I want to thank the authors for their significant effort to address the comments of both reviewers in this revision. The changes have significantly improved the manuscript, both in content and organization. There are a few aspects of the original comments which could be more fully accounted for, and a few minor issues arising in the revisions themselves. Once these are addressed, I recommend publication.

Notes on responses to reviewer 1
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1. Re: the comment "In terms of providing context/motivation for this work, I would [be] interested in seeing discusson on topics such as..."

The sentences added by the authors are a good start in addressing this comment, but could go farther in addressing the broader impact of the work. The added text says that "The TROPOMI retrieved XNO2/XCO ratio is useful for estimating mid-day OH over isolated localized sources..." but does not explicitly tie that to a larger science question or societal benefit. An additional sentence or two explaining what scientific question(s) this could help answer (could it provide information on the dominant NOx chemical regime for example?) or how it will help e.g. improve future air quality would more completely address the reviewer's comment.

Author Response:

This sentence is added to the discussion section:
"The TROPOMI retrieved XNO_2/XCO ratio is useful for estimating mid-day OH over isolated localized sources, such as the city of Riyadh, showing a clear contrast between the urban plume and the background. Such TROPOMI derived OH estimates offer a new opportunity to evaluate urban photochemistry in chemistry transport models. OH depends non-linearly on NOx and VOC emission, meteorological conditions, etc. (Sillman et al., 1990), which vary substantially between cities that are monitored by TROPOMI. Therefore, the application of our method to the global and multi-year dataset that is available could contribute substantially to the understanding of urban photochemistry and the development of effective pollution mitigation strategies. "

Additionally, the discussion of what cases this method could be applied to is fairly abstract. One of reviewer #2's comments pointed out that Riyadh is an ideal case for this technique because it is a large, isolated point source with minimal clouds, and suggested testing the percentage of clear sky pixels required by withholding increasing percentages of data and seeing how the answer changed. If done with the OSSE study added in section 3.3, then the authors could quantify the change error with decreasing data availability. I strongly recommend including this test, which would help quantity the statement "isolated tropical and subtropical cities are best suited for this method", as cities in tropical Amazonia (for example) will frequently lose significant amounts of data due to clouds.

Author Response:

As suggested by the reviewer, we applied a bootstrapping method to the data of 17th August and 18th November, 2018 to quantify the impact of a reduced data availability on the estimation uncertainty for OH, NOx and CO emissions. 500 images were generated repetitively while randomly reducing the TROPOMI data size (N) to 50 % of the original datasets (N_R). For each image, the OH concentration, NOx and CO emissions were estimated using our least square optimization method.

From the ensemble statistics we infer that the uncertainty of optimized OH is <15 % for summer and <21 % in winter due to the reduced data availability, which is smaller than the total estimation error (listed in Text S6, Tables S1 and S2). Similarly, for NOx and CO emissions, the uncertainty is < 15 % for summer and winter.However, this outcome depends on the random sampling process in the boot strapping method. In reality, the data gaps due to cloud cover are distributed differently. Since we select cloud free days, the bootstrapping test is not representative for our method and was therefore not included in the paper. A tropical case, cloud free days would obviously be less abundant than over Riyadh. However, we have shown that a single cloud free day may suffice to obtain a useful OH estimate.

2. Re: the comments about adding statements on the iterative optimization and CAMS use to the abstract

The authors have addressed these comments adequately, however the abstract has grown quite long and dense. I might suggest that the authors (a) make the results their own paragraph, and (b) reduce the number of results presented in the abstract to focus on the 1 or 2 most important ones.

Author Response:

We condensed the abstract as follow:

A new method is presented for estimating urban hydroxyl radical (OH) concentrations using the downwind decay of the ratio of nitrogen dioxide over carbon monoxide column mixing ratios (XNO_2 / XCO) retrieved from the Tropospheric Monitoring Instrument (TROPOMI). The method makes use of plumes simulated by the Weather Research and Forecast model (WRF-CHEM) using passive tracer transport, instead of the encoded chemistry, in combination with auxiliary input variables such as Copernicus Atmospheric Monitoring Service (CAMS) OH, Emission Database for Global Atmospheric Research v4.3.2 (EDGAR) NOx and CO emissions, and National Center for Environmental Protection (NCEP) based meteorological data. NO_2 and CO mixing ratios from the CAMS reanalysis are used as initial and lateral boundary conditions. WRF overestimates NO_2 plumes close to the center of the city by 15 % to 30 % in summer and 40 % to 50 % in winter compared to TROPOMI observations over Riyadh. WRF simulated CO plumes differ by 10 % with TROPOMI in both seasons. The differences between WRF and TROPOMI are used to optimize the OH concentration, NOx, CO emissions and their backgrounds using a iterative least square method. To estimate OH, WRF is optimized using a) TROPOMI XNO_2/XCO, b) TROPOMI derived XNO_2 only.

For summer, both the NO_2/CO ratio optimization and the XNO_2 optimization increase the OH prior from CAMS by 32 ± 5.3 % and 28.3 ± 3.9 % respectively. EDGAR NOx and CO emissions over Riyadh are increased by 42.1 ± 8.4 % and 101 ± 21%, respectively, in summer. In winter, the optimization method doubles the CO emissions also, while increasing OH by ~52 ± 14 % and reducing NOx emission by 15.5 ± 4.1 %. TROPOMI derived OH concentrations and pre-existing Exponentially Modified Gaussian function fit (EMG) method differ by 10 % in summer and winter, confirming that urban OH concentrations can be reliably estimated using the TROPOMI-observed NO_2/CO ratio. WRF optimization method can be applied to single TROPOMI overpass, allowing to analysis day to day variability in OH, NOx and CO emission.

3. Re: the comment about v2.2.0 of TROPOMI data

Thank you to the authors for going back and incorporating the newer data version at least in the error calculation. Two notes:

- Please include a reference to Text S6 in the captions for tables S1-S3 so that the reader can find the methodology for the error calculation more easily.

Author Response:

Changed as suggested

- One potential issue in using v1.2.x and v1.3.x of TROPOMI data in the main analysis but v2.3.x in the error analysis is if any of the changes between the two TROPOMI data versions affect the tropospheric slant column. Based on my reading of sect. 5 of the NO2 readme (https://sentinels.copernicus.eu/documents/247904/0/Sentinel-5P-Nitrogen-Dioxide-Level-2-Product-Readme-File/3dc74cec-c5aa-40cf-b296-59a0f2140aaf), that is not the case - it looks like most changes affect the AMF or the stratospheric column. However, it would be good to acknowledge this inconsistency in Text S6 and include links to the NO2 readme and the PAL page (https://data-portal.s5p-pal.com/product-docs/no2/PAL_reprocessing_NO2_v02.03.01_20211215.pdf) so readers can understand the differences themselves.

In our opinion the use of v2.3.x does not introduce an inconsistency as we only use the difference with v1.2.x and v1.3.x to obtain an, admittedly crude, estimate of systematic error. We agree, however, to specify the differences between these retrieval versions using the suggested references.

4. Re: the comment "The authors assessed NO2 data quality vs. ground-based measurements...is there a similar analysis that can be done for CO?"

I concur with the authors' explanation in the response. I'd suggest adding something to the end of Sect. 2.2 that points the reader to Text S6 where this is discussed.

Author Response:

- ***In Line 131, this sentence is added: "The comparison of TROPOMI derived XCO to the 28 different TCCON ground based station suggest that difference between TCCON and TROPOMI is in the range of 9.1 ± 3.3 % (Shah et al., 2020). Such difference is used to estimate the uncertainty in the NOx emission and life time (see Table S1, S2, S3 and Text S6). "***

5. Re: the comment "I'm not sure I agree with this justification for not allowing XNOx,Bg to be lost by OH..."

Here, I don't follow the authors' justification that because WRF and CAMS are within 10-20% this means that the background NOx is in photochemical equilibrium. I'm not sure from this response whether the comparison is WRF vs. CAMS or summer vs. winter. It's also not clear to me whether the "WRF" in this response is the passive tracer simulation or the full chemistry test (though only the latter would make sense to me). If the statement "The application of OH to the NOx background results in much smaller background NOx concentrations in WRF than in CAMS" means that the authors' applied the CAMS OH concentrations as fixed data to the passive tracer WRF background NOx and compared it to colocated NOx from a full-chemistry CAMS simulation, I'm not surprised that there is such a large difference as the fixed OH fields in this case wouldn't respond to the changing NOx-HOx equilibrium in the WRF simulation.

But, I think Table R1 communicates the point needed - that even if OH is treated as a fixed concentration and applied to the background NOx tracer, the difference in both derived NOx emissions and OH concentrations is reasonably small. (I am surprised that NOx emissions decrease in the summer with Bg OH-NOx loss test - I would have expected lower background to need greater emissions to make up the plume magnitude. But the w/BG OH NOx emissions value is within the uncertainty of the no BG OH NOx value.) I would recommend the authors' include Table R1 in the supplement and reference it in the discussion of the treatment of background NOx.
Author Response:

This sentence is added in the section 4, line 525 to 527: "A sensitivity test has been performed in which $XNO_{x,Bg}$ is lost by OH. In this case the optimized NOx emission and OH for summer and winter differ by < 7 % from the default method where the background is treated as an inert tracer (see Table S6)."

6. Re: the comment "Please state why this model simulation is well suited to evaluate emission changes..."

I'm unclear on Fig. R3 - in the right panel, is XCO_WRF_opt the optimization result using the prior reduced by the factor of 10? If so, this is a nice sensitivity test and should go into the supplement (with the meaning of the plot series in the right panel clarified), not just the response.

Author Response:

This sentence is added to the discussion section: "Furthermore, a sensitivity test has been performed in which the prior emission has been changed. The optimized emission varied by < 5 %, demonstrating robustness of the method to the choice of prior (see Fig S24). This also indicates that the optimization method can be used to study emission changes. "

7. Re: comments on various URLS:

- The https://cophub.copernicus.eu/s5pexp link in the data availablility statement still gives a "Not found" error
Author Response:

The link has been modified to " https://s5phub.copernicus.eu ; http://www.tropomi.eu "

- The Zenodo link given in the data availability statement *does* require a login, this is just a link to your personal account's list of deposited data. If I follow it and log in, it shows my data, not yours. Please provide the DOIs or DOI URLs given in the Zenodo page for each dataset (and please test these links in a private browser window - that is the best way to ensure they are truly public).
Author Response:

The Zenodo link is as follows: " https://doi.org/10.5281/zenodo.5752219 ". To make sure the link is public, I sent it to 10 different persons who had no problem accessing the data without login.

Notes on response to reviewer 2
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1. Re: the comment "The larger issue is the choice to use passive tracers..."

Thank you to the authors for clarifying that this was to reduce the computational cost. Two new comments on the added sentences:

- It's not clear how this method helps separate out the effect of meteorology, OH concentration, NO2 + OH rate constant, and NO2/CO ratio. The OH concentration and NO2/CO ratio is clear, but given that the model meteorology and NO2 + OH rate constants will still have errors and there is no optimization of wind direction or kinetics, I don't think you can say that this method isolates the effect of meteorology and kinetics.

Author Response:

We agree with the reviewer that transport model uncertainties influence the comparison with satellite data in a way that is difficult to quantify. However, as TROPOMI measures the NO_2 and CO from same platform and taking their ratio reduces the impact of transport model uncertainties. The model allows to compute these influences on the NO_2/CO ratio. There is no way our optimization method can separate the uncertainty in OH from uncertainties in reaction kinetics, but we do not claim this.

In Section 3.2, line 354 this explanation is provided as follow: " As expected, $Ratio_{without\ OH}$ shows an approximately straight line when the background is removed, because transport influences NO_2 and CO in the same way and therefore cancels out in the ratio (see Fig. 3b). The $Ratio_{with\ OH}$ however, shows an approximately gaussian relation with distance due to the influence of the sink on NO_2. This comparison demonstrates the sensitivity of the relation between XNO_2/XCO ratio and downwind distance to the NO_2 lifetime, which we want to exploit to quantify OH. "

- The statement that this method is an important improvement over the EMG method because it includes the use of a transport model isn't quantified. Introducing a transport model isn't an automatic improvement if it doesn't result in better estimates of OH, better estimates of emissions, or some other quantifiable improvement. Please be specific and quantitative about how it is an improvements - either show with the new simulations in Sect. 3.3 that the WRF optimization produces more accurate OH or emissions, or focus on how this method enables analysis of day-by-day OH and emissions, whereas the EMG method needs longer time periods.

Author Response:

The updated version of the manuscript includes this explanation (section 3.6, line 476): "In contrast to EMG method, the optimization method can be used for a single TROPOMI overpass (see Section 3.6) and does not require yearly averaged NO_2 data, allowing analysis of day-by-day OH, NOx and CO emission (see Section 3.3). Segregation and averaging of NO_2 urban plume by wind sector is not required in the optimization method. "
In the conclusion section (line 567) we explain the difference as follow: "With our method, single TROPOMI overpasses can be used to estimate OH whereas the EMG method requires averaging of urban NO_2 plumes by wind sector.

2. Re: the comment about showing that this method works for single-day overpasses

Thank you for including a single day results. In combination with the new Figs. S17 and S18, this is very promising. Two notes:

- Why in the new text do you use r2 for the initial WRF XNO2 and XCO comparisons, but X2 for the optimized comparisons? It would be nice to use X2 for both to be consistent.

Author Response:

r2 and X2 quantify something different and their use serves different purposes. r2 is used as measure of spatial correlation, whereas X2 is used to test the goodness of the optimized fit. Since these numbers were not meant to be compared, we do not see the need to make them consistent.

- The original reviewer comment also mentioned testing what percentage of data must be cloud free for this method to work by essentially bootstrapping smaller and smaller percentages of data and checking the results. This would work especially well with the new synthetic tests in Sect. 3.3, and having a quantification of how much clear sky data is necessary would strengthen the discussion in Sect. 4 about how widely this method could be used. There the authors suggest that this method will work best for tropical and subtropical cities, but cities in e.g. the Amazonian tropics will often have a high fraction of cloudy pixels. Could such a bootstrapping analysis be added to the supplement to support Sect. 4?

Author Response:

This question is addressed in the answer to question 1 of reviewer 1.

Other notes on revised text

Line 12: why are emissions not mentioned along with OH concentrations as an output of this method?

Author Response:

The main objective of this paper is to estimate OH. As suggested by reviewer 1, we tried to reduce the size of the abstract, prioritizing the main objective. For this reason, we decided to keep it as is.

- Line 113: there are numerous citations you could use to support the statement that high resolution NO2 priors better resolve the NO2 gradients, to name a few:
* www.atmos-chem-phys.net/11/8543/2011/
* https://acp.copernicus.org/articles/14/3637/2014/
* https://acp.copernicus.org/articles/15/5627/2015/

Author Response:

The citations are added in line 113.

- Line 271: is the PBL height here taken from the WRF simulations?

Author Response:

In line 271, we added: "The PBL height at the time TROPOMI overpass has been taken from WRF"

> Line 311: could clarify the interpretation of f_emis, f_OH, f_Bg by specifying whether f = 0 or f = 1 means that the prior was correct. Normally, I think of "scale factors" as being multiplied by the prior (so f = 1 would mean the prior was correct) but here it looks like f = 0 means that instead.

Author Response:

The scaling factor f_{emis}, f_{oh} and f_{bg} represent the modification of the prior in percentage change. The scaling factors f_{emis} ,f_{OH} and f_{Bg} obtained from the ratio optimization have been divided by 10

because $\frac{\Delta F}{\Delta emis}$ ***,*** $\frac{\Delta F}{\Delta OH}$ ***and*** $\frac{\Delta F}{\Delta Bg}$ ***are defined as the change in F due to modification of emission, OH and background by 10 %.***

Line 463: what does "For winter, the dissimilarity between the EMG method and the prior reduces after optimization," mean? Do you mean that the difference between the EMG and WRF-optimized results are smaller than the difference between the EMG results and the prior? As written, it sounds like both the EMG and prior are being optimized somehow.

Author Response:

The sentence is modified to "For winter, the difference between the EMG and WRF-optimized results are smaller than the difference between the EMG results and the prior "

Text S1: in step 2, please specify which reaction the second-order rate constant in question is for

Author Response:

The sentence is modified into: "First, we calculate the IUPAC second order rate constant for the reaction between NO_2 ***and OH, using the pressure and temperature for each vertical level.***

- Table S4: please add to the caption what the quantities in parentheses are

Author Response:

Changed as suggested.