

Responses to CASAC Questions on the Ozone PA from Consultant Dr. Dan Jaffe

Thank you for the opportunity to assist this round of the NAAQS review.

Response to questions from Dr. Sabine Lange:

- 1) Yes, I agree with your statements: Annual averages have changed much less than the design values due to the NO_x disbenefit. How this impacts health is a question for epidemiologists, so I am not able to answer.

Response to questions from Dr. Corey Masuca:

- 1) The mechanism for S-T exchange is accurately described. However stratospheric O₃ does not only impact high elevation sites. Its impacts these locations the strongest, but stratospheric O₃ also mixes in and becomes part of the USB.
- 2) Yes near road monitoring sites should be mentioned. I assume since these data are already included in AQS.
- 3) I am not following the question. The discussion on the calculation of the MDA8 and design values appears to be correct.
- 4) Diurnal patterns are controlled by many factors. This includes photochemical production, emissions, temperature and especially meteorology. At night, shallow boundary layers give rise to surface O₃ depletion due to both NO titration and surface reactivity. The discussion mentions most of these factors, except for the role of meteorology and surface deposition.
- 5) On average, US background contributes about 30 ppb to the total, but there are significant regional, daily and seasonal variations. Thus, background is a significant contributor to O₃ concentrations, even in urban areas. Locally generated O₃ will build on these background concentrations. In general, chemical-transport models have made huge progress in their ability to model O₃ due to improvements in grid resolution, chemistry, meteorology, etc. Nonetheless, there are still uncertainties in the magnitude O₃ on the order of 10 ppb (Jaffe et al 2018). Natural sources (lightning, stratosphere, soil emissions, wildfires, etc) are all included the definition of US background O₃. (see Figure 2-15). Sections 2.5.1 discuss these natural sources. Interstate transport of O₃ would not be considered part of the US background so it would not be discussed in this section. That said it should be mentioned somewhere and I do not see it anywhere in the PA document.
- 6) As with US background, methane contributes to a global scale enhancement of O₃ of around 5 ppb. Thus it is a small, but significant contribute to urban O₃ everywhere.

Response to questions from Dr. James Boylan:

I have all of section 2 on air quality. Overall I think it is well down and accurately conveys the major sources of surface O₃ in the U.S. I do have the following comments/suggestions:

Page 2-3, line 20: add soils.

Page 2-4, line 9: “volume” ?

Page 2-7, line 23: Really? I don't think a lot of companies are making these instruments yet so when and how will SLAMs implement the chemiluminescent monitoring instruments?

Page 2-18, Figure 2-10: The high elevation site is a bit unusual in showing no diurnal pattern. Typical high elevations sites do show usually show a pattern but can differ from low elevation sites.

Page 2-21, End of section. As noted by Dr. Lange (see her questions), it is also important to discuss changes at the lower end of the distribution and the annual average O₃ associated with changing NO_x. It suggests that the overall annual average has not changed much, while the high concentrations have declined.

Page 2-26, line 9: “...global natural AND INTERNATIONAL sources..”

Page 2-27, lines 25+26: I find this sentence confusing.

Page 2-28, line 14: Statement about CO is out of context and maybe misleading.

Page 2-28 line 23: “... are generally small.” ??

Page 2-29, lines 1-2: I find the discussion on VOCs a bit simplistic. Not everywhere is swamped by biogenic VOCs. See example in Qian et al 2019 (<https://doi.org/10.1021/acs.estlett.9b00160>, 2019)

Page 2-29, line 7: Order of magnitude is too large. Maybe factor of 2 or 3.

Page 2-29, line 20: A new analysis by Buysse (DOI: 10.1021/acs.est.9b05241) strengthens this point.

Page 2-29, line 25: The exceptional event rule should also be mentioned here, as it was for strat-trop exchange.

Page 2-32, line 9-10: I think what's important here is the breakdown between natural and anthropogenic methane. We know the current and historical concentrations fairly well.

Page 2-33, line 13: A good references here is Lin 2015 (DOI: 10.1038/ncomms8105)

Page 2-33, line 21: Really need to define “baseline O₃” here.

Page 2-36, line 11: But a lot of the satellite data is associated with the “apriori” which is another model. Do you distinguish based on the fraction of the apriori in the column?

Page 2-37, lines 1-5: This discussion on model errors and bias really needs at least one figure.

Figures 2-16 and 2-17: I think the captions can be improved here. Might be better to say “Modeled MDA8 from all sources and the components”

Figure 2-20: Is there a difference between MDA8 and 8-hour contributions? Also, clarify this is “modeled MDA8”. Can you simplify to “contributions of each sources to the modeled MDA8”.

Figure 2-22: Change “Base ppb” to “total modeled MDA8 ppb”

Page 2-50, line 12: “Error”

Figure 2-25: Hard to read this figure. Suggest focusing on period between May-September.

Figure 2-26: Wrong caption. Suggest focusing on period between May-September.

Page 2-59, line 23: Its probably important out that fire emissions and chemistry are a very large uncertainty, larger than the other natural sources. I also suggest to point out the need for better tools to identify these contributions for exceptional event analyses.