Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-1174-RC1, 2018

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimating background contributions and U.S. anthropogenic enhancements to maximum ozone concentrations in the northern U.S." *by* David D. Parrish

Anonymous Referee #1

Received and published: 9 December 2018

This paper fits long-term trends of ozone design values (ODVs) in the northeastern and rural western US to exponential decay forms with a pre-derived decay scale (21 years) from prior work for Los Angeles, and infers US background ODVs from the asymptote. It concludes that the ODV in the northeastern US is 45.8 ppb, points out that it represents a large fraction (65%) of the current NAAQS over which the US has no regulatory authority, and that it is much larger than models implying that models have large errors.

I have a number of problems with this paper, and not sure that they can be fixed, so ACP may need to do arbitration or seek another reviewer. As I see it, there is no reason

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that the 21-year ODV decay time scale from LA would apply to other regions. The rural western US is mostly flat, and the northeastern US has a very different trend history initiated with the early 2000s NOx SIP Call. In fact, it seems from Figure 6 that the trend in the Northeast since 2000 could be fit to a linear decrease just as well as to an exponential decrease, and the linear decrease would imply a zero backgound ODV – which does not make sense of course but makes the point that there is no robustness to the estimate of background ODV presented here from the aymptote to the exponential decay curve. As the paper points out, a 10% change in the decay time scale would lead to a 5 ppb change in background ODV – but there is much more than 10% leeway to the fit in Figure 6.

There is also no physical rationale for a single time scale in the exponential decay of ODV, and in the absence of such a rationale any interpretation or extrapolation can be very foolish. The decrease of ODV in the Northeast is thought to be driven mainly by US NOx emissions, which have decreased linearly since 2000 according to EPA although Jiang et al. (PNAS 2018) suggest that they have been flat since 2009 – in any case, I don't see how either scenario would drive an exponential decay of ozone. Even if the response of ozone to NOx emissions was exponential, we would need a sum of exponentials to describe the ozone decrease because different anthropogenic NOx sources have decreased at different rates, and NOx emissions from fertilizer use and small industries have not decreased at all according to EPA. The effect of anthropogenic emissions on ozone is thus much more complicated than can be explained with a single exponential, and even if one can achieve such a fit to the data there is no rationale for extrapolation without understanding. Considering that NOx emissions from fertilizer use and small industries have not decreased, and that some VOC emissions have not decreased (reference in the paper to McDonald), one must conclude that the background ODV derived in this paper is biased high, possibly by a large amount.

Indeed, a punch line of the paper is that the background ODVs inferred from the exponential fit are 65-90% of the NAAQS. That seems like a big fraction, but the background

ODV estimates are biased high (see above). In addition this is misleading, considering that the ODV is depleted under polluted conditions. In the northeastern US in particular, the ozone background is highest in subsiding northerly flow, whereas the ODV exceedances are under stagnant conditions with southerly flow where background ozone is much lower.

As the paper points out, model estimates of background ozone are much lower than what is presented in this paper. The paper attributes this to error in the models. It is fair to say that there is a $\pm$ 10 ppb uncertainty in model estimates, as quoted in the paper. But that uncertainty is not a bias, whereas the background estimate in this paper is unarguably biased high. The paper does point out to some extent the uncertainties in its estimate of background ODVs, but that disappears in the abstract where the message is about the high contribution of the background to the NAAQS and how the models need to be corrected.

Aside from these basic issues of scientific content, I found the paper to be much longer than it needs to be. Figures 1 and 2 show the Pacific Northwest and the Midwest but these then drop from the radar screen, why even bother? Descriptive discussions of population, topography, etc. don't seem necessary. There's a lot of chattiness and repetition.

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