

## Response to Reviewer

# “Direct observations of NO<sub>x</sub> emissions over the San Joaquin Valley using airborne flux measurements during RECAP-CA 2021 field campaign”

Qindan Zhu<sup>1,\*</sup>, Bryan Place<sup>2,\*\*</sup>, Eva Y. Pfannerstill<sup>3</sup>, Sha Tong<sup>4,5</sup>, Huanxin Zhang<sup>4</sup>, Jun Wang<sup>4</sup>, Clara M. Nussbaumer<sup>1,6</sup>, Paul Wooldridge<sup>2</sup>, Benjamin C. Schulze<sup>7</sup>, Caleb Arata<sup>8</sup>, Anthony Bucholtz<sup>9</sup>, John H. Seinfeld<sup>7</sup>, Allen H. Goldstein<sup>3,8</sup>, and Ronald C. Cohen<sup>1,2</sup>

<sup>1</sup>Department of Earth and Planetary Sciences, University of California, Berkeley, Berkeley, CA 94720, United States

\*Now at Department of Earth, Atmospheric and Planetary Sciences, Massachusetts Institute of Technology, Cambridge, MA, United States of America

<sup>2</sup>Department of Chemistry, University of California, Berkeley, Berkeley, CA 94720, United States

\*\*Now at Office of Research and Development, U.S. Environmental Protection Agency, Research Triangle Park, North Carolina 27711, United States

<sup>3</sup>Department of Environmental Science, Policy, and Management, University of California, Berkeley, Berkeley, CA 94720, United States

<sup>4</sup>Department of Chemical and Biochemical Engineering, Center for Global and Regional Environmental Research, and Iowa Technology Institute, University of Iowa, Iowa City, Iowa 52242, United States

<sup>5</sup>Collaborative Innovation Center on Forecast and Evaluation of Meteorological Disasters (CIC-FEMD), Key Laboratory for Aerosol-Cloud-Precipitation of China Meteorological Administration, Nanjing University of Information Science Technology, Nanjing 210044, People's Republic of China

<sup>6</sup>Department of Atmospheric Chemistry, Max Planck Institute for Chemistry, Mainz 55128, Germany

<sup>7</sup>Department of Environmental Science and Engineering, California Institute of Technology, Pasadena, CA 91125, United States

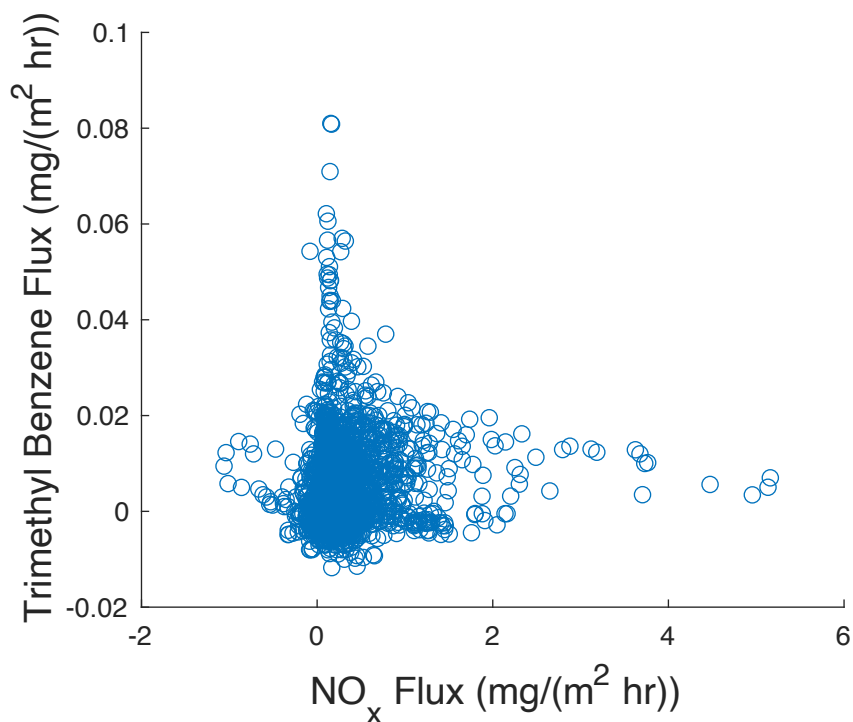
<sup>8</sup>Department of Civil and Environmental Engineering, University of California, Berkeley, Berkeley, CA 94720, United States

<sup>9</sup>Department of Meteorology, Naval Postgraduate School, Monterey, CA 93943, United States

**Correspondence:** Qindan Zhu (qindan\_zhu@berkeley.edu) and Ronald C. Cohen (rccohen@berkeley.edu)

We thank the editor for very careful reading of the manuscript. Below we respond to comments. The reviewer's comments will be shown in red, our response in blue, and changes made to the paper are shown in black block quotes. Unless otherwise indicated, page and line numbers correspond to the original paper. Sections, figures, tables, or equations referenced as “R*n*” are numbered within this response; Figures, tables, and equations numbered normally refer to the numbers in the original  
5 discussion paper.

The authors have adequately addressed the major concerns of the two primary referees. The have also acknowledged the concerns of the unsolicited comments and addressed them to the extent possible with the available data. This paper is sufficient for publication after consideration of the following minor revisions. Below, line numbers refer to the author response document, not the manuscript.



**Figure R1.** The scatterplot of  $\text{NO}_x$  flux and trimethylbenzene fluxes with footprints exclusively covering soil land cover type.

10 Thanks so much for the recognition of our response.

L40: Is there any correlation of soil  $\text{NO}_x$  fluxes with TMB flux, especially at the high  $\text{NO}_x$  tail? If not, worth stating in paper.

We do not find any correlation between soil  $\text{NO}_x$  fluxes with TMB flux (Shown in Figure R1). We add it to Sect 3.4:  
 “No correlation between trimethylbenzene flux and  $\text{NO}_x$  flux is found over croplands. Among all observations ...”

15 L52: Won't filtering out fluxes below LOD introduce a high bias in any resulting averages? If so, maybe better to keep them in.

We do not filter all fluxes below LOD. Instead, we scan through the segments and only filter out fluxes in the segments which have high LODs and hence all fluxes are below the LOD. To address this comment, we calculate the average fluxes with and without this filtering. It turns out that the filtering does not affect the resulting average.

20 L81 and Fig. R2: Are these uncertainties in 500m-average fluxes? Please clarify.

Yes. We clarify it in the last paragraph of Sect 3.6:

“We propagate the total uncertainty from each component using Eqn. 16 and the distribution of total uncertainty of **500m average NO<sub>x</sub> flux** is shown in Figure 3 (b).”

L161: Are these details regarding BL depth described in the text or SI? Fig. R3 would be good to include in the SI.

25 Yes, we have included the description of BL depth in Sect 3.1 and add Fig. R3 in the supplementary (Fig. S2):

“While most of the measurements are within the planetary boundary layer (PBL), the airplane arose above the boundary layer occasionally and these observations above PBL are removed in later analysis. **The PBL heights are determined using the sharp gradient in the dew point, water concentration, toluene concentration and temperature at the soundings conducted during the voyage, and we interpolate the PBL heights to the full duration of the flight. The PBL heights agree well against the hourly PBL heights from the High-Resolution Rapid Refresh (HRRR) product (Figure. S2).**”

L165: It is still not clear to me why you are normalizing by the standard deviation (which is a scalar for each leg). You’d then just have to multiply it back in to get the right units in the flux. This operation isn’t detrending, it is normalization and is not, to my knowledge, standard practice for wavelet fluxes. So, why do it? If you have a good reason, please justify in the text.

Sorry for the confusion. After reviewing the wavelet decomposition algorithm, we agree with the reviewer that this operation is normalization, not detrending. Moreover, normalization is not the requisite step for wavelet decomposition. The normalization is only used to produce Figure 1(a). We changed the word “detrend” to “normalize” in the context:

“For two simultaneous time series of NO<sub>x</sub> ( $W_c(a, b)$ ) and vertical wind speed ( $W_w(a, b)$ ), we ~~first detrend them by subtracting out the average followed by dividing the standard deviation of a scalar time series. Then we~~ obtain the wavelet cross-spectrum following Eqn. 1. The Morlet wavelet-specific reconstruction factor  $C_\delta$  is 0.776. We then sum up over the full frequency scales to yield a time series of flux (Eqn. 2).

$$E_{c,w}(j) = \frac{\delta t}{C_\delta} \frac{1}{N} \sum_{n=0}^{N-1} [W_c(a, b) \cdot W_w^*(a, b)] \quad (1)$$

$$F(t) = \overline{c'w'} = \frac{\delta t}{C_\delta} \frac{\delta j}{N} \sum_{n=0}^{N-1} \sum_{j=0}^J \frac{[W_c(a, b) \cdot W_w^*(a, b)]}{a(j)} \quad (2)$$

Figure 1 exhibits an example of CWT flux calculation. Figure 1 (a) shows the ~~detrended-normalized~~ NO<sub>x</sub> and vertical wind speed in a straight segment of  $\sim 50$  km. The ~~detrending-normalization~~ is realized by subtracting out the average followed by dividing the standard deviation of a scalar time series...”

L464: “Reviewer 3” opens with hostility. The authors handled this rather gracefully.

Thanks. We endeavor to address the reviewer’s concerns and improve our manuscript.

L507: This discrepancy between what “controls” the slope of the divergence is, I think, an open question (or at least a confusing one). Paradoxically, the reviewer and the authors may both be correct. An analysis of the budget equation at any point in the atmosphere shows that the flux is related to chemistry, advection, and storage:  $dC(z)/dt = \text{chemistry} + \text{advection} + dF(z)/dz$   $dF(z)/dz = \text{chemistry} + \text{advection} - dC(z)/dt$  integrating from  $z = 0$  to  $z = z_m$  (measurement height) gives  $F(z_m) = (\text{integral of chemical production/loss}) + (\text{integral of advection}) + (\text{storage}) + F(z = 0)$  It is also true that the flux at the top of the PBL is “fixed” by entrainment, which is defined as the product of the concentration gradient and the entrainment velocity ( $w_e$ ).  $F(z = z_i) = (C(z < z_i) - C(z > z_i)) * w_e$  If we is purely determined by micrometeorology, the system would seem to be over-determined. Perhaps the solution is that  $C(z < z_i)$  is inherently a function of the terms in the budget equation – I honestly am not sure of how to resolve this. Regardless, this paper is not the appropriate place to resolve it, and my opinion is that the authors have made a strong effort to constrain divergence as well as the data allows while also being honest about the uncertainties.

Thanks so much for this comment. We actually agree with the reviewer that the treatment of vertical divergence is quite confusing in current studies related to flux analysis. That is the main reason for us to conservatively account for the uncertainty from the vertical divergence.

L518: Are NO<sub>x</sub> mixing ratios in the free troposphere higher or lower than in the boundary layer? It should be possible to put some constraint on the entrainment flux in this way (at least the sign).

NO<sub>x</sub> mixing ratios are lower in the free troposphere than in the boundary layer, so the entrainment flux is negative. It is consistent with our calculation ( $-0.09 \text{ mg N m}^{-2} \text{ h}^{-1}$ ) at the boundary layer top. However, we have to emphasize that the uncertainty in the calculation can be large.

L537: It is also worth mentioning that Sha (2021) is entirely model-based using a parameterization derived from Oikawa. Not sure it is fair to present it as an independent estimate on par with actual measurements.

Since Guo et al. (2020) and Almaraz et al. (2018) are also model-based, we think it is fair to present Sha et al. (2021) on par with Guo et al. (2020) and Almaraz et al. (2018).

L540: It is worth stating in the text why you cannot compare directly to Trousdell and citing the paper. This will help address Reviewer 3’s concern about the lack of citation of considerable prior work.

It is a good point. In Sect 5.3 describing the limitation of our study, we add:

“Second, as our measurements only cover limited cropland areas in SJV over a short time period and it is around the time of fertilizer use, we cannot scale the estimated soil NO<sub>x</sub> emission to the whole year or to the total cropland areas in California. **Therefore, we cannot directly compare our estimate of soil NO<sub>x</sub> emission against other studies reporting soil NO<sub>x</sub> emissions on an annual basis or on a larger spatial scale.**”

80 L585: So, is there a way to estimate friction velocity from measurements in the mixed layer? If not, do you have any way to validate the HRRR friction velocity, or does it not strongly impact your results? This is something the airborne flux community needs to consider carefully with respect to accurate footprint estimates. And if it's something that needs more work, that'd be worth mentioning in conclusions.

85 Friction velocity accounts for shear stress in the turbulent boundary layer and it can be parameterized from measurements. For instance, Foken and Napo (2008) and Amiro (1990) presented the calculation of friction velocity using the instantaneous wind vectors ( $u'$ ,  $v'$ ,  $w'$ ). Hannun et al. (2020) calculated the friction velocity using the mean flux parameterization. Camuffo (2014) showed four different parameterizations of friction velocity under different turbulence conditions. We use the friction velocity from HRRR since the roughness layer, another parameter used in the footprint calculation, is also from HRRR. We also test out the effect of friction velocity on footprint by varying the friction velocity by 50%, and it does not affect the result.

L666: "Statistical analysis is applied and the increase among three bins is statistically significant." Please quantify this in your response and in the revised text.

90 We quantify the temperature dependence of soil  $\text{NO}_x$  emission using the slope from a linear fit between soil temperature and estimated soil  $\text{NO}_x$  emissions. We add it in Sect 5.2

95 "A range of soil temperature between 295K to 304K is observed. **We apply the linear regression fit between soil temperature and estimated soil  $\text{NO}_x$  emissions and show a positive temperature dependence of soil  $\text{NO}_x$  emissions with the slope of 0.02 ( $\pm 0.008$ )  $\text{mg N m}^{-2} \text{h}^{-1} \text{K}^{-1}$ .** We then bin observed soil  $\text{NO}_x$  emissions to three soil temperature categories..."

## References

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