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

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The author is grateful for the time and thought reflected in the referee's comments regarding this paper. They will lead to improved discussion in the revised paper. However, I believe that these comments are incorrect in some respects, as detailed below, and very importantly miss the significant value of the analysis presented. U.S. policy makers must set national ambient air quality standards (NAAQS) for criteria air pollutants including ozone. A major uncertainty they face is the contribution to measured ambient concentrations made by transported background ozone. Currently policy makers must rely on estimates of the background ozone contribution calculated by models

of atmospheric transport and chemistry. However, these estimates vary widely among different models, and are recognized to be so uncertain that their utility to policy makers is limited. The value of the reviewed paper is that it presents an observationally based estimate of the background contribution that policy makers can consider in their work. As noted by the referee, this observationally based approach is not perfect, but I believe that the results are more accurate than the model results. Each of the referee's comments is reproduced below (*in italics*) followed by my response (in plain text).

The analysis is designed to separately quantify the U.S. background ozone design values (ODVs) and the enhancements of the ODVs above that background contribution due to U.S. anthropogenic precursor emissions. The U.S. background ozone design value is assumed to be the maximum ozone DV that would exist in these regions in the absence of U.S anthropogenic precursor emissions. The US background and US anthropogenic increment are derived from a simple exponential function, analogous to the function derived for California subregions in Parrish et al. (2017).

Although the idea of a simple model to describe design value behavior is appealing, there are several problems with this approach.

(1) The simple exponential function has been applied to separate the contributions of anthropogenic US precursor emissions to the ozone design values from the contributions by US background ozone (i.e., ozone that would be present in the absence of US anthropogenic precursor emissions). This formulation of the ozone problem is based upon a chemical transport modeling definition of US background ozone; the US background ozone can be estimated by "zeroing-out" US anthropogenic emissions in a chemical transport model. In areas far from the Pacific Coast, where US background concentrations enter the country, it is difficult to see how this simple observational model can untangle the interactions between US biogenic emissions (part of the background) and US anthropogenic emissions (part of the anthropogenic component). It is not at all clear that the asymptotic value approached by the exponential equation in this manuscript represents US background, or some mixture of US background (e.g.,

biogenic VOCs) combined with an especially persistent US anthropogenic (NOx) component that hasn't yet been substantially reduced by control strategies.

The referee speculates that there may be "an especially persistent US anthropogenic (NOx) component that hasn't yet been substantially reduced by control strategies," which can confound the analysis presented in the manuscript. As we discuss in the paper, this is a concern. However, there is no evidence of such a component with an NOx emission magnitude comparable to the well-known emission sectors (mobile, industrial and power plant sources) that have been effectively reduced by control strategies. The influence of such an emission component must be kept under consideration, but in that absence of any evidence of such a component, there is no justification for rejecting the results of the analysis presented in the manuscript.

Notably, the emission inventories typically used for regional photochemical modeling do not include any such "especially persistent US anthropogenic (NOx) component". If such a component did exist, photochemical modeling results would be biased. Such a possibility further emphasizes the value of the present observationally based estimate of U.S. background ozone, so that biases in either the model results or the observationally based estimate can be understood through comparison of results, and the results obtained from each approach improved.

(2) Interannual variation of ozone data is smoothed because three years of data are averaged together to get a design value. The rationale for using the three-year average of the fourth high is that attainment of the ozone standard is linked to a three-year average, and hence, it is important to study the behavior of this somewhat unwieldy metric in order to reach policy-relevant conclusions.

But a design value is defined for a specific metropolitan area. The highest three-year average of the fourth high at any monitoring site within the metro area is the design value. Since the analysis does not examine ODVs for individual metro areas, or select the fourth high for each metro area for each year, the ODVs described in the paper

are not actually the ODVs used in regulatory applications. It can be argued that this distinction is scientifically trivial, but in this case we are discussing policy, not science, so the distinction is important. The author could redefine the regions according to the EPA's definition of nonattainment areas to match the policy definition.

The referee's description of ozone design values is not entirely complete. As discussed in the manuscript, an ODV is defined for each monitor in the U.S. The ODVs analyzed in this paper are indeed the ODVs that are considered in regulatory applications. EPA's process for defining nonattainment areas involves many considerations beyond the tabulated ODVs. One of these considerations is the ODV for the area; the area ODV is simply the highest ODV of those recorded at all of the sites within the area during each year. Since the highest site ODVs are included in our analysis, we are analyzing the ODVs actually used in regulatory applications. To assuage the referee's concern, Figure 1 below illustrates the analysis for nonattainment area ODVs, and Table 1 gives the derived parameters. (The four nonattainment areas that lie entirely within the northeastern states are included; the fifth nonattainment area is only partially in the area under consideration, so it is not included.) These nonattainment areas are described in the manuscript. As can be seen from comparing Table 1 below with Table 2 of the manuscript (and properly considering the parameter confidence limits included in the tables), the analysis presented in the manuscript is directly relevant to the nonattainment area ODVs. The results for the NY-NJ-LI-CT and Greater CT nonattainment areas in Table 1 are in close accord with the Connecticut and New York/maximum results given in Table 2 of the manuscript. Similarly, the results for the Jamestown, NY and Dukes Co, MA nonattainment areas in Table 1 are in close accord with the New York/rural upwind and Massachusetts/coastal results given in Table 2 of the manuscript. The analysis for the nonattainment areas is entirely consistent with the analysis and discussion included in the manuscript; nothing new would be gained by including this additional analysis in the manuscript; however, it could be included in the Supporting Information of a revised submission.

But the statistical analysis would still be somewhat clumsy; the three-year averages smooth out much of the interannual variability, and can cause autocorrelation issues, as the author notes. There are, however, other metrics just as relevant to policy as the ODVs used in this study. A better metric for individual monitoring sites would be the 4th high (98th percentile) maximum daily eight-hour ozone concentration for each year at each site. If the fourth-high/98th percentile metric for each year at each site were analyzed, there would be no overlap among years, eliminating problems with autocorrelation and excessive smoothing of interannual variations among years, yet the analysis would be at least as relevant to regulatory status as the current analysis. The physical interpretation of the data would be simplified as well, because the metric itself would be more closely tied to the observations of a single site and single year, instead of being smeared over three years.

Using a different metric would help resolve an issue related to the smoothing of interannual variation. The author asserts that the simple exponential model of ODV trends has achieved a degree of success in describing the variation, based upon the confidence intervals. These confidence intervals have been modified to account for covariance due to lack of independence among ODVs. But the interannual variation would be larger if the analysis had been performed on annual 98th percentiles rather than ODVs for each site, and it is unclear whether the modification of confidence intervals to account for covariance also accounts for the reduction of interannual variation. The results would be more compelling if the interannual variation had not been shaved down by using three-year running averages.

The preceding two paragraphs critique the statistical fitting technique utilized in the manuscript; however, the referee's discussion is incorrect. The goal of the statistical analysis is to extract the systematic, long-term change in a set of observed ODVs (or fourth-high/98th percentile) as accurately as possible, given the interannual variability about those long-term changes. No statistically significant information regarding the long-term change is lost by working with 3-year means (i.e., the ODVs) rather than

annual mean data (i.e, fourth-high/98th percentile). A qualitative explanation for this can be given. Deriving 3-year means from annual data involves an averaging process, which minimizes the sum of the squares of the deviations of the annual data from the derived 3-year means. The fitting procedure employed in the analysis to derive the long-term change minimizes the sum of the squares of the deviations of the 3-year ODVs from the derived long-term change. The final result is independent of whether the sum of the squares of the deviations is minimized in two steps (3-year mean calculation followed by the fit to the long-term change), or in one step (extracting the long-term change directly from the higher frequency annual data). We work with 3-year mean ODVs because they are of most policy relevance, and we have properly dealt with autocorrelation issues.

Additionally, working with annual mean data (i.e, fourth-high/98th percentile) would worsen a separate autocorrelation concern. Interannual meteorological and climate variations can drive differences in ozone photochemical production and transport; such differences can persist over multiple years, so that annual mean data may be autocorrelated. Working with 3-year mean ODVs reduces the influence of this autocorrelation.

(3) Increasing the interannual variation, however, would probably worsen another issue: the inability of the model to converge on a solution for the three model parameters. As the author notes on page 7, lines 10-16, the shorter data record for the northern regions appears to be preventing estimation of the three parameters of the exponential function. To resolve this issue, the author has assumed that one of the parameters can be set at the same value as derived for California. As the author notes, the value of tau = 21.9 years derived from California implicitly assumes that control strategies have produced approximately equal relative reductions in anthropogenic ozone enhancements throughout the country. This assumption is questionable, and the results for the northeastern states seems to show that it is unwarranted.

As discussed in my response immediately preceding this comment, no statistically significant information regarding the long-term change is lost working with 3-year means.

Increasing the interannual variation by working with annual means would not worsen the uncertainty in deriving the three model parameters; the greater number of independent data available for the fitting procedure would closely compensate for the larger interannual variation.

The referee is correct that the assumption of tau = 21.9 years is questionable, and that question is addressed in detail in my response to Comment 2 of referee 1. Notably, the referee does not state in what way "the results for the northeastern states seems to show that it (the assumption) is unwarranted"; thus I am unable to respond to the last part of this comment.

(4) Table 2 shows the derived values of y_0 (US background ozone) and A (US anthropogenic component) for subsets of monitoring sites. The values of US background ozone for low altitude sites vary from 41±10 ppb in suburban Massachusetts to 61±6 ppb in coastal Connecticut. This is a large variation over a short distance for a value that is supposed to reflect relatively unvarying background ozone.

The referee correctly points out that he values in Table 2 derived for y_0 for the low altitude sites vary from 41±10 ppb in suburban Massachusetts to 61±6 ppb in coastal Connecticut and suggests that this variation is large. This large variation arises from two sources. First, as reflected in the 95% confidence limits given in the table, there is statistical uncertainty in deriving the parameter values from the fits of Equation 1 to the ODVs; the confidence limits for the two extreme results that the referee quotes indicate that the statistically significant portion of the variation is not as large as the referee suggests. Second, as discussed in the paper, some of the variation does arise from real departures of the derived y_0 values from the true U.S. background ozone. Figure 2 below illustrates an analysis of the distribution of the derived y_0 values that allows these two sources of variation to be examined separately. The ordinate scale of this figure is designed so that a normal distribution defines a straight line. In this figure, 13 of the 17 derived y_0 values do define a normal distribution with a median of 47.7 ppb and a standard deviation of 4.5 ppb. We believe that these 13 values accurately reflect

U.S. background ozone; this result is in close accord with the 45.8 ± 1.7 ppb estimate of this quantity given in the manuscript. The 4 of 17 derived y_0 values in Figure 2 lie in a high value tail of the distribution; these are the 4 y_0 values that we discuss separately in the manuscript. This figure nicely illustrates the value of our analysis based upon the simple model encapsulated in Equation 1 – the large majority of the results provide a robust estimate of the regional U.S. background ozone, and deviations from a uniform result point toward issues that deserve further investigation.

One interpretation is that the Connecticut "background O3" includes a lot of ozone generated in the New York City area, and that therefore the simple exponential model cannot determine US background.

This interpretation suggested by the referee is not valid. Connecticut ODVs certainly do reflect a lot of ozone generated in the New York City area. However, this New York City generated ozone has been decreasing over the years; thus, our analysis would properly include it in Connecticut's time varying term (A*exp[-(year-2000)/tau]), not in Connecticut's constant y_0 term. The simple exponential model can indeed determine U.S. background ozone, within the caveats thoroughly discussed in the manuscript.

The author chose a different interpretation, and re-set the US background to a lower value, which increased the US anthropogenic component to more acceptable levels. This portion of the analysis is not convincing, and seems to be an attempt to compensate for the simple model's shortcomings. In fact, it is essentially an admission that the original simple model cannot be used to distinguish between US background ozone and US anthropogenic ozone.

The referee incorrectly describes the reasoning in our manuscript. We followed two clearly distinct steps. The first step is the purely mathematical fitting of Equation 1 to the observed ODVs. The success of this step is judged by the good agreement between the fits and the observations. The second step is the physical interpretation of the A and y_0 parameters. As discussed in the manuscript and illustrated in Figure 2 below, the derived y_0 values provides a great deal of information regarding the U.S. background ozone, but we do not argue that the two quantities are identical in all situations. Hence, the four values in the high value tail, which includes the Connecticut y_0 value. We do not re-set the U.S. background ozone to a lower value; rather we discuss why the derived Connecticut y_0 value is not equal to the U.S. background ozone. This is not an attempt to compensate for the simple model's shortcomings; rather it is a discussion of an issue identified by the simple model that deserves further investigation. It is not an admission that the original simple model cannot be used to distinguish between US background ozone and US anthropogenic ozone; in the majority of cases the model provides a robust distinction.

Ultimately, I have concluded that the assertions claiming that US background and anthropogenic increment can be derived from the simple exponential function are not compelling, especially for the northeastern states. It is possible that the analysis could be re-worked, by changing the ozone metric to the annual 98th percentile, but the failure of the method to derive the three parameters of the model even with three-year running averages, and its inability to distinguish between US background and US anthropogenic in the northeastern states suggests that this simple approach is flawed for the regions to which it has been applied in this study. I thought that the study was interesting, and the author did a commendable job in explaining the uncertainties and possible shortcomings of the approach. This admirable transparency in describing the methods is worthy of emulation, but I do not think the study should be published in its present form.

The author appreciates the referee's positive comments. However, as discussed in detail above, I believe that the objections raised by the referee are generally not valid. Hence, the overall conclusion stated here is not justified.

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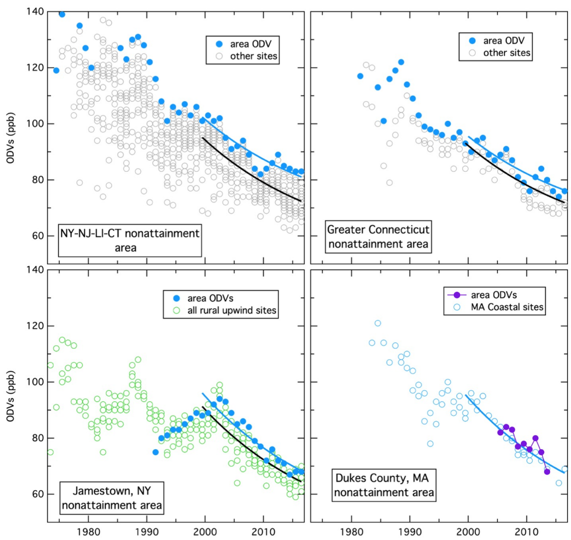

Fig. 1. Time series of nonattainment area ODVs (solid symbols) and the ODVs reported from all monitoring sites (open symbols) in the four nonattainment areas included within the northeastern states.

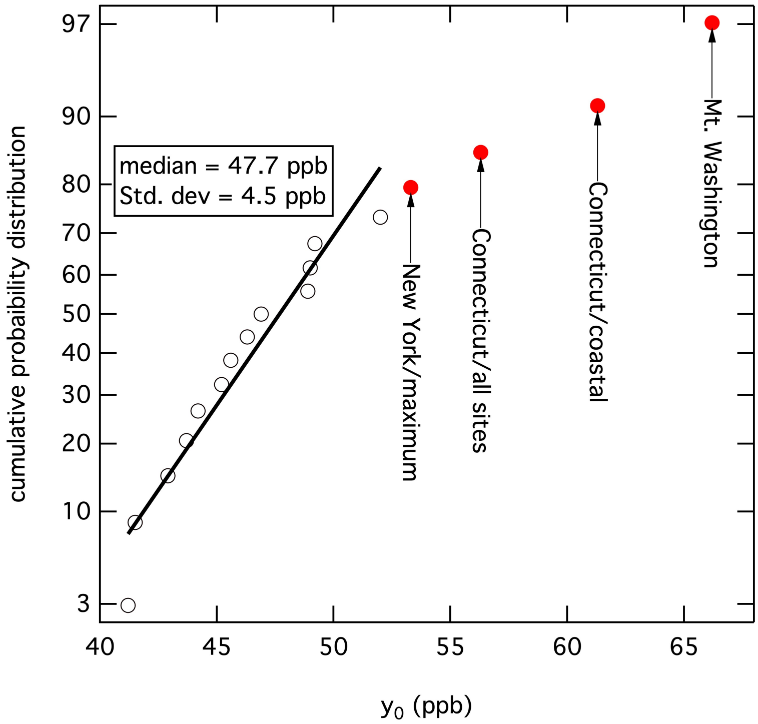

Fig. 2. Cumulative probability plot of the y0 determinations listed in Table 2 of the manuscript. The line is a linear regression fit to the open points, which defines a normal distribution.

Table 1. Results of least-squares fits to Eq. 1 illustrated in Figure 1; RMSD indicates the root-mean-square deviation between the observed ODVs and the derived fit.

Nonattainment area/sites	y_0 (ppb)	A (ppb)	RMSD (ppb)	years fit
NY-NJ-LI-CT area/ODVs	63 ± 12	40 ± 16	3.0	2000-2017
NY-NJ-LI-CT/all sites	53 ± 4	42 ± 6	5.8	2000-2017
Greater CT area/ODVs	59 ± 12	36 ± 17	3.2	2000-2017
Greater CT/all sites	54 ± 7	39 ± 10	4.1	2000-2017
Jamestown, NY area/ODVs	45 ± 14	51 ± 19	3.7	2000-2017
Jamestown, NY/all rural upwind sites	42 ± 7	50 ± 10	5.1	2000-2017
Dukes Co, MA/all sites	44 ± 9	52 ± 13	3.2	2000-2017

Fig. 3. Table 1