

## Response to the Reviewer #1 comments

Dear authors,

Thank you for taking into consideration the well meaning suggestions posed both myself and another reviewer. While your manuscript is improved at places, I continue to have reservations about certain parts of your methodology, as well as its presentation.

I read your new manuscript before reading your reply-to-reviewers, to have a fresh view. I remain of the strong opinion that the material is too much for one piece of work, and as a result the main aim/take away message remains muddled. I strongly, again, recommend that you split your work into Part A and Part B. I firmly believe that this will aid you as well in presenting your technique/findings in a clearer way. The annotated text contains numerous points for further discussion/clarification.

Best wishes.

We would like to thank the reviewer again for a very thorough and detailed review, although many comments are related to the original text and could be included in the previous review. We made most of the corrections suggested by the reviewer.

As to the question of splitting this paper into two. There are pros and cons with both one vs. two papers. Pros of two papers is that it might be clearer; on the other hand, having both method and application together is also advantageous. Our feeling is that since (i) only one reviewer commented on this, and (ii) it would be a large amount of work by authors, but also reviewers and editorial staff, to continue with the one paper approach.

Additionally, this manuscript is focused on the COVID-19 impacts, but it was necessary to develop a method to study these impacts on different components (urban, industrial, background) of the NO<sub>2</sub> distribution. The method is based on previously developed approaches (e.g., “wind rotation”) and algorithms (e.g., for point, multi-source, and area source emissions estimates). In this study, the algorithm is designed for a specific task that allowed us to use some simplifications. For example, we used a constant, not seasonally dependent, lifetime and made a few other simplifications. So, it is not a universal method, but a version that tuned to deal with a specific task. This is another reason why we prefer not to split it into two parts.

Specific comments:

p. 1, l. 26. Since you mention the coefficient, but it is not too high, you might wish to also mention the statistical significance of this value.

It is available in the text, but we do not think it is necessary to mention it here. It is expected to be significant and included in the abstract.

p. 1, l. 27 Isn't this slightly self-evident? since NO<sub>x</sub> are emitted from traffic in pure urban environments? if the urban locations you study are affected by industry, the game changes of course.

We added the word “estimated” to highlight that we isolate the urban component from background and industrial NO<sub>2</sub> and compare only the urban emissions estimated from satellite data.

p. 1, l. 27 Does this imply that the background NO<sub>x</sub> emitted around urban locations is due to a different, and stable source? a natural source?

One of our goals is to isolate the background component in TROPOMI data and to show that its response is different to that from the urban component. This has not been done before. As for the interpretation, it is discussed in the paper, but we do not want to include it in the abstract.

p. 1, l.30. This phrase is rather stand alone. Is this the mean of 2020? then I find the -6% of the background rather significant, not so?

We added that it is in 2020 and reminded again that we are talking about the mean for all areas. We highlighted the 20-σ deviations because it is a very large deviation. The confidence intervals are given in any case.

p. 3, l. 28, This is rather vague. Which NO<sub>2</sub>-related parameters were not affected by the lockdowns?

We changed the text to “*The variability of urban, industrial, and background NO<sub>2</sub> components due to meteorological or observational conditions was studied by comparing the estimates of the three components for 2018, 2019, and 2021 that were not affected by lockdowns.*”

p. 5, l. 4. The titles of these plots are misleading. The plots show S5P NO<sub>2</sub> but this information does not appear anywhere on the plots. Re-title and add a subtitle on the wind speed separation. Also add a title on the colour bar, as is the norm.

The figure caption starts with “Mean TROPOMI VCDs...” and we added “TROPOMI NO<sub>2</sub> VCD” to the colour bar.

Furthermore, the resolution of these maps is low. Please update.

Corrected.

It would be beneficial for non-US citizens to add the state-lines. From these maps, one cannot know where the megacities/power plants are. While I realise that it would make the maps too busy, you might wish to add a separate map showing these locations.

We can obviously add the state lines, but this would indeed make the maps too busy. NO<sub>2</sub> distribution does not follow these lines. And, they are shown in Fig. 5 anyways. As for megacities, they are also shown in Fig. 5. The power plants and other sources are shown in Fig.6 where we discuss individual areas. Fig.1 is just an introductory figure that shows main features of the NO<sub>2</sub> VCD distribution.

The local topography is also taken on faith since we cannot expect readers to know where the Rocky Mountains or the Alognquin Provincial Park are.

Four geographical areas mentioned in the paper are labelled on the map and described in the figure caption. However we prefer not to “overload” the maps with additional labels. We believe that the ACP readers know about the Rocky Mountains.

p.5, l.17. This is not a small point to sweep under the carpet. You have one, maybe two due to your large 3x4 deg area of study, L.T. of observations. The diurnal variability of tropo NO<sub>2</sub> during those L.Ts is strong since S5P senses the sharp decline of the NO<sub>2</sub> peak.

You have to discuss all these points in detail.

We already explained in the original response, that the diurnal variability of NO<sub>2</sub> VCD is not large. Moreover, the paper is focused on relative changes in 2020 vs. the levels of previous years that make the impact of diurnal variability on the final results (i.e., relative differences) even smaller.

p. 6, l. 10. This is really vague. How many are most? in the next sentence you mentioned sources not available. How many/big were these sources?

We added that data from oil refineries are not available in CEMS.

p. 6, l. 21. So this applies to India and China basically?

It also includes sources from all other regions.

p. 6, l. 26. Why did you not choose the previous year as baseline? then, you could have included these months as well. Since traffic also depends on meteorological conditions, wouldn't this provide a more "climatological-like" proxy? discuss.

Google mobility data are available only as deviations from a specific baseline. We added this to the data set description.

p. 7, l. 29 IT would be far easier to the reader if this paragraph included examples of each of the steps described, as the text flows. There are results discussed in the text, results from work that has not been shown yet.

I strongly recommend that you consider changing your presentation manner to include example results in this section. Especially since you yourselves have found the need to discuss results in order to explain the method.

The technique used in this section is a further development of the methods that are about 7 years old. There are other papers that describe the technique, and they provide more information and examples. We add a reminder about this at the beginning of the section. The first part of this

section describes the statistical model used in the analysis and gives the necessary formulas. Then we have Fig. 3 where we give examples of all components of that model and explain them.

p. 8, l. 5. This is all very nice, but of course the lockdown did not last 3 months anywhere. How do you reconcile this? already at this point, the reader is a bit puzzled at the huge 3x4 deg grid compared to the SSP spatial resolution.

It was explained in several places, that we need a time interval of several month for our method to work reliable. The impact of lockdowns is shown in Fig. 1. It shows a decline in mobility in all analysed areas (except China) that lasted 3 month or even more.

As for “the huge 3x4 deg grid”, it looks that we have a major misunderstanding here. Nowhere in the paper we use or even mention a “3x4 deg grid”. 3x4 deg is the size of an area around major cities. All calculations were done for each of such area individually using all TROPOMI L2 observations in that area.

p. 8, l. 26 Is it common to have such a strong, well-defined plume of vehicular NO<sub>x</sub> emissions? SO<sub>2</sub> plume from power plants which are emitted above 100m already are a different issue. Discuss.

Yes, it common for plumes averaged over a long period, e.g., for a season or year. Again, the technique is not new. Originally, this approach was suggested by Beirle et al., Science, 2011, as discussed in the Introduction. Note that this sentence starts with “*For typical plume characteristics (discussed below)...*” The next paragraph explains that the NO<sub>2</sub> lifetime is about 3-4 hours. At the wind speed of 20 km per hour, we will have a substantial plume on a distances of 120-160 km.

p. 9, l. 14 How were these values chosen? a sensitivity study? discuss.

Yes, it is based on a sensitivity study and the results of that study are discussed in the next sentences. We added a sentence about a sensitivity study.

p. 9, l. 32. How big are these locations? already at page 9 and the actual spatial analysis of this work is not mentioned. Discuss.

We changed “each location” to “any place”.

The statistical model is used to estimate **parameters**. Then, using these parameters (and the wind, elevation and population density data), the background, urban, and industrial components can be calculated for any time and location including, for example, every TROPOMI pixel. I.e., the NO<sub>2</sub> value at that pixel can be represented as a sum of the three values. Then, any operation that can be applied to TROPOMI data, can be also applied to these components.

p. 10, l. 4. I am keen to guess that this is indeed the case. However, you could have picked a 1x1 deg around your sources. If the computational cost was the only reason you chose 3x4, then this cannot be assumed a valid reason to produce results. Discuss extensively.

There is some misunderstanding here. We clearly stated that “*The fitting was done for all satellite pixels centered within 3° by 4° areas around large cities and collected during a three-month period by minimization of the squares of the residuals ( $\epsilon$ )*”. We are talking about 336 sources (0.2° by 0.2° grid) **within** that 3x4 deg. area. So, we assume that there are 336 sources plus a number of industrial sources within that area. We added a reminder that the grid is “*within the analyzed 3° by 4° area*”

p. 10, l. 17. But also in the extremely well populated East and West coasts of America, most of Europe, for e.g. the Benelux region, etc.

We do not understand the comment. Yes, there are areas with very high population densities. But the estimates are done for emission per capita. They are approximately the same for a highly populated New York area and for a much less populated area of Minneapolis. It will be later shown in Fig. 12.

p. 10, l. 22. For spring/autumn, sure. Winter? you were rather vague as to the winter lifetime values in your previous section.

The paper uses data only for the period from March 16 to June 15, so we do not to worry about the winter lifetime values.

p. 10, l. 27. am still not convinced as to this "therefore". How many NOx emitting locations have such a huge orography change within 3x4? apart from the Po Valley, i.e the Alps, for e.g.

If the analysed area is small, then the correlation coefficient between the elevation and population density could be high because the cities tend to be in valleys. This correlation declines if we increase the area size. But the area should not be too large for a different reason. Again, 3x4 deg. area is where we run our analysis. It is not a global grid.

p. 11, l. 10. This would be so much more clear if an actual example was given. Consider this.

An example is given in Fig. 3.

p. 11, l. 18. This paragraph contains too many detailed information to be followed without an actual example. For much of the text previously, strong suggestions that this method works on individual sources was given. Now we read a lot about clustering of industrial sources. Discuss.

A few lines above we stated that “*We would like to emphasize, that the factor analysis, described in the next two paragraphs, was used to improve emission estimation for individual sources or clusters of sources. It is not required if only total emissions from all point sources in the area are estimated in order to separate them from urban emissions or if all industrial sources are isolated remote sources.*” This text is an explanation how multiple emissions sources in close proximity were handled. It is a new part of the algorithm. It is not important for many readers but could be significant for those who will use this method to estimate emissions.

p. 11, l. 31. All this should be demonstrated with an example. This section reads not like an ACP paper but a GMD paper.

Please, see the previous comment.

p. 12, l. 30. Is this number from the bottom-up emission inventory or a result of this work?

It is from our estimates. We added that.

p.13, l. 20. Successfully for a correlation coefficient of 0.55? what was the mean and median R2 you found for all the locations you studied? the question arises to the mind of the reader here.

It looks that there is a major misunderstanding here. The 0.55 correlation coefficient describes how well the statistical model (Eq. 1) can reproduce the NO<sub>2</sub> values for **individual TROPMI pixels**. It is clearly stated in Section 3: *“The fitting was done for all satellite pixels centered within 3° by 4° areas around large cities and collected during a three-month period...”* Correlation of 0.9 here would mean that there is no need for satellite measurements at all. A couple of Pandoras in Europe would be enough to establish the model coefficients and then the statistical model would produce daily NO<sub>2</sub> maps over Europe from the wind, population density, and elevation data. We do not claim that we can reproduce individual TROPMI pixels. Instead, we are using that statistical model to reproduce the 3-month mean values with high spatial resolution within the analysed 3° by 4° area as shown in Fig. 3a. I.e., we can estimate that mean value for any location within that area and the correlation coefficient between our estimates and the real TROPOMI 3-month averages is 0.96 (for Montreal). **It is explained in the next sentences.** Please see also our comment to p. 9, l. 32.

p. 13, l. 24. In variance, yes, correlation of more than 0.90, but in absolute 0.55? how do your reconcile these two? and what of the other locations?

Please see the comment above.

To “reconcile these two”, let us give a simple example: you are measuring a highly variable atmospheric parameter for many years with a temporal resolution of 1 hour. There is a long-term linear trend in your data. You can calculate that trend by fitting the original 1-minute data by a linear function or, for example calculate the monthly averages and then fit them. The trend estimates and their uncertainties are the same in both cases. However, the correlation coefficients between the fitted and predicted (by that linear function) data could be very different. They are likely would much lower in the case of hourly data (for example, 0.55) than for monthly means (that could be even 0.99). This is exactly what is happening in our analysis. The only difference that the fitting is done not in an 1D space (time), as in our example, but in a 4D space (lat, lon, u- and v- wind components).

The correlation coefficient is above 0.9 for all analysed areas in Canada and U.S. This correlation coefficient is above 0.8 for 241 of 261 analysed areas.

p. 13, l. 1. Great. Please provide the correlations and the correlations of the variances for this example as well.

They are 0.96 and an 0.94 for Minneapolis and Seattle respectively. We added that.

p. 14, l. 1. Title extremely vague. Re-phrase.

Corrected to “*NO<sub>2</sub> VCDs estimation results for urban areas*”

p. 14, l. 15 Please add which part of which figure shows which city, in the text here.

We added that “*The Fig. 6 is divided in 4 sections with the area name shown on the top of each area.*”

p. 15, l. 1. You mean, in absolute levels? state this clearly, state the levels.

Yes, corrected.

p. 15, l. 9. This is not very small. Discuss.

Correlation of 0.54 does not mean that we cannot estimate the emissions. It just means that the uncertainties are higher than for uncorrelated plumes. The emissions uncertainties are given in the next sentence.

p. 16, l. 7. Any physical explanation for this? discuss.

It is discussed in the next paragraph and later in Section 5.

p. 16, l. 21. A CTM might give you a proper indication of this. Have you considered this option? studying the relative contributions from one of the well-established CTMs that run for the US?

This may be possible, but it is outside the scope of this paper. Also, the CTM resolution should very high to match our calculations on a 0.2° by 0.2° grid.

p.16, l. 25. Is there a strong agricultural presence in the locations you looked into? discuss.

We are considering 261 area around the world. It is very likely that at least some of them are located near agricultural areas. We are just describing potential sources.

p. 16, l. 26. I should think that a larger discussion on the known S5P background issue should be included in this text. There exist statistical methods by which this "unphysical" background may be removed, especially in 3x4 grids.

One way, suggested by the S5P data providers, evaluates a zonal mean of all the minimum values at every latitude of the domain and then subtracts this mean from every daily gridded pixel of the domain.\

Has something similar been attempted here? discuss.

The biases are discussed in a recent paper by van Geffen et al., 2021 (van Geffen, J., Eskes, H., Compernelle, S., Pinardi, G., Verhoelst, T., Lambert, J.-C., Sneep, M., ter Linden, M., Ludewig, A., Boersma, K. F., and Veefkind, J. P.: Sentinel-5P TROPOMI NO<sub>2</sub> retrieval: impact of version v2.2 improvements and comparisons with OMI and ground-based data, Atmos. Meas. Tech. Discuss.) There are two effects contributed to the bias. Both are expected to **scale roughly linearly** with the column amount and that is consistent with the validation results. Therefore, the relative differences (2018-2019 vs. 2020) are not affected by them. We added this comment with the reference to Section 2.1. It also means that the background, urban, and industrial components are affected by a scaling factor. Their absolute values may change in the analysis is done with a better version of TROPOMI, but not so much their relative contributions.

p. 16, l. 27. This discussion is valid but I fail to see how it would explain such a big background level which comes from the daytime S5P observations. Discuss.

It does not explain the background level, but it may contribute to it. If the night lifetime is longer than TROPOMI-based lifetime estimates, then it is possible that a part of the plume could travel farther than expected by the used plume dispersion functions based. Then that part would be counted in the background. This does not really need to be mentioned in the text.

p. 17, l. 5. As you rightly state, these estimates are quite low. Especially considering the statistical errors introduced by your model, the S5P measurements, the fact that one h of observations results in an annual emission estimate, etc. I hence fail to see why this discussion is included in the main text and not the supplement. Discuss.

The reviewer again is mixing up the uncertainties of the residuals (that are rather high) and uncertainties of the model parameters that are very low. Please see out reply to p.13, l. 20 and p.13, l. 24 comments. The uncertainties here are directly linked to the parameter uncertainties. The uncertainties here show how well we know the values of individual components in different years. The interannual variability shows how one year is different from the other.

As it was suggested by both reviewers to have the uncertainties in the main text (before they were in the Supplement) and they are included here.

p. 17, l. 18. See my point above.

Here we are talking about uncertainties due to **interannual variability** for **individual** areas. Yes, they are high and the baseline, estimated from just two years, is not very accurate. This is why we also consider regional averages later in section 4.4.

p. 17, l. 19. Reading the paper again, with a fresh eye, I still consider it more appropriate to split the work into two parts. There are a number of improvements that may be applied to the discussion above, where the technique is presented, alongside the general results. With these, the paper would increase even more.



I strongly recommend splitting the work into two, maybe making the COVID part into a short communication/letter-type article.

Please see our reply at the beginning of this document.

p.17, l. 21. For urban and industrial this makes perfect sense, but does it for the background? considering all the issues you yourselves have already discussed previously? I am not as convinced.

To our best knowledge, this study is the first attempt to divide tropospheric NO<sub>2</sub> into three components related to industrial and urban emissions and the background distribution. Therefore nobody a priori knows how they react to a major changes urban emissions caused by COVID-19 lockdown.

p. 17, l. 23. Is this really the most important part of this work, and you show it first? I would expect first to see the urban/industrial results and then - if needed - the background.

As the background components contains most of the NO<sub>2</sub> mass, it is discussed first.

p. 17, l. 23. It looks to me like this paper has too many goals. To demonstrate the robustness of the method, to show it on different examples AND to show that it can sense the COVID-related changes. These goals sometimes get confused, such like in this case.

Another reason for which I am recommending splitting of the paper.

Please see our reply at the beginning of this document.

p. 18, l. 2 Comparing two years cannot possible result in the reader understanding what the "normal" is for Alberta. Both these years could be outliers! Rethink and comment.

It is a discussion about possible factors affecting the background component. The previous sentence stated that "*In 2020, Alberta had a "historically low" level of forest fires*". From this, the reader should expect that the **natural** NO<sub>2</sub> emissions (where forest fires one of the main sources) in 2020 that should be lower than in the other years. We modified the sentence slightly to make this clear.

p. 18, l. 21. So, this means that for industrial emissions, this technique cannot sense the COVID effects. Discuss.

For the analyzed areas, the 2020 percentage change in emissions from industrial sources were typically within the limits of interannual variability. However, the 2020 values were below the baseline in almost all analyzed areas. If we calculate the mean value of percentage changes for the entire Canada-US region by averaging percentage deviation in individual areas, then such mean value is statistically significant.

All of this was discussed in the last paragraph of Section 4.4. We moved that paragraph up and joined it with the sentences at p. 18, l. 21.

p. 18, l. 29. Which are for a different year, to begin with. And come from different inventories. Discuss.

Figure 9 shows industrial emissions for 2018 and 2019. They are from the same years as it is stated in the text: “*Each dot on the plot corresponds to industrial emissions from one area in either 2018 or 2019 with the total of 40 data points.*” Emissions data are from the same EPA emissions inventory, although some of the emissions are measurement, while the other are estimates.

We modified the text to make this message clearer.

p. 18, l. 30. On which timescale? 3 months? what does this level say about your technique? can it or can't it be applied to derive annual NO<sub>x</sub> emissions from S5P? discuss.

The reviewer criticized us many times (e.g., p. 17, l. 23 ) that we are discussing the advantages of the method instead of focusing on COVID impacts. And here the reviewer is asking us to do exactly that. We added a sentence that “*Then, the annual NO<sub>2</sub> emissions are expected to be estimated with uncertainties of about 2.5 kt y<sup>-1</sup> that is twice less than about 5 kt y<sup>-1</sup> for SO<sub>2</sub> emissions uncertainties (Theys et al., 2020).*”

p. 18, l. 33. Extremely vague and in contrast [for the industrial sources] to what was stated above. Rephrase, discuss.

We modified this paragraph and moved it up. Please see our reply to p. 18, l. 21.

p. 19, l. 25. Provide proper reference. What is this anomaly about? how was it studied? and what about Belgrade?

It is just a comment on a possible explanation for interannual variability in a particular case. We added a reference.

p. 19, l. 29. Phrase too large, too confusing. Rephrase carefully.

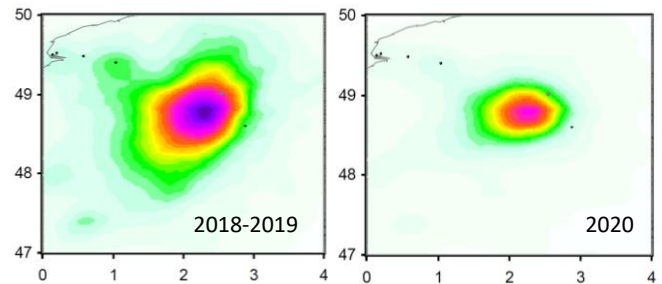
Corrected.

p. 20, l. 16. The colour scheme for the first two columns should change, it is nearly impossible to see the variation since all values fall in the red/purple part of the colour bar.

This was done intentionally to highlight that the background component is so large in these areas, that it often exceeds the signal from the cities themselves. Note that all NO<sub>2</sub> figures in the paper use the same color scale.

p. 20, l. 28. Not clearly seen at all I'm afraid. Please change the colour bar limits.

In this sentence, we are comparing the two periods for the urban component in Paris : “...while the decline in the **urban** component is about  $-57\% \pm 1.5\%$  clearly seen on the plot.” The urban component is in Fig. 11, column e, and these plots are also shown below:



We hope that the difference is evident from this plot. We added that the urban component plots are “in column e”.

p. 21, l. 6. In numbers?

We added the decline values

p. 22., l. 9. Do these show in the Fioletov SO<sub>2</sub> emission inventory?

They are discussed in Section 2.4. We determined “hotspots” that appear on NO<sub>2</sub> maps, then run them against known emissions source databases that includes the SO<sub>2</sub> catalogue (Fioletov et al., 2016). For sources that are not in these databases, we use satellite images. For example, most of cement factories are not included in these databases.

p.22, l.15. Again, since you discuss annual estimates in the end, surely you have to consider that this lifetime is not the same for all 12 months of the year. Discuss.

All calculations in this paper are based on data for the period from March 16 to June 15. It was stated in several places. Therefore, the emissions are also only for that period. They are expressed as annual emissions rates in  $\text{kt y}^{-1}$ , so it would be easier to compare them with reported emissions that are in  $\text{kt y}^{-1}$ . Similarly, for example, the speed is reported in  $\text{km h}^{-1}$  even if you walk for only 5 minutes. Now we explicitly explain the emissions units in Section 3.

p. 22, l. 18. Extremely vague statement. Discuss or omit.

We removed the sentence

p. 22, l. 22. Siberian pipelines?

See here:

[https://www.esa.int/Applications/Observing\\_the\\_Earth/Copernicus/Sentinel-5P/Connecting\\_the\\_dots\\_nitrogen\\_dioxide\\_over\\_Siberian\\_pipelines](https://www.esa.int/Applications/Observing_the_Earth/Copernicus/Sentinel-5P/Connecting_the_dots_nitrogen_dioxide_over_Siberian_pipelines)

No, the pipeline is not located close enough to Siberian cities with population more than 1 million to be inside the areas used in our analysis.

p. 22, l. 27. Paragraph vague. Provide numerical findings.

It is a description based on Figure 12. We reminded about that and added some numbers.

p. 23, l. 3. Yes, but are these changes low enough to show the COVID effect of 2020? again, this paragraph reads like the goal is to discuss the methodology and not the COVID effect.

Fig. 13 is just another way to demonstrate that the COVID-19 lockdown has a different impact on the background and urban components. Here we are talking about geographical distribution of the changes.

p. 25, l. 2. What are the statistics of this correlation? the C.I.?

It is given in the **next** sentences: *“The correlation coefficient between the percent changes in per capita emissions and “retail and recreation” mobility is 0.62 (the probability that there is no correlation is less than 0.0003). There is no statistically significant correlation (the correlation coefficient is -0.08) between the background NO<sub>2</sub> and mobility data (Fig. 15 right).”*

p. 25, l. 14. Well, no. This suggests that your method cannot see the 2020 as an outlier, since for individual areas "the observed 2020 decline of the urban component in many areas is within that uncertainty" whereas you have some signal [possibly and for some areas] when performing regional analysis.

This should clearly be stated in your conclusions.

Furthermore, this creates questions as to the usability of this emission dataset by other scientists.

Yes, the 3-month decline in 2020 in some areas is within the interannual variability. However, if we group these areas into regions and calculate regional averages, then for **all regions** (except China), the regional decline is **outside 2- $\sigma$  limits**. Then if we calculate the average of all these areas, that average is **outside 10- $\sigma$  and 20- $\sigma$  limits** for background and urban areas respectively. This is exactly what is stated in the conclusion.

As for emission dataset, we focused on relative changes in 2020 compared to the previous years. The comparison was done for a 3-month period and 3-month emissions were calculated. It is not an emission inventory, although the method can be used to create a such inventory.

p. 25, l. 31. A good effort, but you still have to deal with the known background issue on the S5P data before being able to say that the background you result in is a "true" background NO<sub>2</sub> field.

Please see out reply to your comment at p. 16, l. 26. We also added that “*this study included the background component as a function of the elevation in the analysis*”. This was not done in previous studies.

We also implemented all technical corrections suggested by the Reviewer (not listed here).

## Response to the Reviewer #2 comments

The study shows a new method to distinguish background NO<sub>2</sub> and NO<sub>2</sub> 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 NO<sub>2</sub> pollution and quantifying the impact of the COVID-19 lockdown on the different components.

Most of the comments from the first review were implemented into the manuscript, which improved the quality of the manuscript. In particular, the classification and comparison of the results with previous studies and the uncertainty and interannual variability analysis have been necessary and important additions. The requested additional information on the calculations of the used wind profiles from ERA5 data indicate that they may not take into account for the correct heights of the analyzed sites (see general comments). Clarification or replacement of the wind data is needed. I recommend publication in ACP with minor revisions.

We would like to thank the reviewer again for very useful comments.

### General comments:

Page 5 Line 5: “The ERA5 wind components at 1000, 950, and 900 hPa were averaged (that approximately corresponds to the mean winds between 0 and 1 km).”

I think that the ERA5 wind data at 1000, 950, and 900 hPa correspond to an elevation of approximately 0-1km but above sea level, and the considered source regions include some elevated sites which are > 1 km asl (like Denver with 1600 m asl), where wind profiles estimated from the 1000, 950, and 900 hPa levels are not appropriate. If this is the case and is not corrected, replacement of the wind data may be needed, as they are important for emission estimates.

We used one of the features of ERA5 reanalysis: All of ERA5 is in regular pressure levels. If the surface pressure is lower than the pressure (e.g. 1000hPa) it will simply duplicate the winds at the lowest pressure available. For example, if the site was at a high altitude, e.g. Denver at ~1.6 km, the surface pressure will be roughly 800hPa, so the winds at 900hPa, 950hPa, and 1000hPa are in the ERA5 files, but these will be exactly the same as the 800hPa winds.

We added this explanation to the text.

### Specific comments:

Page 8 Line 1: You often use quotation marks although it is often not necessary, especially in this sentence.

Corrected.

Page 13 Line 14: “The Pearson correlation coefficient I”, typically R not I, it is also R in the figure.

It was a typo. Corrected.

Page 13 Line 27: Instead of Fig. 4 h it should be Fig. 4 g.

Corrected

Page 14 Line 15: Introduction to the following detailed analysis but most of this is repeated in the following, could possibly be deleted, and just keep the first sentence. In case this paragraph is not deleted change “TROPOMI-base” to “based”

We were asked to have this paragraph by the other reviewer. We corrected “based”.

Page 18 Line 9: “The reason for that is not immediately clear but may be related to unusual meteorology and persistent cloud cover there in 2020.” Shouldn't it then be apparent when analyzing the interannual variability (table 1), but I don't think the interannual variability for Vancouver is very high?

The 15% deviation for Vancouver in 2020 is within the natural variability. We changed the sentence to “*This Vancouver anomaly is within 2- $\sigma$  limits of natural variability as discussed in section 4.3 (Table 1) and reason for that is not immediately clear but may be related to unusual meteorology and persistent cloud cover there in 2020.*” And removed the next sentence about statistical significance.

Page 18 Line 10: “Information about statistical significance of such “outlier” is discussed in section 4.4.” This is section 4.4, where is it discussed?

It should be section 4.3. We removed this sentence as described above.

Page 18 Line 14: This does not fit well here the previous paragraph and the following one should follow each other. Besides, it has nothing to do with the covid lockdown effect, either take it completely out or possibly shift it to the discussion of figure 6a.

We removed this sentence.

Page 20 Line 15: Please add a comment on if the decrease is within the interannual variability/if it is significant. For Europe2 I think this is not the case for all three components (background, urban, industrial) this should be mentioned.

We added the following text: “ For Europe-1, the decline of the background component is within the 2- $\sigma$  level for all the areas and the decline of the urban component outside the 2- $\sigma$  level for all the areas. For Europe-2 however, the decline of the urban component is inside the natural variability limits. For the industrial component, the variability is high and the 2020 decline is within the 2- $\sigma$  level for most of the areas.”

Page 22 Line 1: Which maps are meant, give a link to the figure (12 or 13 top)?

We are referring to the maps of the residuals such as shown in Fig. 3c. We added this to the text.

Technical corrections:

Often emission is used in the singular, but it should be emissions (Page 10 Line 2, Line 15, ...)

Page 2 Line 29: "(Gkatzelis et al., 20021;," the year should be changed to 2021.

Page 3 Line 18: NO<sub>2</sub> emissions should be NO<sub>x</sub> emissions.

Page 3 Line 23: Change "related" to "related"

Page 3 Line 26: Change "backgroiuund" to "background"

Page 5 Line 8: I think "and lower over high winds" should be changed to "and lower under high winds".

Page 6 Line 8: Change "Note that CEMS database" to "Note that the CEMS database".

Page 6 Line 19: "(accessed on March 2, 2021) And" missing dot.

Page 10 Line 12: "were converted to a 0.2° by 0.2° by averaging", "grid" should be added.

Page 10 Line 21: "these components", the relation has been lost, because the corresponding sentence has been deleted. Something like "The proxy plume functions of the main components (background, urban and industry) used in the model..." could be added to the sentence before this sentence.

Page 14 Line 6: "and urban I components" replace I with (e).

Page 16 Line 22: "into free troposphere" change to "into the free troposphere"

Page 16 Line 29: Change "nighs" to "nights"

Page 17 Line 10: Change "The 2020 data are not used this estimate since they were largely affected the lockdowns." to "The 2020 data are not used in this estimate since they were largely affected by the lockdowns."

Page 19 Line 25: Change "cause" to "caused".

Page 20 Line 5: Change "Italy" to "in Italy"

Page 23 Line 11: Delete one "also".

Page 24 Line 9: "For example, a large uncertainties in the industrial emission" I think "a" should be changes to "the".

Page 24 Line 13: "where largest urban emissions decline was observed" would be clearer if it is changed to "where also the largest...".

We implemented all technical corrections suggested by the Reviewer.