

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)The paper by Trousdell describes new aircraft measurements, which, combined with the mixed layer budget equation, attempts to constrain entrainment, advection, and the emission/production of ozone, methane and water. The dataset and analysis could be suitable for ACP, but as it stands the paper tries to address too many disparate issues: entrainment, the ozone budget, the methane budget, surface heat fluxes and the water cycle. In my opinion the paper needs to be significantly modified before publication. As a consequence the findings are often not discussed in depth

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and put into context of uncertainties.

(Response)Thank you for spending the time to go through our paper, your comments and critiques are greatly appreciated. I understand how you could see that we attempt to take on disparate issues, but our goal is precisely that: to bridge dynamics and chemistry. We feel that the atmospheric chemistry and boundary layer communities can benefit from each other, and the use of this simple mixed layer model demonstrates that. This journal attracts those interested in atmospheric dynamics as well as chemistry so we believe it is the perfect fit for our manuscript. To clarify, our intention when detailing these various topics like the ozone budget, the methane budget, entrainment, etc. is not to necessarily go into great depths on each but to show how a simple mixed layer budget equation sufficiently closed by in-situ flight data, including a detailed calculation of entrainment, can be used to uncover useful and novel estimates of emissions and photochemical rates. With this in mind, and in light of the complex mesoscale environment, we feel it is inadvisable to add all of the details from these various topics, yet it is important to present them together. The crux of this study is really the computation of dynamic quantities, like entrainment and linking them to the chemistry of the boundary layer.

On the other hand, we can see how our treatment of the uncertainties of these estimates could come across as lacking depth, so we have rewritten that entire section (4 Error Analysis) to clarify our estimates of the uncertainty of this approach. A major uncertainty, that needs more evaluation, is the fusion of in-situ observations with large scale reanalysis data. What are the uncertainties of this approach? E.g. when extracting mean vertical wind speed or surface fluxes from NARR, and plugging these data into eqs. (4),(6), etc., to extract small residuals of the observed quantities. To be clear, we do not put surface fluxes nor mean vertical velocities into equations 4 and 6. The only reanalysis data we incorporate is into equation 2, the inversion height budget equation. To answer the question directly, I would refer the reviewer to Table 1, wherein it appears that the very conservative uncertainty we assign to the mean vertical ve-

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locity of the NARR (0.5 cm/s or approximately 50%) leads to large uncertainties in the derived entrainment velocities: ~ 1.0 cm/s for each project average (1.5 and 3.0 cm/s averages). We feel this is a reasonable estimate of this uncertainty and do propagate it through the entrainment terms in the other budget equations (4 & 6) where that term is not always the leading one, however the large uncertainties in methane emissions are a direct result of these assumed uncertainties. Therefore, we disagree with the reviewer in that this is not a case of trying to tease out a small residual from large terms with large uncertainties.

With regards to the suitability of our estimated uncertainty in the NARR vertical velocities, we refer to a study by Albrecht et al. (2016). They also utilized reanalysis data in the form of omega, which was later transformed to vertical velocity and subsequently used in the exact same inversion height budget equation we use. They estimated the error of the average vertical velocity derived in this fashion to be ± 0.1 cm s⁻¹, a full factor of five times smaller than ours. In addition, they conclude that the majority of variation from this budget equation is reflected in the local time rate of change of inversion height when compared to the variations in the advection of inversion height and vertical velocity combined. We also found the budget equation of inversion height to be dominated by time rate of change on average, so feel that we are measuring the most important term in the z_i governing equation (2). We have added these details to our error analysis section to help clarify these points.

(Referee) Generally the paper lacks a consistent analysis of error propagation, which makes it hard to follow the uncertainty of the complex method of extracting tracer budgets. We have expanded our error analysis section (Section 4) to include a more detailed analysis. Some of the errors were calculated formally using a standard error which is a residual from the linear fit normalized by the number of data points, but other error terms are not subject to such statistical formalism. For instance, the error in the scalar jump, which is diagnosed by eye from vertical profiles is given what we deem a conservative estimate of its error. We also note that our estimated error for vertical

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velocity obtained from NARR is five times greater than that used by the Albrecht et al 2016 study for the reanalysis data they used (ECMWF). We try to be careful and we include errors with every term in the budgets. For cases when terms were not subject to a rigorous mathematical analysis of error we attempt to be conservative and overestimate the potential error. In Tables 1-3 we have now included the standard deviations of the mean values from each campaign to give a sense of the natural, background variation relative to the observational uncertainty estimates to help place these in context. Section 3.2.1: The ozone budget has to be corrected and time-shifted due to rapid photochemistry. Is this done arbitrarily to minimize residuals?

(Response) Our ozone budgets were not time shifted or corrected for rapid photochemistry. The only instance in the manuscript where we corrected the ozone levels was to make plots of horizontal gradients and advection, which were corrected by way of the secular linear time rate of change to a common time stamp. This reduces the spatial 'noise' of the aircraft measurements which are sweeping over the region throughout the day when the mean ozone is on the rise. Ozone production: methane is used as a VOC tracer to demonstrate that P(O₃) is NO_x-limited. Yet methane is not a very good tracer, because it has quite different sources compared to VOCs emitted from transport and combustion processes (e.g. aromatics). In addition biogenic VOCs are not considered at all by this approach. Methane is a fugitive emission and therefore does not represent the variation of VOC reactivity properly. To make a more convincing point the authors should use data from the parallel SEACRS mission or ground based observations in combination with a photochemical model to show what fraction of OH reactivity is due to methane (likely very small) and whether methane significantly co-varies with the local VOC reactivity. 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

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there is a suggestive relationship with our inferred ozone production rates that is consistent with past studies of the ozone production regime.

With respect to the SEAC4RS dataset we found only one boundary layer leg within the Central Valley of California during that mission. With that we have about one hour of data taken in the early evening containing 28 data points from the dataset of Don Blake showing a correlation of 0.6 or greater with CH₄ for; CO, DMS, HCFC-124, HFC-134a, HFC-152a, CH₃I, CH₂Cl₂, C₂HCl₃, C₂Cl₄, MeONO₂, EtONO₂, i-PrONO₂, n-PrONO₂, 2-BuONO₂, 3-Methyl-2-BuONO₂, 3-PeONO₂, 2-PeONO₂, Ethane, Ethene, Ethyne, Propane, Propene, n-Butane, 1-Butene, i-Butene, i-Pentane, n-Pentane, 1-Pentene, 2_3-Dimethylbutane, 2-Methylpentane, 3-Methylpentane, n-Heptane, Benzene, Ethylbenzene, and beta-Pinene. From this mix of hydrocarbons we maintain that CH₄ is a decent, although imperfect, tracer for other reactive hydrocarbons. Trying to use this limited data set in a photochemical model seems well beyond the scope of this paper, and the NO_x-limited nature of the ozone environment has been confirmed in other studies (Pusede et al. [2012], Brune et al., [2016]).

(Referee)In the following section (3.2.2) methane emissions are discussed, but given the un- certainty of the local methane budget (e.g. 100 +/-100Gg/yr), one wonders about the significance of the results. Again, without proper error propagation it makes it hard to follow the validity of the approach, especially uncertainties originating from the model- data fusion. The reader is left with the impression that the approach relies on luck and a fair wind.

(Response)It is true our methane emission errors are of a similar order of magnitude as the overall flight-to-flight spread. This is caused by the fact that we estimate our errors in entrainment to be nearly the same order of magnitude as the results. But this magnitude of uncertainty in entrainment is common for measurements of such a difficult, yet important, parameter (de Arellano et al. [2004] de Roode & Duynkerke, [1997]; Bretherton et al, [1995]; Wolfe et al., 2015). So, naturally emission estimates that are derived directly from this parameter are going to have similarly large uncertainties. But

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we believe that it is still a valid measurement, and when repeated over many flights, the mean measurement is indeed meaningful. Furthermore, this is a very important result to the methane community, which is faced with a paucity of such estimates. It also might be useful to note here that inverse modeling techniques used to derive a posteriori emission estimates likely have similarly large uncertainties, but these are rarely, if ever, explicitly treated in modeling papers (Cui et al. 2015).

(Referee)Section 3.2.3: Surface latent heat flux: In my opinion this part of the paper presents the most interesting aspects, as it shows a significant bias of surface fluxes obtained from re-analysis data. Why do the authors not present a more in-depth analysis of this finding?

(Response)The calculation of the water budget was an easy addition for us because our payload measures water vapor, and since the budget equation for water does not have any internal source terms under our flight conditions. The results are included to show the robustness and wide applicability of the budget method. We agree the findings are interesting, but we leave them as general warnings to the community about the latent heat calculated in NARR, and that this is certainly going to have an effect on ABL heights due to partitioning of latent vs. sensible heat fluxes. But to probe this result more deeply would require a lot more information about the land-surface and we feel would distract from the main objective of the manuscript.

(Referee)Section 4: Rather arbitrarily 5 lines of error analysis are presented here, but only address a very small part that would be necessary for the entire paper.

(Response)We agree with you that our error analysis was overly concise, and we hope that the expanded error analysis section will assuage many of your concerns. Generally, in my opinion the paper tries to address too many disparate issues and therefore lacks in depth analysis of the individual pieces. For a focus on ozone, the authors should definitely combine their results with a more comprehensive set of chemistry observations, which seem to be available. We believe that estimating net O₃ production

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is a significant feat, and we have done so with equal or better uncertainty than other reports of in the literature (Pusede et al, 2014; Brune et al., 2016). Furthermore, without a vast array of chemical species and meteorological data to constrain a model, we do not feel that all that much would be gained in such an exercise.

(Referee) For a focus on entrainment and PBL dynamics, a PBL model should be used in conjunction with the budget equation. The paper would also greatly benefit from a more thorough discussion of the associated uncertainties when closing the PBL budget. Perhaps a useful resource to better constrain the thermodynamical and dynamical properties of the PBL during the research flights, and address the propagation of errors and uncertainties, can be found here: <http://classmodel.github.io/>

(Response) We hope that in light of our responses here and above the reviewer will reconsider their conclusion that the absence of applying more complicated models to this data set is a sign of a superficial treatment of the subject. Our intention here is to present an empirical study of surface emissions, ozone photochemistry, and entrainment in the San Joaquin Valley, and the wide applicability of the airborne budget method we have applied. Perhaps our use of the term 'model' in the title is a bit misleading, because by 'model' we really mean a simple analytical tool that can be applied to airborne data. We do not wish to resort to any higher order models in this analysis, because such models necessarily require boundary conditions and initial conditions that were not constrained by observation – specifically, OH reactivity and/or speciated VOC data in the ozone analysis, and surface heat fluxes in the entrainment analysis. We do not believe that the particular model referred to above will help us better understand uncertainty, but will rather add more. The Dutch slab model is based on many inputs including, but not limited to, surface heat fluxes, drag coefficients, initial boundary layer height, free tropospheric stability, and of course subsidence.. Additionally, the model does not include advection so we would still need some way to account for the uncertainties of this term and the subsidence term, but would have no way to know the uncertainties in the surface heat or momentum fluxes. We feel the method presented

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here is more direct because it is not driven by all of these unknown parameters and more closely tracks the uncertainties in the governing equations themselves, as we hope is now more clearly presented in the new section 4.

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