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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 #2

Received and published: 15 March 2013

Overall I find the science and analysis conducted for this manuscript to be acceptable (with a few minor exceptions described below) and I think that the information provided will be valuable to the scientific community. I recommend that the authors conduct a major revision of the manuscript according to my comments below.

Before I begin my detailed review of the manuscript I will first list two general complaints.

1) My first complaint is directed towards the ACPD policy on copy editing and I ask the editor to pass my comment on to the relevant department. I have been a peer-reviewer for ACPD for 10 years and I have complained many, many times in the past that some

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of the figures are too small to read. For this paper Figures 2 and 7 are far too small. Illegible figures make the review process very difficult and ACPD needs to implement procedures that ensure all figures are legible. This is a complaint that I rarely make to other journals such as JGR, Atmospheric Environment or Nature.

2) My second complaint is directed towards the co-authors of this manuscript. The standard procedure for submitting a manuscript to a peer-reviewed journal such as ACPD is that all co-authors read the manuscript and correct any errors prior to submittal. The manuscript is not to be submitted until all co-authors approve of the final version. It is clear to me that the co-authors of this manuscript did not fulfill their responsibilities in terms of proof-reading the manuscript. The result is a paper that is riddled with grammatical errors and also contains some factual errors that easily could have been corrected. This dereliction of duty is completely unacceptable and places an undue burden on the editor and referees; it is also a disservice to the lead author. I will not spend my time correcting the many grammatical errors as this is the responsibility of the co-authors. Should the editor ask me to re-review this paper at a later date I will decline if the co-authors have not performed their duty of correcting the manuscript. The editor will then have to find another referee which will delay the review process.

In my comments below, if no explanation is given, please insert the recommended text into the appropriate place in the manuscript

Page 3137 line 4, change to: We examined observed and inventoried trace gas emissions,

Page 3137 line 9 Here and throughout the manuscript the use of the term “column contents” is not grammatically correct. A grammatically correct and more accurate description would be to say something like: “The decreasing trend of the lower tropospheric CO column is ~ 8.0 Dobson Units(DU) decade -1 ” where lower tropospheric is defined as 0-1500 m above ground level

Page 3137 lines 12-13, would be more clear as: “as indicated by aircraft measurements

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above the regions upwind of the Baltimore/Washington airshed."

Line 15 When discussing daily net ozone production you need a time unit, such as ~20 ppbv day-1

Page 3138 lines 7-9 Thus ozone is one of the six criteria pollutants regulated by the US Environmental Protection Agency through National Ambient Air Quality Standards (NAAQS).

Page 3138 line 10 As a secondary air pollutant the majority of tropospheric ozone is produced through...

Page 3138 line 13 You need to be more specific about the contribution of the stratosphere to tropospheric ozone. In the upper troposphere the stratosphere is a major source of ozone. Also the reference to Levy et al. 1985 is out of date. There are far better estimates of strat-trop exchange such as: Stevenson et al., Multimodel ensemble simulations of present-day and near-future tropospheric ozone, JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 111, D08301, doi:10.1029/2005JD006338, 2006

Page 3138 line 17 The EPA 2012c reference says nothing about attributing the decrease in ozone in LA to emission reductions. Please find an appropriate reference.

Page 3138 line 20 The reference to Oltmans et al 2006 is outdated. Instead please see: Oltmans, S. J., A.S. Lefohn, D. Shadwick, J.M. Harris, H.E. Scheel, I. Galbally, D.W. Tarasick, B.J. Johnson, E.-G. Brunke, H. Claude, G. Zeng, S. Nichol, F. Schmidlin, J. Davies, E. Cuevas, A. Redondas, H. Naoe, T. Nakano, T. Kawasato, 2013: Recent tropospheric ozone changes – A pattern dominated by slow or no growth. Atmospheric Environment, 67, 331-351.

Page 3138 line 20 The reference to Yorks et al 2009 is not appropriate as it is not about long term trends. See Oltmans et al 2013 and Cooper et al 2012. Then revise the following statement accordingly: "however long-term trends are not clearly observed"

Page 3138 line 24 The following paper should also be referenced: Parrish, D. D., D. B.

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Millet, and A. H. Goldstein (2009), Increasing ozone in marine boundary layer inflow at the west coasts of North America and Europe, *Atmos. Chem. Phys.*, 9, 1303–1323, doi:10.5194/acp-9-1303-2009.

Page 3138 line 25 Need to give more details on where the NAAQS exceedances occur. How bad are they?

Page 3138 line 29 Which universities and agencies created RAMMPP?

Page 3139 line 14 The claim that previous studies focused on rural areas is not correct. The Lefohn studies relied on ozone monitors that were mainly in urban areas, as did the following paper: Fiore, A. M., D. J. Jacob, J. A. Logan, and J. H. Yin (1998), Longterm trends in ground level ozone over the contiguous United States, 1980–1995, *J. Geophys. Res.*, 103(D1), 1471–1480, doi:10.1029/97JD03036.

Page 3139 line 29 Use upwind and downwind instead of upstream and downstream

Page 3140 line 15 Why not also use NEI 1999?

Page 3143 Here you use background when you should use baseline, as you did on page 3138. HTAP has defined the terms background and baseline, see the following reference: Dentener, F., T. Keating, and H. Akimoto (Eds.) (2011), *Hemispheric Transport of Air Pollution 2010: Part A: Ozone and Particulate Matter, Air Pollut. Stud.*, vol. 17, U. N., New York.

Page 3143 line 27 “...back trajectory analyses are capable of representing the general transport pathway of a hypothetical air parcel (references)”

Page 3144 line 5 If a site is northwest of Baltimore, how can it be downwind?

Page 3144 line 4 You need to provide a description of the trajectory calculations, briefly describing HYSPLIT and the type of meteorological data uses (resolution etc.)

Page 3144 Here, or at some other point in the paper you need to cite earlier studies that establish the link between reduced powerplant emissions and reduced ozone in the

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eastern US: Frost, G., et al. (2006), Effects of changing power plant NOx emissions on ozone in the eastern United States: Proof of concept, *J. Geophys. Res.*, 111, D12306, doi:10.1029/2005JD006354. Kim, S.-W., A. Heckel, S. A. McKeen, G. J. Frost, E.-Y. Hsie, M. K. Trainer, A. Richter, J. P. Burrows, S. E. Peckham, and G. A. Grell (2006), Satellite-observed U.S. power plant NOx emission reductions and their impact on air quality, *Geophys. Res. Lett.*, 33, L22812, doi:10.1029/2006GL027749.

Page 3144 line 25 To be consistent with the use of the term “baseline” change this to say: (using 1997 emissions as the reference values)

Page 3145 line 7 You need to give more background information on the policies that led to the reduction in NOx emissions. The so-called “NOx SIP Call” may have paved the way for power plant NOx reductions but the actual program that reduced NOx is the NOx Budget Trading Program (NBP) later superseded by the Clean Air Interstate Rule (CAIR) NOx ozone season program. You need to replace the NOx SIP Call statements with NBP, and please also provide a brief description of NBP. For more details see the following references:

Butler, T. J., F. M. Vermeylen, M. Rury, G. E. Likens, B. Lee, G. E. Bowker, and L. McCluney (2011), Response of ozone and nitrate to stationary source NOx emission reductions in the eastern USA, *Atmos. Environ.*, 45, 1084–1094, doi:10.1016/j.atmosenv.2010.11.040.

U.S. Environmental Protection Agency (2009), The NOx budget trading program: 2008 environmental results, report, Washington, D. C. [Available at http://www.epa.gov/airmarkets/progress/NBP_3/NBP_2008_Environmental_Results.pdf]

<http://www.epa.gov/cair/>

Page 3146 line 2 Need a reference that states how ozone is affected by weather

Page 3146 line 2 Is this trend statistically significant? Please compare this finding to the Lefohn 2010 and Cooper et al 2012.

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Page 3146 line 5 Are these CO monitors in urban areas?

Page 3146 one 11 Delete the word "suggesting"

Page 3146 line 20 Why scale by 4%?

Page 3147 line 11 Are these metric tons?

Page 3147 This paper has done a nice job of showing changes in air quality based on extensive measurements. Then the paper strays into some areas of speculation, such as the expected decrease in ozone based on NOx reductions and very vague ozone production efficiency values. This back-of-the-envelope calculation is too simplistic and not robust. This type of quantification is best left to chemical transport models. I recommend dropping this paragraph, which is not a loss to the paper as it is already strong from the excellent measurements.

Page 3148 line 16 To my eye the profile in Figure 7c is not uniform as it has a distinct bulge at 1 km.

Page 3148 line 5 The individual profiles in this figure are not visible at all.

Page 3149 line 18 Use baseline instead of background

Page 3150 first paragraph As you say, the RAMMPP profiles are from heavily polluted days while Figure 4 is for all pollution levels. So I don't see how you can specifically conclude anything about the role of transport.

Page 3150 line 20 Cooper et al 2012 show ozone trends of -1 ppbv per year across much of the Mid-Atlantic during summer when considering the 95th percentile of ozone values. These events should correspond to your RAMMPP profiles which occurred on highly polluted days. So according to Cooper et al, the rate of decrease for polluted events is 10 ppbv per decade, similar to your 13 ppbv per decade value.

Page 3151 Here you compare the decrease in CO from the aircraft to the decrease at the surface based on the national average. You then go on to say why this is an apples

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and oranges comparison illustrating why this is the wrong approach. A much better approach is use the actual CO measurements from the surface monitors in your region and calculate the observed trend for your area during the summer months as shown in your Figure 3

Page 3153 Use the term “transport pathways” rather than circulation patterns.

Page 3153 The estimates of the emitted NOx associated with the trajectories uses as crude method that is not up to the standards of ACP. For example this method does not account for the altitude of the trajectories. What if the trajectory crosses an emission source but is above the boundary layer? In this case the trajectory won't pick up any emissions. Furthermore no consideration is given to the speed of the trajectory, with a slower trajectory having a longer residence time and more time to accumulate emissions. The better method would be to use a chemical transport model, or to use inverse modeling, such as the FLEXPART retroplume method. This section should be deleted. The paper does not lose anything by dropping this section. The authors did not provide any description of the weather or transport conditions associated with each cluster. It would also be very useful to plot the typical observed trace gas mixing ratios associated with each cluster. Are pollution events more severe when the transport is from a particular direction? To my way of thinking this would be an appropriate and interesting use of the clusters.

Various pages There are many instances when mixing ratios are referred to as concentrations (for example Figure S5). This is incorrect and mixing ratios must be referred to as mixing ratios.

Figure 1 Please add labels to the map to indicate the locations of the 5 airports with the most profiles.

Figure 2 Far too small to read

Figure 4 The regression line appears to be in error. The way it is drawn there are much

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stronger deviations below the line than above. I used the linear equation provided to check the endpoints and the y-values that I get for 1996 and 2012 are, 66.7 and 57.0, respectively. These numbers are 10 ppbv greater than even the values that are plotted. Please correct. Also, please make it clear that the ozone values are based on afternoon measurements.

Figure 7 Far too small

Figure 8 The mean value at 3 km is over 200 ppbv which seems high. How does this compare to the most recent year of data? Has there been a downward trend at 3 km? If so this would be an important result, showing that emissions changes at the surface have an effect as high as 3 km.

Figure 11 As in Figure 4, the regression line in the top panel seems too high.

Figure S2 I don't understand why the Annual US emissions are scaled by 4% and 5%. What does "better demonstration" mean?

Figure S5 The black dots are too small

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

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