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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Trends in emissions and concentrations of air pollutants in the lower troposphere in the Baltimore/Washington airshed from 1997 to 2011" *by* H. He et al.

Anonymous Referee #1

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SUMMARY & OVERALL RECOMMENDATION. A paper of very high interest to the ACP community combining emissions inventories, some satellite data and aircraft profiles to conclude that large (50% or more) decreases in NOX and CO emissions in the Ohio/Penna region (mostly) with minor contribution from local source reductions have led to significant tropospheric ozone declines in the B/W region. The latter are given in terms of of 2/3 ozone production decrease (in ppbv ozone/day), column amount and absolute near-surface ozone loss (13 ppbv/decade). Take-home messages are (1) emission controls are effective in reducing ozone in the mid-Atlantic; (2) the losses in ozone are roughly consistent with reductions in reported CO and NOX emissions; (3) trajectory analyses coupled with B/W ozone losses suggest the reduction of NOX

in the Ohio/Penna power-plant source region is primarily responsible for the ozone exceedance improvement that is observed over the 1997-2011 period. The analyses are conducted carefully and logically and presented with appropriate support, uncertainties and interpretation. The paper should be published after careful revision. It could be more tightly organized, some new studies (from 2011 DISCOVER-AQ, recently published or in press) should be cited to strengthen background and interpretation; minor corrections are needed for facts, grammar (see "Minor Points" below). The Abstract does not capture the above enumerated points as effectively as it could and should in order that the work carries full impact. The caveat, noted in the paper (see comment below about ozone decrease), that values reported here reflect the sampling bias of the aircraft (toward the most polluted summer conditions), should appear in the Abstract. Indeed, one aspect of confusion in the paper is use of annually averaged emissions amounts and some figures that appear to have continuous or full-year information, when the RAMMP (page 3139) observations come from basically a summertime sampling/modeling project. The results appear to be internally consistent in this respect but the distinctions of summer vs annual need to come out in the Abstract. This could be a really high-impact paper!

SPECIFIC COMMENTS/RECOMMENDATIONS.

Abstract. See general comments above to guide a revision. The CO and ozone reductions are described but analysis and text give ample space to NOX decreases since 2003 as a main driver for ozone improvements. NOX is not even mentioned in the Abstract! Likewise, text shows the importance of including the CPF in trends analysis and this is not mentioned either.

INTRODUCTION. This is somewhat disjointed. A lot of references are listed without mention of the main points relevant to the current study. In particular, a number of RAMMP-related studies are listed. The context from them would be useful to placing the current study in perspective. Which are most relevant? Which gave previous background on ozone, CO or NOX in the B/W region that relate to the current paper?

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Expand as appropriate. If other papers from the UMD group and beyond are less essential, omit them. Somewhat in contrast to the above request to trim, it is recommended to include recently published and (in press) papers from DISCOVER-AQ that are relevant to the current paper because (1) conditions like sea-breeze that affect part of the B/W domain are analyzed; (2) profile data from aircraft and sondes relevant to the RAMMP studies are described and in some cases compared to satellite and ground-based columns. Specific papers as follows: Martins et al., JGR, 2012 (on Hampton, VA sea-breeze); Martins et al, J Atmos Chem, 2013; Reed et al, J Atmos Chem, 2013; Stauffer et al., J Atmos Chem, 2012; Stauffer and Thompson, J Atmos Chem, 2013. D. K. Martins, R. M. Stauffer, A. M. Thompson, M. Pippin, T. Knepp, Surface ozone at a coastal suburban site in 2009 and 2010: Relationships to chemical and meteorological processes, J. Geophys. Res., 117, D05306, doi: 10.1029/2011JD016828, 2012. R. M. Stauffer, A. M. Thompson, D. K. Martins, R. D. Clark, C. P. Loughner, R. Delgado, T. A. Berkoff, E. C. Gluth, R. R. Dickerson, J. W. Stehr, M. A. Tzortziou, A. J. Weinheimer, Bay breeze influence on surface ozone at Edgewood, MD, during July 2011, J. Atmos. Chem., doi: 10.1007/s10874-012-9241-6, 2012 D. K. Martins, R. M. Stauffer, A. M. Thompson, H. S. Halliday, D. W. Kollonige, E. Joseph, A. J. Weinheimer, Ozone correlations between upper air partial columns and the near-surface at two mid-Atlantic sites during the DISCOVER-AQ campaign in July 2011, J. Atmos. Chem., revised in review, 2013. A. J. Reed, A. M. Thompson, D. E. Kollonige, D. K. Martins, M. A. Tzortziou, J. R. Herman, T. A. Berkoff, N. K. Abuhassan, A. Cede, Effects of local meteorology and aerosols on ozone and nitrogen dioxide retrievals from OMI and Pandora spectrometers in Maryland, USA during DISCOVER-AQ 2011, J. Atmos. Chem., in press, 2013. R. M. Stauffer and A. M. Thompson, Bay breeze climatology at two sites along the Chesapeake Bay from 1986-2010: Implications for surface ozone, J. Atmos. Chem., joch-12-0508, in press, 2013. A. M. Thompson, R. M. Stauffer, S. K. Miller, D. K. Martins, E. Joseph, A. J. Weinheimer, G. S. Diskin, Ozone profiles in the Baltimore-Washington region (2006-2011): Satellite comparisons and DISCOVER-AQ observations, J. Atmos. Chem., joch-13-0007, submitted, 2013.

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SECTION 2. Page 3141 lists a number of satellite instruments but some of these are not relevant to the current study. MOPITT is and SCIAMACHY trends in NOX may be. TES? IASI? AIRS? Incidentally, there is an error; AIRS is on Aqua not Aura (line 14).

Page 3141- Lines 24-28 - this is confusing - can you simply refer to the MOPITT version and characteristics in the data as used in the present study?

Page 3142. Mentioned are 1000 flights along east coast but only a certain # in Balto/Wash area. Specify how many profiles used in the current analysis.

Page 3143. Although not as large in domain as the LLJ, sea breeze has a large impact at some B/W sites. At line 9, suggest revising to .. Two important transport processes in the mid-Atlantic region that affect summertime pollution are the low-level jet (LLJ) and sea-breeze. The LLJ (Corsemeier, etc) transports ozone and its precursors from the south. .... Due to the proximity of the Chesapeake Bay to Baltimore, sea breeze is significant in certain locations within the B/W study region (Stauffer et al, 2012; Stauffer and Thompson, 2013; Thompson et al., 2013). Indeed, as emissions of ozone precursors and as shown below, ozone itself, decline significantly in the mid-Atlantic region, the presence of sea-breeze appears to play a decisive role in air quality exceedance episodes (cf Martins et al., 2012).

SECTIONS 3 & 4. MINOR POINTS (Sections 1-2) . Page 3138 Line 20 delete (and ozonesondes, etc) - Four years in Yorks et al is not “long-term” and extension of the B/W (Beltsville) record in Thompson et al (2013) is inconclusive (still only 8 years total).

Page 3139 Line 22 study “of” not “on”

Page 3139 Line 24. Spell out “years”. Because so many studies and data have been referred to, be explicit after this sentence by stating “Federal (or national) emissions information and RAMMP aircraft information collected by the UMD airborne group form the bases of the investigation” or words to that effect.

Page 3139 Line 25 spell out “section 3"

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page 3141 - Line 7 insert ‘of “the” actual ambient....’

page 3143. Line 7 Do you mean on a typical westerly transport “day” or “conditions” ? looks as if a word is missing

Page 3144. Lines 24 and 25 - reference(s) needed for the decline in emissions

Page 3145 for the entire US; (need period or semi-colon) consequently, (need comma)

Page 3147 Lines 20-24. You mention “complicated” chemistry as responsible for a less than expected ozone reduction. How about the CPF? Is this the best place to mention or reference Bloomer et al for the first time [it appears on page 3149]?

Page 3148 Line 12. End sentence “previous days” with reference to Yorks et al (2009)

Page 3148 Lines 17-19. Grammar. Air quality; however, ...

Page 3150 Lines 18-24 - this is a major point of the paper, ie column ozone cleanup is greater than surface (!) But this figure might be biased by sampling conditions of flights. This key take-home message is *also* missing from Abstract.

Page 3151 Lines 25-27. End sentence before 2011. Thus, the hypothesis of... [exhaust is singular]... verified. Omit “and future studies...”

Page 3152 Lines 15-18 The argument about magnitude of CO reduction makes sense but does this assume that VOC oxidation to CO is not a significant source in this biogenically rich area (given that mobile VOC emissions have also probably declined)?

Page 3154 Last line - improved, not improving

Page 3155 Line 1 - declined, not decline

Page 3155 Line 14 ..quality days improved..” (Delete “has been”)

Page 3155 Line 29 from “the” USEPA NEI....

Pointer - Check Figure labels and captions carefully. Figure 2a - especially illegible

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Figure 3 - caption should mention observed in the B-W area “based on ... number of sites or databases used.” Because so many possible sources of data are listed in Section 2, data sources should be listed in figures (as, for instance, in Figure 4)

Figures 5 and 6 - tons/day - clarify the unit

Figure 7 - illegible without a lot of on-screen magnification

Figure 9 - bubble size relates to # of measurements but no scale given

Figure 10 - column contents from aircraft (xx to yy km range, depending on year). Add this information to caption

Figure 11 - panel c (lowest) presumably NEI CO emissions. Be explicit

Figure 13 - caption grammar - do you mean ?was conducted with ozone columns vs ..

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 3135, 2013.