

Interactive comment on “The impacts of precursor reduction and meteorology on ground-level ozone in the Greater Toronto Area” by S.C. Pugliese et al.

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We thank the review for their consideration of our manuscript. Our response to their comments are below (original comment in small indented text).

Major Comments

1. It would be more appropriate to analyze trends in terms of %/year changes. This is likely to better fit the data and better represent the underlying relationship, and would allow for easier comparison across sites and pollutants.

We agree that it would be more appropriate to analyze trends in terms of percent per year changes; this has been updated in Section 3.1 and throughout the rest of the manuscript.

2. Results are presented primarily in terms of summertime averages, which is appropriate. However, two other temporal metrics are of interest as well: (1) fourth-highest daily max 8-h ozone, to better align with the form of the Canadian regulatory standard, and (2) looking beyond summer, since some studies indicate health impacts of ozone extend to low concentrations and sensitivity relationships are likely to be very different in other seasons. In addition, it would be informative to comment on differences between conditions on high and low ozone days within the summer.

Thank you for the suggestion regarding the use of other temporal metrics.

(1) We do agree that there is value in using a metric that aligns with the Canadian regulatory standard. In Figure 4 we include the Design Value for each year and we think this is adequate to give the reader a sense of how the Canadian regulatory standard is being met (or exceeded). We feel that using summertime averages allows us to analyze correlations between ozone and meteorological parameters (for example: when air is from the W-NE, ozone concentrations are typically lower than when air is transported from the W-SE) and these correlations would not be visible if we solely looked at the 4<sup>th</sup> highest daily max 8-hr ozone.

(2) It is true that the health impacts of ozone can extend to low concentrations (indeed this supports our focus on summer averages above) and therefore other seasons could be considered. However winter ozone levels in the GTA are generally much lower between October and April, and the balance of factors governing its abundance may be very different than during the summer months. For example, throughout the 13 year study period, Toronto North exceeded the 65 ppb 8-hr maximum once in the non-summer seasons in 2007 and 2008, with no exceedances for all other years. We chose to focus the scope of our analysis of chemical and meteorological factors on the most photochemically active time of the year.

To discern the differences between conditions of high and low ozone days within the summer, we considered a number of meteorological parameters (such as temperature, wind direction and incoming solar radiation) and found, for example, that days with wind from the W-SE and/or with higher levels of incoming solar radiation, ozone concentrations were higher (Figures 5 & 6).

3. Despite its usefulness in some other contexts, it's unclear that  $O_x$  is an appropriate metric for characterizing long-term trends in ozone, especially given the ozone regulatory context. Here,  $NO_2$  declined far more steeply than  $O_3$ , so most of the  $O_x$  trend reflects reduction in  $NO_2$ .

We think the reviewer raises an interesting point and agree that  $O_x$  may not be an appropriate metric given it reflects the large reductions in  $NO_2$ . We have changed our emphasis to focus on the trends of ozone, not  $O_x$ , during the 13 year study period. This includes a new revised discussion on the percentage decreases of ozone during the study (in Section 3.1) as well as focusing the net wind vector analysis on ozone concentrations.

4. In Section 3.4, is there evidence to indicate where the transition lies between  $NO_x$ - and VOC-limited ozone formation regimes, in terms of the ratios of OH reactivity to  $NO_2$  and to VOCs that are reported here?

A study done in 2009 by Geddes et al. analyzed long-term changes in nitrogen oxides and VOCs in Toronto and used a computational model to generate a contour plot of instantaneous ozone production as a function of VOC reactivity and  $NO_2$  concentrations. This contour plot was produced under conditions of optimal ozone production. According to this plot, in order for Toronto to be  $NO_x$ -limited,  $NO_2$  concentrations must be below  $\sim 2$  ppb and VOC reactivity can range from  $\sim 1$ - $6$   $s^{-1}$ . While for the suburban sites, such as Newmarket, some hours of the day reach these low levels of  $NO_2$ , averaged over the midday and throughout the summer,  $NO_2$  concentrations are higher than this and therefore production of ozone is consistently on the VOC-limited side of the contour plot at all eight sites. This is a result consistent with Figure 7 in the manuscript which outlines in the "late" period of the study, the GTA is in a VOC-limited regime. This information has been included in the manuscript (Section 3.4, lines 361-364).

5. The text and Figures 2c have helpful delineation between types of VOCs, but then it becomes unclear which VOCs are being included in Table 2, Figure 7 and some portions of the text. This should be clarified.

We thank the reviewer for pointing this out, we have clarified in the text that the original suite of  $\sim 40$  NAPS compounds are included in Table 2 and Figure 7.

6. The authors adopt a thoughtful technique to use VOC ratios to gauge trends in OH, and find evidence for increasing OH. Was this expected? Has modelling been conducted to examine whether it predicted a rise in OH under large-scale  $NO_x$  and VOC reductions as have occurred in Toronto?

Yes, an increase in OH concentration throughout the study was expected. Assuming that the production rate of OH has not changed significantly, which is reasonable if ozone has stayed nearly constant and  $O^1D + H_2O$  is the main OH source, then a large decrease in the sinks of OH (VOC and  $NO_2$ ) would likely lead to an increase in its concentration. To our knowledge, no modeling has been conducted to examine whether a predicted rise in OH under large-scale  $NO_x$  and VOC reductions has occurred in Toronto.

### Minor Comments

- p. 10211, line 3: Pointing to a \$9.6 billion impact in Ontario is somewhat misleading in a paper focused on ozone, since most of that impact presumable arose from particular matter.

We thank the reviewer for this suggestion. To ensure the reader is not misled and to highlight that ozone is responsible for only a *fraction* of the \$9.6 billion in economic losses, we specified that the 2014 Toronto Public Health Burden of Illness Update reports that ozone is responsible for 14 % and 29 % of the premature mortality and hospitalizations, respectively, in the city (Section 1, lines 40-43).

p. 10222, line 1: change “was” to “were”

We have changed “was” to “were”.