This paper by Fioletov et al. is quantifying urban, industrial and background changes in NO2 during the COVID-19 lockdown in Spring 2020 based on TROPOMI NO2 tropospheric vertical column density data (VCD). A statistical regression analysis is used to sperate urban, industrial and background components of the observed tropospheric NO2 VCD. The total NO2 mass of the three components is estimated and converted to emissions assuming a constant lifetime. The analysis was done for 263 urban areas around the world with a focus on urban areas from the US and Canada. To study the impact of the Covid-19 lockdown on the different components the analysis is done for the lockdown period in spring 2020 and is compared with the average of the same period in 2018 and 2019.

It was found that changes in the background component are rather negligible but that the urban component declined over most regions by 18%-28% and for some regions even up to 60%. The decline in the industrial component varies more between the regions and declines up to 40% were found.

The study showed a new method to distinguish background NO2 and NO2 from urban and industrial sources by fitting of satellite data by a statistical model with empirical plume dispersion functions using wind data, data about population density, location of industrial sources and elevation data.

The paper is of significance for identifying and separating different components of NO2 pollution and quantifying the impact of the COVID-19 lockdown on the different components. On the one hand, the paper and the appendix are already very long, and the authors should think about where they can shorten or compress (concentration on certain regions, leaving out the analysis with night light data etc.). On the other hand, the evaluation, comparison, and discussion of the results is to short in several sections, and this should be given more emphasis. There is also a lack of error consideration in the graphs and text. The chapter on the relative contribution of the different components needs further explanation and discussion on the representativeness of the results and the chosen area size. Due to the significance, the actuality of the topic and the new method for isolating various components of NO2 pollution from satellite data, I recommend publication in ACP with major revisions.

We would like to thank the reviewer for a very thorough and detailed review. We made the corrections suggested by the reviewer. We added several paragraphs that provide additional information on estimating uncertainties that were previously in the Supplement and removed/shortend some other parts.

We removed the night light section and moved some results to The Supplement. Supplement was also revised and is much shorter now. However, we were not able to make the main text shorter because we added more comparisons and interpretation results as well as the variability estimates that were requested by the reviewers.

Note that we also repeated the analysis with the more recent TROPOMI data set (the used versions are described in the text) and changed the criteria for industrial point sources in the urban areas (used population density directly instead of the correlation coefficients). All of that slightly affected some of the results. We also excluded 2 areas. In one case, the population

density data were clearly unreliable, in the other case the noise was too high, probably, due to biomass burning.

General comments:

Can you comment on how much your study results are influenced by meteorology (wind, temperature, sun, clouds)?

Winds are included in the plume dispersion model and therefore are accounted for. Interannual variability in general was discussed in the original version of the Supplement. To address this we moved that section to the main text and added two tables with information about the uncertainties and variability. We also added a figure to the Supplement (Fig. S3) that demonstrates TROPOMI is very stable.

More numbers and error estimations are needed and should be added to the figures and values in the text.

In the original manuscript, the uncertainty estimates and justifications were in the Supplement to make the main text shorter. In the revised version, we moved them to the main text (Tables 1 and 2 and related discussions) and added more information as requested by reviewers.

More comparisons to existing publications, especially to discuss declines of NO2 found with your method, are needed.

This was also requested by the other reviewer. We added such comparisons and discussions.

Chapter 4.2 Relative contribution of different components (Page 12 Line 8/24):

The values you are giving ("urban and industrial sources are only responsible for a quarter of the NO2", ...) are not really representative, they are largely depending on the size of your area. I have not seen a clear argument for choosing an area of 3°x4 °. The results depend on size of the chosen area and size of the cities. If the areas are chosen smaller or in relation to the city size the contribution of background to the NO2 mass would be reduced. Comparisons between 2020 and 2018/2019 are reasonable, but general statements about the relative contributions of the different components are difficult. Wouldn't it be possible to do the analysis without area size playing a role? If this is not the case, it should be better explained and discussed how the calculation was performed.

The reviewer is correct. The relative contribution of the components depends on the size of the area. The area should be neither too small nor too large. However, the characteristics such as the mean background value and annual emissions per capita are much less dependent on the area size. We emphasized that we used that relative contribution statistics only to highlight the importance of the background component. All of this is mentioned in the text and expanded in the revised manuscript. In the revised version, we focus on the characteristics that do not depend on the area size such emissions per capita and moved the relative contributions figure to the Supplement.

All references to the appendix should be consistent and clear, sometimes they are not. (P 3, L 22; P 6, L 24; P11, L31, and more)

We moved some sections from the Supplement to the main text and corrected the references to the remaining sections.

Specific comments:

Page 3 Line 32: Specify the TROPOMI NO2 product. OFFL or NRTI and which versions?

We added this information.

Page 3 Line 11: Can you better explain where these remote areas with background NO2 are located?

We gave some examples in the text and figure caption: National Forests in Montana and Algonquin Provincial Park in Ontario.

Page 3 Line 24. Comment somewhere in this chapter on how the different data sets were brought together in terms of spatial and temporal resolution.

We added a few sentences about this in the Data Sets section. All such procedures are very common

Page 4 Line 17: I think wind profile data are not directly available from ERA5, you probably computed them, a brief comment on that would be helpful.

We averaged winds at 100, 950, and 900 hPa. We added a sentence about this.

Page 4 Line 22: It would be clearer if each dataset gets its own small chapter, since quite a lot is written about the individual datasets.

Corrected

Page 7 Line 18: That sounds very general, perhaps just misleadingly formulated. Is the emission per capita factor coupled with the population density data?

We provide additional details about the urban emission estimates and the use of population density data.

Page 7 Line 48: “high population density zones typically occupy a small part of the area and industrial sources are typically located away from such highly populated zones” Not in general, for example in the Ruhr area in Germany, the Po valley or Riyadh this is not the case.

Actually, in all these cases, industrial sources are not located in the highly populated areas, but 10-20 km away. In the revised version, we specifically excluded industrial sources, that are in the areas with population density above a certain limit (600,000 people per 0.2° by 0.2° greed cell).

Page 9 Line 13: “We monitored the correlation coefficients between industrial and population density-related plume functions and, in some cases, excluded certain sources or even certain urban areas from the analysis.” How was decided which areas were excluded, with a certain correlation coefficient?

In the revised version, we specifically excluded industrial sources, that are in the areas with population density above a certain limit as mentioned above. It solved the issue with high correlation coefficients.

Page 10 Line 12: Explanation for (g) is missing.

It was in the next paragraph. We rearranged the text to have the explanation earlier.

Page 10 Line 16: This explanation would have provided a better understanding about the used algorithm a bit earlier in the text, maybe include something similar in chapter 3 (The Emission fitting algorithm).

We moved the text up as suggested.

Page 10 Line 26: Why do you only give a value for one city (Boston) and not for the others? Please also add values for the other discussed cities. In addition, error estimation is needed and should be added to the figures and values in the text

The values were in the Fig. 8, but we added them to the text. We also added two tables with the uncertainty budget with related text.

Page 11 Line 5: Are the values comparable to results from other studies? For Boston but also interesting for the other cities.

We added such information

Page 11 Line 5/6: Error margins are missing, please discuss and add to the figures and in the text.

There are two types of errors: related to interannual NO2 variability and to measurement uncertainties. We added a table with the uncertainty values.

Page 11 Line 10: Is the airport visible on the map, where is it located, please add a description in the text.

We added a description.

Page 11 Line 26. Why are oil refineries not included in the EPA NEI inventory, please comment.

We added more information about US inventories. Unlike power plants, emissions from most of the other sources are not measures but estimated and such estimates are available for annual emissions.

Page 11 Line 31: Please include how additional sources not included in the inventories were discovered and added for the analysis.

In the revised version, we used an additional EPA eGRID inventory to locate the sources.

Page 12 Line 1: I think this is true for some of the cities. At least for Seattle, the changes are not large, but for some of the other cities, changes in background values (especially Boston) and structure (especially Houston) are visible. Is this really negligible compared to changes in mean VCDs? Where do these changes come from?

We added some discussion about the background values here. Our goal was to isolate and quantify that background component and to show its importance. At this point, we can only speculate about its origin. In case of Houston, the structure became more uniform after the processing with the latest available versions. It is likely related to some "bad" values that get through the filtering in the original calculations.

Page 12 Line 12: Rephrase sentence, right now just the two facts (sharp gradients and short lifespan) are mentioned, make a better connection.

Corrected

Page 12 Line 21: Add a reference here.

A reference was added.

Page 13 Line 5: Did you calculate the average over the deviation of the background values of 2020 compared to 2018/2019? Yes on average not much happened between 2020 and 2018/19, but there are cities with a decrease and some with an increase, that should be mentioned.

We added some comments here. However, it in not always clear what is driving the background variability and more research is required.

Page 13 Line 18. If you think that these error bars also apply to the other areas, then they are maybe nearly all within the range of natural variability. How can you exclude that the weather in 2020 was not different from 2018/2019 for also other areas than Vancouver just in the other way?

We added some discussion about this. The NO2 changes over individual areas are affected by natural variability. We used the 2018, 2019, and 2021 values to estimate this variability. Obviously three years are not enough to estimate variability for a particular site. Instead, we assume that standard deviation of the variability is the same over different areas within one region and that the impact of meteorology is independent from one area to another. Two publications are cited to support the latter.

Page 13 Line 27: Could it also be the different strength or length of the lookdown?

Yes. We changed the sentence.

Page 14 Line 1: "Note that 2020 US EPA NEI reported emission were incomplete at the time of this study." What does that mean, how does it influence your comparison?

It just meant that we did not use the 2020 reported emissions in this study. We removed that sentence and changed the text.

Page 14 Line 13: "The NO2 decline was particularly large, more than 50%," The decline of only the urban or industrial component or in the mean?

In the urban component. Corrected.

Page 15 Line 2: What is about the strong increase for Sofia in the industrial component? In the urban component there is a strong decline. Any ideas about the different behavior of the two components?

The uncertainties for the industrial component are large and the industrial component for Sofia is small. Therefore, the difference in the industrial component for Sofia may be related to the noise. We added more discussion about the uncertainties. We also added one example where we were able to establish a source of the emissions increase (Cuba).

Page 15 Line 8/11: Please give standard deviation.

Random (or independent) uncertainty that is typically given is very small, 1%-2% The main problem is the natural interannual variability that is hard to estimate from just two years, but this can be done by analyzing multiple areas in one region. In the revised version, we also added 2021 to better estimate the interannual variability. We added a section about this issue and a table.

Page 15 Line 30: Is this induced by COVID-19 or by environmental policy decisions or weather?

It is probably both, but we do not want to speculate. We just stated that there was a 60% decline.

Page 16 Line 7: What are the consequences for your method? Is the consequence that you cannot use your method for this case, how have you decided which cities are still good enough, have enough NO2?

It was used just to show that emissions per capita are very different from region to region. It does not affect the method. We moved this paragraph to the discussion about emissions per capita in various regions of the world to avoid confusion. Figure 12 was moved to the Supplement.

Page 16 Line 10. "Another obstacle is in Western Africa, where biomass burning made it difficult to estimate "background" levels as they were very different from year to year." How do you handle this problem? Are such cases excluded and according to what criteria?

Yes, several areas with population over a million in Western Africa were not included in the analysis. Biomass burning areas appears as large positive anomalies on the maps of the residuals. We added this explanation.

Page 16 Line 28: It comes to mind that this might not just be background, but that background and urban/industrial components cannot be perfectly separated. Add one or two sentences of discussion.

We added a few sentences to discuss this issue.

Page 17 Line 18: "China shows the smallest and not significant decline in the urban component," Add that for China the strongest lockdown was before your chosen time window.

It was mentioned later, but we moved that sentence up as suggested.

Page 18 Line 7: Why now back to the uncertainties in Figure 14? Add this earlier in the text before Figure 15 was discussed in detail.

We changed the order of the paragraphs here

Page 18 Line 23: Add which time periods are analyzed and compared for the mobility data. Same periods as before, so the 2020 period with 2018/2019?

We added this to the dataset description in Section 2.

Page 19 Line 3: The Comparisons to Lange et al. doesn't fit in this chapter (4.5 The global COVID-19 lockdown impact).

We moved it to next section.

Page 19 Line 20: Nice that it worked, but is there really a new benefit from it, think about to skip it or only mention at some point that it is also possible to use night light data.

Based on suggestions from both reviewers, we removed that part. One of the main reasons why we tested the nightlight data set is that population density data are unreliable over some regions.

Page 19 Line 28: “On a scale of several hundred km (as we analyzed 3° by 4°areas), most of the NO2 mass is typically related to the background component.” See general comment about the representativeness of this results.

We changed the discussion about the total mass and contribution of the individual components.

Page 21 Line 8: Add information about wind data, emissions (EPA, NPRI), world powerplant database, elevation and population data.

Corrected

Figure 8: Figure description is not fitting very well to the figure.

The description was edited

Add error bars to the figures. Adjust the scaling of the y-axis according to the values for all plots, even if you consider the following plots, it can be adjusted. Make clearer that this are the three components background, urban and industrial, especially the title “mean NO2” over the background component plot is misleading.

We added a section on variability and uncertainties. We also added the grey dashed line that represents the uncertainly due to interannual variability. We also corrected the issue with the components labeling.

Figure 9: Add regression line etc. Are the TROPOMI-based estimated emissions the sum of industrial and urban emissions? Please add a comment to the figure description.

Corrected as suggested

Figure 10a, b: Same as for Figure 8.

Corrected

Figure 14: Same as for Figure 8 and 10.

Corrected

Figure 16: What is meant by “pre-lockdown period”, which periods are compared with the google mobility data?

We added this information

Technical corrections:

Page 1 Line 18: Add that you used tropospheric NO2 vertical column density.

Corrected.

Page 2 Line 22: “One common approach is based on a rotation of satellite NO2 pixels around the source to align the wind data to a common direction,”

“to align the wind data to a common direction” is misleading, should it not better be something like “to align the NO2 data with wind data to a common wind direction.”

Corrected to “… so the NO2 data would appear if the wind is from one common direction”

Page 6 line 23: delete “as”

Corrected

Page 6 line 24: shows instead of show

Corrected

Page 9 Line 13: Correct “density-relate signal” to “density-related”

Corrected

Page 10 Line 19: Correct ”x” with the correct sign (dot) for multiplication

Corrected

Page 10 Line 27: Why starting with Boston, when it is not the first city in the figure? Could be changed in the text or in the figure.

Corrected

Page 13 Line 10: Delete first per.

Corrected

Page 15 Line 16: “over the mountains in valleys in the Milan area.” Replace “in” with “and”.

Corrected

Page 18 Line 7: “uncertainty values” instead of “uncertainties values” and delete “the regions”

Corrected

Page 18 Line 19,20: The two sentences are very similar. Add “(background, urban, and industrial)” to the first sentence and delete it in the second one.

The first sentence was changed to “The three components of NO_2 distribution (background, urban, and industrial) were calculated for every country and compared with various characteristics of mobility data.” And the second sentence was deleted.

Page 20 Line 8: “Abrupt changes and urban and industrial emissions” change “and” to “in”.

Corrected

Page 20 Line 15: “than in the boundary layer and a relatively small amounts of NO2” delete “a”.

Corrected

Page 21 Line 5: “to complement and emissions improve available “bottom-up” inventories” delete “emissions”

Corrected

Figure 2: “Panel (b) is the sum of panels (d), (e) and (f).” Shift it after the description of (f) so before “Emission point sources are…”

Corrected

Why is the second source in (g) a grey and not a black dot, could be changes to make it consistent.

Slightly larger gray dots were used to mark airports. We added this to the text.

Figure 4: “(c) and well as individual components” replace “and” with “as”

Corrected

Mention the highlighting of the airport in the text.

Corrected

Figure 7: In the figure the label says “Population” in the text below it is called “urban”, make it consistent.

Corrected. The figure is now in the Supplement.

“The contribution from industrial sources and cities” could be also changed to “industrial and urban sources”

Corrected

Figure 13. Change legend title in the top figure from “Environs Component” to “Background Component” to make it consistent.

Corrected