOMI data was available). We selected these two periods spanning equal number of days based on the patterns seen in TROPOMI data ~~, the amount of data per day,~~ and the weather conditions (see 4.3). In the first period, TROPOMI data show high CO pollution over the whole IGP. The second period shows lower XCO, due to changing weather conditions (see Section 4.2). In some comparisons, results are averaged over two regions of India: the IGP and the area south of the IGP: non-IGP, as defined in Fig. 1b.

We divided the ground-level measurement stations in India into two groups: one group consisted of stations directly in the city and the other group of stations was at the city edge in surrounding rural background regions (Fig. 2 ~~respectively~~ red

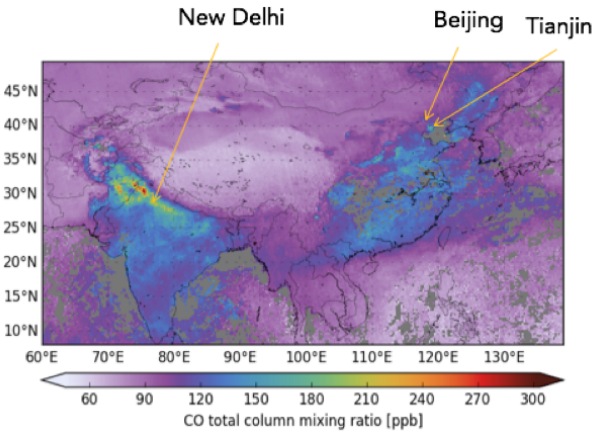

Figure 3. XCO over India and China as observed by TROPOMI, 13 November 2017, comparing New Delhi with large CO emitting cities in China, Beijing and Tianjin.

and black labels indicate inner and outer city, respectively). This distinction was used to investigate differences in the source signature of CO inside and outside of cities.

3 Results

3.1 TROPOMI and CAMS over South-East Asia

In some of the first TROPOMI observations collected in the first half of November 2017, the northern part of India, more specifically, the IGP, stood out by its high XCO values (see Fig. 3). XCO values were even significantly higher over the IGP than over any region of South-East Asia, even higher than over China. This is remarkable, since in earlier studies, China ~~used to be~~ was the most polluted region of the world (e.g., Baldasano et al., 2003; Kan et al., 2012). On the other hand, China has recently been active in reducing air polluting emissions, including CO (Zheng et al., 2018), while in India, emissions continued to increase over the past years (Krotkov et al., 2016).

It was estimated that China reduced its CO emissions by 23% between 2013 and 2017 (Zheng et al., 2018). India only took its first steps to improve the air quality in December 2017 by implementing the National Clean Air Program (NCAP), i.e., after the high pollution event studied in this paper. This makes it plausible that the New Delhi region was more polluted in this period than any region over China. NCAP does not yet include strict targets for emission reductions and rather focusses on setting up an effective ambient air quality monitoring network and making plans for prevention, control and abatement of air pollution (Ganguly, 2018; Ministry of Environment, Forest and Climate Change (MoEF&CC), 2018).

~~However, how unique were~~ To determine how unique these high CO values were during this time of the year ~~in India~~ over the IGP~~? We~~ , we analysed the last four years of ~~Copernicus Assimilation and Monitoring Service (CAMS) datato investigate this. These~~ CAMS data. The results confirm that the high pollution episode of November 2017 was exceptionally long with

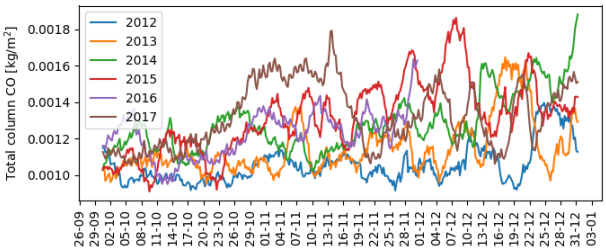

Figure 4. CAMS CO columns (kg/m^2) from October to December (2012 - 2017) averaged over the IGP domain (see Fig. 1)

more than two weeks of CO column amounts exceeding 0.0015 mg/m^2. However, according to the CAMS model, CO columns reached short-term values that were higher during December 2014 and 2015 than in November 2017 (Fig. 4). The high CO columns of November 2017 are therefore not unique for this part of India. ~~However,~~ These high pollution episodes during the post-monsoon period occur more frequently in recent years (Fig. 4). As long as emissions are not reduced, India will probably encounter such events more often in future post-monsoon and winter seasons.

3.2 Comparing WRF to TROPOMI

3.2.1 Agreement between WRF and TROPOMI

We compared our WRF results with the TROPOMI data, and found that WRF could reasonably well reproduce the high XCO values spread over the whole IGP during 11-14 November 2017 and the lower XCO values during 15-19 November. Fig. 5 shows that both modelled and TROPOMI retrieved XCO are very high in the north-west of India (Fig. 5, panels a, c, and e). The highest spatial correlation between WRF (including standard GFAS emissions) and TROPOMI is found on November 11, 12, and 13 (r=0.87, 0.88, 0.88 respectively). On November 14 and 15 poorer correlations of 0.78 and 0.76 are found, respectively. The model captures the transition between the two periods slightly different from the observations, with TROPOMI showing ventilation of CO to the south-east earlier than WRF. The spatial correlation went up again to 0.81 for 16 and 17 November and 0.85 for 19 November. The WRF simulation underestimates XCO during the 11-14 November period over the IGP (Fig. 5a). Adding either 20% extra MACCity emissions or adding substantial amounts of GFAS fire emissions: between 500% (Fig. 5e) and 1000% of regular GFAS emission, gave XCO values that are more similar to the TROPOMI values without notable changes in the spatial patterns over India. In all cases, WRF overestimated the CO levels at the border of Pakistan south of the IGP. For 15-19 November, however, the simulations with MACCity and standard GFAS emissions already overestimate the XCO measured by TROPOMI (Fig. 5b). This might have to do with a deviation in the changing meteorological conditions in WRF. ~~Not~~ The simulations in WRF were not including the atmospheric chemistry, but this is probably only playing a minor role (see section 4.3).

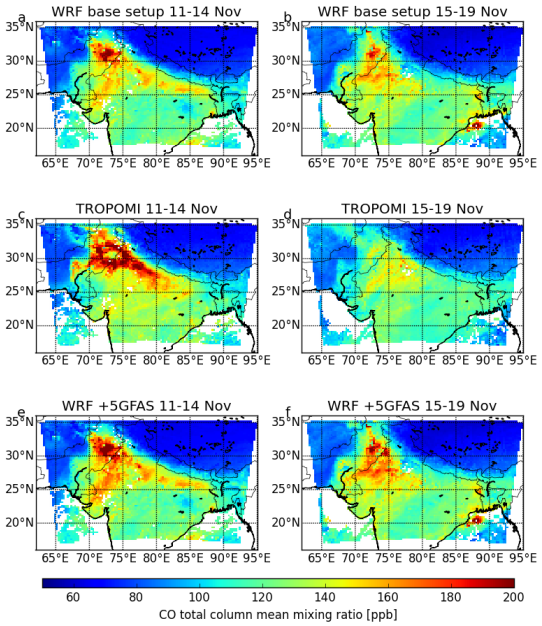

Figure 5. (a.) WRF simulated total columns with MACCity emissions, with 1x GFAS emissions, 11-14 Nov, (b.) same as a., but for 15-19 Nov, (c.) TROPOMI total columns 11-14 November, (d.) same as c. but for 15-19 November (e.) WRF simulated total columns with MACCity emissions including 5x GFAS fire emissions 11-14 November, (f.) same as e. but for 15-19 November.

3.3 ~~CO columns over and outside of the Indo-Gangetic Plain~~

3.2.1 CO columns over and outside of the Indo-Gangetic Plain

The XCO levels measured by TROPOMI and modelled by WRF are clearly enhanced during 11-14 of November over the IGP compared to more southerly regions of India (Non-IGP, Fig. 6). The IGP CO total columns are on average 30 ppb higher than ~~then~~ over non-IGP regions (see Fig. 1b for areas of IGP and non-IGP). When we average over 15-19 November, this difference between the IGP and the non-IGP mostly disappears; the column average XCO over the Indo-Gangetic Plains is now lowered from 162 to 129 ppb for TROPOMI and from 152 ppb to 124 ppb for WRF, while the non-IGP XCO only slightly decreased for TROPOMI (124 to 118 ppb) and remained nearly equal at 129 ppb for WRF. A WRF simulation based on MACCity without GFAS (green bars), shows the same XCO pattern. Since the emissions of MACCity are not changing day-by-day, the difference between the periods is solely caused by different meteorological conditions (see also section 4.2).

3.3 Comparing WRF to ground-level measurements

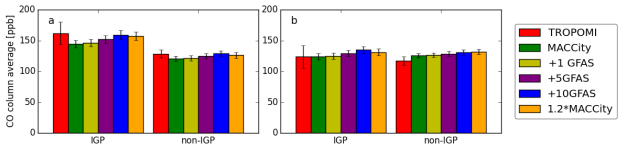

Figure 6. CO averaged over Indo-Gangetic Plains and non-IGP area. Panel (a.): for the period 11-14 Nov, panel (b.): for 15-19 November. The 1σ error bars denote the spread over the different days in the averaging period.

~~The~~ CO concentrations at ~~all~~ the ground-level measurement stations that are used are ~~enhanced between the 3rd of Novemberand the 13th of November~~generally increasing until 14 November, compared to earlier measurements. This is in accordance with the total column CO levels seen in the 2017 timeseries from CAMS. This is in accordance with the total column CO levels seen in the 2017 timeseries from CAMS. The CO diurnal cycles of the observations and the simulation show clear night-time accumulation in the stable nocturnal boundary layer, which vanishes during day-time with increasing boundary layer depth. The CO concentrations generally reach lower levels after ~~the~~ 16 th ~~of~~ November (Fig. 7). The WRF model largely follows the CO enhancement and reduction pattern, although the diurnal cycle seems delayed by 3 hours compared to the ground-based measurements. This might be due to the hourly time profiles that were used for the emissions, which were derived for Europe (van der Gon et al., 2011), but do account for the local time shift. In Fig. 7, we zoom in on November 11-20, the days for which also TROPOMI data are available. Averaged time series are shown of the measurements collected at stations in the provinces of Delhi, Punjab, and Uttar Pradesh and the corresponding averaged WRF concentrations. The stations inside the cities (Fig. 7a) show a clear reduction in mixing ratio during the latter half of this period (1050 ppb, 15-19 November), compared to the first half (1700 ppb, 11-14 Nov). The observed reduction, which we observed also in TROPOMI XCO, is reproduced by WRF (1400 to 880 ppb). At locations outside cities (Fig. 7b) this pattern is less pronounced, both in WRF and in the measurements. WRF largely follows the measured CO mixing ratios, but slightly underestimates the CO values after the 16 th of November. WRF shows enhanced XCO during ~~the~~ 15 th and 16 th ~~of~~ November, which is not observed.

To further investigate the origin of the XCO variations, the contribution of different emission categories in WRF is shown in Fig. 8. We show here the inner-city stations, as these are the areas were most people live, but the picture is not very different for outer-city stations (see also Table 2, section 4.1). As can be seen, the surface concentrations are much less sensitive to the background CO (black) compared to the total column mixing ratios. On all days the category *residential and commercial combustion* contributes most to the total CO concentration (on average 67% for ground-level and 35% for the total column including the background). Other large contributors are *industrial processes and combustion* and *traffic*. Surprisingly, we find a rather small contribution from fires to the total mixing ratio of 1-2% in our simulation with MACCity and standard GFAS emissions (see section 4.1, Table 2). Even with strongly enhanced GFAS emissions, the contribution remains on average within 20%. The larger XCO measured at inner-city stations -compared to the stations outside the cities- also point to large contributions from urban emissions.

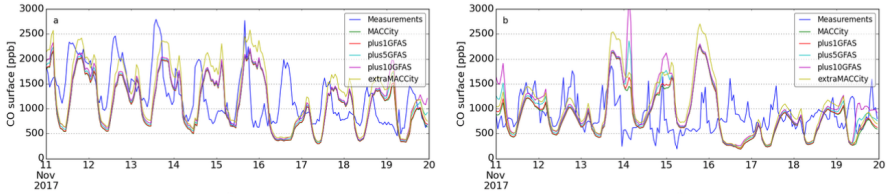

Figure 7. (a.)Ground-level measurements and WRF model averaged CO over *inner-city* stations (~~mg/m^3~~ ppb, see Fig. 2), time is in UTC (b.) Ground-level measurements and WRF model averaged over stations outside cities (~~mg/m^3~~ ppb, see Fig. 2)

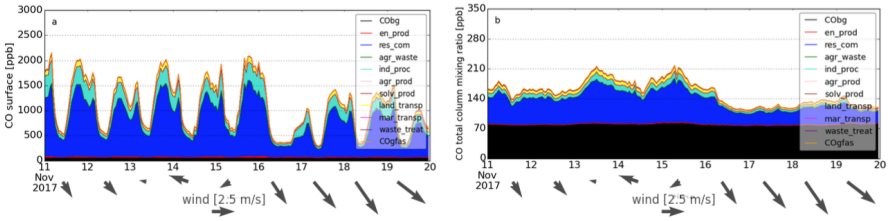

Figure 8. Average inner-city source contribution to (a.): ground-level, ~~time is in UTC~~ (b.): ~~inner-city~~ total column mixing ratio, time is in UTC. Below the mixing ratios, the 10-meter wind speed and direction are depicted, longer arrows ~~are~~ indicate higher windspeeds (see legend)

4 Discussion

We found XCO values of over 200 ppb in substantial parts of Northern India, in both the TROPOMI and model simulations. From the satellite data and total column WRF mixing ratios, it is clear that CO is not only enhanced directly around Delhi, but over the whole Indo-Gangetic Plain, with very high values west of Delhi.

~~Some ground stations were excluded because of the proximity to large CO sources, leading to very high observed CO concentrations. At the station of Harayana Rohtak, north-west of Delhi, the measured CO concentration was on average of 20 mg/m^3 from 11-14 November, but a value as high as 402 mg/m^3 was reached during a short peak. Our model could not resolve these high CO values, and also other nearby measurement stations did not record these high values. They are therefore very likely caused by a local source that cannot be reproduced at the resolution of WRF. For this reason, also several other locations were excluded, where short-term CO peaks of several tens of milligrams per cubic meter were measured (stations: Bihar Gaya, Bihar Patna, Madya Pradesh Dewas, Madya Pradesh Ujjain, Rajasthan Jaipur Adarsh Nagar, West Bengal Durgapur). Although these measured enhancements are likely traffic related, it is well possible that some of these enhancements are caused by local biomass burning. Overall, however, our results point to a relatively small role of biomass burning in the enhanced CO concentrations over the largest part~~ This means that not only the people living in Delhi, but also a large part of the hundreds of millions of people inhabiting the rest of the IGP ~~, as shown in Fig. 8 for inner-city stations. A similar contribution of biomass burning is found out of the city (not shown)~~are affected by the bad air quality.

4.1 Contribution of different sources

According to the tracer simulations in our model that are based on MACCity and GFAS emissions, more than 50% of the CO near ground-level in November 2017 was caused by residential and commercial combustion. Other main contributors are industrial combustion and traffic. For the total column mixing ratios the background CO, entering from the boundaries of the domain, was clearly more important and responsible for around a 30%-50% of the total column mixing ratio (see Fig. 4). The background CO is however rather constant, and day-by-day variations in XCO are caused by residential and commercial combustion, similar to what we observed at ground-level. The CO emission contribution from fires has certainly a more episodic nature than the anthropogenic sources in MACCity. In this section, we do not only include the average contribution, but also consider the maximum contribution of fire emissions to investigate short-term variations in the contribution of fires to CO.

At the measurements stations that we considered, except for Punjab at ground-level, only a minor contribution from fire was found of 1% to 2%, both for total column and ground-level CO (see Table 2). The steadily increase in CO levels started before November 11; to make sure we did not miss part of the biomass burning contribution, we included in Table 2 also a longer period of the post-monsoon season: 1 October - 19 November. At ground-level in Punjab the average and maximum contributions were respectively 6% and 44%, ~~respectively~~ over the whole modelled period of 1 October ~~to~~ - 19 November. In the 11-19 November period, the maximum contribution of biomass burning to the ground-level contribution there was 23%, with an average of 2% (see Table 2).

Some ground stations were excluded because of the proximity to large CO sources, leading to very high observed CO concentrations. Although these measured enhancements are likely traffic related, it is possible that some of the enhancements are caused by local biomass burning. Overall, however, our results point to a relatively small role of biomass burning in the enhanced CO concentrations over the largest part of the IGP, as shown in Fig. 8 for inner-city stations. A similar contribution of biomass burning is found out of the city (not shown).

There are strong indications, however, that GFAS might severely underestimate the fire emissions (Mota and Wooster, 2018). Cusworth et al. (2018) concluded in their recent paper on biomass burning in India that the resolution of the MODIS satellite instrument, on which GFAS fire emissions are partly based, misses many small fires. In addition, thick smoke from fires might lead to an underestimation of fire emissions from GFAS, as MODIS might identify these as clouds, as was found in a recent study over Indonesia (Huijnen et al., 2016). The results of increasing the fire emissions by a factor 5 - 10 in WRF are shown in Figs. 5 and 6. Adding biomass-burning emissions in the WRF simulation does not lead to a higher spatial correlation between WRF and TROPOMI but CO levels get closer to TROPOMI values in the 11-14 November period, so it might be that the GFAS fire emissions were indeed underestimated in this period. Or a substantial amount of fires is missed in GFAS in this period. However, the mixing ratios during the 15-19 November period are overestimated with respect to TROPOMI when higher GFAS emissions are assumed (see Figs 5,6). Alternatively, MACCity already explains a very large part of the observed CO levels, and increasing the MACCity emissions by 20% gives rather comparable results to increasing the GFAS fire emissions by a factor 5-10 for the total columns (Fig. 6). Biomass burning would still be a minor contributor to the average CO levels even if the emissions are enhanced by these factors.

Table 2. Contribution of standard GFAS biomass burning to the total CO levels per region at ground level (GL) and in the total column (TC) between 11-19 November 2017.

Period		Delhi GL	Delhi TC	Punjab GL	Punjab TC	Uttar Pradesh GL	Uttar Pradesh TC	Inner city GL	Outer city GL
11-19 Nov	maximum	12%	11%	23%	10%	3%	3%	14%	23%
	average	1%	1%	2%	1%	1%	1%	1%	2%
1 Oct - 19 Nov	maximum	17%	15%	44%	16%	15%	11%	40%	44%
	average	2%	2%	6%	1%	1%	1%	2%	5%

For Delhi and Uttar Pradesh the contributions of fire emissions to the total CO levels are on average minor, even if the GFAS emissions are increased. For Punjab, we find that fire emissions might have contributed significantly to the ground-level concentrations for a few days in the 1 October ~~to~~ - 19 November period. From the total column, and other stations, however, we conclude that MACCity already explains a very large part of the observed XCO and ground-based CO levels, and fire emissions can only have played a very minor role.

In this paper, however, we assume that the emissions of MACCity do not grossly overestimate CO emissions over the IGP. Compared to TROPOMI and the amount of emissions that might come from fires based on GFAS and GFED, this assumption seems legitimate. When comparing the total emissions of MACCity to the EdgarV4.3.1 emission database of the most recent year 2010, however, EdgarV4.3.1 gives for November a circa 20% lower emission estimate, when taking emission factors of van der Gon et al. (2011) to convert from yearly emission to monthly emission. This gives more space to add extra emissions of GFAS, although we should keep in mind that in that case we have to increase the emissions of GFAS even more radically, in the order of >30x the original emissions.

4.2 Meteorological conditions

In general, the post-monsoon and winter season are the seasons with the worst air quality in the IGP. The photochemical loss is low and other meteorological variables, e.g., the absence of rain and low wind speeds, contribute to high levels of pollution. Several studies have been performed studying this relation for Delhi based on PM (e.g. Gani et al., 2018; Tiwari et al., 2015; Guttikunda and Guttikunda and Gurjar (2011) for example, found that even with constant emissions over each month, the estimated tracer concentrations were 40% - 80% higher in November, December, and January and 10% to 60% lower in May, June, and July, compared to the annual average.

For November 2017 we identified meteorological conditions as the most important reason why the CO mixing ratios at ground-level and in the total column increased as observed. Although not extreme, the meteorological conditions were favourable for the accumulation of air pollutants. The wind speeds near the surface were low for several weeks: < 2.5 m/s at

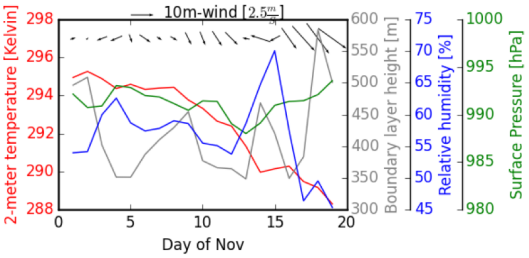

Figure 9. WRF 24-hour average meteorology over an area of 70x40 km^2 around Delhi and Agra in November 2017: 2-meter temperature (red, left axis), Boundary layer height (gray, right axis), relative humidity (blue, right axis), Surface pressure (green, right axis) and 10m wind speed and direction (black arrows, key gives length of 2.5 m/s wind). The transition from high to low CO levels is taking place around ~~the~~ 15 ~~th of~~ November.

10 meter height, limiting the advection of CO away from the sources (Fig. 9). The temperatures were relatively low, decreasing from 22°C to 16°C from 1 Nov to 19 Nov, thus limiting vertical convection. The planetary boundary layer heights were low with daily averages between 350 and 580 m, diagnosed from WRF, while the air pressure (around 990 hPa) and relative humidity (up to 70%) were relatively high (Fig. 9). The most important changes that we found in meteorological parameters around ~~the~~ 15 ~~th of~~ November when the CO concentrations started decreasing, are in the wind speed, the wind direction, the relative humidity and the boundary layer height. ~~The~~ As can be seen in Fig. 8, the wind speeds clearly increased after ~~the~~ 15 ~~th of~~ November and the wind direction changed from a north-westerly direction to easterly winds in this period of the highest CO concentrations and the start of the ventilation. The relative humidity (RH) went up from 55% to 70% on November 15 and decreased afterwards to 45%. The boundary layer was highest on ~~the~~ 18 ~~th of~~ November (580 meter, see Fig. 9) but we found that more to the north-west of the IGP, closer to the Himalayas, boundary layer heights were also exceeding the height of this mountain ridge on November 14 and 15 (not shown). The highest CO values around Delhi were found during 13-16 November, so just before the winds were turning and increasing. In our WRF simulation, the most important contributors to the decrease in CO were both the ventilation of the IGP with clean air from the Himalayas followed by advection of the pollution to the south-east, which took place over all days after 15 November, and the outflow of CO towards the Nord-West, around the Himalayas, in the upper troposphere. We could clearly observe this outflow of CO in the upper layers of WRF on November 14 and 15 and it was also visible in the TROPOMI measurements on the same days (see also: Borsdorff et al., 2018a). The emissions of MACCity that went into the WRF simulation with only MACCity emissions were the same every day of November, which means that the increase and decrease in CO levels in the MACCity-only run (Figs. 6, 7) were due to the meteorological conditions.

Meteorological variability in relation with ~~air~~ CO pollution was studied before by Verma et al. (2017), who focused on Agra, Uttar Pradesh, 100 km South East of New Delhi. The meteorological conditions they reported for November 2015 were similar to what we found for November 2017. However, the RH peak of 70% we found was in the upper 25% of their measurements for November, and the boundary layer was shallower than the lowest level of 450 meter they found in their study.

We should remark here that the meteorology in our study is based on WRF model simulations, while in Verma et al. (2017), the meteorology came from automatic weather stations, except for the boundary layer height, which was obtained from the MERRA (Modern Era Retrospective-analysis for Research and Applications) reanalysis. ~~Just looking at the data that we have, however, we can conclude that compared to~~ From the meteorological and observational data we conclude that 2017 shows a similar build of pollution as in the 2015 study, however, circumstances for CO accumulations were slightly ~~more favourable for accumulation~~ stronger in November 2017. In our study, as in Verma et al. (2017), stagnant weather conditions are an important cause of the high pollution levels.

4.3 The effect of chemistry and VOC emissions

In the base setup of WRF we did not include indirect CO sources from e.g., VOC oxidation in the analysis. Also, we did not include the OH oxidation of CO, and the only sink of CO was outflow at the lateral boundaries of the model. To assess the importance of VOC oxidation to the XCO and ground-level concentration, we performed a few sensitivity simulations, including indirect CO sources from VOCs and methane and the oxidation of CO by OH. The most important differences were found over the region directly around New Delhi. XCO increased by up to 4% due to the oxidation of VOCs and methane. Oxidation of CO by OH lead to a decrease of up to 7% and, combining both mechanisms, a net decrease in XCO of up to 4% was found in the New Delhi region. In the non-IGP area, the effects of VOC oxidation, OH removal, and the two combined were respectively +2%, -5% and -2% on the XCO. Over the Indo-Gangetic Plain, effects are somewhere in-between. Patterns of XCO enhancement over India were hardly affected by VOCs and OH oxidation. Compared to the uncertainty in the emissions, we consider these model simplification unimportant and therefore justified for the goals of this study.

5 Summary and conclusions

TROPOMI showed very high levels of XCO (>280 ppb) over Northern India during the high pollution event in India in November 2017. TROPOMI captured the spatial pattern of the pollution, covering not only Delhi, but rather the whole IGP. This demonstrates the high importance of investigating the sources and the transport of pollution as hundreds of millions of people are living in the IGP and their health is likely affected by the bad air quality. CO is very suitable for investigating air pollution, not only because of the negative health impact of CO itself, but also as tracer to track the dispersion of other pollutants, due to its life-time of several weeks. November is in the post-monsoon crop burning season, and media and scientific papers pointed to emissions from crop residue burning as the main reason for the high pollution levels over the IGP (Jha, 2017; Vadrevu et al., 2011; Liu et al., 2018; Cusworth et al., 2018).

In this study, we analysed two consecutive periods in November: 11-14 November with the highest CO levels and 15-19 November, when CO levels decreased. High CO levels and a subsequent drop in CO were observed by TROPOMI, in ground-level measurements, and in our WRF simulations. The meteorological situation, characterized by low wind speeds and shallow atmospheric boundary layers, was most likely the primary explanation for the temporal accumulation of regionally emitted CO in the atmosphere. The increase in wind speed and change of wind direction around 14 November led to the subsequent

dispersion. The dominant role of meteorology, rather than emission variations, is supported by the fact that the WRF simulations that used constant emissions during the whole period, showed a similar temporal dependence, including decreasing CO levels after ~~the~~ 15 ~~th of~~ November.

After analysing the contribution of specific emission sectors to the simulated and observed CO levels over India, we conclude that residential and commercial combustion explain the largest fraction of the high CO pollution over the IGP. Biomass burning only plays a minor role in the CO enhancement: on average 1-2% at ground-level, and only 1% to the total column pollution level. In earlier studies, it was found that the GFAS biomass burning data, used in our analyses, likely underestimate the actual emissions of CO (Mota and Wooster, 2018; Cusworth et al., 2018; Huijnen et al., 2016). The comparison of TROPOMI data with our WRF simulations, based on MACCity and GFAS data, confirms that CO emissions are underestimated in the 11-14 November period. The difference could be accounted for by increasing the GFAS emissions ~~to~~ by 500%-1000%~~of the value in GFAS~~, a rather large increase compared to the ~~findings of~~ 200%-600% increase found in the last named studies. Fires missed by GFAS observations might explain part of this increase. In that case, the contribution of biomass burning to the observed pollution levels becomes more important: in the order of 5%-20%, but it would still remain smaller than the contribution of urban CO emissions. Therefore, unless urban MACCity emissions are largely overestimated and GFAS emissions are underestimated even more, which we consider a less likely scenario, the contribution of urban CO emissions is the most important contributor to the CO pollution inside and out of the cities. These findings have important implications for emission mitigation efforts to avoid extreme pollution levels over the IGP during the post-monsoon period.

Our results have implications for ongoing winter time pollution mitigation efforts in India. Meteorology is found to be key driver of the extreme pollution episodes, ~~which cannot be altered cheaply with the current state of geoengineering technologies~~however, in conjunction with strong CO emissions. Hence, to mitigate the pollution, reducing the largest CO emission sources (residential and commercial combustion) remains the best solution, short-term and long-term.

Data availability. Data used in this study can be found under ftp://ftp.sron.nl/open-access-data-2/TROPOMI/tropomi/

Author contributions. Iris Dekker, Sander Houweling, Maarten Krol, Thomas Röckmann, Sudhanshu Pandey and Ilse Aben were active in the conceptualization of the study, and designed the methodology. Iris Dekker performed the model simulations, carried out data analysis, and wrote the manuscript. Sander Houweling mentored Iris Dekker. Sudhanshu Pandey, Tobias Borsdorff and Jochen Landgraf helped with the resources: Sudhanshu Pandey with the acquisition of the Indian surface measurements and Tobias Borsdorff and Jochen Landgraf provided the TROPOMI data. All authors contributed to revising the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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