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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Observing Entrainment Mixing, Photochemical Ozone Production, and Regional Methane Emissions by Aircraft Using a Simple Mixed-Layer Model" *by* Justin F. Trousdell et al.

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(Referee)Review: Observing Entrainment Mixing, Photochemical Ozone Production, and Regional Methane Emissions by Aircraft Using a Simple Mixed-Layer Model

This paper describes the design and execution of two flight experiments in the San Joaquin Valley of California to quantify entrainment rates and then uses these entrainment velocities to solve for: (a) ozone production rates, (b) methane emissions, and (c) evapotranspiration. The authors are attempting numerous things here, which makes the paper difficult to read and, at times, the results difficult understand. The work is

interesting, but paper would benefit from better organization around a clear goal prior to publication. Adding clarity may be as simple as removing the excessive inessential detail.

General comments:

The Introduction should be reorganized to better frame the work. Some specific issues are as follows. In paragraph 2, the text does not define "tracer method" or "budget of the inversion base height" when describing what is done in the forthcoming analysis. This makes it difficult for the reader to know what is done here and how this work is different from past work.

(Response)We have added some clarification clauses to describe these methodologies, but exact details have to be postponed to the method descriptions of Section 2.

(Referee)The sentence, "by way of targeted airborne campaigns we are able to probe the regional ABL vertically and horizontally and calculate entrainment rates and mesoscale advection," seems key, but is placed awkwardly in the middle of paragraph 3.

(Response)This statement is made after introducing the concepts of entrainment and advection, and therefore does not seem awkward in its placement to us. We have attempted to make a more clarion statement of the paper's overarching goal at the end of paragraph 3, keeping in mind that positional emphasis is typically carried by the end sentence of a paragraph (The Elements of Style, by Strunk & White [1999]):

The central goal of the work presented here is to show how, by way of targeted small-scale airborne campaigns, it is possible to probe the regional ABL vertically and horizontally to calculate entrainment rates and mesoscale advection, and thereby shed light on all of the processes that change the concentrations of trace gases in the boundary layer throughout the day. This methodology thereby reveals the quantitative origins

of chemical constituents measured in near-surface air, by comparing direct observations of all but one of the leading terms of the scalar budget equation, and inferring the unknown term as a residual.

(Referee)The fourth paragraph returns to the idea of scalar budgeting, but still does not define, instead suggesting I should already be familiar with the concept (done through the particular way the references are discussed).

(Response)We have defined a scalar budget in an added subordinate clause in the second paragraph, and a new sentence at the end of the third paragraph as per earlier suggestions. Then we devote the entirety of Section 2.7 to defining exactly what the methodology is. We do not see how to further clarify the technique in the introduction without burdening the section with excessive detail.

(Referee)While I agree with the content in paragraph 5, this paper is not actually about, "better understand[ing] the diurnal behavior of the wintertime boundary layer in the San Joaquin Valley."

(Response)We think that reporting observed entrainment rates in the winter, which have never been reported, does in fact help to better understand the ABL's diurnal behavior.

(Referee)The discussion in paragraph 6 should more relevant to the analysis performed. For example, the paper never significantly discusses PM, but investigates ozone production, methane emissions, and evapotranspiration. While there is some text on ozone and drought here, methane is absent entirely.

(Response)We have added a concluding sentence to this paragraph that helps to establish the importance of the work: Entrainment aloft becomes an even more important factor during stagnant conditions in the SJV because it represents the principal mode of ventilating the air pollutants in the ABL, and therefore its quantification is crucial to predicting the intensity and duration of an air quality episode. Although the work does

not explicitly address PM issues, the results are directly applicable to the wintertime PM problem in the SJV and we hope will be used by others working on the DISCOVER-AQ data set. Also, because methane is not directly an air quality concern, we leave it out of this paragraph. We have removed a couple of sentences in the hopes that they might be considered "excessive inessential details."

(Referee)The last paragraph presents an outline of the paper, but the preceding text has not setup these goals, nor does the outline mention the ozone production, methane emission, or evapotranspiration applications.

(Response)We have expanded the outline paragraph in an attempt to state the goals of our work more clearly, as per the reviewer's earlier suggestion.

(Referee)Most of Section 2.1 is irrelevant. The authors should relate the descriptive information directly back to their analysis and delete superfluous detail. (Response)We have condensed much of the information originally presented in Section 2.1 as it was also suggested by reviewer 2. However, we disagree that this discussion of the dynamic environment is irrelevant. We chose to include a clear survey of mountain-valley dynamics to set the stage for this unique mesoscale environment in which the experiments took place and because we do not find such a concise description anywhere in the extant literature. This dynamic complexity lies at the heart of why the region endures some of the poorest air quality in the nation. For others working on recalcitrant air quality issues in this area, or similar ones such as the Po Valley in Italy, we feel this information is essential for consideration.

(Referee)Sections 2.6 and 2.7 should be framed around what was done here, rather than as done currently, as a general discussion of the two methods using the author's dataset as an example. The last sentence of Section 2.7, "ultimately the approach using the budget of boundary layer inversion height, outlined in Section 2.6 was taken to calculate the entrainment rate," should be given to the reader up front. Additionally, the last paragraph in 2.7 is described almost narratively of how the analysis was done.

Please reorder such that results are presented to convey the logic of the analysis to the reader.

(Response)We have restructured/rewritten Section 2.7 to better coordinate the general discussion of the scalar budget equations with how they were used in these experiments.

(Referee)What are the results for Ox, as opposed to O3 and NO2 separately? Use of P(Ox) would be especially important in the wintertime and better suited for a winter/summer comparison. Secondly, has wintertime P(O3) been found to be NOx-limited also? That seems unlikely; please clarify.

(Response)Unfortunately, we did not have measurements of NO2 save for one single flight, and therefore were not able to perform a budget of odd oxygen.

- Yes, the results presented in Fig. 9 indicate that P(O3) is NOx-limited in the wintertime, but the inference is not strong given the limited spread in VOC:NOx ratio, and the uncertainties in using CH4 as a general VOC proxy. Nevertheless, we feel the result is worth presenting, especially since very little is known about winter O3 production because it is not often considered.

(Referee)Broadly, the outline of the paper is to compute the entrainment rate and then use this rate to explore three things: (a) ozone production rates, (b) methane emissions, and (c) water. Adding text or a dedicated section after discussion of the three studies, but prior to the Conclusion, that ties everything back together would do two valuable things. First, it would clarify the narrative and logic of the paper, and second, it would reinforce the significance of the work.

(Response)We have attempted to tie everything together more clearly throughout the revised manuscript and thus do not see the value in repeating this before doing so again in the conclusions.

(Referee)Specific comments:

Page 2, lines 3–4: Citation needed on, “this mixing tends to be a significant contributor to the ABL budget of the scalar.”

(Response)Stull [1990], Arellano et al. [2011], Lehning et al. [1998].

(Referee)Page 3, lines 17–18: Should this be 105 exceedances "per year"?

(Response)We have eliminated this statement as non-essential.

(Referee)Page 7, line 7: w(e) is not defined in the text (it is instead defined on page 8, line 23).

(Response)Defined in both places now.

(Referee)Page 10, lines 18–20: What is the evidence for: “For the purposes of estimating regional source strengths or regional in situ photochemistry, we suggest that the more pertinent mixing process is the dilution of the anthropogenically influenced ABL air mass by the more global ’baseline’ FT air.”

(Response)This is more of a conjecture, claiming that it is the ABL growth rate after its initial ‘encroachment’ through the morning’s residual layer that is key in understanding regional chemistry and surface emissions because the residual layer tends to be made up of mostly recycled air from the region. Of course, in principle, the budgets should still hold during the more rapid growth of the morning ABL, but they become more difficult to accurately measure due to the greater presence of transients and inhomogeneities. We do not feel this detail should be introduced into the manuscript because it is somewhat tangential as we did not perform the budget analysis in the morning hours, and it would not make sense to anyway because of the low O3 production at high solar zenith angles, which does not impact the afternoon O3 maximum very significantly.

(Referee)Page 11, lines 34–35: How is this shown in Fig. 7: “the importance of entrainment mixing on an ozone exceedance day.”

(Response)It is shown in the subsequent discussion where the jumps observed in Fig.

7 are used to estimate a time rate of change of O3 and NO2 concentrations due to entrainment dilution.

(Referee)Page 12, lines 35–36: It is difficult to see that methane is an appropriate proxy for total VOC. Even if dairies and gas production are the dominant source of VOCs, what matters more is that the drivers of methane emission match the drivers of the other VOC, which might not be true even if the sources are the same.

(Response)As discussed in Section 3.2.2 the majority of methane in both studies are believed to be associated with fossil fuel extraction and dairy operations. The studies of Gentner et al. [2014] and Pusede et al. [2014] indicate that methane is fairly well correlated with alcohols (which have strong dairy sources), higher alkanes (natural gas), and CO (other anthropogenic activities.) While we acknowledge that methane is a somewhat crude tracer of reactive VOC, we present the results because there is a suggestive relationship with our inferred ozone production rates that is consistent with past studies of the ozone production regime.

(Referee)Page 13, lines 3–5: Can an estimate of the uncertainty be given?

(Response)We have included an average uncertainty estimate from our experimental results to better frame the comparison, and have done so in all of the Tables as well. There is no estimate of uncertainty in P(O3) made by Pusede et al. (2014).

(Referee)Section 4: I recommend moving Section 4 to precede Sections 3.2.1–3.2.3.

(Response)We feel that a discussion of the errors in the measurements specifics is best delayed until the details of the experimental results are related, so we have kept Section 4 after Section 3, but we have expanded it considerably to make clear exactly how our errors have been treated in our results.

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