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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Background ozone over Canada and the United States" *by* E. Chan and R. J. Vet

Anonymous Referee #1

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General comments:

The manuscript presents diurnal, seasonal, and decadal trends (by season and region) in observed ozone for several regions across the United States and Canada. Ozone data at each measurement location are classified into six clusters based on trajectory analysis. The cluster with the lowest and highest 95th percentile ozone values are taken to represent" background" and "most polluted" conditions, respectively Principal component analysis (PCA) is applied, separately for each season, in order to group sites into regions with similar variations in 6-hour average ozone. This sort of data analysis is useful and adds new information to a growing body of work attempting to diagnose trends in surface ozone under both background and polluted conditions. Identifying trends in observed ozone distributions under these conditions at the broad

suite of locations examined here is an important step towards better understanding how, and ultimately why, the ozone distribution has been changing in recent years.

My main concerns are with the approach and interpretation of results:

1. The approach defines air masses associated with one wind direction that contain the lowest 95th percentile ozone values as representing "background" conditions, for each site. More support is needed for the choice of the "lowest 95th percentile". Are the conclusions robust to using other metrics (e.g., median)? The trajectory-based approach could select for air masses with chemical (titration by NO) or depositional (exposure to a forested area) processing that do not necessarily reflect a hemispheric-scale background, though the second main message of the conclusions implies that this is not occurring: "background ozone teased out from the entire non-urban dataset are spatially consistent and possibly hemispheric in nature". This statement seems to conflict with discussions elsewhere in the text (e.g., Section 4.4 and first paragraph of conclusions) and in prior work (e.g. Parrish et al 2009 and others) discussing seasonal and regional variability in background. More problematic, Parrish et al [ACP, 2009] find that "it is not adequate to simply select a wind direction window" to remove continental emission influences at several sites they examine for trends in background ozone; their conclusion seems supported by the results here and should be addressed.

2. The PCA to group sites that vary similarly was based on 6-hour average ozone data at each site. These groupings are then applied for "clean" and "polluted" air masses. I don't understand why the PCA isn't applied to the dataset after screening based on trajectory analysis (if the trajectory analysis is effective in separating background and polluted conditions). Might "background" variability be similar across a wider grouping of sites? The figures indicate that this does occur in some cases (e.g., PC1 and 5 in Fig 8a). In contrast, PC13 groups 2 sites with different seasonal cycles, suggesting some problems with the approach.

3. More generally, the current framing of the manuscript is confusing by defining "back-

ground" one way in the introduction, and then using a different definition for the analysis without clearly acknowledging the transition. The introduction defines background as "the combination of naturally and anthropogenically produced ozone from outside of Canada and the US, plus naturally formed ozone within Canada and the US" whereas the first main message in the conclusions states "decreasing trends must be due to regional ozone decreases caused by decreases in ozone precursor emissions since the mid-2000s" which implies that anthropogenic emissions inside the US (known to have decreased) contribute to "regional background" . More precise language e.g., referring to the metrics used, in place of "background", might begin to reconcile the contradictory statements throughout the text.

Specific comments:

Abstract. The first sentence could include that this analysis uses observations. The discussion on the variations in trends across seasons and regions could be expanded since these are the central findings of the study.

Section 1.2. This section should make a clearer distinction between surface ozone (the focus of this study) and tropospheric ozone (relevant for climate forcing). I disagree with L18 since work to date is not clear on whether tropospheric ozone will increase or decrease in a warmer climate as increased water vapor shortens the ozone lifetime [Johnson et al, Journal of Geophysical Research-Atmospheres, 1999 and other studies]. The increase in tropospheric ozone in the IPCC chapters cited is largely driven by increasing NOx and CH4 emissions rather than a climate feedback [e.g. Prather et al, Geophysical Research Letters, 2003].

Section 1.3. The results shown later (e.g., Section 4.1) suggest that it is not possible to separate out the background as defined here from measurements, so it seems misleading to state that this is the definition used here. This is addressed somewhat in Section 1.5 (L6-8 p. 21115). It would be better to address the differences in definitions clearly in one place.

Section 1.7. The previous studies have to some degree addressed the questions laid out here and could be acknowledged (or combine this section with 1.6). Section 1.8. L24-26 is incorrect. Previous work (those studies cited, as well as Parrish et al., 2009; the Jaffe et al studies cited; Lin et al., 2000; Wang et al., 2009) does address seasonal and day-to-day variability. Parrish et al also address the diurnal cycle. Further, several studies referenced examined background ozone across the entire United States: e.g., Lin et al., 2000; Fiore et al., 2002 and 2003; Wang et al 2009. Lefohn et al, 2001 (Journal of Geophysical Research-Atmospheres) should be included.

Section2. Why are 6-hour averages used for the seasonal but then only daytime 6-hour averages for decadal analysis? More explanation should be given for the expected influence of temperature on trends in the background . How does the trajectory model represent turbulent mixing within the PBL and convective mixing?

Section 3.2. How was the 75% minimum total variance arrived at?

Section 3.5 What is the percentage of data retained for the decadal trends analysis? Does this population of background air masses contain ozone levels that tend to fall in a certain percentile range of the overall observed distribution at a site for a given season? The physical meaning behind the choices of fitting parameters (one day autoregression; 3-5 year periodicities) should be discussed. Section 4.1 P21122 L2-3. This is confusing as the introduction states that anthropogenic emissions are not included in the background. The limitations of the approach need to be more clearly laid out from the beginning, including efforts taken (e.g., using the diurnal cycle as a diagnostic for local influence makes sense) to determine the extent to which the approach is successful in isolating background ozone.

Section 4.3. Given the lack of difference between C4 and C6 in the Egbert example, why not group these together?

Section 4.4 The first sentence doesn't seem to address seasonal variations (the section title). More generally, this question seems somewhat ill-posed if the background

and ozone produced from domestic emissions are considered earlier to be separate contributions to the ozone distribution.

Section 4.5. The logic behind PC9 defining an upper limit needs to be more clearly laid out.

Section 4.6. In the final paragraph, are fires considered part of the background? Jaffe et al (Environmental Science and Technology, 2008) is also relevant.

Section 5. Some of the statements here have been concluded in prior studies (e.g., variability in background is discussed e.g. in Lefohn et al, 2001 and Fiore et al 2003, both in Journal of Geophysical Research-Atmospheres), though this study certainly extends beyond prior work. More generally, some rewriting is needed to address the three major concerns above.

Technical Comments

P21121 L 16-21. Consider illustrating with an example from one region.

Section 4.2 This section should be shortened and clarified. The key point seems to be

P21124 L1-3 and that local photochemistry dominates the regional variability

Table 1. Are the ranges representing spatial averages over different years, or seasonal means at different sites within the region? Is the diurnal range spatially and seasonally averaged?

Table 2. Is this for the subset of data selected for the trajectory cluster with the lowest 95th percentile ozone?

Figure 1. Symbols need to be associated with groups to facilitate interpretation of Table 1.

Figures 6-9 are difficult to read. The numbers on the map should be explained. I don't see the need to repeat the population density, site groupings, or altitude figures

multiple times – these could probably be condensed into 1-2 Figures (e.g., with Figures 1 and 2). That would leave more space for legible panels. What are the multiple lines representing in Figures 7 -9 panels?

Figure 10. How are the shapes determined for the regional boundaries? What are the numbers corresponding to (this applies to other figures too)? The definition of "cleanest air" should be given in the captions.

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