# Final author reply to editor (second revision)

## **Dear handling editor:**

Based on both referees' comments we have made the following new revisions to the manuscript:

- 1. Adding an analysis to estimate how much of the differences between the ozone season and exceedance day composites are due to intra-seasonal variations versus episodic changes on exceedance days,
- 2. Expanding discussions of: (a) possible satellite retrieval biases due to vertical profile and temperature differences on exceedance days, (b) the possible impact of the coarse albedo climatology dataset on the TROPOMI high HCHO artifact over the Great Lakes, and (c) "noise" levels of 4 km and 12 km versions of the TROPOMI composites,
- 3. Including a comparison of AQS surface NO<sub>2</sub> measurements to our TROPOMI-based results, and
- 4. Emphasizing new insights gained from and the value of this study in the abstract and the conclusions sections.

In the "tracked changes" version of the manuscript, we highlight changes/additions in red text. Our responses to the individual referee comments begin on the next page.

## **Response to referee #3**

### **Referee's introductory comment**

In this paper, Acdan et al. analyzed the TROPOMI HCHO and NO2 VCDs and their ratio (FNR) over the Lake Michigan region. They compared 3-year (2019-2021) composite meteorology, HCHO, NO2, and FNR maps between days with ozone exceedance and other days during the ozone season (May to September). They found that on ozone exceedance days, HCHO, NO2, and FNR tend to be greater over the region, and lake breeze circulation also tends to be stronger. This points to the importance of meteorology in ozone pollution episodes in the area. Similar comparisons were also made between weekdays and weekends. Overall, this paper demonstrates the application of TROPOMI data in regional air quality study and should be of interest to the readers of ACP. The results are largely qualitative and subject to limitations owing to uncertainties in the TROPOMI data products. The authors addressed some of the comments from both reviewers, but there are still major concerns (see specific comments below) about the data analysis method and the robustness of the results presented. I feel that major revisions are necessary before the paper can be considered for publication in ACP.

### **Response to introductory comment**

We thank referee #3 for providing thorough feedback on our revised manuscript. Based on both referees' comments, we have made the following new revisions to the manuscript:

- 1. Adding an analysis to estimate how much of the differences between the ozone season and exceedance day composites are due to intra-seasonal variations versus episodic changes on exceedance days,
- Expanding discussions of: (a) possible satellite retrieval biases due to vertical profile and temperature differences on exceedance days, (b) the possible impact of the coarse albedo climatology dataset on the TROPOMI high HCHO artifact over the Great Lakes, and (c) "noise" levels of 4 km and 12 km versions of the TROPOMI composites,
- 3. Including a comparison of AQS surface NO<sub>2</sub> measurements to our TROPOMI-based results, and
- 4. Emphasizing new insights gained from and the value of this study in the abstract and the conclusions sections.

Our responses to referee #3's specific comments are as follows:

## **Referee's specific comments**

(1) Most of the O3 exceedance days are in June and July as shown in the supplemental material, whereas the non-O3 pollution days are probably more evenly distributed from May to September (give that >450 days were used for the composite). One may argue that the differences between the two composites in meteorology and chemical composition can well be due to seasonal changes in meteorology, emissions, and chemical processes (as well as seasonal changes in TROPOMI retrieval performance). How do you separate the effects of seasonal changes vs. episodic events?

### Response

This is a very insightful comment, and we agree that it is important to separate the effects of intra-seasonal changes and episodic events.

Because 94 % of ozone exceedance events occur in June, July, and August (**Table S1**) in the Chicago metropolitan area (CMA), it is possible that the differences we see between the ozone season and CMA exceedance day composites are due to intra-seasonal changes. More specifically, the inclusion of May and September data in the ozone season composite may be the cause behind the composite differences because the data used in the exceedance day composites mostly come from June–August TROPOMI observations.

To estimate the effect of intra-seasonal changes, we created boxplot distributions by month for TROPOMI NO<sub>2</sub> and HCHO composite values and NAM temperature composite values. Then we compared the June–August mean values (when most exceedances occur) to the May–September mean values (entire ozone season). We deemed the difference between the June–August mean and May–September mean the amount of change we expect to see in our difference composites (**Figs. 2f, 3c, and 5c**) due to intra-seasonal changes. Finally, we compared this difference to the mean difference we see in the main text difference composites (**Figs. 2f, 3c, and 5c**). Dividing these two values gives us an estimate of how much (%) of the change shown in our main text figures are due to intra-seasonal differences, while the remaining amount of difference we prescribe as due to changes in environmental conditions on exceedance days.

#### Changes to manuscript

In the main text, we added the following discussion to Section 3.2.1 on lines 406–419:

It is important to discuss here whether the differences between the O<sub>3</sub> season and CMA exceedance day composites are caused by intra-seasonal changes or episodic changes inherent to O<sub>3</sub> exceedance days. Because 94 % of ozone exceedance days occur in June, July, and August (**Table S1**) in the Chicago metropolitan area (CMA), it is possible that the differences we see between the ozone season and CMA exceedance day composites are due to intra-seasonal changes. More specifically, the inclusion of

May and September data in the ozone season composite may be the cause of the composite differences because the data used in the exceedance day composites mostly come from June–August TROPOMI observations. To estimate the effects of intraseasonal changes, we created boxplot distributions by month for TROPOMI NO<sub>2</sub> and HCHO composite values and NAM temperature composite values (**Figs. S5–S7**). Both TROPOMI HCHO VCDs and NAM 2-meter air temperatures follow a strong intraseasonal cycle, but TROPOMI NO<sub>2</sub> VCDs do not. By comparing intra-seasonal differences in these monthly composites (**Figs. S5–S7**) to the differences we see between the O<sub>3</sub> season and CMA exceedance day composites (**Figs. 2f, 3c, and 5c**), we estimate that about 50 % of the HCHO and temperature changes are due to intraseasonal changes (and the other 50 % due to O<sub>3</sub> exceedance day conditions) while 100 % of the NO<sub>2</sub> changes are due to exceedance day conditions (**Table S7**). More information about our methodology to separate intra-seasonal and episodic changes can be found in the text below **Table S7** in the supplemental information document.

In the supplemental information document, we added Figs. S5, S6, and S7, which show the intra-seasonal cycles of NO<sub>2</sub>, HCHO and temperature, respectively. NO<sub>2</sub> does not appear to have a strong intra-seasonal cycle, while HCHO and 2-meter air temperatures do:





seasonal changes (and the other 50 % are due to exceedance day conditions):

	NO <sub>2</sub> [× 10 <sup>15</sup> molec. cm <sup>-2</sup> ]	HCHO [× 10 <sup>15</sup> molec. cm <sup>-2</sup> ]	Temperature [K]
(a) June–August mean	1.747 (Fig. S3)	8.96 (Fig. S4)	297.83 (Fig. S5)
(b) May–September mean	1.766 (Fig. S3)	7.73 (Fig. S4)	295.41 (Fig. S5)
(c) Difference [a – b]	-0.02	1.22	2.42
(d) Mean difference between OS and Ex composites [Ex – OS]	0.37 (Fig. 3c)	2.52 molec. cm <sup>-2</sup> (Fig. 5c)	4.31 (Fig. 2f)
	NO <sub>2</sub>	нсно	Temperature
(e) Percent of (d) due to monthly differences during the ozone season (intra-seasonal changes) approximated as: $[(e) = \frac{c}{d} \times 100 \%]$ $if (e) < 0 \%, report as 0 \%$ $if (e) > 100 \%, report as 100 \%$	0 %	48 %	56 %
<ul> <li>(f) Percent of (d) due to exceedance day differences (episodic events) approximated as:</li> <li>[(f) = 100 % - (e)]</li> </ul>	100 %	52 %	44 %

(2) If lake breeze circulation plays an important role in ozone pollution in the area, one may also expect differences in the vertical distribution of NO2 and HCHO between exceedance days and non-O3 days. This would lead to different biases in retrievals between the two composites and should be discussed.

#### Response

We agree that this is a valuable discussion to add to the manuscript.

## **Changes to manuscript**

We added the following discussion to Section 3.1.1 on lines 272–277:

One important thing to note is that because the strength of the lake breeze is different, it is possible that the vertical profiles of NO<sub>2</sub> and HCHO are different between exceedance days and non-exceedance days. The TROPOMI NO<sub>2</sub> and HCHO retrieval algorithms rely on forecasted model vertical profiles to produce VCD data (De Smedt et al., 2018; Van Geffen et al., 2022a). Therefore, the satellite retrievals used to create the ozone season and CMA exceedance day composites below (**Figs. 3-5**) may have different biases depending on how well the model forecasts vertical profiles of NO<sub>2</sub> and HCHO on exceedance days versus non-exceedance days in which the strength of the lake breeze circulation varies.

(3) Similarly, given the large differences in temperature (and the temperature-dependence of gas absorption cross sections), one would expect biases in retrievals on ozone exceedance days (if fixed cross sections are used in the slant column density fitting). This should be pointed out in the paper.

#### Response

We agree that this is valuable information to add to the manuscript.

According to the TROPOMI NO<sub>2</sub> ATBD (Van Geffen et al., 2022; <u>https://sentinel.esa.int/documents/247904/2476257/Sentinel-5P-TROPOMI-ATBD-NO2-data-products</u>), a correction factor is applied to the NO<sub>2</sub> absorption cross section to account for temperature sensitivity during the air mass factor step. The difference between the effective temperature of the NO<sub>2</sub> (in a specific layer) and the temperature of the baseline cross-section (220 K) is used to determine the correction factor assuming the temperature dependence is linear (equation 18 in the ATBD). Other gaseous species involved in the retrieval (O<sub>3</sub>, H<sub>2</sub>O, and the O<sub>2</sub>-O<sub>2</sub> collision complex) use fixed cross sections at reference temperatures. However, it says in the ATBD that variations of these cross sections have little effect in the retrieval of NO<sub>2</sub> slant columns, which is why a correction factor is only applied for NO<sub>2</sub>.

According to the TROPOMI HCHO ATBD (Hilboll et al., 2022;

https://sentinels.copernicus.eu/documents/247904/2476257/Sentinel-5P-ATBD-HCHO-TROPOMI.pdf/db71e36a-8507-46b5-a7cc-9d67e7c53f70?t=1658313806426), the HCHO absorption cross section is used at a fixed reference temperature of 298 K. BrO, NO<sub>2</sub>, and the O<sub>2</sub>-O<sub>2</sub> collision complex also use cross sections at fixed temperatures. Only the O<sub>3</sub> cross section is adjusted by fitting two absorption cross sections at different temperatures and assuming a linear dependence on temperature.

### **Changes to manuscript**

We added the following discussion to Section 3.1.1 on lines 290–302:

Another important thing to note is that the significant differences in temperature between the ozone season and CMA exceedance day composites may also lead to different biases in the satellite retrievals used to create the NO<sub>2</sub> and HCHO composites below (**Figs. 3-5**). The absorption cross sections of various chemical species used in the TROPOMI retrieval algorithms are temperature dependent. This can lead to retrieval biases if the cross sections are not adjusted for temperature. To mitigate the potential bias for NO<sub>2</sub>, a correction factor is applied to the NO<sub>2</sub> absorption cross section by calculating the difference between the effective temperature of the NO<sub>2</sub> and the temperature of the baseline cross section and assuming the temperature dependence is linear (Van Geffen et al., 2022). The other species used in the NO<sub>2</sub> retrieval algorithm (O<sub>3</sub>, the O<sub>2</sub>-O<sub>2</sub> collision complex, and H<sub>2</sub>O) use fixed cross sections, but the temperature dependence of these cross sections has little effect in the retrieval of NO<sub>2</sub> (Van Geffen et al., 2022). In the HCHO retrieval algorithm, the cross sections of most of the species (HCHO, BrO, NO<sub>2</sub>, and the O<sub>2</sub>-O<sub>2</sub> collision complex) are fixed while the

O<sub>3</sub> cross section is adjusted by fitting two absorption cross sections at different temperatures and assuming a linear dependence on temperature (Hillboll et al., 2022). Retrieval biases stemming from using absorption cross sections at fixed temperatures may be larger in the CMA exceedance day composites of NO<sub>2</sub> and HCHO (**Figs. 3-5**) since temperatures tend to be warmer than usual as shown in **Figure 2**.

(4) Line 117: how does the change in spatial resolution of TROPOMI in 2019 (near the end of the ozone season) affect your results?

#### Response

We do not believe the change in TROPOMI spatial resolution affects our results in a substantial way because we composited data onto a 12 km  $\times$  12 km grid, which is a coarser spatial resolution than the TROPOMI pixel footprint (both before and after the upgrade to higher resolution). Therefore, we did not make any changes to the manuscript based on this question.

(5) Line 170: what are the noise levels of HCHO, NO2, and FNR at 4x4 km<sup>2</sup> and 12x12 km<sup>2</sup>?

### Response

We have provided calculations of relative "noise" levels between the 4 km and 12 km composites in the manuscript. The estimated "noise" level was 3.6 times higher for HCHO, 2.6 times higher for NO<sub>2</sub>, and 3 times higher for FNR in the 4 km composites compared to the 12 km composites.

### **Changes to manuscript**

We added the following text to the manuscript in Section 2.2 on lines 170–175:

Note that we used a grid with a coarser spatial resolution than the original TROPOMI pixel footprint based on a sensitivity test using a  $4 \text{ km} \times 4 \text{ km}$  grid. This sensitivity test involved identifying a region of fairly uniform HCHO and NO<sub>2</sub> in the 12 km composites and then comparing absolute differences between nearest neighbors ("noise") to the mean value within that region in both the 4 km and 12 km composites. The estimated "noise" level was 3.6 times higher for HCHO, 2.6 times higher for NO<sub>2</sub>, and 3 times higher for FNR in the 4 km composites compared to the 12 km composites.

(6) Line 183: Are there differences in meteorology and chemical composition between more localized and more widespread ozone exceedance events?

#### Response

Yes, there may be differences. However, given the limited number of exceedance events, we do not feel there is enough data to produce statistically robust results/composites separately for more localized and more widespread ozone exceedance events. Therefore, we did not make any changes to the manuscript based on this question.

(7) Line 305: the coarse resolution of Kleipool climatology could be a bigger issue for urban areas – how does this affect your interpretation of the results over urban cores?

#### Response

This is a good question, and it is possible that the Kleipool climatology could be a bigger issue for urban areas. We do not know exactly how the coarse resolution affects retrievals over urban cores. However, the TROPOMI HCHO validation paper by De Smedt et al. (2021; <u>https://doi.org/10.5194/acp-21-12561-2021</u>) did compare TROPOMI HCHO to surface-based MAX-DOAS network column measurements, which includes instruments in major urban cores (e.g., Mexico City in Mexico, Madrid in Spain, Munich in Germany, and Beijing in China). The bias between TROPOMI and the MAX-DOAS measurements vary between sites, but it is found that on average TROPOMI HCHO observations are biased by -25 % for columns greater than  $8 \times 10^{15}$  molec. cm<sup>-2</sup>. We already noted this in Section 2.1 lines 140–141.

Our belief that there is an over water high HCHO bias for Lake Michigan is based on the fact that we see higher HCHO VCDs over all of the Great Lakes (Fig. S2). Our speculation that the Kleipool albedo climatology might be part of the cause of this artifact comes from a previous reviewer as well as the TROPOMI HCHO ATBD (Hilboll et al., 2022; https://sentinels.copernicus.eu/documents/247904/2476257/Sentinel-5P-ATBD-HCHO-TROPOMI.pdf/db71e36a-8507-46b5-a7cc-9d67e7c53f70?t=1658313806426), which states that the spatial resolution of the albedo dataset is coarser than the resolution of TROPOMI, which can induce errors in VCDs for coastal regions and is definitely something we should mention in the main text.

### **Changes to manuscript**

We have expanded the discussion of the Kleipool climatology and its possible impact on HCHO observations over Lake Michigan in Section 3.1.2 on lines 332–342:

Therefore, we believe this over water bias is unrealistic, especially since it is found over all the Great Lakes (**Fig. S2**). As noted in the TROPOMI HCHO algorithm theoretical basis document (Hilboll et al., 2022), the coarse resolution of the OMI-derived surface albedo climatology dataset used in the retrieval (Kleipool et al., 2008) can induce errors in VCD calculations for coastal regions. The high HCHO bias over

the Great Lakes may be in part due to the Kleipool dataset being too coarse to fully resolve the complex surface albedo properties that are common to lake surfaces. We also acknowledge that the resolution of the Kleipool climatology may also affect TROPOMI HCHO observations over urban cores since urban areas can also have complex surface albedo properties. However, TROPOMI validation studies (e.g., De Smedt et al., 2021) have been performed for urban sites, providing estimates of the TROPOMI retrieval biases for such areas (see **Section 2.1**). Validation over water surfaces is more difficult to conduct since the necessary instruments are not routinely deployed over lakes. Further research is needed to assess the impact of albedo changes between lake surfaces and the surrounding coastal areas on TROPOMI HCHO retrieval performance.

We additionally added **Figure S2** to the supplemental information document that shows the high HCHO artifact over the Great Lakes:



(8) Section 3.2: the weekday vs. weekend analysis appears to be only loosely connected to the previous section. I'm not sure if it actually adds any significant value to the paper.

## Response

We consider the weekday versus weekend analysis to be a complement to the previous section because it shows how differences in NO<sub>2</sub> vertical column densities can also lead to substantial changes in FNR values (e.g., in the urban core of Chicago). This is opposite to the ozone season versus exceedance day analysis in which changes in FNR values are largely dominated by HCHO differences as opposed NO<sub>2</sub> differences.

### **Changes to manuscript**

We have added the following text to the conclusions section on lines 569–575 to highlight the value that the weekday-weekend analysis adds to the paper:

This weekday versus weekend analysis complements the ozone season versus exceedance day analysis because it shows that differences in NO<sub>2</sub> VCDs can also lead to substantial changes in FNR values and O<sub>3</sub> chemistry sensitivity (e.g., in the urban core of Chicago). This is opposite to the ozone season versus exceedance day analysis in which the largest changes in FNR values and O<sub>3</sub> chemistry sensitivity are dominated by HCHO differences. Additionally, opposite to the ozone season versus exceedance day analysis, we find no significant differences in 2-meter air temperature and 10-meter wind speed, direction, and divergence between weekdays and weekends.

(9) Section 4: it is not quite clear to me if there are any new insights gained from this study. Perhaps authors can emphasize any new results in the conclusions (and the abstract as well).

### Response

We thank the referee for this comment, but we respectfully disagree due to the following:

- This work is important because the Lake Michigan region contains Chicago, which is one of the largest metropolitan areas in the United States that experiences ozone nonattainment associated with lake breeze circulations. The Lake Michigan region as a whole has been understudied compared to other regions (e.g., the Northeast U.S./New York City), especially regarding the use of satellite data. To our knowledge, our study is the first that utilizes TROPOMI data to assess changes in FNR values and inferred ozone chemistry sensitivities between ozone exceedance days and average ozone season days for the Lake Michigan region as a whole.
- One particularly new insight gained is that NO<sub>2</sub> concentrations appear to be concentrated in the urban core of Chicago on exceedance days due to the convergence of the wind field along the western Lake Michigan coastline. This result was discovered by connecting changes seen in the satellite based results to wind

divergence/convergence values calculated from model analysis meteorological data, which is a methodology not often performed in FNR studies. This study demonstrates the potential of using the model analysis wind data included in the TROPOMI data files to gain new insights into the transport patterns underlying the changes in chemical vertical column densities observed by TROPOMI.

- Another insightful finding is the possible high HCHO artifact over the Great Lakes, which deserves further investigation (already highlighted in the conclusions).
- Our results are comparable to another study conducted for New York City, which suggests that the results are applicable to other coastal urban environments with O<sub>3</sub> exceedance problems (already highlighted in the conclusions). Our study can be the basis of future work for other researchers who wish to conduct similar analyses for their areas of interest.

# Changes to manuscript

To emphasize the value of this study and highlight some of the new insights gained, we added the following to the abstract:

Lines 14-16

Despite being a highly populated region with coastal O<sub>3</sub> air quality issues, the Lake Michigan region in the United States, including the Chicago, Illinois, metropolitan area (CMA), remains relatively understudied, especially from the satellite perspective. In this work, we present the first study that utilizes TROPOspheric Monitoring Instrument (TROPOMI) satellite data over the Lake Michigan region...

# Lines 23-29

Utilizing 10-meter wind analysis data, we show that the lake breeze circulation is stronger on exceedance days. The strengthening of the lake breeze causes stronger convergence of the wind field along the southwestern Lake Michigan coastline, which can concentrate NO<sub>2</sub> emissions originating in this area. This finding provides a possible explanation for the higher TROPOMI NO<sub>2</sub> VCDs over the urban core of Chicago on exceedance days. Investigation of 2-meter air temperature analysis data reveals that temperatures are higher on exceedance days, which explains the stronger lake breeze circulation and provides a possible cause for the higher TROPOMI HCHO VCDs over the entire region (due to increased temperature dependent biogenic VOC emissions).

Similarly, we added the following to the conclusions:

Lines 539–543

Despite being a highly populated area that experiences coastal O<sub>3</sub> air quality problems, the Lake Michigan region is relatively understudied, especially from a satellite perspective. To address this research gap, we created mean formaldehyde to nitrogen dioxide ratio (HCHO/NO<sub>2</sub>; "FNR") composites using 2019–2021 S5P TROPOMI

satellite data over the Lake Michigan region to assess changes in ozone precursor levels and the inferred O<sub>3</sub> chemistry sensitivity between: (1) O<sub>3</sub> season days and Chicago metropolitan area (CMA) O<sub>3</sub> exceedance days, and (2) weekdays and weekends.

Lines 552-560

Ten-meter wind analysis data shows that the lake breeze circulation along the southwestern Lake Michigan coastline is stronger during CMA exceedance days, which causes stronger convergence of the wind field and the concentration of NO<sub>2</sub> emissions originating in the area. Thus, the strengthening of the lake breeze is a possible cause for the higher TROPOMI composite NO<sub>2</sub> VCDs in the urban core of Chicago on exceedance days. This analysis demonstrates the potential of using the model analysis wind data included in the TROPOMI data files to gain new insights into the transport patterns underlying the changes in chemical vertical column densities observed by TROPOMI. Both higher TROPOMI HCHO composite VCDs and the stronger lake breeze can be explained by higher temperatures on exceedance days, which we showed to be true using model analysis 2-meter air temperature data.

# **Response to Referee #4**

## **Referee's introductory comment**

The revised manuscript is much improved over the original version. Major areas of improvement include the following:

- Use of PAMS data has been eliminated from the manuscript. The 6-day integrated HCHO measurements are of little or no value for use in HCHO/NO2 ratio analysis.

- Explanation of the uncertainty in use of the J20 criteria values for NOx sensitive and VOC sensitive regimes has been included. Use of these criteria are especially justified in examining high O3 exceedance days.

- The same number of weekday and weekend days are now used in the analysis.

- It has been demonstrated that useful results are obtained even though the analysis period covers the Covid-19 pandemic lockdown. The same general results for ozone sensitivity are found for each of the three years.

- Section 2.4 has been added, which covers the J20 criteria uncertainty, the differences in OMI vs. TROPOMI, and the Covid-19 effect.

It is good to see that reprocessed TROPOMI NO2 data have been used in the revised version of the manuscript. Now all NO2 data used in the calculations are from the same version of the retrieval algorithm.

The paper should be published after minor revision.

## **Response to introductory comment**

We thank referee #4 for reviewing our revised manuscript. Based on both referees' comments, we have made the following new revisions to the manuscript:

- 1. Adding an analysis to estimate how much of the differences between the ozone season and exceedance day composites are due to intra-seasonal variations versus episodic changes on exceedance days,
- Expanding discussions of: (a) possible satellite retrieval biases due to vertical profile and temperature differences on exceedance days, (b) the possible impact of the coarse albedo climatology dataset on the TROPOMI high HCHO artifact over the Great Lakes, and (c) "noise" levels of 4 km and 12 km versions of the TROPOMI composites,
- 3. Including a comparison of AQS surface NO<sub>2</sub> measurements to our TROPOMI-based results, and
- 4. Emphasizing new insights gained from and the value of this study in the abstract and the conclusions sections.

Our response to referee #4's specific comment is as follows:

## **Referee's specific comment**

(1) Concerning the larger NO2 seen over the whole domain on ozone exceedance days: I would recommend that the authors determine if surface NO2 monitoring network data show the same result. This analysis could very easily be done.

# Response

We agree that looking at surface NO<sub>2</sub> monitoring network data would be a great comparison to the TROPOMI-based results.

We analyzed U.S. EPA Air Quality System (AQS) surface NO<sub>2</sub> data at 13:00 local time (approximately matching the TROPOMI overpass time) for 8 monitoring sites within the study domain. These 8 sites were selected as they were the only ones that had data for our entire 3-year study period from 2019–2021.

### **Changes to manuscript**

We added **Table S5** in the supplemental information document that compares the ozone season mean values to the exceedance day mean values. For 7 of the 8 sites, surface NO<sub>2</sub> levels are higher on exceedance days compared to the ozone season. When the 8 sites are averaged together, we find that surface NO<sub>2</sub> levels on exceedance days are higher by about 19 % compared to the ozone season average. This closely matches the TROPOMI composite results in **Figure 3c**, which shows a domain-wide average NO<sub>2</sub> vertical column density increase of about 21 % on exceedance days:

	AQS surface NO <sub>2</sub> : ozone season vs. CMA exceedance days					
Site*	Ozone season mean (OS) [ppb]	Exceedance days mean (Ex) [ppb]	Ex – OS [ppb]	Ex % change from OS		
A (17_31_119)	13.01	17.69	+4.68	+36 %		
B (17_31_219)	8.94	10.21	+1.27	+14 %		
C (17_31_3103)	10.77	13.16	+2.39	+22 %		
D (17_31_4002)	6.65	7.20	+0.55	+8 %		
E (17_31_76)	5.65	5.98	+0.33	+6 %		
F (18_141_15)	2.17	1.45	-0.72	-33 %		
G (18_89_22)	2.71	2.76	0.05	+ 2 %		
H (55_79_56)	7.69	9.04	1.35	+18 %		
All sites	7.27	8.62	1.35	+19 %		
FROPOMI composite – full domain	1.75 molec. cm <sup>-2</sup>	2.11 molec. cm <sup>-2</sup>	0.36 molec. cm <sup>-2</sup>	+21 %		

We additionally included a map in the supplemental information document (**Fig. S1**) that shows the locations of the 8 surface monitors:



Finally, in the main text we added the following sentence to Section 3.1.2 on lines 315–317:

We note that surface NO<sub>2</sub> observations at 13:00 local time from the average of eight AQS monitoring sites also indicate higher NO<sub>2</sub> levels on CMA exceedance days compared to the average across all O<sub>3</sub> season days during the 2019–2021 study period (**Table S5; Fig. S1**).