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Atmospheric Measurement Techniques Discussions

Interactive comment on "Traffic-related air pollution near roadways: discerning local impacts from background" *by* Nathan Hilker et al.

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Response to Anonymous Referee 1

Of course, it makes a big difference if a roadside station is located 6 m away from the nearest traffic line or 10 m. Actually, any different position may lead to different concentrations, because the variability of air pollutant concentrations varies largely spatially. The authors are aware of that. However, the tone sometimes suggests that the results (concentrations!) are transferable to other locations or situations. For example, the last sentence of the abstract ("Downwind conditions enhanced local concentrations by a factor of 2 relative to their mean, while upwind conditions suppressed them by a factor of 4") is, one the one hand, perfectly fine. On

the other hand, there is no caveat saying: "This applies to this very specific situation, don't generalize!" Also, referring to lines 289-299 and Table 2, the absolute number of C_L are not comparable to each other between sites, even though drastic differences between the sites are apparent. The authors are asked to go through their manuscript and find more cautious wording in this respect.

Reply: Thank you for underlining this point, and we agree that the wording should be as explicit as possible so as to not imply generalizability where it may not be applicable. The expectation with this analysis is that, since the local components of the concentrations are normalized with respect to their mean values, the shape of the curves of these normalized concentrations w.r.t. wind direction and wind speed ought to be somewhat generalizable for receptors near roadways with varying rates of emission. I.e., the areas above and below unity for these curves is always equivalent thanks to the following property:

$$\int_0^N \left(\frac{x(\theta)}{\bar{x}} - 1\right) \cdot d\theta = \frac{1}{\bar{x}} \cdot \int_0^N x(\theta) \cdot d\theta - \int_0^N d\theta = \frac{N \cdot \bar{x}}{\bar{x}} - N = 0$$

However, as is pointed out, the distance of the receptor from the roadway along with the height of the sampling inlet will almost certainly impact the shapes of these curves, even if they are less impacted by source strength.

Action: The manuscript will be revised carefully to be as explicit as possible in its wording to properly address this caveat.

In the eyes of this reviewer, the data set allows much more interesting analysis *of emission factors, for example between NOx and CO2. How do NOx/CO2 ratios or UFP/CO2 ratios compare o the results of similar studies? Similar applies to CO. It is however acknowledged that this is outside the scope of this study.*

Reply: Yes, we agree. In fact, emission factor analysis from this study has already been performed and reported by Wang et al. (2018) (see: doi.org/10.1021/acs.est.8b01914). This was the originally intended use for the background subtraction algorithm.

Action: The text will be updated to refer readers to this analysis.

Section 4.3 and Table 4: Why not did authors apply his method for ozone? When using maxima instead of minima for the time series analysis this should be no problem to do. The justification given in lines 362-364 is no convincing. The urban background concentration of O3 could have been quantified that way, and be compared with the respective results of methods 1 and 2.

Reply: This is a great suggestion and results will be updated accordingly to include it. For added simplicity, the same method of rolling minima interpolation can be applied to -1*O3(t), and once calculated the sign can be flipped again, thereby allowing the same algorithm to be applied. Figure 1 shows an example of this algorithm applied to O3 at NR-TOR-2. We can see that the difference between near-road O3 and inferred background is generally greatest when there are larger concentrations of NOx, which should be expected.

Eq. 2 seems screwed. Probably, a parenthesis is missing on the right-hand side, opening before $C_{NR}[i]$ and closing after $C_{BG}[i]$.

Reply: Agreed.

Action: Changed accordingly.

Line 187: A justification should be given as of why M and N are typically not identical. It is a bit counterintuitive.

Reply: The reason why M and N are typically not identical is that there will be a prevailing wind direction at most air quality monitoring locations. For example, if sampling is done continuously and data are not excluded based on wind direction, it should be expected that downwind data will occur more frequently than upwind data, for example, if it aligns with the prevailing wind direction of the site. The important point here is that N and M constitute sets of data that are inherently mutually exclusive (i.e. one cannot sample upwind and downwind of a road simultaneously with a single receptor) and may occur under different conditions (e.g. time of day).

Action: The text will be updated to elaborate on this point as it is perhaps not obvious in its current state. Further, wind rose plots will be included for each near-road location in the SI.

Line 194: Why did you chose 75 % here? Likely, the results are more reliable if 100 % is used. See also line 413.

Reply: Thank you for pointing this out. This was actually a mistake in the text. Hourly averages in general were included only if $\geq$ 75% of minutely data were available, and this applied also to the meteorological data. For classifying whether a given hour was downwind or upwind, the hourly vector averages were used directly.

The only hours omitted were stagnant hours in which the wind speed was < 1.0 [m/s].

Action: The text will be corrected accordingly.

The PM2.5 results are puzzling indeed. It could be the precision and accuracy of the analyzers not being able to resolve the small differences in concentrations between stations and within time series. If so, the results are not statistically robust. This issue should be analyzed in more detail and presented and discussed in the manuscript.

Reply: You raise a good point regarding the precision between instruments, especially the PM2.5 monitors, and whether the background-subtraction methods are able to distinguish what is likely a very minor contribution to the total near-road signal. Regarding PM2.5 specifically, more in-depth results have been reported already by Sofowote et al. (2018) at NR-TOR-1 using an Aerodyne ACSM, XACT 625, AE33, and SHARP 5030, and their findings suggested the major component of PM2.5 responsible for these “local” fluctuations was black carbon, which is measured also using an AE33 here. Indeed, Table 2 would also suggest that BC is a major subset of this “local” PM2.5.
The SHARP 5030 manual specifies an hourly precision of “$\pm$2 μg/m3 < 80 μg/m3; $\pm$5 μg/m3 > 80 μg/m3”, and a precision between two monitors of “$\pm$0.5 μg/m3 (2-σ, 24-hour time resolution)" (Thermo Scientific, 2013). So, the average site differences between near-road and background sites, which are all around 2 μg/m3 or less and calculated over 2 years of data, are likely statistically significant results (Table 2). However, this raises an important issue as to why methods of background-subtraction applied to an hourly near-road time-series fails to properly pick out the local component: if the average local component is 2 μg/m3, and the hourly precision of the instrument is $\pm$2 μg/m3, then the signal-to-noise ratio of this local component on an

hourly time scale is likely quite small and perhaps not detectable.

Action: This raises an important caveat regarding instrument precision limits and the application of background-subtraction algorithms to near-road data. Naturally, it brings to question the precision of all other instruments used in this study. Thus, instrumental precision will be reported in the methodology section, and reasons why the background subtraction algorithm for PM2.5 has seemingly failed will be discussed in the results.

Thank you for the feedback!

References

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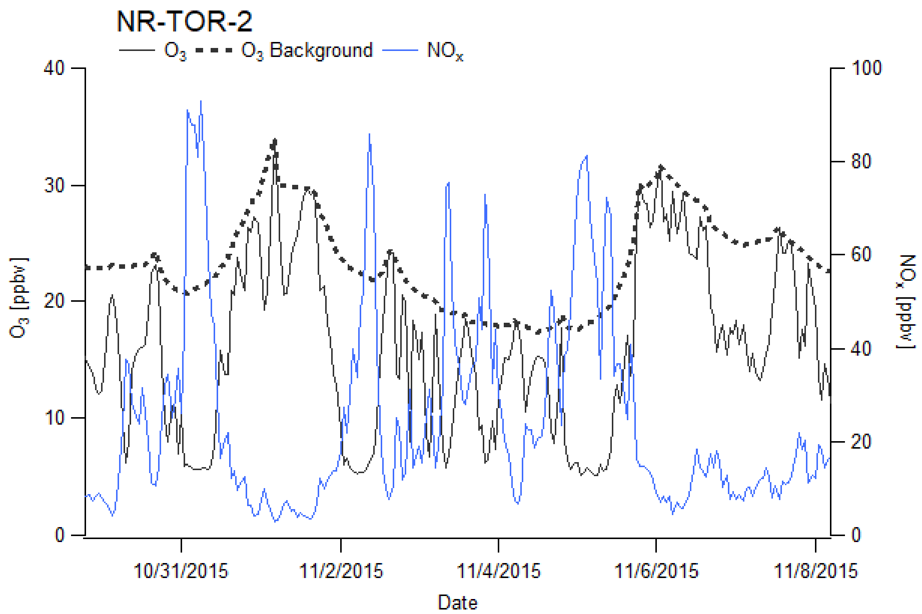

Fig. 1. Example of background-subtraction algorithm applied to ambient O3 concentrations at the near-road downtown Toronto site, NR-TOR-2, wherein a rolling maximum is calculated rather than a rolling minimum