Note\* all responses are in blue type.

Thank you for your time and critiques of our work; they are greatly appreciated. We feel it will be clearest to respond to each bullet point made by including it as a blue comment beneath each respective point.

Review of "Observing Entrainment Mixing, Photochemical Ozone Production, and Regional Methane Emissions by Aircraft Using a Simple Mixed-Layer Model," Trousdell et al., ACP (2016)

### Summary

This paper presents results from two small flight campaigns in California. Observed trace gas concentrations and profiles are used to derive entrainment velocities and examine the boundary-layer budgets of ozone, methane and water vapor. Results are used to evaluate photochemical ozone production, regional methane emissions and evapotranspiration.

The presented data is new, and the analysis of boundary layer budgets is a useful technique that is perhaps under-utilized in our field. The paper is generally well-written, although the embellished language is distracting at times and some sections provide an over-abundance of contextual details. Revisions are necessary before publication.

### **General Comments**

Section 2.1 provides a wealth of interesting but non-essential details on the topography and meteorology of the SJV. The first three paragraphs could probably be condensed down to one by removing such details –particularly those regarding specific orographic effects, which get confusing unless one constantly refers to a map or is familiar with the area. Indeed, the third paragraph (page 4, line 13) seems totally irrelevant given that the data presented is all daytime. The last paragraph in this section reads like a primer on mountain-valley flows and again seems only tangentially relevant to the results presented later.

We understand the referee's point here, and we have condensed much of the information originally presented. We chose to include a clear survey of mountain-valley dynamics to set the stage for this unique mesoscale environment in which we are working and because we do not find such a concise treatment in the extant literature. It is exactly this dynamically complex environment which has exacerbated the markedly poor air quality in the region. For others working on the

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.

The conclusions section is just a summary of main findings. It would be useful to add some discussion of needs for future work, in particular how some of the findings (such as dramatically incorrect emission inventories) could be further verified and eventually incorporated into better emission parameterizations. Is the ABL budget method a practical technique for grounding-truthing regional emissions on a model-relevant scale?

We have add two paragraphs to the conclusions in order to suggest further research that may build on the accomplishments of this study.

## Specific Comments

P2/L27: Wolfe et al. (2015) is another relevant and recent citation.

Thank you, yes, we have added that reference at this point. We had already included it in our paper elsewhere but had neglected it here.

Equations 4-7 and discussion thereof: Seems inconsistent. For example, the surface/entrainment terms are given different symbols for O3 and water. And the entrainment flux sign seems wrong – a higher concentration of stuff in the ABL should give rise to a positive entrainment flux (stuff leaving the ABL) and a negative contribution to dX/dt. It might be more straightforward to show a generic budget equation for any scalar, and then discuss specific treatments for water, ozone and methane.

You are correct, equation 4 had a sign inconsistency from our other equations, and there was substantial inconsistency in the symbols we had used. We have more systematically applied consistent symbols for the scalar budget equations and corrected the sign mistake. In response to a perceived misconception apparent in the reviewer's comment, we further added some discussion to clarify the role of entrainment in the ABL budget equations. A higher ABL concentration with everything else fixed would give rise to a dilution of the boundary layer concentrations, and yes this drives a negative dC/dt. However, this is not due to "stuff leaving the ABL" as the reviewer states. Entrainment in an actively turbulent ABL is an irreversible mixing process that incorporates free tropospheric (FT) air into the ABL, not vice versa. The positive scalar flux at the ABL top is the equivalent to a downward flux of concentration deficit (when the FT possesses a lower concentration), and we have explicitly stated that in the text now. We thank the reviewer for bringing this to our attention.

Page 8, Lines 16-22: suggest deleting.

Advice taken; we have removed these lines from the manuscript. We originally wanted to emphasize that in principal different scalars could be used in their respective budget equation to expose entrainment rates, i.e. water, ozone, or methane, and have made that point up front during the discussion of equations 4-7 as per reviewer's suggestions.

Eqn. 5: How are the BL concentrations determined for this calculation? Is it an average over the whole ABL, or just the upper portion? Same question for FT? Are uncertainties from this averaging (e.g. std of mean) propagated through to entrainment flux?

The scalar jump is determined from looking at vertical profiles and making the best eye judgment of the difference in concentrations between the top half of the ABL and the lowest ~100m of the FT. Often it is quite clear as can be seen in our example from fig. 7. We have included a brief description of how these values are determined and their estimated uncertainties, which are like all the terms propagated through to the final results. The error analysis section (4) has been greatly expanded so this should be much clearer now.

P12/L7: how is this map generated? Is it an interpolation of ground site data? Please expound. Also, another way of stating the opposing O3 and NO2 advective terms is that Ox=O3+NO2 is conserved.

The NOx and O3 advective maps are interpolated to a 2D grid from aircraft data taken in the ABL. All data is corrected for the calculated mean regional time rate of change back to a common time stamp of 13:30. This has been more clearly explicated in the text.

As for the odd oxygen interpretation, we do not agree. The gradient of ozone is an order of magnitude greater than that of  $NO_2$ . This is not simply a titration situation, but is intimately linked to rapid ozone production. We feel that the discussion of odd oxygen in this study would not serve to illuminate because it introduces a further unknown variable of the NOx emission rates. Also, we only had the  $NO_2$  measurement on one single flight.

Section 3.2.3: These findings seem to suggest that NARR has serious flaws and should be adjusted, at least coarsely, to more accurately represent agricultural practices in some broad sense. A naïve question: would such issues impact the subsidence velocity derived from NARR?

We do not believe that large scale vertical motion would be all that susceptible to partitioning of surface heat fluxes among latent to sensible, but it certainly affects the convective activity and entrainment and boundary layer depths in the model. Subsidence is generally believed to be controlled by synoptic flow conditions. Although we do suspect that subsidence can modified a good bit due to mesoscale orography. A better representation of agricultural practices would lead to a better estimate of the latent heat flux, which affects the partitioning in the surface buoyancy flux, and for a constant net radiation forcing this would lead to lower ABL heights for greater latent heat fluxes. This is why the NARR ABL depths are so much higher than measured, for instance.

# Table 3: The third column is technically not a flux, but a flux divergence. Also, please give CH4 production in ppmv/h for easy comparison with other terms.

The third column is the entrainment flux contribution to the flux divergence. We report it that way to have it in comparable units to the other terms. But reporting the surface emission similarly would not make sense to us, as the units most people are familiar with are something like the chosen ones of Gigagrams per year. The CH4 production (surface emission) term is simply the numerical sum of the other columns, so we thought it would be redundant to see it in the same units.

# Figure 9: is there any physical rationale behind a power-law fit?

The short answer is no. We know that the ozone chemistry is non-linear, and the simplest non-linear relationship is a power law.

## **Technical Comments**

Fig. 2: Please label flight regions 1 and 2 as referenced in section 2.1.

We have changed the legend of Figure 2 to indicate the region numbers 1 and 2.

RASS is defined twice.

Got it.

P6/L32: delete ", which"

Deleted

P6/L35: "as per the Fundamental Theorem of Calculus" is a gratuitously pretentious statement.

We did not consider that such a foundational mathematical principle could be considered pretentious, but have eliminated the wording to protect the common reader.

Equations 1-3: subsidence is referred to as both  $W(z_i)$  and W. Pick one.

Okay, thanks we will stick with just, W, with the implicit understanding that it can be a strong function of height.

P9, L13: delete "the 5 hour period of late morning to early afternoon from" P10/L17: delete "a remove of"

Done.