Author's Response to interactive comments by Editor and 4 Anonymous Referees on "Estimating background contributions and U.S. anthropogenic enhancements to maximum ozone concentrations in the northern U.S." by David D. Parrish

The author greatly appreciates the many comments regarding this paper, the time and effort that clearly went into these reviews, and the Editor's recommendations. We have taken these comments to heart, and have made significant changes to the analysis and the manuscript. Responses to all comments follow, and where appropriate the manuscript has been revised as described herein, and indicated in the "tracked changes" manuscript copy at the end of this response.

To organize this response, we first give an overview of the four major issues raised in the reviews, and briefly summarize how we have addressed each. This is followed by more extensive discussion of each major issue. Finally, point-by-point responses to all reviewer comments are given, often simply referring back the more extensive discussion of the major issues.

Overview: Major Scientific Issues Raised by the Reviewers and Our Response

A. ***Reviewers questioned our choice of Equation 1, which incorporates an exponential decrease above a constant offset, to quantify urban ODV changes.***

   Response and Changes Made: We agree that additional support for using Equation 1 will strengthen the paper. We discuss this choice from four perspectives that support its use, provide context for the analysis, and guide the interpretation of the derived parameter values. We have expanded the discussion in Section 2.2 of the paper to address this comment.

B. ***Reviewers questioned our decision to set τ equal to 21.9 years (i.e., the value derived in a southern California ODV analysis).***

   Response and Changes Made: The reviewers raised valid concerns and questions about this choice of τ. In response to their comments, we have now included two approaches to estimate τ in the northeastern U.S. to provide support for the assumed value of τ of 21.9 years. One approach includes a linear analysis, similar to that suggested by Referee #1 in the first round of reviews. These 2 approaches provide values of τ that, within uncertainties, overlap the original value of 21.9 years for τ. We describe these analyses in Section 2.3 and the results in Section 3.3. The original analysis remains in the main text as our primary choice, but we hope the reviewers will agree that this choice is now is strengthened by the additional analyses. However, we acknowledge that these additional analyses do support other reviewer comments suggesting that the uncertainties in our results are likely larger than we stated. Therefore we have provided additional uncertainty analysis of the magnitude of τ and the derived value of y_0 in the northeastern U.S. The result is a larger confidence interval (± 3.0 ppb for y_0), which is incorporated into the uncertainty discussion in the revised manuscript (new Section 3.3.4).

C. ***Reviewers point out that our analysis neglects that temporal changes likely occur in the background ODV contribution (i.e., our assumption of a constant y_0 may not be valid).***

Response and Changes Made: The discussion in Section 3.1 that shows y_0 has remained nearly constant in the western rural states over the past 3 decades has been clarified. The variance due to varying background ODV contributions is shown to be small relative to that from the changing anthropogenic contribution in the northeastern states. A detailed comparison of the ODVs at western rural sites with a baseline ozone site on the U.S. west coast is included in the expanded discussion below. These comparisons show that applying Equation 1 with an assumed constant background is well justified for the northeast U.S.

D. ***Reviewers were critical of the success of Equation 1 in describing spatial and temporal ODV distribution.***

Response and Changes Made: U.S. ODVs result from a very complicated system of chemical and physical atmospheric processes. Equation 1 provides a basis to quantify the role of only one of these processes - changing anthropogenic emissions. We agree with the reviewers that judging the degree of success (and failure) of this description is important for the informative interpretation of the derived parameters. Section 3.3 of the paper discusses this judgment, and that discussion is further extended below.

Discussion of Major Issues

Issue A: Choice of exponential decrease above a constant offset (Equation 1) to quantify urban ODV changes

Four considerations guide our choice of Equation 1, which contains an exponential decrease to quantify long-term changes of U.S. urban pollution ozone contributions:

1) Examination of ozone observations in U.S. urban areas reveals similarity of trends throughout the country, with general decreases in all areas, albeit occurring at different rates and after varying onset times. Our goal is to quantitatively evaluate these decreases in order to elucidate the processes controlling U.S. urban ozone concentrations. Fitting observational data to a simple functional form is a common tool utilized for quantitative observational analysis; linear trend analysis (i.e., fitting observational data to a linear function) is one example. In this work we select a functional form that a) is consistent with our general understanding of urban ozone, and b) is as simple as possible (i.e., has the fewest unknown parameters). There are three general features of ozone in U.S. urban (and rural) areas that must be consistent with the selected functional form: first, there is a background contribution, below which ozone cannot be reduced by U.S. precursor emission controls; second, maximum ozone concentrations have been enhanced above that background due to pollution-generated ozone; and third, the pollution enhancements are presently generally decreasing due to on-going emission reductions. To the best of our understanding, Equation 1 of our paper,
$$O_3 = y_0 + A \exp\{-(\text{year}-2000)/\tau\}, \qquad (1)$$
with three undetermined parameters, is the simplest possible functional form consistent with these three features. (A linear fit with only two undetermined parameters – slope and intercept – is simpler, but a linear fit to a decreasing trend will eventually go negative, and therefore cannot fit a positive background contribution).
2) Equation 1 gives excellent fits to the last two or more decades of ozone observed in U.S. urban areas. Parrish et al. [2017a] show that Equation 1 captures 98.4% of the total variance in the maximum ODVs in 7 southern California air basins over the 1980 to 2015 period. Figure 9b of the revised manuscript shows that Equation 1 captures 89% of the total variance

in the maximum ODVs in the 8 northeastern U.S. states from 2000 to 2017. These fits suggest that the derived parameter values must contain useful information regarding the processes driving temporal evolution of U.S. urban (and rural) ozone concentrations.

3) Since Equation 1 is chosen to be consistent with our general understanding of urban ozone changes, the parameter values can be directly related to the processes controlling U.S. ozone concentrations. For example, y_0 in Equation 1 is designed to quantify the background ozone contribution. However, this design does not necessarily mean that a parameter value can be directly interpreted as Equation 1 implies; the simple, direct interpretation of y_0 (as well as the other two parameters) must proceed with careful consideration of potentially complicating factors.

4) A simple intuitive argument suggests that an exponential decrease in the pollution ozone contribution is to be expected. When emission controls are initiated, early progress can be rapid, since there are large emission sources that evolved with no plans for their control. As an illustrative example, it might be possible to reduce the pollution ozone contribution by half in the first 15 years of control efforts. After that period reducing emissions will be harder, since the most easily controlled emissions have been addressed. During the next 15 years, it might be possible to again reduce the remaining pollution ozone contribution by half (i.e., to 25% of the original). If this example were realistic, then the emission reductions would follow Equation 1 exactly with $\tau = 21.6$ years, close to the value of $\tau = 21.9 \pm 1.2$ years reported by Parrish et al. [2017a]. Simply put, the expected increasing difficulty in reducing emissions by an absolute amount implies an approximately exponential decrease in the impact of those emissions. Equation 1 captures this expectation.

As the above discussion makes clear, Equation 1 can only be applied to time periods after maximum urban ozone concentrations were reached, and emission controls had advanced to the point that ozone concentrations began to decrease consistently. It is true that this decrease began ~1980 in California, but not until ~2000 in many regions of the northeast U.S.; thus the present analysis is primarily limited to post-2000. Four comments are relevant here. First, the single exponential fit must fail at some point if extended to earlier times, as a maximum urban ozone concentration must have occurred at some time in the past in any given region. Second, some regions in the northeast do approximately follow Equation 1 to earlier times, e.g., Connecticut data in Figure S3, where the ODV maximum of 169 ppb (off scale in figure S3) occurred in 1982. Other northeastern U.S. regions are fit for years before 2000 (see Table 2). Third, it would be of interest to investigate the timing of the ozone maxima in different regions (and to understand the possible role played by evolution of urban monitoring site networks), but such investigation is beyond the scope of this paper. Finally, the literature contains many examples of analyses based on linear fits to ozone trends after maxima were reached [e.g., see Fig. 17a of Lin et al., 2017; Fig. 5 of Jaffe et al., 2018]; the inability of the linear trend to describe data before the maxima is not interpreted as casting doubt on the geophysical interpretation of the mathematical fitting parameters for the period after the maximum – the analysis presented in this paper must be considered similarly.

This discussion leads to the conclusion that fitting to Equation 1 is a valid approach for observational analysis of ozone in both California and the northeast U.S., but that complicating factors must be carefully considered in geophysical interpretation of the derived values of the fitting parameters. In response to reviewer comments, our revised paper goes further to acknowledge and explain these complications and assumptions.

Issue B: Choice of τ set equal to 21.9 years (i.e., the value derived in a southern California ODV analysis).

It is difficult to precisely determine the three parameters of Equation 1 from the relatively short (~2000-2017) record of consistently decreasing northeastern U.S. ODVs. The primary analysis (retained in the revised paper) is based on the simplest approach – assuming that τ in the northeastern U.S. is the same as derived for southern California ($\tau = 21.9 \pm 1.2$ years). To test this assumption and address reviewer comments, we now include two analyses to investigate the τ value appropriate for the northeastern U.S.; both analyses assume constant τ and y_0 values across that entire region.

Table 1. Results of iterative, non-linear regression analysis

τ (years)	26.0 ± 6.0
y_0 (ppb)	41.8 ± 3.0
State	A (ppb)
Maine	51 ± 10
Massachusetts	56 ± 10
New Hampshire	46 ± 10
New Jersey	66 ± 10
New York	61 ± 10
Rhode Island	55 ± 10
Vermont	39 ± 10

The first analysis is an iterative, non-linear regression analysis similar to that described in Section 2.4 of Parrish et al. [2017a] that simultaneously fits seven data sets to Eq.1 to allow extraction of nine parameters. The data sets are the 2000-2017 maximum ODVs recorded in seven states shown in Figure 10 of the revised paper. (Note that since its recent ODV behavior is different from the other states, as discussed in the paper, Connecticut is not included here.) This fit gives a value for τ, a value of y_0, and values of A for each of seven states (Table 1). The derived τ value is larger than the California value, although they agree within the derived 95% confidence limit. Correspondingly, the derived y_0 value is smaller than derived earlier (45.8 ± 1.7 ppb), and the A values are larger (compare to A^* values in Table 3 of manuscript).

The second approach uses linear fits to the initial period of decreasing ODVs. Figure 1 shows three of the ODV data sets selected to give the largest possible contrast in the contribution of pollution ozone and the smallest variability about the fits to Equation 1. Linear fits are shown for the initial periods (2000-2017 for two, and 1991-2017 for the data set with a large gap in the ODVs near 2000). The absolute value and the time derivative of Equation 1 when evaluated at year 2000 are $y_0 + A$ and $-A/\tau$, respectively. Four parameters (τ, y_0, A_1 and A_2) are required to fit two ODV data sets if the τ and y_0 values are common. The slopes and intercepts (given in Table 2) of the two linear fits in each panel of Figure 1 provide 4 relationships that allow the calculation of the 4 exponential fit parameters. Algebraic manipulation gives expressions for $\tau = -\Delta_{\text{intercept}} / \Delta_{\text{slope}}$, where Δ indicates in the difference in the subscripted parameter between the two linear fits, and $y_0 = (\Sigma_{\text{intercept}} + \tau * \Sigma_{\text{slope}})/2$, where Σ indicates the sum of the subscripted parameter from the two fits. These equations allow calculation of estimates of τ and y_0 from the two linear fits in each panel of

Table 2. Results from linear fits to early years of three ODV data sets

Data set	intercept (ppb)		slope (ppb/yr)	
	original	corrected	original	corrected
New York City	108.6 ± 3.8	110.1 ± 3.8	-2.16 ± 0.38	-2.91 ± 0.38
Vermont	78.2 ± 1.8	79.0 ± 1.8	-1.07 ± 0.18	-1.44 ± 0.18
Maine NE coast	70.6 ± 1.5	69.8 ± 1.5	-0.89 ± 0.13	-1.05 ± 0.13

Figure 1. One complication is that the linear fits give biased estimates of the year 2000 slopes and intercepts required in the above derivation. First order corrections for these biases are made from numerical evaluation of linear fits to an exponential function, and included as corrected parameters in Table 2. The resulting estimates of τ are 21.1 ± 5.9 and 21.7 ± 5.0 years for the fit parameters in the upper and lower Figure 1 panels, respectively.

In summary, the *τ* estimates from these two analyses do agree with the southern California value within their confidence limits, indicating that there is no evidence for a different exponential rate of decrease of U.S. anthropogenic ODV enhancements between these two U.S. regions. Importantly, these two analyses give estimates of y_0 that are independent of the assumed *τ* value.

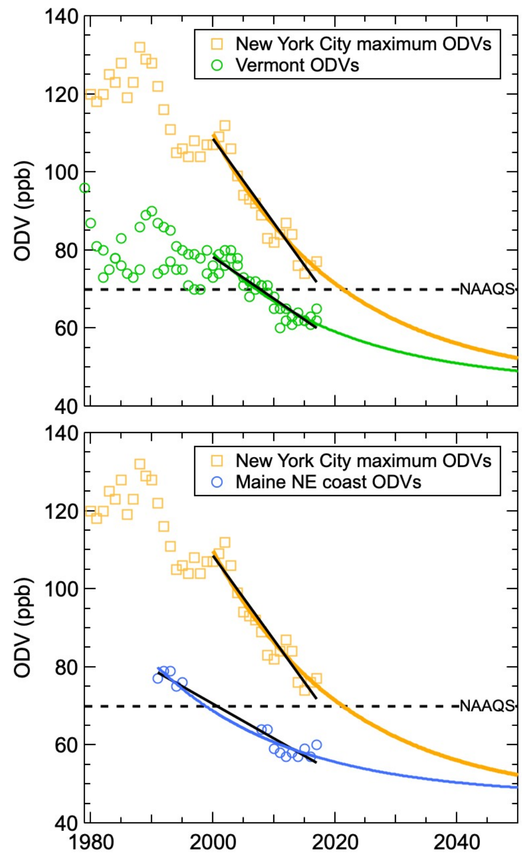

Figure 1. Comparison of linear and exponential fits to three ODV data sets.

Issue C: Neglect of temporal changes in the background ODV contribution (y_0).

Parrish et al. [2017b] report that seasonal average baseline ozone concentrations at the U.S. west coast were increasing before a maximum was reached in the mid-2000s, and are now decreasing. Since transported baseline ozone is the primary source of the U.S. background ODV contribution, assuming a constant y_0 contribution as done in the present analysis may compromise the results. Figure 2 compares the ODVs recorded at Lassen Volcanic NP [one of the sites considered by Parrish et al. 2017b] with those at two of the rural western state sites. Table 3 gives the coefficients of quadratic polynomial fits, such as Parrish et al. [2017b] utilized to quantify these long-term changes. There are no statistically significant differences in the temporal trends of these ODVs, as quantified by the *b* and *c* parameters (although there are differences in the absolute concentrations as quantified by the *a* parameters.) Figure 2 is consistent with Section 3.1 of the paper, where we conclude that the ODVs in the rural western states are dominated by the U.S. background contribution.

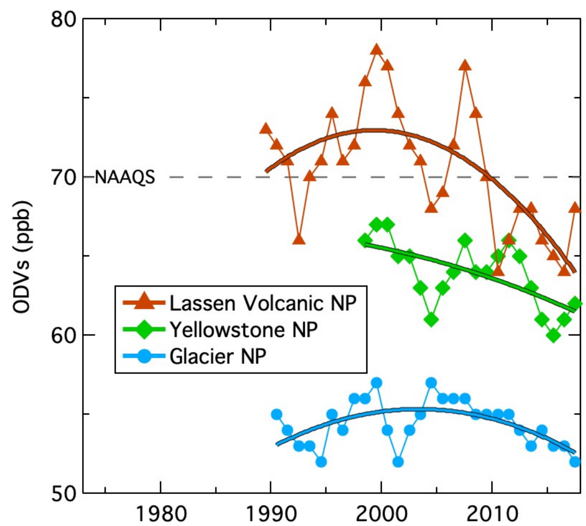

Figure 2. Comparison of ODVs recorded at three relatively isolated rural sites.

The impact of neglecting background temporal changes can be judged by comparing the ODV variance over the 2000-2017 period between the northeastern states (Figure 3 of the manuscript), where anthropogenic ODV contributions dominate, and North Dakota (Figure 4 of the manuscript), where only the U.S. background contribution contributes significantly. The sample variance in each region is calculated from the square of the standard deviation - 107.3 ppb^2 and 3.7 ppb^2 in the northeastern states and North Dakota, respectively. The North Dakota variance can be taken as a rough approximation for the background ODV contribution to the ODV variance in the northeastern states, which would amount to a contribution of no more than a few percent. Table 3 compares the variance of ODV series in Figure 2, which are similar to that of North Dakota. In summary the neglect of temporal changes in y_0 in the northeastern states is of negligible consequence in the determination of the y_0 and A values in that region.

Table 3. Comparison of quadratic fits to ODVs at a baseline site on the U.S. west coast and two sites in the rural western states

Site	a (ppb)	b (ppb yr^{-1})	c (ppb yr^{-2})	RMSD (ppb)	Variance (ppb^2)
Lassen Vol. NP	73.0 ± 1.6	-0.01 ± 0.20	-0.027 ± 0.019	3.0	9.9
Yellowstone NP	65.6 ± 1.8	-0.14 ± 0.49	-0.005 ± 0.027	1.7	4.4
Glacier NP	55.1 ± 0.7	0.11 ± 0.10	-0.013 ± 0.008	1.2	2.0

Issue D: Judging the success of Equation 1 in describing spatial and temporal ODV distribution

U.S. ODVs are determined by a very complicated system of chemical and physical atmospheric processes. Equation 1 ignores nearly all (including varying influences from stratospheric intrusions, increasing Asian anthropogenic emissions, increasing frequency of wildfires, variable meteorological conditions, increasing methane concentrations, a warming climate, etc.), while considering only two ozone contributions - background ozone and anthropogenic ozone pollution. Further, these two contributions are treated in a simplified manner – the background contribution is assumed constant, and the anthropogenic contribution is assumed to have decreased exponentially since the time that urban ozone concentrations began decreasing consistently. This simple approach is only justified if it successfully quantifies the spatial and temporal distribution of U.S. ODVs.

The discussion of Figure 9 in Section 3.2.2 of the paper demonstrates the success of Equation 1 in capturing the variability of 18 years of ODVs recorded in 8 northeastern U.S. states. These ODVs include 1719 individual values. Notably, there are no free parameters in the two fits shown in Figure 9: constant values of y_0 and τ were derived from other analyses, and the A values are interpolated from Figure 9 (for the individual monitoring sites) and taken from Table 3 for the state maxima. Equation 1 then captures 89% and 71% of the variance in state maxima and all 1719 ODVs, respectively. This indicates that no more than the remaining 11% and 29% of the variance in state maxima and all 1719 ODVs, respectively, can be accounted for by the combined influences of:

- The atmospheric processes listed above, and ignored by Equation 1.
- Departures from the assumed constant background contribution and the assumed simple exponential decrease of the anthropogenic contribution at the assumed California τ value.
- Spatial variability of the A parameter not captured by the contour plot of Figure 9.
- Any measurement issues such as temporally evolving monitoring networks giving inconsistent temporal changes, instrumental errors, etc.

The success of Equation 1 in quantitatively describing the spatial and temporal distribution of U.S. ODVs is a strong indication of the utility of the derived parameters for estimating aspects of the ODV distribution.

Point-by-Point Responses

Notes:

- The comments from the Editor and the Referees are reproduced below in black regular font with our responses in *blue italic font*.
- Our revisions have been guided by several of the constructive comments and suggestions offered by Referee #4; therefore we have responded to that review first in our discussion below.
- Some of the Reviewers' comments have been divided into separate paragraphs to facilitate responses; *(Para)* indicates where such divisions have been made.

Anonymous Referee #4

Review of acp-2018-1174

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

There is a considerable amount of interesting and potentially valuable information and analysis in this manuscript, but it is not presented in a form which allows the reader to appreciate its value.

In addition to the difficulty I had in reading this work and trying to extract its main points, I, like the other two reviewers, think the geophysical interpretation of the mathematical analysis may be overstated beyond what is truly warranted by the data presented. Perhaps this feeling is exacerbated by the repetition of sections of text, but for me it fundamentally derives from the significant difference in the temporal evolution of ODVs over the entirety of the record for the regions studied here in comparison to the "reference" region of southern California.

I have four points of concern with regard to the content, which I think any resubmitted manuscript should address:

1) The inability of the single exponential analysis to describe the NEUS and MWUS data before 2000 seems to cast doubt on the geophysical interpretation of the mathematical fitting parameters and/or on the rigid adherence to the California tau parameter, which is derived in Parrish et al. 2017 (JGR) from observations that are well-described back to 1980. Pg. 13, line 4 ascribes this change in the temporal behavior in the NEUS to regulatory efforts around the turn of the millennium in the eastern US, thereby suggesting that the California behavior (or at the very least the time constant) should not be used as a model for the present region(s).

*Please see the **Issue A** discussion above regarding the use of the single exponential and the geophysical interpretation of the derived parameters. Our revisions acknowledge the complications that must be considered when applying the exponential. Please also see **Issue B*** *and the next point below, which describes our additional tests of the primary approach of using the California tau parameter.*

It seems that finding y0 values is a primary goal of this work, so the author must present a more complete sensitivity analysis than 10% change in tau mentioned briefly in section 4.2. Given the shortness of the data record being fit, what is the range of tau values that could reasonably describe the observed decay? If the time constant were set to, e.g., 10 yrs. or 40 yrs., instead of 21.9 yrs., how much would y0 change?

*Please see the **Issue B** discussion above regarding the appropriateness of application of the California time constant to the northeastern U.S. We now incorporate two additional analyses that provide insight into the range of tau values that apply to the northeastern U.S., and an expanded confidence limit for y_0 is now discussed in the revised manuscript.*

The alternative analysis approach described in AC-3 may be worth including in a revised version of this manuscript, if doing so can be accomplished in a concise and compelling way. (Could a revised manuscript be structured around the alternative method as the primary analysis approach?) This alternative method seems to provide similar insight into the data without requiring the use of the CA tau parameter. The author would, however, still need to discuss the implications and limitations of analyzing only the recent portion of the data record, regardless of the method used.

*Please see Issue B and the discussion of additional analyses completed in response to this comment and comments of the other Reviewers. The alternative analysis approach is included in our revised manuscript (Section 3.3.1). We have worked to improve the concise and compelling way the material is described. Given that each approach involves assumptions, and considering that the alternative approaches give values of tau that overlap the assumed California tau value (21.9 years) of our original analysis, we have retained our original analysis as the primary analysis in the paper. We hope Referee #4 agrees that the additional analyses have strengthened the case for our primary approach. Please see the **Issue A** discussion above regarding the implications and limitations of analyzing only the recent portion of the data record. We have briefly discussed this issue in Section 3.2 of the revised paper.*

2) In light of the author's 2107 GRL paper showing that the transported contribution to NA background ozone is now decreasing, I would like to see a discussion of how those findings do or do not affect the interpretation here, given that a constant y0 value is a fundamental presumption of the present analysis.

*Please see the **Issue C** discussion above regarding the how changing background ODV contributions affect the interpretation of the present analysis. This issue is now specifically discussed in Section 4.2.*

3) The three low-altitude "exceptions" should not be excluded from discussion. If they are not failures of the method but rather sites in a category of their own (where the "y0 value is not equal to the U.S. background ozone" –AC-2), they should be explained and analyzed, not buried in a sentence in section 4.1. Why is y0 different at these sites than others in NEUS (or at least for the

CT sites)? What does y0 represent in these cases, if not US background ODV? Or is this just merely evidence that the US background ODV has different values in different locations?

The second paragraph of Section 4.1 now explains and analyzes these exceptions as fully as possible: "The cause of this difference is not understood. Whether this difference is simply a statistical fluctuation cannot be determined at this time; however, random fluctuations of similar magnitude are only rarely apparent in the temporal records of ODVs in the states discussed. McDonald et al. (2018) have recently discussed a class of ozone precursor emissions, i.e., volatile chemical products - including pesticides, coatings, printing inks, adhesives, cleaning agents, and personal care products - that have not been addressed by emission controls to the same extent as other emission sectors. The impact of this emission sector on ODVs has not been quantified, but is expected to be most significant in areas of largest population density, exactly the regions where the significant differences in temporal evolution of ODVs are noted." Unfortunately, further discussion is not possible without undue speculation.

Why does NY/max (y0=53) get singled out as an exception, but Maine/Cadillac Mtn (y0=52) does not? To my eye, Fig. 2 in AC-2 suggests Maine could equally belong to the category containing Maine/Cadillac Mtn. What criteria were used to determine which groups of sites belonged in the category of "exceptions"?

This raises a good point. Figure 2 of AC-2 (now included as Figure S11 in the revised Supplementary Information) indicates no clear reason to separate Maine/Cadillac Mtn from NY/max as their y_0 values are similar. However, the separation must be made somewhere, and these two data sets lie near the "knee" of the cumulative probability distribution plot where the separation naturally belongs. The differentiation was made for two reasons. First, the other two low-altitude "exceptions" are either highly urbanized or just downwind from highly urbanized areas, so it makes sense to include NY/max with these two exceptions. In contrast, Maine/ Cadillac Mtn is one of the furthest downwind areas, so it makes sense to include it with the other areas downwind form the major urban center. Second, the confidence limits on the Maine/Cadillac Mtn y_0 are so wide that it may well lie in the range of the y_0's found at the other downwind areas. In the interest of brevity, this discussion has not been added to the revised paper.

4) Why are all the remaining USNE y0 values then averaged together, despite the fact that they have a wider spread than the Rural West values which are never averaged together in Section 3.1? The discussion of coastal sites in the text led me to expect a subcategory that included NH/coastal, MA/coastal, and ME/coasts, rather than just having all those sites lumped into a single average y0 value with the inland locations.

Figure 2 of AC-2 (now included as Figure S11 in the revised Supplementary Information) shows that remaining USNE y_0 values approximately define a Gaussian distribution with a median of 47.7 ppb and a standard deviation of 4.5 ppb. The 95% confidence limits of the remaining USNE y0 values in Table 2 range from 4 to 10 ppb (16 ppb including the Maine/Cadillac Mtn confidence limit). This rough correspondence between the standard deviation of the y_0 distribution and the standard deviation that corresponds to the 95% confidence limits suggests that there is no statistically significant difference between the remaining USNE y_0 values, so they are all lumped into a single average y_0 value. In contrast, the Rural West y_0 values do exhibit *statistically significant differences, so it is appropriate to discuss them separately rather than averaging. In the interest of brevity, this discussion has not been added to the revised paper.*

Comments related to Presentation:

We agree with the Referee's suggestions below to reduce redundancy, focus attention more fully on the main points, and reduce distractions of secondary information. We have made the following revisions to address these comments and suggestions (answers interspersed with Referee's comments):

Substantial revision of the text will allow the reader to focus on the important messages the author wishes to convey. Perhaps a colleague with "fresh eyes" can help the author frame the discussion for a scientist, rather than for a local expert at a regulatory agency.

This was an extremely helpful suggestion. A colleague not previously involved in the manuscript was asked to join as a coauthor. After working through the paper to understand all aspects of the analysis, she led the reorganization of the entire manuscript, Supplement and this response to the comments.

I recommend that the author focus first on presenting only the western and northeastern US data, followed by the method in brief. The caveats, while important, are distracting when presented in the Introduction. The sections which discuss limitations should all be combined together somewhere in the body of the manuscript and only be enumerated once.

We have focused the discussion on the two regions as suggested, which involved eliminating text from the Introduction and moving Figures 1 and 2 of the prior manuscript to the Supplementary Information.

Caveats are now collected and discussed in one section (4.2).

There is a tremendous amount of detail presented in the Introduction that does not really build the author's case but which does consume the reader's attention and capacity to manage information (e.g., page 3, lines 17-32 present many names and numbers that seem to require a deep understanding and attention. Yet only a few pieces of information from this paragraph are truly critical to understanding and appreciating the message.)

We agree and we have eliminated this paragraph.

Similarly, the other two geographic regions do not seem critical to present in such detail. Figure 1 and Figure 2/top panels are not the most important figures, but by placing them so early in the manuscript the implication is that they should be read carefully and all four panels digested fully.

As mentioned, we agree and we have eliminated Figures 1 and 2 from the main manuscript and moved them to the Supplementary Information.

After spending so much effort to read the early sections of the manuscript, I had little patience or focus left by the time I got to the discussion of A and A* values – which surely are more central to the message of the study than are the nuances of the geography of Martha's Vineyard, for example.

The Referee's point is well taken. We hope we have removed the distractions successfully so that the paper is now more focused on the central messages of the study.

There are many examples of redundancy throughout the text where content is repeated in essentially the same format as presented in earlier sections.

We agree with the Referee's suggestion. We have made the discussion more concise and removed redundancy wherever possible. Specifically, we only briefly mention caveats *in the introduction, and collected the discussion in one section (4.2)*.

<u>In conclusion</u>, in its present form, I do not believe this manuscript meets the standards of *Atmospheric Chemistry and Physics*, nor the expectations of its readers. But I do believe it could become an interesting contribution to the community's understanding of ozone trends if the author addresses the substantive concerns identified above (as well as some or all of those identified by R1 & R2) and invests in crafting a more streamlined manuscript that is much easier for the reader to understand and digest.

We appreciate the very constructive criticisms of Referee #4 on the scientific issues and the presentation of the manuscript. We are also appreciative of the Referee's statement that the essence of the paper can be an interesting contribution to the community's understanding.

Anonymous Referee #3

Parrish uses a simple mathematical fit and an overly specific interpretation. The author has responded to previous comments by adding an acknowledgment of weaknesses of the technique and interpretation. The acknowledgment is not sufficient given the reliance of the manuscript on the specific interpretation.

As noted in our Overview, other responses above, and further responses below, we have revised our manuscript to provide alternative analyses as well as additional discussion of the assumptions of those analyses and the primary exponential analysis. Any analysis of anthropogenic and background ozone, whether observation-based or model-based, involves assumptions and simplifications. We acknowledge, as do all such studies, that the full complexity of "reality" is not represented in the analyses. Nevertheless, the application of new approaches such as the ones we have presented here serve as a useful way to advance understanding of the scientific issues and concurrently shed new light on the uncertainties inherent in all studies. We have widened the confidence limits of our results as a result of the scientific issues raised by the referees and based on our alternative analyses.

In response to previous comments, the author asserts that there is likely no persistent emissions. The NEI shows a decay of emissions from 2002 to 2014 (a comparable period) with a lifetime of 18 years (see https://gispub.epa.gov/neireport/2014/) -- which is in reasonable agreement with the authors tau. However, the interpretation requires two more assumptions. The first assumption is that the trends are explainable by single exponential decay function. The second major assumption is that ozone trends will continue responding to future reductions as they have now.

First assumption: The interpretation by Parrish is based on exponential decay of local contributions, which are related to emissions. If the emissions reduction is not reasonably fit by a single exponential function, then it is over interpretation to conclude that y0 is background. The NEI trends data shows that some sectors are increasing while others are decreasing. Individual exponential fits would create tau values ranging from -50 to 22 years. For example, "Industrial and Other Process" category has gone up Nationally by 25% over the time period being analyzed (tau=-50years). If each sector is separately allowed to grow to 66 years (3 x tau in this manuscript), using the single exponential function would predict just 12% of the sum of individual functions. The "Industrial and Other Process" sector is likely linear not exponential; even holding it constant would mean that the single exponential function would predict just 34% of the future emissions. This highlights that the exponential decay is actually the sum of equations. Even the exponential decay of certain sectors could be due to an exponential decay in controlled processes and a constant or increasing component from uncontrolled processes.

We make no assumption regarding the time dependence of ozone precursor emissions. Given the shortcomings of emission inventories (e.g., Miller et al. 2006), quantifying emission changes is uncertain. We do know from ambient measurements in Los Angeles that emissions have decreased approximately exponentially, but at different rates for different species (and likely for different sectors as well). For example, in Los Angeles over the past 50 years, ambient VOC concentrations have decreased at about 7.5% yr^{-1} or a factor of ~50 over 5 decades (Warneke et al., 2012) while NOx concentrations have decreased at about 2.6% yr^{-1} or a factor of ~4 over 5 decades (Pollack et al., 2013). It is widely recognized that the response of ozone to changes in precursor emissions is complicated; we do not rely upon any quantitative information regarding precursor changes.

*Please see the **Issue A** discussion above regarding our choice of the exponential decrease of the U.S. anthropogenic ODV enhancements.*

Suggesting that an exponential decays that corresponds with specific sector controls can be used to characterize all sector contributions is over interpretation. There is evidence of this over-interpretation in the manuscript itself. Figure 7 is an excellent example of where the interpretation is obviously flawed. If this approach were robust, you could apply it to a time interval of observations and get the same result as if you applied it again after accumulating more data. If you had applied the model in 2003 (before the NOx SIP call took effect), the "Background ODV" appears as though it would have converged on 80-90 ppb. The exponential decay after 2003 highlights that the decay is in response to controls put in place over that time period. During that same time, motor vehicle emissions have also decreased. If controls targeted all sectors, the interpretation would be more reasonable. There is no evidence provided by the author in the manuscript.

Responding to this comment is difficult, as it is not clear to which data set the referee is referring. Taking the Massachusetts coastal data set as an example, Figure 3 shows fits for three time intervals with the derived y_0 values annotated: 4 years (2000-2003) as the referee suggested, 10 years (2000-2009), and the full 18-year period (2000-2017). The 4-year period gives physically unrealistic results; interannual variability gives an overall increase in ODVs over that period. Nevertheless, the derived y_0 value agrees (nearly) with the other two results within the derived confidence limits, despite the implication of a negative pollution ozone contribution. When 10 *years of data are considered, there is close agreement with the 18-year period, but the confidence limits are so large that the results are of limited usefulness. This exercise does not show an obvious flaw in the interpretation, nor does it present any evidence of an over-interpretation – close attention must be paid to the confidence limits in this (or really in every) observational analysis. The exercise does show that one can apply this approach to a time interval of observations, and then get a statistically consistent (not the same) result if applied again after accumulating more data. Thus, the approach does pass this "robustness" test suggested by the referee, and this exercise demonstrates the value of additional years of data.*

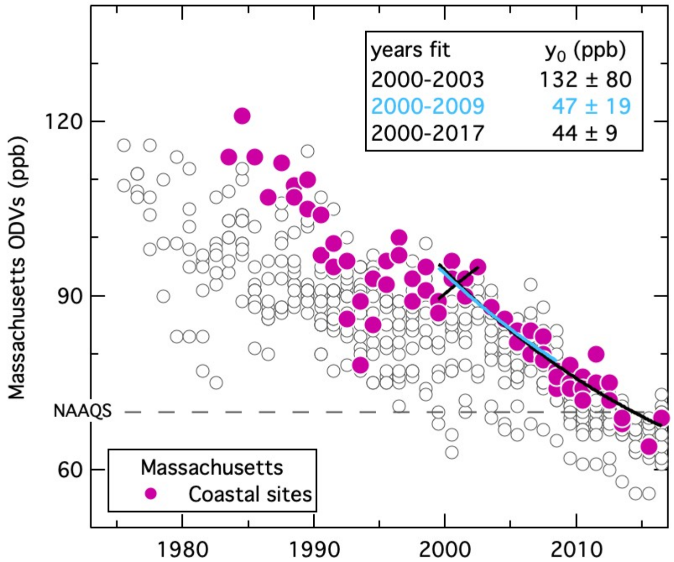

Figure 3. Analysis of the Massachusetts coastal data set (violet points from Figure 6 of the revised manuscript) over the time periods indicated in the figure annotation. The fits to Equation 1 are indicated by the superimposed curves.

Second assumption: If the sensitivity of ozone to NOx is not expected to remain constant, then this extrapolation is over-interpretation. As NOx-limitation becomes more sever, it is expected that the per unit NOx ozone production efficiency increases. This means that the remaining NOx will contribute more per unit than the NOx that has been controlled to date. Thus, any remaining NOx (the constant or increasing sectors) will contribute more than expected from present day inventory fraction.

We do not make any assumption regarding how ozone responds to changing emissions of NOx or VOC ozone precursors. Indeed, this is one of the strengths of our approach. Our assumption regarding the temporal evolution of ODVs can be simply stated, as in the first paragraph of Section 4 of the manuscript: "… the second term (of Eq. 1) is interpreted as an estimate of the regional U.S. anthropogenic ODV enhancement (i.e., the amount that ODVs are enhanced above the U.S. background ODV by photochemical production of ozone from existing U.S. anthropogenic precursor emissions." Thus, as the ***Issue A*** *discussion above reflects, we do choose an exponential decrease to describe the temporal behavior of the U.S. anthropogenic ODV enhancements, but do not assume any particular relationship to precursor concentrations.*

Finally: The author uses uncertainty in USB to suggest that USB estimates from this technique are reasonably within uncertainty. Much of the uncertainty in USB comes from natural process like stratospheric intrusions and wildfires. These processes are not expected to make substantial contributions in New York on exceedance days that are often typical local ozone production.

Stratospheric intrusions and wildfires are believed to make substantial contributions in urban areas such as New York, as indicated by the successful exceptional event demonstration (based on wildfire influence in urban areas) submitted to the U.S. EPA by the Connecticut Department of Energy and Environmental Protection as we discuss in the paper. The U.S. EPA has formulated the 2016 Exceptional Events Rule (https://www.epa.gov/air-quality-analysis/final-

2016-exceptional-events-rule-supporting-guidance-documents-updated-faqs) so that potential exceedances (generally occurring in urban areas) due to natural processes can be excluded. This rule specifically mentions stratospheric intrusions and wildfires. These processes occur on an episodic basis and contribute to the magnitude of interannual variability in observed ODVs, which does affect the accuracy and precision of our U.S. background ODV estimates. Similarly, the difficulty of accurately modeling the sporadic influences of these natural processes affects USB estimates from models. We believe that the most productive way forward is to work to get agreement between observationally based results and model results, and then to focus on reducing the uncertainties in both sets of results.

The author has received much of this feedback at various meetings. Before resubmitting again, consider that the feedback is consistent and from many sources.

The author has indeed received much of this feedback (as well as other feedback) at various meetings from many sources. However, the feedback has not been consistent; it has included supportive feedback as well as various scientific disagreements. In keeping with the peer-review process, I have responded to each argument raised, either with a useful revision of the paper's discussion and/or approach, or a carefully reasoned rebuttal. This rigorous review by colleagues is much appreciated and has contributed usefully to an improved manuscript, which I hope addresses the concerns of the referees.

Anonymous Referee #5

This paper attempts to determine values for the US background Ozone Design Value (ODV) as well as its anthropogenic enhancement, by fitting observed ozone timeseries with a simple exponential decay function, representing a reduction of anthropogenic ozone enhancement due to reducing emissions, with an asymptotic approach to a "true" background ODV.

The rationale for doing this is that numerical models of ozone are currently not able to provide satisfactory estimates of these values, which are important for policymaking in the US. Having an alternative, measurement-based approach for estimating these values would be of great interest to policymakers. Exactly how well this can be done is an open scientific question.

Two anonymous referees have both recommended that this paper be rejected after its initial review phase. Both argue that the approach used by the author is overly simplistic, and lacks a physical basis. While I can sympathise to some extent with their criticisms of this paper, on balance I think that the underlying issue (the ozone problem in the US) is important enough that alternative approaches should be explored.

The author has generally done a reasonable job of explaining the limitations of his approach, and provided an extensive discussion of how the results can and should be interpreted. He has generally anticipated the criticisms of the reviewers, but this has not stopped them from making them anyway. I think the author could perhaps have done more to highlight the uncertainties and potential biases in his revised version, but he is also justified in sticking to his guns, as he has done, since his original submission already covers these issues well enough.

Given the importance of the underlying issue, I have no doubt that future work will continue to develop different approaches for observation-based estimation of background ODVs and their anthropogenic enhancement. The current paper provides a good basis on which such future work can build.

Thank you for these thoughts. During this second manuscript revision we have paid particular attention to improving the discussion of uncertainties and potential biases.

Anonymous Referee #6

Review of "Estimating background contributions and US anthropogenic enhancements to maximum ozone concentrations in the northern US" by D. Parrish

This manuscript attempts to provide observationally-based estimates of background ozone and anthropogenic ozone production in the northern US using time series of ozone design values (ODVs) for the rural Northwest and the northeastern US. The analysis is based on multiple unsubstantiated and likely incorrect assumptions. I agree with the editor's decision to not publish precisely because of the author's argument that this is likely to be a high-impact paper given that it estimates background ozone to be significantly higher than previous analyses in most areas. While the author argues that the potential importance of the paper means it should be published so that it can become part of the open debate regarding background ozone, the assumptions that underlie the analysis are, in my opinion, so problematic that it should not be published until they can be justified via modeling or some other formal analysis. The discussion of the potential weaknesses of the analysis does not provide sufficient caution regarding the interpretation of the results and the uncertainties associated with the estimates of background ozone and anthropogenic enhancements to ODVs are misleadingly small given the number of assumptions and the disregard for temporal and spatial covariations among the ODVs. I believe that the manuscript has a likelihood of skewing the conversation about background ozone because it seems to provide highly significant, observation-based estimates of the background but in fact is based on an overly-simplistic view of ozone chemistry and makes so many "leaps of faith" regarding ozone's response to changing emissions that its results are meaningless without significant work to lay the grounds for those assumptions.

Our analysis indeed contains assumptions and simplifications. It has good company in doing so—all modeling analyses, upon which current policy formulation rests, do the same. The imperfect nature of the state of the science is well illustrated by the current generation of photochemical models, which cannot reproduce observed ODVs. To our knowledge, no urban ODV time series of significant length has yet been accurately modeled. Indeed, photochemical modeling cannot generally reproduce an observed urban ODV in even a single year. Hence, the U.S. EPA has resorted to the use of Relative Reduction Factors (RRFs) (see https://doi.org/10.1016/j.aeaoa.2019.100029) in an attempt to compensate for the inaccuracies of photochemical models. Thus we do not agree that modeling can be used to as a way to validate our analysis.

On the contrary, our analysis offers a different (but not claimed to be perfect) approach that is based on observations and that provides results that can be usefully compared to model results. As we assert and as other referees have agreed, our analysis is worthy of joining the open scientific debate. Broadening the scientific analyses available to policy makers supports science-based policy formulation.

The first assumption is that ODVs everywhere in the northern U.S. have decreased following an exponential curve in response to emissions controls. While this exponential decrease appears to be fairly well-defined for Los Angeles, it is so poorly defined for the regions in this analysis that a second and more objectionable assumption must be made that the timescale (tau) is the same for the rural northwest, the highly variable northeast, and Los Angeles. The need for this assumption arises from the fact that the ODVs have only been decreasing since 2000 in many of the time series analyzed and the decrease has been small relative to that in Los Angeles, so that the exponential decay is shallow and not well-defined. In fact, it is clear that in many cases a linear decrease would fit the ODV data as well if not better than an exponential. Assuming the exponential form and the same tau everywhere implies that 1) anthropogenic precursor emissions have decreased exponentially in response to emissions controls in all regions analyzed and that 2) "control strategies have produced approximately equal relative reductions in anthropogenic ozone enhancements through the country." This is such an overly-simplified view of the highly heterogeneous system of emission control and of ozone's highly nonlinear response to emissions changes that it is difficult to overstate how breathtaking it is.

*Please see the discussion **Issues A and B** above regarding the choice of the single exponential to describe the ODV response to decreasing precursor emissions, and the appropriateness of the application of the California time constant to the northeastern U.S. We agree that in the rural western states the exponential decay is shallow and not well-defined, so we have removed the application of the exponential analysis to this region.*

We know of no evidence that the anthropogenic ODV contribution has actually decreased at differing rates in different U.S. locations. We have examined the time evolution of ODVs throughout the country, and can find no region with a rate of decrease statistically significantly different from that in California (and the regions examined in this paper), over the past two decades. If such differences could be found, then the analysis could be modified to account for those differences, which would allow extraction of more information regarding the U.S. ODV distribution; however, at present information regarding regional differences in ODV evolution is lacking.

(Para) There are two throwaway statements in the paper regarding the possible influence of "volatile compounds", but the role of VOCs in ozone chemistry, the fact that ozone can be either VOC- or NOx-limited and that the response to emissions controls can be opposite for two nearby locations depending on the NOx / VOC mix are completely ignored. So too are the differences in emissions controls that have been applied in Los Angeles and those that have been applied in the Northeast (there is a brief discussion of this issue on page 18, lines 20-26, but the author appears to believe that the question of whether precursor emission reductions have been similar between Los Angeles and the Northeast is unknowable, when in fact a simple analysis of emissions inventories would provide a great deal of relevant information) and the transition of substantial regions of the Northeast from VOC-limited to NOx-limited during the period from 2000 to the present.

Since our analysis is based solely on the time evolution of observed ODVs, there is no need to consider (or even the possibility to incorporate) information regarding the NOx / VOC mix. Similarly, any information gleaned from emission inventories would not be relevant. Our goal is to examine the measured temporal and spatial distribution of ODVs to see what is revealed.

(Para) Furthermore, by the author's own description, the equation used to describe the decrease in ODVs "assumes that all sectors of anthropogenic US ozone precursor emissions have been reduced by emission controls at approximately the same rate. However, in some respects this is a poor approximation in that some emission sectors have received lesser efforts than others. Any emissions that have not been reduced would tend to lead to an overestimate of US background ODV." In fact, we know with absolute certainty that all precursor emissions have not been reduced at the same rate. Given this and the fact that any increase in tau also leads to a lower estimate of background ozone, the confidence limits provided for the background ozone estimates are ridiculously small and the estimates are not meaningful.

*We agree with the referee's point that the confidence limits must be realistic. Please see the **Issue B** discussion above regarding the uncertainty of tau value; a wider confidence interval for y_0 and an improved discussion of the tau uncertainty have been incorporated into the discussion in the revised manuscript.*

Another objectionable assumption is that background ozone has remained constant over the entire observational record and that all of the processes that might change background ozone act merely to produce variability in ODVs rather than trends. In fact, we have good reason to believe that there should be trends in background ozone associated with wildfires and international emissions, and that these should be highly regionally variable.

*Please see the **Issue C** discussion above regarding our choice of assuming that the background ODV contribution (y_0) is temporally constant. Failure of this assumption is indeed indicated by the small temporal trends in background ozone documented in that discussion and in Parrish et al. [2017b]. As discussed in Section 4 of the revised manuscript, these trends make significant contributions to the small ODV variance in the rural western states, and presumably make contributions of similar magnitude to the variance in the northeastern U.S. Importantly, every observational analysis fails to capture 100% of the variance of the analyzed data set, but if a large fraction of the variance of the data set is captured, then the analysis results can provide the basis for useful understanding of the system observed in the collection of the data set. As we discuss in the **Issue C** response above and in Section 4.2 of the revised manuscript, our analysis captures a large fraction (two-thirds or 89%, depending on the ODV data set used) of the total ODV variance in the northeastern states. The unaccounted for variance (including varying background ozone) does not compromise our results, but it does limit the precision (i.e., it increases the confidence limits of the results).*

The author argues that two additional analyses demonstrate the robustness of the results despite the assumptions inherent in the equation describing ODV changes. The first such analysis is that shown in Figure 10, where ODVs are reconstructed from Equation 1 using the tau value from Los Angeles and a single, regionally-averaged value of background ozone (yo). The only thing proven by this analysis, in which there is only one free parameter remaining (A), is that a very shallow exponential curve reasonably fits ~15 years of data for the Northeastern US. Given that there has been a relatively steady decline in ozone in the Northeastern US since 2000, this is a given once you fix a value for tau and yo. The fit improves if you consider only the maximum ODVs in each state each year, but no justification is provided as to why this should be the case.

Figure 9 of the revised manuscript demonstrates the success of Equation 1 in capturing the variability of 18 years of ODVs recorded in 8 northeastern U.S. states. Please see the ***Issue D*** *discussion above for more details of this demonstration. The improvement of the fit when considering only the maximum ODVs in each state each year is a direct consequence of the removal of the variability of the ODVs within each state – smaller variability of ODVs about the fit yields improved fits; the discussion of this issue has been clarified in the revised manuscript.*

(Para) The second analysis that is meant to demonstrate the robustness of the results is, to my mind, a direct argument against their robustness. This analysis also relies on unsubstantiated assumptions that 1) all ODV time series follow the same functional form (it is not clear why this should be the case given the heterogeneity of ozone precursor emissions in the region and the nonlinearity of ozone chemistry) and 2) all of the time series will approach a common US background ODV (we have no a priori expectation of this given expected gradients in influence from international transport from Canada, variability in natural emissions, etc). Furthermore, one of four time series that differs substantially from all of the other time series in the Northeast (i.e. the New York City urban area, which has a higher background ozone value that most of the rest of the region in Table 2) is used as the reference. The regression analysis produces background ozone estimates ranging from 25 to 62 ppb, but a single, averaged value is calculated and compared to the result from the exponential fit analysis. Yet if one were to assume that the variability in the derived background ODV is real (and no justification is provided as to why it should not be) then the derivation of A values (or the taus – it is impossible to say which) in the exponential fits would be completely different.

The New York City reference for this analysis is the ODV time series included in Figure 11 of the revised manuscript, not the New York/maximum listed in Table 2 and discussed as an exception. We apologize for this confusion, and have clarified this distinction in the revised manuscript. The regression analysis does produce background ozone estimates ranging from 25 to 62 ppb (Table S2). The reason for the wide range of estimates is that the analysis is imprecise because it is based on quantifying the intersection of two linear fits; each of these fits is uncertain due to the scatter in the fitted ODVs. We discuss this uncertainty in Section 3.3.1. Importantly, the relatively large number of independent determinations does provide averages with relatively small confidence limits. This analysis does provide a useful consistency test of the primary analysis.

(Para) The uncertainties of 3.9 ppb and 1.7 ppb on the background ozone estimates do not consider regional and temporal coherence of the ODVs nor the uncertainties associated with the assumptions underlying each analysis and are likely grossly underestimated. While the author is correct that there is not a good way of accounting for these additional uncertainties, the low values provide a misleadingly precise estimate of background ozone.

There is no reason to believe that either regional or temporal covariance of the ODVs (not accounted for in the uncertainty analysis) make large contributions to the uncertainties in the background ozone estimates. With regard to temporal covariance, we have accounted for the covariance associated with the three-year averaging period of the ODVs. The time scale of any additional covariance would necessarily be on longer time scales. Wildfire occurrence and stratospheric intrusions are major drivers of temporal variation of background ozone, but their temporal variation is largely removed by the three-year averaging period of the ODVs. Also, *such regional and temporal covariance would be expected to affect the background in the western rural states, and their influence would be obvious there; yet the discussion of that region of the country shows very small variations in background ozone over decades of measurements. For example, we can estimate background ozone in North Dakota simply by averaging all ODVs reported over 28 years (1990-2017). That average of 137 ODVs reported from throughout the state is 58.8 with a 2.1 ppb standard deviation. This estimate compares favorably with our estimate of 57.0 ± 1.5 ppb from Table 1 for y_0 and its 95% confidence limit. The 2.1 ppb standard deviation of the 28-year average accounts for all ODV variations from all sources including all regional and temporal variations. The referee asserts "The uncertainties of 3.9 ppb and 1.7 ppb … are likely grossly underestimated". However, the assertion is clearly not correct for the rural western states since the total variance of the North Dakota ODVs are of the same order or smaller than the uncertainties we quote; there is no reason to believe that it is correct for the northeastern states.*

In Section 4.3, the author treats his "observationally-derived estimates" of background ozone as the gold standard against which model-derived values should be evaluated. This discussion illustrates the danger of this paper. There is a natural inclination to treat observationally-based estimates as "truth", when in fact the unsubstantiated assumptions underlying this analysis and the gross underestimate of uncertainties are not likely to be clear to most casual readers. Until additional analysis has been done to provide a foundation for the assumptions on which the analysis is based, this work should not be published.

In our view there is no "gold standard" estimate of background ozone, either from our "observationally-derived estimates" or from model calculations. The wide disagreements between the model-derived and observationally-derived estimates, and among estimates from different model analyses clearly demonstrate the need for further investigation, and our "observationally-derived estimates" do contribute to meeting that need.

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Estimating background contributions and U.S. anthropogenic enhancements to maximum ozone concentrations in the northern U.S.

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Abstract. U.S. ambient ozone concentrations have two components: U.S. background ozone and enhancements produced from the country's anthropogenic precursor emissions; only the enhancements effectively respond to national emission controls. We investigate the temporal evolution and spatial variability of the largest ozone concentrations, i.e., those that define the ozone design value (ODV) upon which the National Ambient Air Quality Standard (NAAQS) is based, within the northern tier of U.S. states. We focus on two regions: rural western states, with only small anthropogenic precursor emissions, and the urbanized northeastern states, which include the New York City urban area, the nation's most populated. The U.S. background ODV (i.e., the ODV remaining if U.S. anthropogenic precursor emissions were reduced to zero) is estimated to vary from 54 to 63 ppb in the rural western states, and to be smaller and nearly constant (45.8 ± 3.0 ppb) throughout the northeastern states. These U.S. background ODVs correspond to 65 to 90% of the 2015 NAAQS of 70 ppb. Over the past two to three decades U.S. emission control efforts have decreased the U.S. anthropogenic ODV enhancements at an approximately exponential rate with an e-folding time constant of ~22 years. These ODV enhancements are relatively large in the northeastern U.S. with state maximum ODV enhancements of ~35-64 ppb in 2000, but are not discernible in the rural western states. The U.S. background ODV contribution is significantly larger than the present-day ODV enhancements due to photochemical production from U.S. anthropogenic precursor emissions in the urban as well as the rural regions investigated. Forward projections of past trends suggest that average maximum ODVs in northeastern U.S. will drop below the NAAQS of 70 ppb by about 2021, assuming that the exponential decrease of the ODV enhancements can be maintained and the U.S. background ODV remains constant. This estimate is much more optimistic than in the Los Angeles urban area, where a similar approach estimates ~2050 for the maximum ODV to reach 70 ppb (Parrish et al., 2017a). The primary reason for this large difference is the significantly higher U.S. ODV background (62.0 ± 2.0 ppb) estimated for the Los Angeles urban area. The approach used in this work has some unquantified uncertainties that are discussed. Models can also estimate

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U.S. background ODVs; some of those results are shown to correlate with the observational-based estimates derived here (r^2 values for different models are ~0.31 to 0.85), but they are on average systematically lower by 4 to 12 ppb. Further model improvement is required until their output can accurately reproduce the time series and spatial variability of observed ODVs. Ideally, the uncertainties in the model and observational based approaches can then be reduced through additional comparisons.

1 Introduction

The U.S. has a long-standing air quality problem associated with elevated ozone concentrations (e.g., NRC, 1991). Fortunately, this problem has been greatly improved over the past 3 to 5 decades, particularly in urban areas. For example, through the 1960s and 1970s the Los Angeles urban area (i.e., California's South Coast Air Basin – SoCAB) endured maximum 1-hr average and maximum daily 8-hr average (MDA8) ozone mixing ratios that exceeded 500 and 300 ppb, respectively (ppb = nmole ozone per mole air) (Parrish and Stockwell, 2015). The National Ambient Air Quality Standard (NAAQS) is based on the ozone design value (ODV), which is defined as the 3-year average of the annual fourth-highest daily maximum 8-hour average (MDA8) ozone concentration; in 2015 the NAAQS was lowered, now requiring that ODVs not exceed 70 ppb. A fit to the long-term trend of the maximum ODVs recorded in the SoCAB indicates that these highest ozone concentrations decreased from 289 to 102 ppb over the 36-year, 1980 to 2015 period (Parrish et al., 2017a). This decrease demonstrates that controls on U.S. ozone precursor emissions have been remarkably effective in reducing maximum ambient ozone concentrations. However, much additional emission reduction effort is required to reach the NAAQS of 70 ppb. A critical question has relevance to policy development for managing U.S. ozone concentrations: What is the limit to which ODVs can be reduced by controlling U.S. anthropogenic emissions? One goal of this work is to provide an observation-based estimate of this limit.

Both natural and anthropogenic processes interact to determine the temporal and spatial distribution of surface ozone concentrations in both urban and rural areas. Thus, even if U.S. anthropogenic emissions of ozone precursors were completely eliminated, ambient ozone concentrations throughout the U.S. would still be well above zero due to contributions from natural sources of ozone, enhanced by anthropogenic contributions from other countries. Parrish et al. (2017a) estimate that this remaining ODV (denoted as U.S. background ODV) would be 62.0 ± 1.9 ppb in the Los Angeles urban area. This contribution is the limit to which the ODVs can be reduced by U.S. emission controls alone; it is so large that there is little margin for enhancement of ambient ozone concentrations by photochemical production from U.S. anthropogenic precursor emissions before the NAAQS of 70 ppb is exceeded.

Two northern U.S. regions (maps in Figures 1, 2 and S1) are the focus of this work; eight northeastern states, which include the most populated U.S. urban area (New York City metropolitan area), and three sparsely populated, rural western states.

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(Montana, North Dakota and South Dakota), containing no cities with >260,000 population. The temporal histories of ODVs measured in these two regions (Figure 3) correlate with the degree of urbanization - in the rural western states they remained approximately constant at relatively small values over the 39 years of measurements, while the largest ODVs with temporally decreasing values have been in the northeastern states. The northern tier of U.S. states also includes three Pacific Northwest states and three midwestern states (map in Figure S1) with intermediate ODV behavior (Figure S2); these regions are not examined in detail but are included here for comparison. Notably, none of the ODVs in these regions have approached the maximum ODVs recorded in the SoCAB (indicated by blue lines in Figures 3 and S2). There are three designated ozone nonattainment areas in the northern U.S. states (based on the 2015 ozone NAAQS - U.S. EPA's "Green Book" https://www.epa.gov/green-book, last accessed 8 July 2019), which include 38 counties in three of northeastern states - Connecticut, New Jersey and New York.

In this paper we apply the approach of Parrish et al. (2017a) to examine the temporal and spatial variability of the highest ozone concentrations (i.e., the ODVs) observed over the past three to four decades in the two contrasting regions of the northern U.S. representing extremes in anthropogenic influence. We separately estimate the U.S. background ODVs and the enhancements of the ODVs above that background contribution due to photochemical production from U.S. anthropogenic precursor emissions. The U.S. background ODV estimates quantify the maximum ozone concentrations that would exist in these regions in the absence of U.S anthropogenic precursor emissions. We also aim to quantify the temporal evolution and spatial variability of the US anthropogenic ODV enhancements, and based on past trends, project the expected time required for the maximum ozone concentrations to decrease to the 70 ppb NAAQS in the northeastern U.S.

Photochemical modeling systems are generally utilized for quantifications and projections of ODVs (e.g., Dolwick et al., 2015; Emery et al., 2012; Fiore et al., 2014). However, present model quantifications of U.S. ozone concentrations have large uncertainties (Jaffe et al., 2018; Guo et al., 2018). An observational-based approach such as presented here provides useful comparisons for the results of modeling efforts, and differences between the two approaches identify needs for further research.

The analysis approach in this paper relies on differences in the temporal behavior of the U.S. background ODV (demonstrated in this work to be approximately constant) and ODV enhancements resulting from US anthropogenic precursor emissions; these enhancements have greatly decreased over recent decades in response to U.S. emission controls. Previously published studies have identified a multitude of additional processes that potentially can make systematic contributions on a variety of time scales to the variability of ozone concentrations at U.S. surface sites; however, there has been little in the way of systematic, quantitative analysis of their effects on ozone concentrations across the U.S. In this work, we first quantify the U.S. background ODVs and the temporal decrease of U.S. anthropogenic ODV enhancements,

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and then discuss the influence of other processes through examination of the fraction of the ODV variance not accounted for by decreasing U.S. anthropogenic ODV enhancements.

Papers investigating U.S. surface ozone trends (see Lin et al., 2017 and references therein) have treated a variety of statistics (medians, means, and various percentiles) to characterize ozone concentrations. In this work all trends are based on ODVs. The reason for this choice is that the NAAQS is based on this statistic, and thus it is most relevant for policy considerations. The ODV corresponds to ~98th percentile of the MDA8 concentrations during the ozone season. As a consequence, the U.S. background ODVs that we discuss are significantly larger than average or median background ozone concentrations examined in other studies. Given these different choices, care must be taken in comparing trends derived in this work with those from other analyses.

The sources of data and the analysis methods are discussed in the next section, followed by the applications of those methods to quantify the U.S. background ODVs and the U.S. anthropogenic enhancements in the rural western region (Section 3.1) and the northeastern U.S. (Section 3.2). The larger temporal ODV trends and the greater spatial variation of those trends in the northeastern U.S. provide the basis for the elucidation of several features of regional ozone concentrations. Section 3.3 examines the uncertainty of the analysis approach used in Section 3.2. Section 4 gives a summary of the approach and the results, discusses implications of those results, and identifies needs for further research.

2 Data and Methodology

2.1 Ozone Design Values analyzed

This work considers Ozone Design Values (ODVs) reported from the beginning of U.S. ozone monitoring in the mid-1970s through 2017 in seventeen northern U.S. states. An ODV, the statistic upon which the U.S. NAAQS is based, is calculated every year for each ozone monitoring station in the U.S. if the measurements achieve the specified completeness criteria. Each year all recorded ODVs are added to EPA's AQS data archive (https://www.epa.gov/aqs last accessed 23 June 2019). All ODVs reported for the northern states were downloaded from this archive; only the ODVs marked as valid were retained for analysis. Exceptional events that have concurrence from the U.S. EPA were excluded. Table S1 summarizes these archived ODVs for each state, including the number of monitoring sites, the years spanned by the reported ODVs, and their maximum and minimum values. The reported ODVs span the range from 169 ppb to 41 ppb. Yellowstone National Park (NP) in another state (Wyoming) is also included because its measurement record has been examined in previous analyses of long-term trends of U.S. background ozone concentrations (e.g., Lin et al., 2017). It should be noted that very few sites have continuous measurements over the indicated time spans, and that many sites operated for only short periods. All reported ODVs are included in this analysis, even if only a single ODV was reported for a particular site. It is implicitly assumed that

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the temporal discontinuities associated with initiation or termination of individual sites does not prevent an accurate quantification of temporal trends of ODVs within the regions selected for analysis.

2.2 Exponential ODV trend analysis

A well-established conceptual model (e.g., Parrish et al., 1986) guides our analysis. Ambient ozone concentrations at U.S. surface sites are composed of two contributions: 1) background ozone and 2) enhancements resulting from ozone produced from photochemical processing of U.S. anthropogenic emissions of ozone precursors. The first contribution is the ozone that would be present in the absence of U.S. emissions of ozone precursors from anthropogenic sources; this ozone is transported into the U.S. or produced over the U.S. from naturally emitted precursors. The U.S. Environmental Protection Agency (EPA) has defined this contribution as U.S. background ozone (e.g., Dolwick et al., 2015). The first contribution has remained relatively constant, while the second contribution has greatly decreased over the past 2 to 4 decades in response to reductions in anthropogenic emissions of ozone precursors.

In this work we focus on the time period of decreasing ODVs. Fitting observational data to a simple functional form is a common tool utilized for quantitative observational analysis; linear trend analysis (i.e., fitting observational data to a linear function) is one example. Here we choose to fit observed ODVs to Equation 1,

$$\mathrm{ODV} = y_0 + A \exp\{-(\mathrm{year}-2000)/\tau\}, \qquad (1)$$

with three undetermined parameters. This equation is the simplest possible functional form consistent with the guiding conceptual model of a background contribution and a consistently decreasing anthropogenic contribution. (A linear fit with only two undetermined parameters – slope and intercept – is simpler, but cannot fit a positive background contribution, as a decreasing linear fit will eventually go negative.) We identify the first term of Equation 1, y_0, as an estimate of the ODV that would result from U.S. background ozone alone (i.e., consistently called U.S. background ODV), and the second term as an estimate of the enhancement of observed ODVs above y_0 (i.e., consistently called U.S. anthropogenic ODV enhancement) due to contributions from photochemical processing of U.S. anthropogenic precursor emissions. This second term decreases exponentially with a time constant of τ, and equals A in the reference year, which we choose as 2000.

A simple intuitive argument suggests that an exponential decrease in the anthropogenic ozone contribution is expected to be a reasonable approximation for the response of maximum ozone concentrations to implementation of emission controls. When controls are initiated, early progress can be rapid since large existing emission sources evolved without planning for their control. With time, reducing emissions will become progressively more difficult, since the most easily controlled emissions will likely be addressed first, and the smaller, remaining emissions will be more difficult and/or expensive to control. This expected increasing difficulty in reducing emissions may well lead to an approximately constant fractional

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decrease in anthropogenic ozone enhancements, which corresponds to an approximately exponential decrease in these enhancements.

A previous analysis (Parrish et al., 2017a) quantified the temporal evolution of the maximum ODVs in seven southern California air basins over the 1980-2015 period (shorter periods beginning later and ending in 2015 in two basins). That work utilized fits to Equation 1 (with the reference year 1980 instead of 2000), and showed that a single value of $\tau = 21.9 \pm 1.2$ years, a single value of $y_0 = 62.0 \pm 1.9$ ppb, and a different value of A in each air basin provided an excellent fit ($r^2 = 0.984$) to the ODVs in all of those air basins.

As we will see in the following analysis, in the northeastern states the period of consistently decreasing ODVs (generally 2000 and later, hence the choice of 2000 as the reference year in Equation 1) is too short to allow precise determinations of all three parameters of Equation 1 from fits to individual ODV time series. In face of this difficulty, our primary analysis approach is to assume that the τ value (21.9 years) derived for southern California is also appropriate for the northeastern states. Uncertainty in the value of τ is then the greatest source of uncertainty in the analysis results; this impact of this uncertainty will be addressed in Section 3.3.2.

Equation 1 assumes that decreasing U.S. anthropogenic ODV enhancements is the only cause of ODV variability at a particular location. Other factors (e.g., rising anthropogenic emissions in Asia, variable occurrences of wild fires, interannual meteorological and climate variability, etc.) can also potentially affect observed ODVs. The approach taken here is to interpret the observed ODVs initially on the basis of Equation 1, and to examine the fraction of the ODV variance captured by that interpretation. The remaining fraction of the variance is then attributed to other factors, including those listed above. We use two statistics to quantify the variance. The root-mean-square deviation (RMSD) between the derived fit and the observed ODVs gives an absolute measure (in ppb) of the ODV variability not captured by Equation 1. The square of the correlation coefficient (r^2) between the observed ODVs and the values derived from the fit to Equation 1 gives a measure of the fraction of the total ODV variance that is captured by that fit; the difference between unity and the r^2 value is then a relative measure (as a fraction) of the ODV variance not captured by Equation 1. In the southern California air basins (Parrish et al., 2017a), the derived $r^2 = 0.984$ and RMSD ≈ 4 ppb indicate that all factors not included in Equation 1 account for no more than 1.6% of the total variance in the basin maximum ODVs analyzed in that work, and contribute a RMSD to those ODVs of no more than ~ 4 ppb.

A potential complication in the interpretation of the two terms of Equation 1 arises if there is a significant fraction of U.S. anthropogenic ozone precursor emissions that has not been reduced by emission controls. Ozone produced from such emissions will not have decreased in the same manner as that produced from most U.S. anthropogenic emissions, which could raise the derived value of y_0 above the actual U.S. background ODV. Parrish et al. (2017a) have discussed this issue

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with regard to the emissions associated with the intense agricultural activity in the Imperial Valley of the Salton Sea air basin, where the derived y_0 is higher than in other southern California air basins. The final section of this paper briefly considers the possible impact of this complication in the northeastern U.S. states.

One difference between the application here and that of Parrish et al. (2017a) should be noted. The former work chose 1980 as the reference year, while here we choose the year 2000. The curves derived from the fits to Equation 1 and the values derived for the y_0 parameter do not depend on the choice of reference year, while the values derived for the A parameter do. Consequently, comparing the A parameters derived here with those given for California by Parrish et al. (2017a) requires adjustments for this difference, which can be provided through the second term of Equation 1.

2.3 Additional observation-based analyses of ODV time series

Acknowledging the uncertainty introduced by the assumptions required to implement the exponential analysis described in Section 2.2, we also derive y_0 through two somewhat different approaches that also provide two estimates of τ appropriate for the northeastern states. These τ values offer insight into the uncertainty of the value of 21.9 years assumed for τ in the exponential analysis.

An independent analysis approach discussed in Section 2.3 of Parrish et al. (2017a) can estimate U.S. background ODVs without assuming any specific functional form for the time dependence of the ODV enhancements. Different assumptions underlie this analysis - namely that all of the ODV time series under consideration follow the same functional form, but not necessarily an exponential decrease, and that all time series are approaching a common U.S. background ODV (i.e., y_0 value). These assumptions imply that all of the time series will converge to a common ODV as anthropogenic precursor emissions are reduced to zero; this common ODV is necessarily the regional U.S. background ODV. In practice this analysis uses correlations between time series of ODVs with U.S. anthropogenic ODV enhancements that differ as much as possible. One time series is selected as a reference; in the examples discussed here the time series with the largest U.S. anthropogenic ODV enhancements is selected. Other time series are then linearly correlated with this reference. The intercept of each linear correlation with the 1:1 line then provides an estimate of the U.S. background ODV; at that point the ODVs from the two time series are equal. Parrish et al. (2017a) show that the results of this approach for seven southern California air basins are nearly identical to the results from fits to Equation 1. We apply this approach to estimate U.S. background ODV in the northeastern U.S. and compare the results to those from the exponential analysis.

Two approaches can approximately quantify the value of τ in the northeastern states – analysis of linear fits to segments of ODV time series, and simultaneous least square regression fits to several ODV time series; these approaches help to constrain the uncertainty of the assumed value of τ (21.9 years). Both of these approaches assume that constant values of y_0 and τ are appropriate for all ODV time series included in each analysis. Section 2.4 of Parrish et al. (2017a) describes an

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iterative, non-linear regression analysis that simultaneously derives values for τ and y_0, plus the A parameter for each ODV time series included in the analysis. This analysis will be adapted to the northeastern U.S. ODV time series.

Finally, a linear fit to the initial period of decreasing ODVs provides direct information regarding the magnitude of τ and y_0. The absolute value and the time derivative of Equation 1 when evaluated at year 2000 are $y_0 + A$ and $-A/\tau$, respectively. Fits to two ODV time series provide four parameters (τ, y_0, A_1 and A_2) if the τ and y_0 values are the same for the two time series. Algebraic manipulation gives $\tau = -\Delta_{intercept} / \Delta_{slope}$, where Δ indicates the difference in the subscripted parameter between the two linear fits, and $y_0 = (\Sigma_{intercept} + \tau * \Sigma_{slope})/2$, where Σ indicates the sum of the subscripted parameter from the two fits. A complication with this approach is that the linear fits to time periods of significant length give biased measures of the derivative and year 2000 value of Equation 1; however, this bias can be corrected to first order through numerical comparison of a linear fit to the selected period of the exponential fit.

2.4 Confidence limits and uncertainties

In this work we consistently give 95% confidence limits for derived parameters, unless indicated otherwise. Most of the analysis in this work is based on non-linear, least-squares regression fits of the archived ODVs to Equation 1, and interpretation of the derived values for y_0 and A. In this interpretation it is important to properly consider the uncertainty of these values. We begin with the 95% confidence limits given by the least-squares fitting routines, which are then adjusted to account for the known covariance between the recorded ODVs. Each ODV is a three-year running mean; therefore only every third ODV is independent from the others determined at a given site. Consequently, the number of independent ODVs in each fit is approximately a factor of three smaller than the number of reported ODVs. Thus, all confidence limits derived from the fitting routines have been increased by a factor of $3^{1/2}$ to account for this covariance. Note that the confidence limits are typically 1 to a few ppb; thus results and their confidence limits are often given to 0.1 ppb precision, even though the last significant figure is likely not justified.

There are additional sources of covariance between the ODVs included in any particular fit. The ODVs from different sites within a region can co-vary due to regionally coherent interannual variability, and interannual variability may lead to covariance between ozone concentrations measured in successive years. We are not able to account for the effect of this additional covariance; the derived confidence limits are thus lower limits for the true confidence limits of the derived parameters. However, as discussed in the next section, we can find no indication that additional regional or temporal covariance of the ODVs makes significant contributions to the uncertainties of the results. The influence of often cited major drivers of temporal variation of ozone, which could possibly cause such covariance, is discussed in Section 4, and found to be small.

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3 Results

Here we examine the time series of ODVs from the western rural states (Section 3.1), fit the time series of ODVs from the northeastern states to Equation 1 (Section 3.2) and discuss the results in the context of the conceptual model introduced above. This model considers the recorded ODVs to comprise two contributions: 1) an approximately constant U.S. background ODV identified with y_0 in Equation 1, and 2) U.S. anthropogenic ODV enhancements, which are approximated by the second term in Equation 1. Section 3.2 and Section S1 of the Supplement discuss further details of the spatial and temporal variability of ODVs in the northeastern states. Section 3.3 uses the alternative approaches described in Section 2.3 to examine the uncertainty inherent in the parameter determinations from the exponential analysis using Equation 1.

3.1 ODVs in rural western states

The sparsely populated, three-state, rural western region generally lies on the Northern U.S. Great Plains downwind of more mountainous terrain to the west. Figure 1 shows a topographical map of the region, with the locations of the ozone monitoring sites indicated. This area gradually slopes to the east and north. All of the monitoring sites lie below 1.55 km elevation, with the exception of Yellowstone NP at 2.43 km.

The histories of the ODVs recorded in the region are illustrated in Figures 3 and 4 and averages with standard deviations and variances are given in Table 1. The gaps in the Montana and South Dakota records were caused by extended periods when no valid ODVs were recorded at any site within the respective states. Throughout the ODV record there is little variability due to any cause. The 283 tabulated ODVs recorded over 39 years at 35 sites in the three states average 59.3 ppb with a standard deviation of 3.7 ppb (corresponding to a variance of 13.4 ppb^2) – strong evidence that the ODVs correspond to an approximately constant U.S. background ODV within this region with no evidence for significant U.S. anthropogenic ODV enhancements. At the individual sites and within each state the entire measurement records are all well described by averages with small standard deviations (Table 1): < 3 ppb in Montana and North Dakota, and < 4 ppb in South Dakota, the state whose sampling sites span the largest elevation range (0.34 to 1.55 km). U.S. background ODVs generally increase with the elevation of the sampling site (e.g., see discussion in Jaffe et al., 2018), so larger variability is expected when the monitoring sites within a state span a larger range of elevations. The state averages in Table 1 lie within a range of ~6 ppb, but there are some significant differences: a maximum in South Dakota (61.5 ppb) and a minimum in Montana (55.4 ppb), with North Dakota intermediate (59.3 ppb). Consistent with the site elevation differences, the average ODV at Yellowstone NP is significantly larger than that at Glacier NP: 64.0 ppb at 2.43 km and 54.5 ppb at 0.96 km, respectively. The variances of the data sets vary from 2 to 15 ppb^2; these values indicate that only very small variance in long-term ODV records can arise from variation of U.S. background ozone alone, at least in this particular region of the country.

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3.2 Exponential fits to ODVs in northeastern states

A topographical map showing the networks of ozone monitoring sites in the eight northeastern U.S. states is given in Figure 2. All of the ODVs recorded in four of the eight states are plotted in Figures 5 and 6, along with curves showing fits of Equation 1 to the ODVs from selected groups of sites over selected time periods. These ODV time series are in striking contrast to those in the rural western states (compare Figures 5 and 6 with Figure 4) with much larger concentrations showing strong decreases over the past two to three decades, and much greater variability of ODV values. We attribute this contrast to the much greater influence of U.S. anthropogenic ODV enhancements in the northeastern states. The greater variability is quantitatively reflected in the ODV variance in this region (251 ppb^2), which is nearly a factor of 20 larger than that seen in the rural western states; this comparison shows the dominant influence of the U.S. anthropogenic ODV enhancements in the northeastern states.

The four states included in Figures 5 and 6 are shown for illustrative purposes, with Figures S3-S10 of the Supplement showing detailed ODV temporal plots and fitted curves to the selected groups of sites in all eight states. These groups of sites were selected to represent different environments within each state, with the expectation that similar temporal ODV trends will be found at all sites within each group. The strategy adopted is to fit the ODVs recorded at all sites within each group over the time period beginning when a clear, consistent decrease in ODVs is first established, and continuing through 2017, the most recent ODVs available. This strategy is required since Equation 1 is designed to provide fits to ODVs only during such periods of consistent decreases. In all cases these fits begin by 2000, with some beginning earlier - either at the start of measurement record, 1990 or 1995 determined by the best, consistent fit to the functional form of Equation 1. Figures S3-S10 include maps indicating the locations of all selected groups of sites. In all, seventeen groups within the eight states were selected; they are listed in Table 2 along with the parameters derived from the fits of Equation 1.

There are some consistent general features of the ODV time series and the corresponding fits that inform the following analysis:

- Throughout the measurement record, the largest ODVs are found in the states that contain the New York City metropolitan area (New York, New Jersey and southwestern Connecticut), or that lie directly downwind (coastal Connecticut and Long Island, New York). Such sites compose two of the selected groups of sites in New York and Connecticut (see highlighted points in that area in the map of Figure 2), whose ODVs and fits of Equation 1 are highlighted in Figure 5.
- In several states, the largest ODVs are recorded at coastal sites (i.e., Connecticut, Massachusetts, New Hampshire and Maine in Figures 5, 6, S5, S7, S8 and S10). The large ODVs at coastal sites emphasize the important, widely-discussed (e.g., Wolff and Lioy, 1980; Wilcox, 1996) role of transport in bringing high ozone concentrations from the major East Coast urban areas far downwind, particularly when that transport occurs over the waters of Long Island Sound and the Coastal Atlantic Ocean. Two relatively isolated Massachusetts coastal sites on the offshore island of Martha's Vineyard

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and near the tip of Cape Cod record some of the highest ODVs within that state (see Figure S7). Dukes County, which includes only Martha's Vineyard, with a total population of ~17,000 was once designated as a marginal non-attainment area for ozone.

- In the past, ODVs at rural, generally upwind sites on the western border of New York (green symbols in on the left in Figure 2) were significantly smaller than in the northeastern U.S. urban areas, although in recent years that difference has diminished (Figure 5). These upwind rural areas in New York, and similar sites in Vermont (Figure S9), experienced ozone concentrations exceeding 80 ppb throughout the measurement record until about 2005. These high concentrations caused Chautauqua County, N.Y., with a population of ~95,000, to also once be designated as a marginal non-attainment area, again emphasizing the importance of ozone transport in the northeastern U.S., although in this case the source of the transported ozone is not as clearly established.

Additional systematic features of the ODV time series in the northeastern U.S. are discussed in Section S1 of the Supplement.

All of the curves derived from the fits of Equation 1 to the long-term trends of the ODVs shown in Figures 5, 6 and S3-S10 are compared in Figure 7, with the corresponding parameters included in Table 2. Except for the four fits denoted by the colored dotted and dashed curves, all fits are similar in the sense that they exhibit the same relative long-term decrease and are asymptotically approaching approximately the same value of y_0. The same relative long-term decrease is necessarily forced by the use of the same value of $\tau = 21.9$ years in all fits. However, the derived A and y_0 values do provide information regarding the spatial and temporal variation of ODVs over the past two to three decades. Three of the four curves with noticeably different behavior are from fits to the groups of sites with the highest recently reported ODVs (Connecticut, especially the coastal sites, and the New York sites highlighted in Figures 2 and 5); these are discussed further in Section S1 of the Supplement. The fourth exception is the one high elevation site (Mt. Washington in New Hampshire at an elevation of 1.9 km), which is also discussed separately in in Section S1. The parameters in Table 2 provide the basis for quantitatively comparing the fits throughout the northeastern U.S. in the next two sections.

3.2.1 Estimation of U.S. background ODV in northeastern states from exponential fits

All y_0 values in Table 2 (excluding the four exceptions indicated in Figure 7) agree with each other within their indicated confidence limits. The arithmetic mean of these y_0 values is 45.9 ppb with a standard deviation of 3.2 ppb. The average of these y_0 values weighted with the inverse square of the respective confidence limits is 45.8 ± 1.7 ppb, where the 95% confidence limit of this average is indicated. All of the y_0 values in Table 2 agree (again excluding the four exceptions noted above) with these average values within their indicated confidence limits. Figure S11 of the Supplement shows the distribution of the y_0 determinations; 13 of the 17 derived y_0 values approximately define a normal distribution with a median of 47.7 ppb and a standard deviation of 4.5 ppb. The median is interpreted as representing a common regional y_0 value, and

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the standard deviation as reflecting the uncertainty in determining each y_0 value. This median is consistent with the above averages. The highest 4 of 17 derived y_0 values define a high value tail; these are the 4 four exceptions indicated in Figure 7.

Recalling earlier discussion, we identify the average y_0 = 45.8 as the best estimate of the U.S. background ODV throughout the northeastern U.S.; there is no discernable spatial variability within this region. This value is significantly smaller than the value of 62.0 ± 1.9 ppb derived for southern California (Parrish et al., 2017a); however even at this smaller value, the U.S. background ODV in the northeastern U.S. amounts to 65% of the 70 ppb NAAQS.

3.2.2 Estimation of U.S. anthropogenic ODV enhancements in northeastern states from exponential fits

The fits to Equation 1 with τ = 21.9 years provide estimates of *A*, the U.S. ODV enhancement in the reference year 2000; Table 2 lists these values for the 17 selected groups of sites from two-parameter fits, i.e., fits with y_0 and *A* as independent parameters determined from the least-squares fits themselves. However, the results above show that a constant value of y_0 = 45.8 ± 1.7 ppb is characteristic of the entire northeastern U.S. region. Using this result allows fits of Equation 1 to all groups of sites without the larger uncertainty in the y_0 derived from the individual fits. Consequently, results of one parameter fits of Equation 1 (i.e., with y_0 held constant at the value of 45.8 ppb) are included in Table 1 as the *A** values. (Such a fit is not included for the Mt. Washington results, since U.S. background ODV is evidently greater than 45.8 ppb as discussed in Section S1 of the Supplement). The *A** values generally agree with the *A* values from the two-parameter fits within their confidence limits, which are smaller since only one parameter need be derived. The exceptions to the agreement between *A* and *A** are the fits to the exceptions discussed earlier - the two groups of Connecticut sites and the New York maximum ozone sites, which are the upper three colored curves in Figure 7. In Table 2 the *A* values for these three groups of sites are anomalously small compared to the results from neighboring groups of sites (i.e., New Jersey, Rhode Island, Massachusetts/coastal); the *A** values for all of these neighboring groups of sites agree more closely. In the following discussion we take these *A** values as the best estimate for the U.S. anthropogenic ODV enhancements in the northeastern states.

A contour plot (Figure 8) derived from the *A** values in Table 2 provides an overview of the spatial variation of the U.S. anthropogenic ODV enhancements across the northeastern U.S. The groups of selected sites fit to Equation 1 give only coarse spatial resolution across the region, so the contour plot has uncertainties not apparent from the smooth spatial variability of this figure. This uncertainty has been mitigated in deriving the contour plot by including duplicate *A** values at the site locations in each selected group of sites; these additions ensure that the contouring program reproduces a more nearly constant value over the sometimes large regions covered by the selected groups of sites. Despite the uncertainties, the contour plot does give a useful, semi-quantitative representation of the magnitude and regional variation of the U.S. anthropogenic ODV enhancements in the region. Note that the contour plot and the *A* and *A** values of Table 2 describe the ODV enhancements in the year 2000. As is apparent from Equation 1 and the illustrated temporal trends in the figures, the

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ODVs have decreased throughout the last two to three decades. The e-folding time of τ = 21.9 years implies that between the reference year of 2000 and 2017, the ODV enhancements decreased by a factor of 2.2. Hence, dividing the year 2000 ODVs in the contour plot by that factor gives an approximation of the 2017 U.S. anthropogenic ODV enhancements.

The ability of Equation 1 to accurately reproduce observed ODVs can be judged by comparing the observed ODVs with the values predicted from the fits derived with y_0 = 45.8 ppb and τ = 21.9 years. Figure 9a shows this comparison as a correlation plot. The fits for ODVs recorded at all sites in the eight northeastern states over the entire measurement period are calculated from the A^* values at each site interpolated from the contour plot of Figure 8. The correlation is high (r^2 = 0.71) for the 1719 separate ODV values recorded at the 148 sites over the 2000-2017 period, but significantly lower for earlier years as expected from the figures illustrating the derived fits. A general decrease in ODVs throughout the region did not begin until 2000, which is about the time that the U.S. EPA "NOx SIP Call" began reducing power plant NOx emissions across much of the eastern U.S. (Aleksic et al., 2013). There is significant scatter about the 1:1 line in the comparison in Figure 9a; the RMSD between observed and calculated ODVs is 5.6 ppb for the 2000-2017 period. Much of this scatter is due to variability in ODVs recorded at different sites within a given region, which arises from differences in local photochemical ozone production and transport patterns. This variability can be reduced by comparing state maximum ODVs (Figure 9b), rather than individual site ODVs. Figure 10 plots the time series of the maximum ODVs recorded in each year for the eight northeastern states with respective fits over the 2010-2017 period. The derived A^* values (given in Table 3) are somewhat larger than would be expected from the contour plot in Figure 8, consistent with consideration of only the maximum ODVs recorded in each state. Stronger correlation (r^2 = 0.89) is found for the fits to the state maximum ODVs as expected, since considering only the largest of the states' ODVs in a given year removes much of the regional variability across the state.

3.3 Evaluation of uncertainty of the exponential fits to ODVs in northeastern states

Here the methods described in Section 2.3 are applied to investigate the uncertainty of the results from the exponential fits presented in Section 3.2. Section 3.3.4 provides an overall assessment of this uncertainty.

3.3.1 Alternative approach for estimating U.S. background ODVs in the northeastern U.S. states

The independent analysis approach introduced in Section 2.3 can estimate U.S. background ODVs through correlations between separate ODV time series. The ODVs from each of the 13 groups of sites that give the black lines in Figure 7 are included in this analysis. The reference ODV time series chosen is the maximum observed ODVs in the New York City urban area (NYC urban maximum), which is equated to the maximum ODV observed each year in either New York or New Jersey. These maxima (plotted in Figure 11a) are all recorded near the New York urban area. This reference is selected because these are among the largest ODVs recorded in the northeastern U.S., and after 2000 this time series closely follows an exponential decrease with little interannual variability. Figure 12 shows three example linear correlations (the ODVs recorded at the three sets of Massachusetts sites) with that reference. Figures S12-S18 of the Supplement show all of the

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linear regressions for the 13 regional data. Figure S19 compares all of the fits, and Table S2 collects the results. These results are quite variable (25 to 62 ppb) due to the relatively short 2000-2017 data records and because the slopes are not widely different from unity, preventing a precise determination of the intercepts of the correlations with the 1:1 line. However, the average of the derived background ODVs (49.2 ± 3.9 ppb for ordinary linear regressions and 42.5 ± 5.7 ppb for reduced major axis regressions, where 95% confidence limits of the averages are indicated) bracket the result derived from the exponential fits, and neither average is statistically significantly different from that earlier result. The agreement between these two approaches for estimating U.S. background ODVs shows that the assumption of an exponential decrease in the ODV enhancements is not essential for estimating the background ODV (although that approach does give more precise results), and increases our confidence in the results of each approach.

3.3.2 Estimate of τ and y_0 from linear fits to ODV trends in the northeastern U.S. states

Linear fits to the period of decreasing ODVs for three ODV time series are shown in Figure 13. These three series were chosen so that one (NYC urban maximum introduced in the previous section) includes the largest ODVs, and two have some of the smaller ODVs in the northeastern U.S.; this choice gives the largest contrast in the fitted slopes and year 2000 intercepts in order to provide the most precise τ determinations. Table S3 gives the slopes and intercepts of those fits. We calculate τ and y_0 after correcting for the bias between the instantaneous value and slope of Equation 1 in year 2000, and the intercept and slope derived from the linear fits over the 18- and 26-year periods. Table S3 gives these corrected values, which in turn give τ = 21.1 ± 5.9 and 21.7 ± 5.0 years and y_0 = 48.7 and 47.0 ppb for the fit parameters from the upper and lower Figure 13 panels, respectively. These τ values compare quite favorably to the assumed California value (21.9 years), while the y_0 values are larger than derived in the analysis using exponential fits to Equation 1 (45.8 ± 1.7 ppb).

3.3.3 Simultaneous least-squares regression to fits northeastern U.S. state ODV maxima

An iterative, non-linear regression analysis similar to that described in Section 2.4 of Parrish et al. (2017a) and introduced in Section 2.3 is applied here to simultaneously fit seven ODV time series to Equation 1 to determine nine parameter values. The data sets are the 2000-2017 maximum ODVs recorded in seven states plotted in Figure 10. A simultaneous fit to multiple ODV time series improves the precision of the parameter determinations. Values of τ and y_0 (assumed the same for all seven states) and values of A for each of seven states are optimized in an iterative process that minimizes the sum of the squares of the deviations between the fit and the original time series. The resulting parameter values are given in Table S4, and Figure S20 compares the fit results with the original ODVs. The derived τ value (26.0 ± 6.0 years) is larger than the southern California value of (21.9 ± 1.2 years), although it agrees within the derived 95% confidence limit. Correspondingly, the derived y_0 value (41.8 ± 3.0 ppb) is smaller than derived earlier (45.8 ± 1.7 ppb), and the A values are larger (compare to Table 3), but again all agree within the derived confidence limits. This fit captures 89.6 % of the variance in the seven ODV

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time series, comparable to the result shown in Figure 9b. (Note that since its recent ODV behavior is different from the other states, as discussed in Section 3.2, Connecticut is not included in this analysis.)

3.3.4 Assessment of uncertainty of the results

Section 3.2 presents fits of Equation 1 to ODV time series in the northeastern U.S. derived with an assumed value for τ ; all confidence limits given for the derived parameters are lower limits due to this assumption. The above analyses in this Section 3.3 investigate alternative approaches to better constrain the overall uncertainty of the results. With regard to the value of τ, the analysis of Section 3.3.2 gives two values (21.1 ± 5.9 and 21.7 ± 5.0 years) that agree well with the assumed value (21.9 ± 1.2 years) derived by Parrish et al. (2017a) from analysis of ODVs in southern California, while the analysis of Section 3.3.3 gives a larger value (26.0 ± 6.0). Importantly, all of these derived τ estimates agree within their indicated confidence limits, indicating that there is no evidence for a different exponential rate of decrease of U.S. anthropogenic ODV enhancements between southern California and the northeastern states.

With regard to the value of the U.S. background ODV (y_0), Section 3.2 gave 45.8 ± 1.7 ppb using the assumed fixed τ value. The alternative approach of Section 3.3.1 gives two results, 49.2 ± 3.9 ppb and 42.5 ± 5.7 ppb, depending upon the linear fitting approach used, Section 3.3.2 gives two estimates of 48.7 and 47.0 ppb (without easily defined confidence limits), and Section 3.3.3 gives 41.8 ± 3.0 ppb. The average of these five results is 45.8 ± 3.0 ppb, which agrees well with the Section 3.2 result. This average value with the wider confidence limit is taken as the best estimate of y_0.

4 Discussion and Conclusions

The analysis presented in this paper is applied to ODVs from eight northeastern U.S. states, and contrasted with ODVs from three sparsely populated rural western states in the northern U.S. (maps in Figures 1 and 2); it has two complementary parts. First, time series of the highest ozone concentrations (i.e., the ODVs, the statistic upon which the NAAQS is based) in the northeastern states are fit to Equation 1. This equation has two terms - one constant and one exponentially decreasing – with two variable parameters: y_0, the magnitude of the constant term; and A, the year 2000 magnitude of the decreasing term. The fits are limited to the most recent two to four decades, when the ODVs are consistently decreasing, and we assume an e-folding time of τ = 21.9 years in Equation 1 in these fits. The success of the fitting process is judged through standard statistical tests that quantify how well the fits capture the variability of the ODV time series, and quantify the uncertainty of the derived parameter values. The second part of the analysis is the physical interpretation of the parameters derived from the fits to Equation 1; y_0 is taken as an estimate of the U.S. background ODV (i.e., the ODV that would exist in the absence of U.S. anthropogenic emissions of ozone precursors), and the second term is interpreted as an estimate of the regional U.S. anthropogenic ODV enhancement (i.e., the amount that ODVs are enhanced above the U.S. background ODV by

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photochemical production of ozone from existing U.S. anthropogenic precursor emissions). Several alternative analyses are presented to compare with the primary analysis of the exponential fits.

The northeastern states contain major urban centers while the western rural states contain no large cities, leading to marked differences in the ODV time series. In the rural western states the ODVs recorded at 35 sites over a 39-year period show remarkably little variability (Figures 3 and 4), with an overall standard deviation of 3.7 ppb (variance of 13.4 ppb^2). In contrast, the ODVs recorded in the northeastern states vary from >160 to <50 ppb (Figures 3, 5, 6 and S3-S10) with an overall standard deviation of 16 ppb (variance of 251 ppb^2). The derived U.S. background ODV has significant spatial variability. Within the rural western states ODV averages (Table 1) quantify the U.S. background ODV; the values for the three states (55 to 62 ppb) are similar to the value of 62.0 ± 1.9 ppb derived by Parrish et al. (2017a) for large areas of southern California, including the Los Angeles urban area. The U.S. background ODV in the northeastern U.S. states (45.8 ± 3.0 ppb) is significantly smaller than in any of the western U.S. regions. For context, these U.S. background ODVs account for 65 to 90% of the 2015 NAAQS of 70 ppb. In the northeastern U.S. the A parameter (representing the U.S. anthropogenic ODV enhancement) varies spatially as shown by the contour plot in Figure 8, with the largest values (>54 ppb) immediately downwind of New York City decreasing to <22 ppb over northeastern Maine. Importantly, these derived A parameters quantify the U.S. anthropogenic ODV enhancements in the year 2000. By 2017 these enhancements had decreased by a factor of 2.2 according to our analysis; thus the largest ODV enhancements immediately downwind of New York City have decreased to ~25 ppb. No signficant anthropogenic ODV enhancements are present in the rural western states.

4.1 Implications of the results for air quality

The analysis presented here and the results of Parrish et al. (2017a) demonstrate that throughout diverse regions of the country (i.e., rural western states, northeastern U.S., and southern California) the U.S. background ODV contribution is significantly larger than the present-day ODV enhancements due to photochemical production from U.S. anthropogenic precursor emissions. This comparison is true not only in rural areas, but also in the two most populous U.S. urban areas, New York City and Los Angeles. Since these ODVs, upon which the NAAQS is based, represent the largest observed ozone concentrations, degraded air quality due to elevated ozone concentrations is attributed primarily to the U.S. background ODV, with local and regional photochemical production from U.S. anthropogenic precursor emissions enhancing that background by a significant, but smaller amount.

Forward projections of the fits to the maximum ODVs (shown in Figures 10 and 11a) allow an estimate of future trends of ODVs in the northeastern U.S., assuming that the U.S. background ODV (i.e., y_0) remains constant at 45.8 ppb throughout the region, and that the exponential decrease of the U.S. anthropogenic ODV enhancements can be maintained with an e-folding time, τ, of 21.9 years, by means of continued emission reduction efforts. These projections suggest that the

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maximum ODVs throughout the northeastern U.S. will drop below the 2015 NAAQS of 70 ppb by about 2021. However, these projections do not account for the variability of observed maximum ODVs (i.e., RMSD of 3.9 ppb in the northeastern U.S.) about the fitted curves, so that even after 2021 this variability will likely result in the occasional recording of ODVs above 70 ppb.

These forward projections cannot account for any systematic deviations of the ODVs from the behavior given by Equation 1. The recent temporal evolution of ODVs in Connecticut appears to differ significantly from the general regional behavior (see Figures 5-7 and 10). In the discussion of the fit to Equation 1 of the Connecticut ODVs this difference was noted (see dashed colored curves in Figure 7), but nevertheless the temporal evolution was forced with y_0 = 45.8 ppb in deriving the A^* values given in Table 2 and in deriving the contour plot of Figure 8. The different behavior and fits for Connecticut are due to the most recent five years of ODV values lying above the expected trend, as most clearly shown in Figure 10. The cause of this difference is not understood. Whether this difference is simply a statistical fluctuation cannot be determined at this time; however, random fluctuations of similar magnitude are only rarely apparent in the temporal records of ODVs in the states discussed. McDonald et al. (2018) have recently discussed a class of ozone precursor emissions, i.e., volatile chemical products - including pesticides, coatings, printing inks, adhesives, cleaning agents, and personal care products - that have not been addressed by emission controls to the same extent as other emission sectors. The impact of this emission sector on ODVs has not been quantified, but is expected to be most significant in areas of largest population density, exactly the regions where the significant differences in temporal evolution of ODVs are noted.

The higher U.S. background ODV (y_0) in southern California of 62.0 ± 1.9 ppb (Parrish et al., 2017a) compared to the value of 45.8 ± 3.0 ppb derived here for the northeastern U.S. implies much less difficulty in achieving the 2015 ozone NAAQS of 70 ppb in the New York City (NYC) urban area compared to Los Angeles (LA), because the northeastern U.S. has a much larger margin for U.S. anthropogenic enhancement of ODVs while still attaining the NAAQS. Figure 11 compares the U.S. background ODVs and the maximum ODVs in these two urban areas. In 2015 these curves indicated maximum ODVs of 78 and 102 ppb in NYC and LA, respectively. To lower the maximum ODVs to 70 ppb would require respective decreases in total ODVs of 10% in NYC and 31% in LA. However, only the U.S. anthropogenic ODV enhancements can be addressed by local and regional controls of ozone precursor emissions. In 2015 these enhancements were about 25% larger in LA than in NYC (40 and 32 ppb, respectively). To reach a maximum ODV of 70 ppb requires ODV enhancement reductions of 25% in NYC and 80% (i.e. a factor of 5 reduction) in LA. The exponential term of Equation 1 projects that such reductions of the 2015 ODV enhancements will require 5 years in NYC and 35 years in LA; hence the projected years of 2021 and 2050 in NYC and LA, respectively. From the perspective of lowering maximum ODVs to the ozone NAAQS, the most important difference between NYC and LA urban areas is the higher U.S. background ODV in LA, although the 25% larger anthropogenic ODV enhancements in LA play a secondary role. This comparison provides an insightful context for the consideration of relative anthropogenic enhancements of ozone concentrations across the country.

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Finally, it is important to note that from a human health perspective, continuing efforts to reduce ambient ozone concentrations are beneficial, despite the difficulty of achieving the NAAQS. Recent studies establish human health impacts from long-term ozone exposure over several years (Turner et al., 2016; Di et al., 2017; Berger et al., 2017). Therefore, any reduction in ozone concentrations below present levels will benefit U.S. human health, regardless of whether or not ODVs remain above 70 ppb.

4.2 Implications for our understanding of surface ozone concentrations

In this work we have used Equation 1 to quantify the temporal evolution of ODVs in the northeastern U.S.; this equation incorporates a constant U.S. background ODV and decreasing U.S. anthropogenic ODV enhancements, but makes no attempt to account for any other process that affects observed ODVs. Previously published studies have identified a multitude of additional processes that potentially can make time-varying contributions to ozone concentrations at U.S. surface sites, including: stratospheric intrusions, which can bring particularly high ozone concentrations to the surface (Langford et al., 2009, 2014; Lin et al., 2012a, 2015); increasing Asian anthropogenic emissions, which are believed to raise ozone concentrations over the U.S. (Jacob et al., 1999; Lin et al., 2012b); increasing frequency of wildfires, which can produce episodic ozone enhancements (McKeen et al., 2002; Jaffe, 2008, 2013; Pfister et al., 2016); variable meteorological conditions, which can lead to changes in transport patterns (Wang et al., 2016) or changes in the conditions conducive to photochemical ozone production (Shen and Mickley, 2017; Shen et al., 2017); increasing methane, which is argued to increase global ozone concentrations (Fiore et al., 2008, and references therein); and a warming climate, which has been argued may partially offset air quality improvement from regional emission controls by (Fiore et al., 2015). However, there has been little in the way of systematic, quantitative analysis of the effects of these additional processes on ODVs across the U.S. Parrish et al. (2017b) show that baseline ozone concentrations transported ashore at the U.S. west coast have systematically varied over a limit range, presumably due to some of the above mentioned processes. Also, any systematic departure of average ODV trends from the purely exponential decrease incorporated in Equation 1 could contribute ODV variability not captured by our analysis. Here we approximately quantify the total influence of all these additional processes and effects by equating that influence to the ODV variance in the rural western states and the ODV variance in the northeastern states not captured by fits of Equation 1 to the ODVs.

In the rural western states all ODVs reported from 35 sites over 39 years of measurements have a standard deviation of 3.7 ppb, corresponding to a variance of 13.4 ppb^2. At the individual sites and within each state the ODV records are all well described by averages with generally smaller standard deviations (Table 1). For example, Glacier NP is a single site with a 27-year measurement record that is often utilized for characterizing background ozone concentrations (see Lin et al., 2017 and references therein); the ODVs at this site have a standard deviation of only 1.4 ppb. The northeastern U.S. states contrast sharply with the rural western states, because here variation in the anthropogenic ODV enhancements dominates the much

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larger variance (251 ppb^2 for the entire 1975-2015 period). Fits of Equation 1 capture the large majority of this variance in this region; in Figure 9 the r^2 values for 18 years (2000-2017) indicate that Equation 1 captures more than two-thirds of the variance of the individual site ODVs, and 89% of the variance of the maximum ODVs in the eight states. The difference between these percentages is attributed to interannual variability in the spatial distribution of ODVs within the states, plus spatial variability in the ODV enhancements not accurately represented by the contour plot of Figure 8. The RMSD between observed and calculated state maxima ODVs is 3.5 ppb (variance = 12 ppb^2), which is very similar to the standard deviation of 3.7 ppb (13.4 ppb^2 variance) of the average ODVs in the rural western states. The analyses in the two regions agree that the total influence of all factors affecting ODVs over the regions accounts for RMSD ≤ 3.7 ppb, or no more than ~11% of the total ODV variance over the 2000-2017 period in the northeastern states. In summary, Equation 1 is remarkably successful at capturing a large fraction of the ODV variability in the northeastern U.S. states. Guo et al. (2018) discuss a contrasting result; they suggest that monthly regional mean U.S. background MDA8 ozone concentrations vary by up to 15 ppb from year to year, and that a 3-year averaging period (as is used to define the ODV) is not long enough to eliminate interannual variability in background ozone on the days of highest observed ozone. This is not a direct comparison, but it suggests that Guo et al. (2018) overestimate the actual variability of the observed ODVs in the two northern U.S. regions examined in this work and in southern California examined by Parrish et al. (2017a).

The estimates derived in this work for the U.S. background ODV can be compared with model results. Fiore et al. (2014) compare calculations of the fourth highest MDA8 North American background (NAB) ozone (also called policy-relevant background (PRB) ozone) from two global models. The NAB concentration is that which would be present if anthropogenic emissions were reduced to zero throughout North America, not just in the U.S. NAB ozone concentrations are therefore somewhat smaller than U.S. background ozone concentrations, but for the purposes of this comparison, we can ignore this difference. The color scales in their Figures 2 and 10 allow estimates of the U.S. background ODV from the GEOS-Chem and AM3 models, respectively. Similarly, the color scale in Figure 6 of Emery et al. (2012) allows estimates of results from a different version of the GEOS-Chem model for the fourth highest MDA8 PRB. Figure 14 and Table S5 compare the model results with the observationally based estimates of U.S. background ODV derived in this work. These model results do have some skill in calculating the U.S. background ODVs. For five regions (three western rural states, the northeastern U.S. region, and the South Coast Air Basin) the model-observation correlations give r^2 values varying from 0.31 to 0.85, but the model results are on average systematically lower by 4 to 12 ppb. Importantly, the model results disagree with each other, as well as with the observationally based results.

4.3 Possible shortcomings of the analysis

An uncertainty in the fits of the ODV time series to the exponential decay of the ODV enhancement term in Equation 1 is the determination of the time constant, τ. The clear decrease in ODVs across the entire northeastern U.S. did not begin until

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about 2000; the 18-year period of consistent decreases is not long enough for fits of Equation 1 to accurately derive all three parameters. The primary approach we have taken is to use τ = 21.9 years, the value determined for southern California (Parrish et al., 2017a) in the northeastern U.S. as well. It is not clear how the time scales of reductions in U.S. anthropogenic ODV enhancements compare between California and the northeastern U.S. In California, precursor emission reductions may have been faster, because that state may have had more aggressive emission control measures, but they may also have been slower because controls on eastern coal-fired power plants dramatically reduced NOx emissions. This latter reduction would not have occurred in California where such power plants are located downwind, out-of-state. On the other hand, emission reduction rates could be roughly the same, as most northeastern U.S. states have adopted the California on-road light-duty motor vehicle emission control program, and this is a large source sector both in California and the Northeast. The alternative analysis approaches described in Section 2.3 with results discussed in Section 3.3 do not show evidence for a different exponential rate of decrease of U.S. anthropogenic ODV enhancements between southern California and the northeastern states, but uncertainty in the value of τ remains a source of uncertainty in all of the results. The y_0 and A values derived from the fits are sensitive to the selected τ value, with a larger value of τ attributing a smaller fraction of the ODV time series to y_0 and yielding a larger A value.

Finally, Equation 1 implicitly assumes that all sectors of anthropogenic U.S. ozone precursor emissions have been reduced by emission controls at approximately the same rate. However, in some respects this is a poor approximation in that some emission sectors have received lesser efforts than others. Any emissions that have not been reduced would tend to lead to an overestimate in the U.S. background ODV, since ozone produced from those emissions would not have decreased. For example, Parrish et al. (2017a) note that continuing agricultural emissions in the Salton Sea Air Basin may account for the anomalously high y_0 value derived for that region. Here, the possible influence of volatile chemical products (McDonald et al., 2018) in the northeastern U.S. is mentioned above. It is not possible to account for uncertainties in the results that may arise from this issue.

4.4 Needs for further research efforts

Accurately quantifying the U.S. background contribution to ODVs (i.e., the limit to which ODVs can be reduced through U.S. anthropogenic emission reductions alone) is important from the perspective of determining the extent of emission reductions required to attain the ozone NAAQS. In this work we have determined the value of the parameter y_0 of Equation 1 within relatively small uncertainties (estimated 95% confidence limits of ~3 ppb). These uncertainties are derived from the scatter in the observed ODVs about the fits to Equation 1. However, identifying the value of y_0 as the U.S. background ODVs brings in additional possible uncertainties (see discussion in the preceding section) that have not been quantified. Traditionally, models have been used to estimate U.S. background ozone (see Jaffe et al., 2018 and references therein), but the models utilized in these efforts have significant shortcomings (e.g., see discussion in Parrish et al., 2017a), that lead to

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large uncertainties in the results. Jaffe et al. (2018) estimate an uncertainty in modeled seasonal mean U.S. background ozone of about ±10 ppb, with greater uncertainty for individual days (such as those that define the ODV), and Guo et al. (2018) find biases as high as 19 ppb in modeled seasonal mean MDA8 ozone. Thus, modeling and the observational based approach discussed in this paper are both available for estimating U.S. background ODVs, but each has significant, poorly quantified uncertainties.

In summary, effective air quality management can be usefully informed by quantification of U.S. background ODVs. However, given the relatively small differences between estimated U.S. background ODVs and the 2015 ozone NAAQS of 70 ppb, these quantifications will be of more utility if they are accurate to within a couple of ppb (see Figure 11 and associated discussion). Currently, two general approaches are available for estimating U.S. background ODVs (the observational based method discussed here and in Parrish et al. (2017a), and a variety of modelling approaches), but the limited comparisons of results from these two approaches and between the different model results indicate differences much larger than ideal. However, the magnitudes of these disagreements are within the uncertainty of the model estimates as discussed by Jaffe et al. (2018) and Guo et al. (2018). Further improvement is required in modeling systems until their output can accurately reproduce the magnitude and variability of the time series of observed ODVs discussed here; these model calculations could then provide accurate determination of the U.S. background ODVs, the ODV enhancements from U.S. anthropogenic emissions, and robust interpretations of the parameters y_0 and A derived in this work. Until that model improvement is accomplished, the observationally based approach utilized in this work can provide useful estimates for air quality management guidance, as well as for comparison with evolving model calculations.

Competing interests. The author declares that he has no conflict of interest. He is the sole proprietor of David.D.Parrish, LLC, which has had contracts funded by several state and federal agencies and private companies. One of those contracts funded some of this work.

Acknowledgements. Some of the content of this paper was originally developed as a report submitted in fulfillment of the Technical Services Agreement between the Northeast States for Coordinated Air Use Management (NESCAUM) and David.D.Parrish, LLC funded under Agreement No. 101132 from the New York State Energy Research and Development Authority (NYSERDA). NYSERDA has not reviewed the information contained herein, and the opinions expressed in this report do not necessarily reflect those of NYSERDA or the State of New York. The author appreciates the comments and discussion provided by Paul Miller of NESCAUM and Tom Ryerson, Fred Fehsenfeld, Owen Cooper and Andrew Langford of NOAA ESRL CSD. The scientific results and conclusions, as well as any views or opinions expressed herein, are those of the author, and do not necessarily reflect the views of NESCAUM, NOAA, or the Department of Commerce.

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Table 1. ODV statistics from the rural western states.

State/Site	Avg. ± Std. Dev. (ppb)	Variance (ppb^2)	years of ODV record
Montana	55.4 ± 2.2	4.8	1979-2017
Glacier NP	54.5 ± 1.3	2.0	1991-2017
Yellowstone NP	64.0 ± 2.1	4.4	1999-2017
North Dakota	59.3 ± 2.7	7.3	1982-2017
South Dakota	61.5 ± 3.8	14.6	1990-2017

Table 2. Results of least-squares fits to Equation 1 illustrated in Figures 5-7 and S3-S10; RMSD indicates the root-mean-square deviation between the observed ODVs and the derived fit.

State/sites	y_0 (ppb)	A (ppb)	RMSD (ppb)	A^* (ppb)	years fit
New York/maximum O_3	53 ± 6	43 ± 9	3.9	53 ± 2	2000-2017
New York/rural upwind	42 ± 7	50 ± 10	5.1	44 ± 2	2000-2017
New Jersey/all sites	43 ± 4	57 ± 6	4.6	54 ± 2	2000-2017
Connecticut/all sites	56 ± 5	40 ± 7	5.0	55 ± 2	2000-2017
Connecticut/coastal	61 ± 6	36 ± 8	4.1	57 ± 3	2000-2017
Rhode Island/all sites	49 ± 8	44 ± 12	4.0	49 ± 3	2000-2017
Massachusetts/Boston	46 ± 6	27 ± 6	3.1	27 ± 2	1990-2017
Massachusetts/suburban	41 ± 10	52 ± 14	3.3	45 ± 3	2000-2017
Massachusetts/coastal	44 ± 9	52 ± 13	3.2	49 ± 3	2000-2017
New Hampshire/coastal	49 ± 6	35 ± 8	3.7	38 ± 2	1995-2017
New Hampshire/northwest	45 ± 6	29 ± 9	3.7	28 ± 2	2000-2017
New Hampshire/Mt. Washington	66 ± 7	8 ± 8	2.9	---	1993-2017
Vermont /all sites	46 ± 7	34 ± 10	2.7	33 ± 2	2000-2017
Maine/interior	44 ± 8	23 ± 10	5.8	21 ± 3	1990-2017
Maine/NE coast	47 ± 5	22 ± 5	2.0	23 ± 2	1991-2017
Maine/SW coast	49 ± 5	36 ± 5	4.1	39 ± 2	1990-2017
Maine/Cadillac Mtn.	52 ± 16	36 ± 20	5.2	44 ± 5	1997-2017

Table 3. Results of least-squares fits of Equation 1 to the state maximum ODVs illustrated Figure 10; y_0 and τ were held constant at 45.8 ppb and 21.9 years, respectively. The absolute root-mean-square deviations between the observed ODVs and the derived fits are indicated. $Year_{NNAQS}$ indicates the projected year that the fit to the state maximum ODV drops to the NAAQS of 70 ppb.

State	A^* (ppb)	RMSD (ppb)	$Year_{NAAQS}$
Connecticut	61 ± 7	5.8	2021
Maine	48 ± 4	3.2	2015
Massachusetts	53 ± 5	3.9	2017
New Hampshire	43 ± 4	3.0	2013
New Jersey	64 ± 5	3.7	2021
New York	58 ± 4	3.0	2019
Rhode Island	52 ± 4	3.4	2017
Vermont	35 ± 3	2.1	2008

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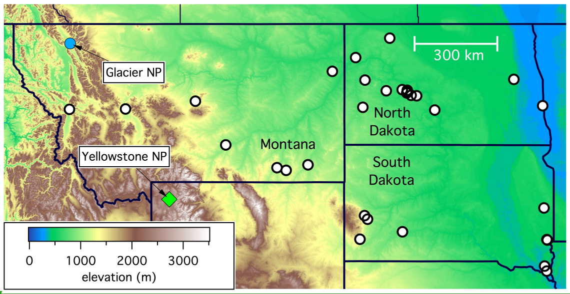

Figure 1: Topographical map of the three rural western states with symbols indicating the locations of the monitoring sites. The two colored symbols indicate two long-term sites in national parks that are discussed in detail. Note that Yellowstone National Park is located in Wyoming, but is nevertheless considered here.

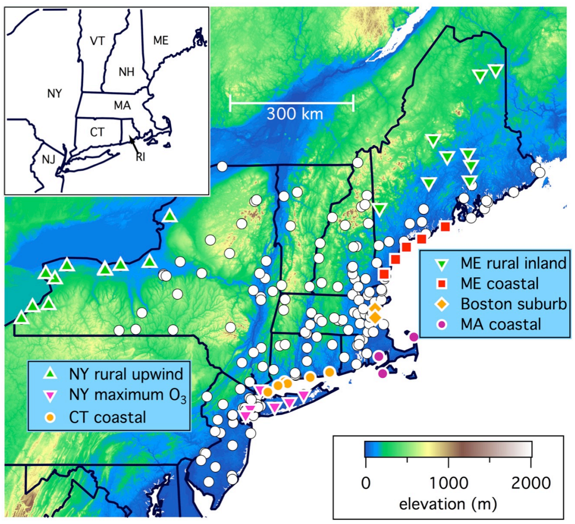

Figure 2: Topographical map of the eight northeastern states with symbols indicating the locations of the ozone monitoring sites. Seven groups of colored symbols indicate groups of sites that are discussed in detail. The inset gives the abbreviations for each of the eight states.

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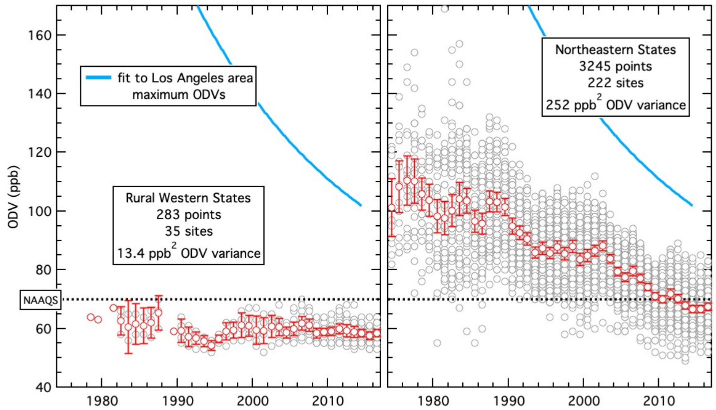

Figure 3: Time series of all ODVs (grey symbols) reported from all monitoring sites in the two northern U.S. regions shown in Figures 1 and 2. The numbers of monitoring sites and reported ODVs and the ODV variance are annotated for each region. The red symbols give the averages and 2-σ confidence limits for all ODVs reported in each year. For comparison, the blue curve in each panel indicates a fit to the maximum ODVs recorded in the Los Angeles urban area (Parrish et al., 2017a). The dotted line indicates the 2015 NAAQS of 70 ppb.

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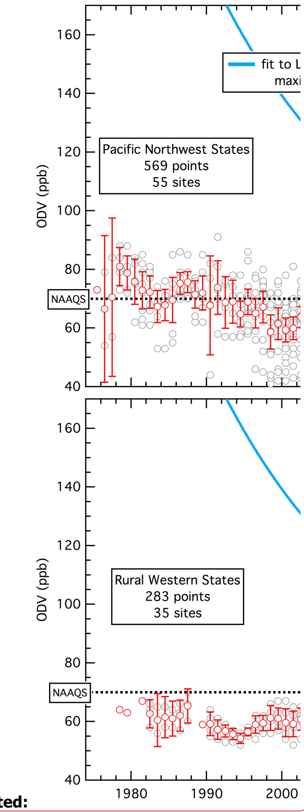

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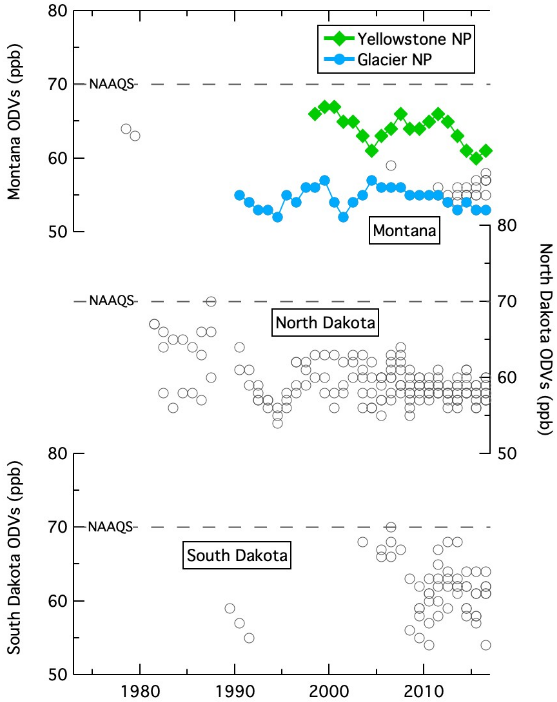

Figure 4: Time series of all ODVs (grey symbols) reported from all monitoring sites in three rural western states, plus Yellowstone NP located in Wyoming. The two sets of colored symbols are results from two long-term sites in national parks.

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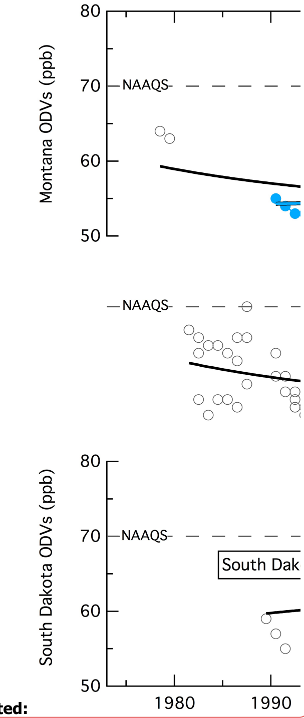

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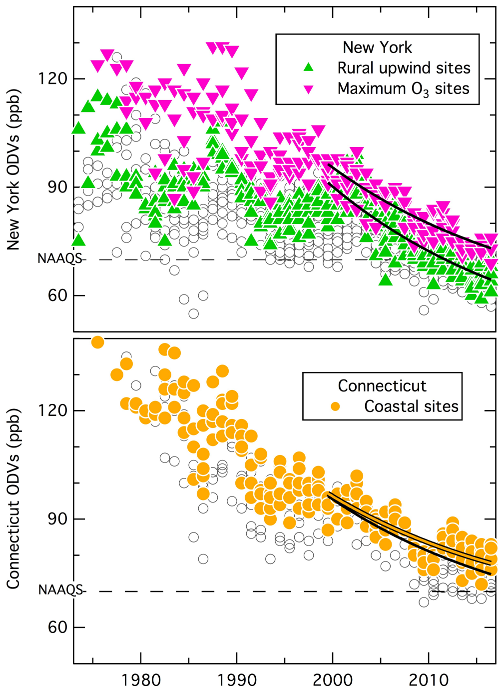

Figure 5: Time series of all ODVs (grey symbols) reported from all monitoring sites in New York and Connecticut. The three sets of colored symbols indicate the results from groups of sites that are discussed in detail. The curves are fits of Equation 1 to respective colored symbols, and to all data points for Connecticut.

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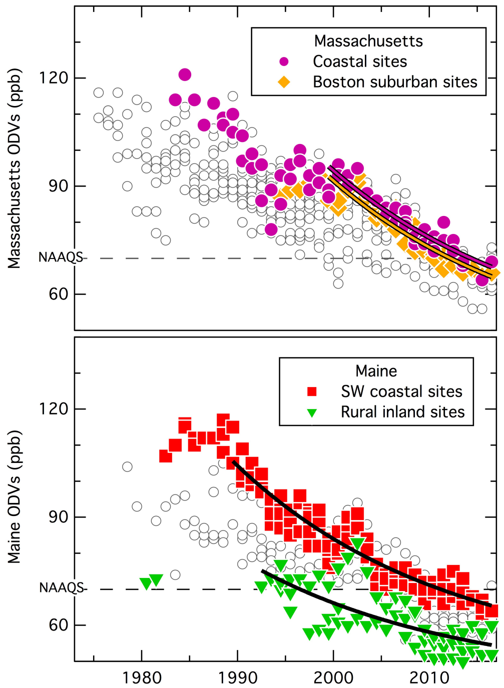

Figure 6: Time series of all ODVs (grey symbols) reported from all monitoring sites in Massachusetts and Maine. The four sets of colored symbols indicate the results from groups of sites that are discussed in detail. The curves are fits of Equation 1 to respective colored symbols.

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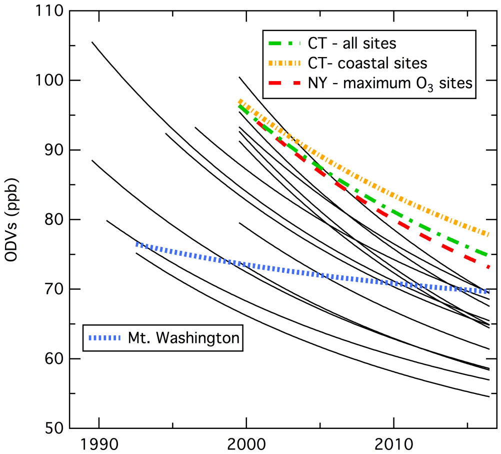

Figure 7: Comparison of fits of the ODVs from 17 groups of sites in 8 northeastern U.S. states shown in Figures 5, 6 and S3-S10 to Equation 1. The parameters of these fits are included in Table 2.

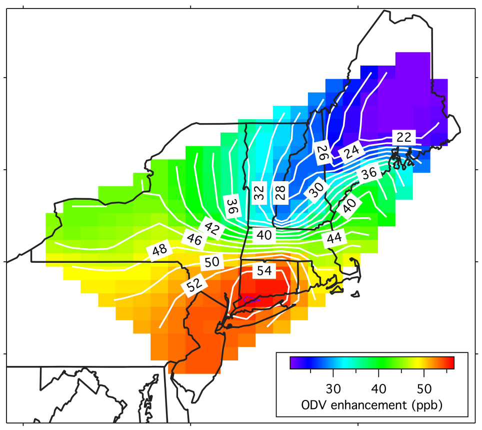

Figure 8: Approximate contour plot of the U.S. anthropogenic ODV enhancement due to photochemical production from precursor emissions in the year 2000, estimated from the A^* values given in Table 2.

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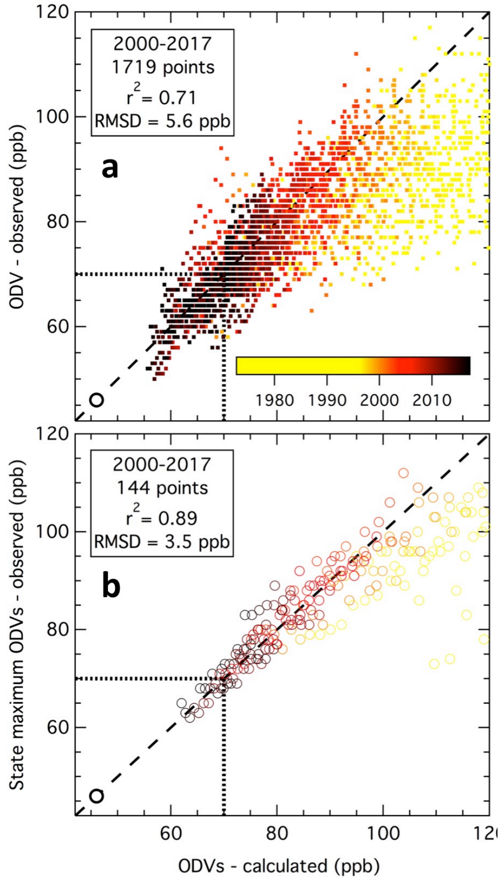

Figure 9. Comparison of observed ODVs color-coded by year with those calculated from Equation 1 for a) all monitoring sites and b) for the maximum observed in each state. The dashed lines indicate the 1:1 relationships with y_0 indicated by the larger circle, and the dotted lines the NAAQS. The number of data, square of the correlation coefficient, and the root-mean-square difference between the observed and calculated ODVs for 2000-2017 are annotated.

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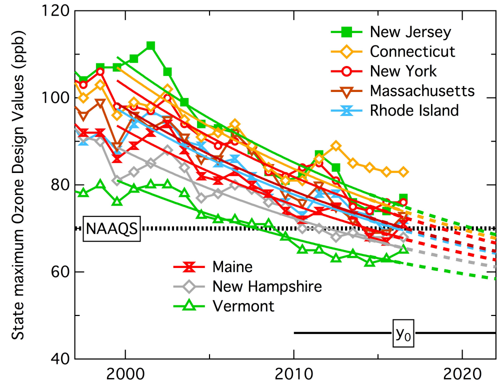

Figure 10: Time series of maximum ODVs reported from any site within each of the eight northeastern states. The solid curves are fits of Equation 1 to the respective colored symbols for the 2010-2017 period. The derived *A values from these latter fits are given in Table 3. The dashed lines are projections of the solid curves.**

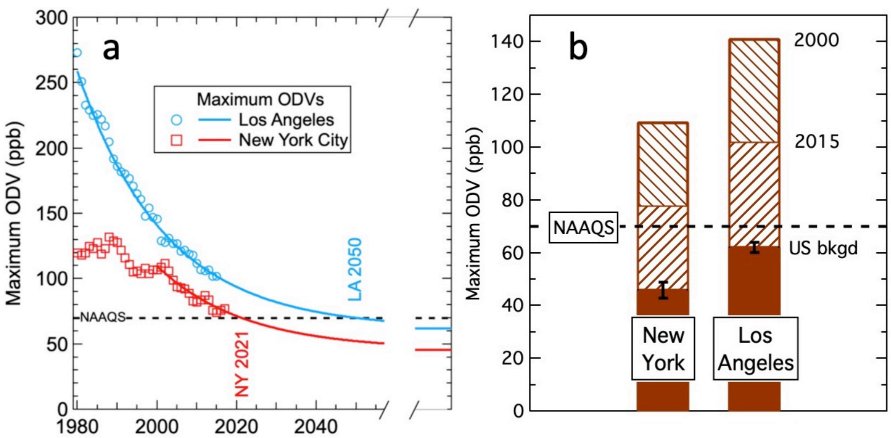

Figure 11. Comparison of maximum observed ODVs in the New York City and Los Angeles urban areas. a) Temporal trend of observations (symbols) and fit to Equation 1, including extrapolation to infinite time; annotations indicate year that extrapolations decrease to 70 ppb. The New York City results are the maxima from either the states of New York or New Jersey, and the Los Angeles results are those for the South Coast Air Basin (Figure 8 of Parrish et al., 2017a). b) Bar graph indicating maximum ODVs in 2000 and 2015 (hatched bars) and the estimated U.S. background ODV (solid bars); the maximum ODVs are derived from the fits to Equation 1 included in a).

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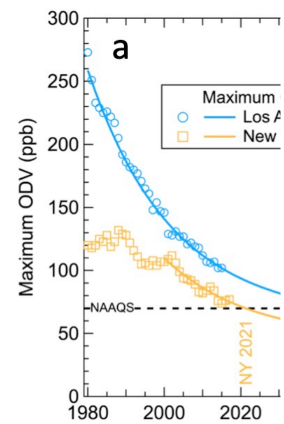

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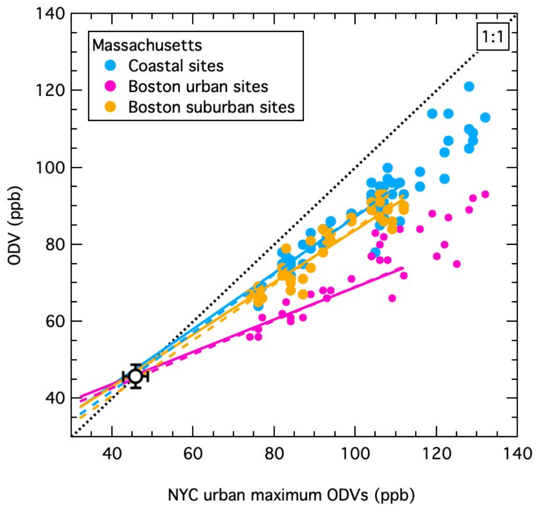

Figure 12. Correlation between the ODVs from three sets of Massachusetts sites and the maximum ODVs recorded in the New York city urban area. Lines of corresponding color show ordinary linear regression (solid) and reduced major axis regression with equal weighting (dashed) fits of the correlated data sets for the ODVs recorded in 2000-2017. The black symbol shows the mean U.S. background ODV derived from the exponential fits to the ODV time series and the dotted line indicates the 1:1 relationship.

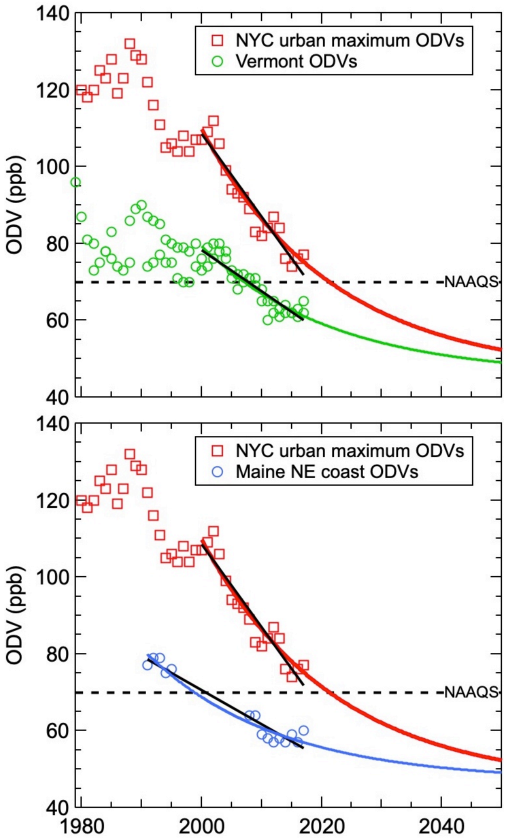

Figure 13: Comparison of linear and exponential fits to three ODV data sets. The black line segments are linear fits to 2000-2017 for two, and 1991-2017 for one. The colored curves are the exponential fits to these time series, also shown in Figures 11, S9 and S10.

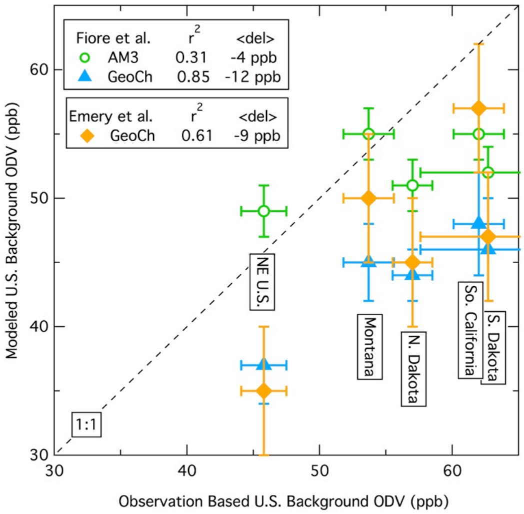

Figure 14: Comparison of U.S. Background ODV estimates from model calculations with those derived in this work from observations. The r^2 values of the correlations and the average differences (<del>) are annotated.

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with τ set to the same value (21.9 years) is used to derive fits to the evolution of ODVs in the northern U.S. states. with τ set to the same value (21.9 years) is used to derive fits to the evolution of ODVs in the northern U.S. states.

As discussed in the Introduction, the conceptual model utilized for interpretation of

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A well-established conceptual model (e.g., Parrish et al., 1986) provides qualitative support for the application of Eq. 1. At any given location in the U.S., we can consider ambient ozone concentrations to be composed of two contributions: 1) background ozone and 2) enhancements resulting from ozone produced from photochemical processing of U.S. anthropogenic emissions of ozone precursors. The first contribution is defined as the ozone that would be present in the absence of U.S. emissions of ozone precursors from anthropogenic sources; this is ozone transported into the U.S., plus that produced over the U.S. from naturally emitted precursors, modified by loss processes. The U.S. Environmental Protection Agency (EPA) has defined this contribution as U.S. background ozone (e.g., Dolwick et al., 2015). The second contribution accounts for ozone produced from U.S. anthropogenic precursor emissions, includes that transported into a region from upwind U.S. sources, and that produced locally and regionally. We identify the first term of Eq. 1, $y_{0,}$ as an estimate of the ODV that would result from U.S. background ozone alone (i.e., U.S. background ODV), and the second term as an estimate of the enhancement of observed ODVs above y_0 due to contributions from U.S. anthropogenic precursor emissions. This interpretation implies that the average of the annual fourth-highest MDA8 ozone concentration in the absence of U.S. anthropogenic emissions would equal y_0, and that in the year 2000 the long-term trend in ODVs is best fit by the sum of y_0 plus A; hence A is identified as the ODV enhancement above y_0 in the year 2000. It should be noted that in the absence of anthropogenic emissions, the four days of highest MDA8 ozone concentrations that would determine y_0 are likely not the same days that determined the actual ODV in any particular year. Indeed, Parrish et al. (2017) show that there has been a systematic shift of the days with maximum ozone concentrations to earlier in the year as the U.S. anthropogenic ozone contribution, with a summertime maximum due to its photochemical source, has decreased, and the U.S. background ozone contribution, which has a springtime maximum, has come to dominate to a progressively greater extent.

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Equation 1 implies that the U.S. anthropogenic contribution has decreased exponentially as emission control efforts reduced ozone produced from all U.S. anthropogenic sources. Associating U.S. background ozone with the parameter y_0 is equivalent to extrapolating long-term trends of observed ODVs to the limit of zero U.S. anthropogenic emissions, when the exponential term in Eq. 1 becomes zero. Parrish et al. (2017) used 36 year ODV records (1980-2015) in southern California to derive a value of τ = 21.9 years for that region of the country. As will be shown in the following discussion, the ODV records in the northern U.S. do not show consistent decreasing trends over such long time periods. As a consequence it is not possible to precisely extract 3 parameters from the regression fits of the measurement records to Eq. 1. Here we take the value of τ = 21.9 years derived from California to apply to all of the northern U.S., which implicitly assumes that control strategies have produced approximately equal relative reductions in anthropogenic ozone enhancements throughout the country. Fixing the value of τ allows precise determinations of the other two parameters (y_0 and A) from the fits to Eq. 1.

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Eq. 1 with the parameters derived in this work and year set to 1980 does give values that can be directly compared to the A parameter values from that earlier work

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Second, the former work considered only the maximum ODV recorded in each year at any of the sites within a given air basin, while here we primarily analyze the ODVs from all sites recorded in a given year in selected regions, although we do also consider the maximum ODVs recorded in each of the northeastern U.S. states. Since y_0 is an estimate of the U.S. background ODV, the value derived for this parameter is expected to be independent of whether the fit of Eq. 1 is made to all ODVs or to only the maximum ODVs in a particular region.

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Mathematically, the first term of Eq. 1, y_0, is the asymptotic value toward which the ODVs approach as the year of the ODV increases, and the second term is the enhancement of the ODVs above y_0, which decreases exponentially with an e-folding time constant of τ years. Thus, A is the enhancement of the ODVs above y_0 in a reference year, defined in this work as the year 2000.

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As discussed in the Introduction, the conceptual model utilized for interpretation of Eq. 1 is simplified; it assumes that decreasing U.S. anthropogenic emissions is the only cause of ODV variability at a particular location. Other factors such as rising anthropogenic emissions in Asia, variable occurrence of wild fires, and interannual meteorological and climate variability can also potentially affect observed ODVs, but they cannot be simply included in Eq. 1. The approach taken here is to interpret the observed ODVs solely on the basis of Eq. 1, and then to examine the fraction of the ODV variability captured by that interpretation. The remaining fraction of the variability is then attributed to other factors, including those listed above. Here we use two statistics to quantify the fraction of the variability not captured by the fits to Eq. 1. The root-mean-square deviation (RMSD) between the derived fit and the observed ODVs gives an absolute measure (in ppb) of the ODV variability not captured by Eq. 1. The square of the correlation coefficient (r^2) between the observed ODVs and the values derived from the fit to Eq. 1 gives a measure of the fraction of the total variability of the ODVs that is captured by that fit; the difference between unity and that r^2 value is then a relative measure (as a fraction) of the ODV variability not captured by Eq. 1. Other factors must then account for the variability quantified by those statistics. In the earlier study of ODVs in southern California air basins (Parrish et al., 2017), the derived $r^2 = 0.984$ and the RMSD $\approx$ 4 ppb indicate that all factors not included in Eq. 1 account for no more than 1.6% of the total variability in the basin maximum ODVs analyzed in that work, and contribute a RMSD to those ODVs of no more than ~ 4ppb.

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A potential complication in the interpretation of the two terms of Eq. 1 arises if there is a significant fraction of U.S. anthropogenic ozone precursor emissions that has not been reduced by emission controls. Ozone produced from such emissions would not have decreased in the same manner as that produced from most U.S. anthropogenic emissions, which could raise the derived value of $y_{0,}$ above the actual U.S. background ODV. Parrish et al. (2017) have discussed this issue with regard to the emissions associated with the intense agricultural activity in the Imperial Valley of the Salton Sea air basin, where the derived y_0 is higher than in other southern California air basins. The final section of this paper briefly considers the possible impact of this complication in the northeastern U.S. states.

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from 2000-2017; the combined influences from all other factors make only relatively minor contributions to the ODV variability across this entire region

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For example, using the Boston data from Figure S5, increasing the assumed value of τ by 10% (from 21.9 to 24.1 years) decreases the derived y_0 estimate by 6% (from 45.8 to 43.3 ppb). Setting τ = 21.9 years means that the confidence limits derived in this analysis are necessarily lower limits. Importantly, the agreement between the results from these exponential fits and the results from the alternative approach discussed in Section 3.2.3 suggests that this additional uncertainty is not large.

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agreement between the results from these exponential fits and the results from the alternative approach discussed in Section 3.2.3 suggests that this additional uncertainty is not large.

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Appendix A. Additional features of ODV time series in the northeastern states

The text of the paper briefly described some consistent general features of the ODV time series and the corresponding fits to Eq. 1 for selected groups of sites in the northeastern U.S. that guided the analysis. Here some additional features of interest are briefly discussed:

New York has two non-attainment areas. In addition to the New York-N. New Jersey-Long Island, NY-NJ-CT moderate non-attainment area with a population of more than 20 million, there is the Chautauqua County (Jamestown), NY marginal non-attainment area with a population of less than 100,000. In Figure S1 the two sites in this latter non-attainment area are highlighted in purple; the

ODVs from these two sites do not differ markedly from the other upwind sites on the western border of the state. In this analysis ODVs from all of the upwind sites are considered together.

Sites in the New York urban area and regions downwind with over-water transport paths from that urban area have recorded the largest observed ODVs. Consistent with this identification, Vermont, the only state with neither major urban areas nor an over-ocean transport path from the New York City area, records the smallest maximum ODVs (see Table 3 and Figure S7).

Although some sites the New York urban area record high ODVs, some other sites in central urban areas in the northeast U.S. are record the lowest ODVs (e.g., New Haven, Connecticut; Providence, Rhode Island, particularly before 2000; and Boston Massachusetts in; see Figures S3, S4 and S7, respectively). This behavior is consistent with fresh NOx emissions in urban areas reducing the ozone concentrations in air masses transported into those areas. This is evidently a very localized phenomenon, as the suburban sites adjacent to Boston (Figure S5) exhibit ODVs similar to other coastal sites in the state.

The farthest downwind coastal monitoring site in northeast Maine (Figure S8) records significantly lower ODVs than other coastal sites, suggesting that ozone concentrations may decrease during transport due to dilution and/or ozone loss to surface deposition.

Interestingly, Connecticut had much higher maximum ODVs than any other state before 1985 (all points above 140 ppb in Figure 2); their cause is unknown. Since 1985 Connecticut ODVs have been similar to those of neighboring states.

Through the measurement record, the differences between maximum and minimum ODVs have decreased, both within individual states and throughout the entire region.

There is one monitoring site at a relatively elevated location in the northeastern U.S. - Mt. Washington in New Hampshire at 1.9 km above sea level (asl). Although the ODV record at this site (Figure S6) is generally not higher than others recorded in New Hampshire, the fit to Eq. 1 shows a much smaller decrease than seen at any other site in the entire region. These ODVs followed a temporal evolution different from any of the other sites in the region (see curves in Figure 8 and parameters in Table 2). The A value (8 ± 8 ppb) is much smaller than that of any other selected set of sites, and the U.S. background ODV (y_0 = 66 ± 7 ppb) is significantly higher than the common y_0 value of 45.8 ± 1.7 ppb derived for the entire northeastern U.S. This difference is attributed to the vertical gradient of ozone over the northeastern U.S. Ozone concentrations in the free troposphere increase with altitude (e.g., see Figure 2 of Fehsenfeld et al., 2006), and it is these higher altitude air parcels that impact Mt. Washington. The value of y_0 derived at Mt. Washington is in reasonable accord with the average ozone concentrations measured over the eastern U.S. by the MOZAIC program in the years near 2000 (Fehsenfeld et al., 2006). The enhancement of the ODVs (i.e., the in A value) in the free troposphere observations at Mt. Washington is much smaller than the enhancements seen at the other sites, which are all located within the planetary boundary layer. Note that the temporal evolution described by the parameters in Table 2 and illustrated in Figures 9 and S6 implies that the Mt. Washington summit site will soon record the highest ODVs in New Hampshire and higher than other sites in the northeastern U.S. outside of and immediately downwind from the New York City urban area; in 2017 Mt. Washington did report the largest ODV in New Hampshire.

The Cadillac Mountain site at in Maine is at a somewhat elevated location (0.47 km asl). In contrast to Mt. Washington, the Cadillac Mountain ODVs (Figure S8) are generally similar to, although slightly higher than, others recorded at the southwest Maine coastal sites. Evidently Cadillac Mountain receives primarily bounday layer air masses. One coastal site is at a relatively high elevation (0.47 km) on Cadillac Mountain.

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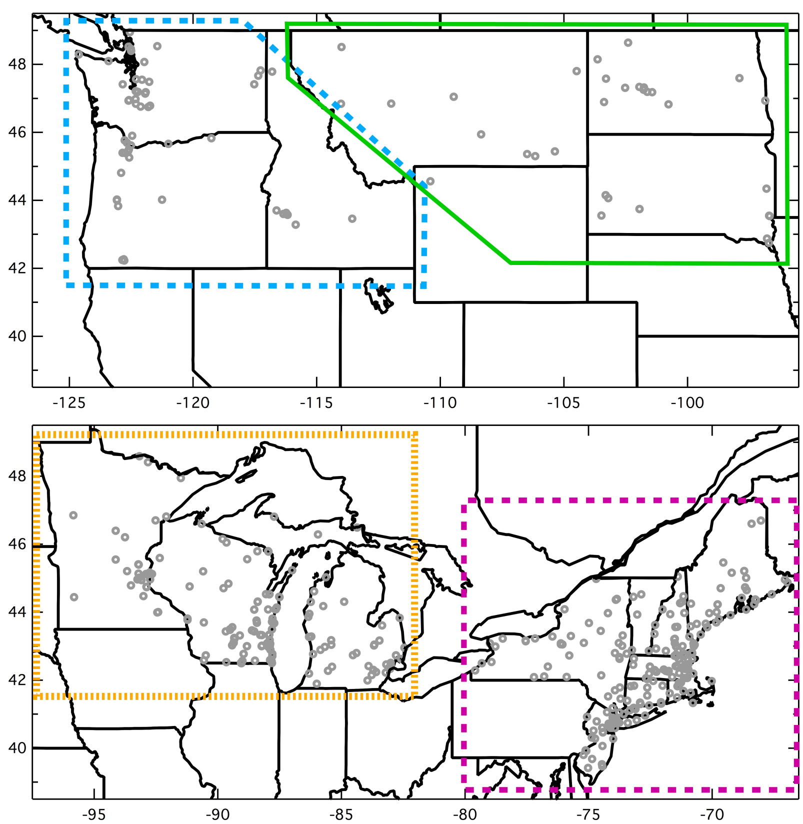

Figure 1: Maps of the northern U.S. with all ozone monitoring sites indicated by grey circles. The colored lines indicate the four regions considered: three Pacific Northwest states (blue dashed), three rural western states (green solid), three midwestern states (dotted orange), and eight northeastern states (purple dashed).

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