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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Airborne flux measurements of biogenic volatile organic compounds over California" *by* P. K. Misztal et al.

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Response to Don Lenschow (Referee 1)

We thank Don Lenschow for providing inspiring and helpful comments which led to major improvements in the revised version of the manuscript. We reply to each comment below in bold text.

Overall Evaluation:

1) While I have rated the manuscript "accept subject to minor revisions," I do have some important concerns that I think should be addressed before the manuscript is approved for publication. I do think that this paper merits eventual publication, as the

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Interactive Discussion

Discussion Paper

techniques and analyses presented here are a significant step forward in the science and demonstrate an approach to quantifying emissions of trace reactive species over horizontally heterogeneous surfaces.

We thank you for your positive evaluation of our research that it presents a significant step forward in science. We also realize that the approach will be refined further as the wavelet analysis becomes more routinely applied to airborne flux measurements. We address all the reviewer's concerns below.

Major Comments:

2) Using a single representative vertical flux divergence to extrapolate all fluxes at flight level down to the surface to estimate surface emission has several assumptions that should be discussed in more detail. The vertical flux divergence is dependent on the rate of isoprene oxidation (which depends on OH concentration and other oxidants), the time rate of change of isoprene concentration, and differential (with height) horizontal advection of isoprene. Since the flights were conducted with a variety of different conditions and locations, one would expect different isoprene lifetimes and horizontal advection scenarios. These effects need to be taken into account or at least evaluated for their impact to be assessed in order to estimate the accuracy of the extrapolation to the surface. Furthermore, I don't see any estimate in 2.7 of the random error contributed by the flight segment length. Thus, the error analysis in 2.7 seems to me to be too optimistic, at least without further justification. This has relevance later on in section 3.2, where it is not clear how much of the variations and differences between e.g. tower and aircraft are due to random errors and how much to real variability.

We have added new text to 2.7 to clarify the uncertainties due to flux extrapolation to the surface: "It is important to note, that the vertical flux divergence is dependent on the rate of isoprene oxidation (which depends mostly on OH concentration during daytime), the time rate of change of isoprene concentration (relevant for conserved species), and differential horizontal advection of iso-

ACPD
14, C2671–C2680, 2014

prene with height (small). Based on directly measured flux divergence in the racetrack gradient flights (Karl et al., 2013) we showed clear linear dependence of the flux divergence with a theoretical concentration gradient (e.g. 1.4 $\times 10^{-4}$ ppbv m^{-1} over a homogenous oak terrain and an OH concentration of 6.6 $\times 10^{6}$ molec/cm^3). Since the flux divergence for isoprene was shown to be primarily controlled by OH concentrations (of which we have a range of estimates), we make an informed assumption here that the divergence coefficients we used to scale the fluxes to the surface are accurate within a factor of two for the entire campaign. Thus a change in the flux divergence coefficients by a factor of two could result in only a ~2% difference to the scaled surface flux for a typical z/zi ratio of 0.3 which is minor relative to other error sources as discussed in Sect. 2.7. As the correction of the fluxes for flux divergence was typically less than 20%, the contribution from less accurate divergence coefficients is assumed to be relatively minor (up to ~2%) for isoprene but could still be more important for other gases (e.g. CO_2), for which more detailed characterization of flux divergence might be needed in future measurements." We minimized the effect of concentration change by performing flights during times when the PBL was well developed and changes in the steady state assumption for chemistry was small, which was evaluated during our profiling flights (Karl et al., 2013). We investigated the effect of horizontal advection during our flux profiles and estimated horizontal advection by taking the isoprene concentration gradient and mean horizontal wind along the horizontal flux leg. We have now also provided an expanded budget of uncertainties in revised Sect. 2.7, following one of the comments by the reviewer #2.

3) In several places, it is claimed that the CWT method can be used to obtain 1 to 2 km spatial resolution in fluxes. This is misleading. True, you can get fluxes to this resolution, but with very large random error. This needs to be considered further and the random error as a function of sample length needs to be discussed and quantified. Among other references, you might take a look at "Errors in Airborne Flux Measure-

ACPD
14, C2671–C2680, 2014

ments," by Mann and Lenschow, 1994, J. Geophys. Res., 99, 14519-14526.

We agree that selecting a short integration scale such as 2 km would result in a very large error (e.g. Mann and Lenschow, 1994), and clarify that we have only evaluated fluxes over longer stretches (> > 2 km). We apologize that our original text may have been misleading regarding this length scale. We now state more clearly that for the wavelet fluxes we actually integrate long segments (e.g. 100 km) and based on wavelet decomposition, we reconstruct the time domain for the wavelet co-spectra to yield time series of discrete coherent structures which are subsequently aggregated to 2-km surface fluxes. Because the variability is much larger at the short time-scales the random error of each 2-km point must be larger (40-50%) than that of the average flux for the whole leg. This error decreases significantly with averaging over 5 km and more and thus it makes sense to use averaged data rather than individual points with models. The 2-km representations provide more flexibility for averaging, for example, individual points can be useful for a regression of isoprene flux versus LAI for all of the 2km data. This would provide a large number of data points to ensure good statistics and gives us information that we would not have if we just had the average over a larger transect. Nevertheless, we think that the wavelet decomposition is another step forward to reduce greatly the systematic errors for high spatial resolution fluxes and the way forward in the future to minimize random errors would be to repeat each leg once or twice.

Other Comments:

4) abstract, l. 6: spell out PTR-MS

Done.

5) abstract, l. 13: the statement, " Vertical flux divergence of isoprene is expected due to its relatively short lifetime..." is misleading. Vertical flux divergence is expected for almost all atmospheric species. In conserved species it is a reflection of time changes

ACPD
14, C2671–C2680, 2014

in the mean concentration. More accurately, you should say that there is a major contribution to vertical flux divergence of isoprene due to its relatively short atmospheric lifetime.

We have corrected the sentence as follows: "Vertical flux divergence is expected for all atmospheric species, but a major contribution to vertical flux divergence for isoprene is due to its relatively short atmospheric lifetime.

6) abstract, l. 15: I can't make sense of a vertical flux divergence expressed as a percent. The units of species vertical flux divergence should be concentration/time. Do you mean that the this is the percent difference between flight-level flux and surface flux?

Yes, we meant the maximal percent difference between flight-level flux and surface flux due to the flux-divergence correction. This has been clarified.

7) p.7968, l. 15: spell out BEIGIS. Also earlier MEGAN and BEIS

Done.

8) p. 7969, l. 9: "Stacked," instead of "Vertical."

Done.

9) p. 7975, l. 8: ...capable of eddy flux measurements.

Corrected.

10) p. 7975, l. 10: What is meant by area "ratio of about 2?" Does this mean that the diameter changes from 2.047 in. to about 2.89 in.? Same comment applies later to the area ratio of about 5. Also, I think that you should use metric units for these dimensions.

We have removed these specifics of the inlet system which were thoroughly described in Karl et al., 2013.

ACPD
14, C2671–C2680, 2014

11) p. 7975, l. 16: "unaffected" is a bit too strong. Perhaps something like "minimized" would be more appropriate.

"Unaffected" has been replaced with "minimized".

12) p. 7976, l. 7: "is a fast sensor which" is redundant.

This has been deleted.

13) p. 7976, l. 13: I'm not clear what 10 Hz separated by a relatively longer gap of 2 Hz means in this context. Can you elaborate more on what this means? Do you really mean 0.1 s dwell time and 2 samples/s?

This is exactly what we meant. This has been clarified as "...10 Hz disjunct sampling corresponding to 0.1 s dwell time and approximately 2 samples/s".

14) p. 7977, l. 8: "sawtooth" rather than "tooth"

Done.

15) p. 7977, l. 14: So, the sensitivity is 10 pptv/17 s, where 17 s is the averaging time?

Yes, we meant to say that the detection limit is dependent on the amount of averaging and is dependent on the sensitivity which is independent of the amount of averaging. We rephrase for clarity: "These high sensitivities ensured low detection limits (e.g. <10 ppt for isoprene at 1-km averaging (~17 s)).

16) p. 7977, l. 17: I'm a bit confused by the comparison between absolute sensitivity and normalized sensitivity. Do they really have the same units?

This is mostly directed to the PTR-MS community. The idea is that the normalized sensitivities (which account for primary ions and water clusters in the drift tube) can be used to compare with other instruments independently of their settings and primary ion count rates.

17) p. 7978, l. 28: ...measurements of concentration profiles in the mixed layer over-

ACPD
14, C2671–C2680, 2014

lying the surface layer of the daytime convective boundary layer...I suggest using CBL instead of MBL, as "mixed boundary layer" is not accepted terminology.

We have changed to CBL to comply with the classical terminology.

18) p. 7979, l. 1: This top-down bottom-up method applies only to a conserved species not VOCs in general. You should say "a conserved species" not a VOC. p. 7979, l. 25: ...on all eight research flights, and MVK+...

We agree and have incorporated the suggested change.

19) p. 7980, l. 3: 150-300 m is very deep for an assumed surface layer. What is this based on? Certainly not on 10% of the PBL depth, nor on applicability of surface-layer parameterizations.

The reviewer is right that the surface layer (SL) was actually less deep. Based on the 10% of the PBL depth and given the PBL depth we measured was typically between 1000 and 2000 m, the typical SL during CABERNET was extending typically up to 100-200 m rather than 150-300 m as we stated before. We apologize for this inaccuracy and have corrected the typical range to reflect the shallower SL.

20) p. 7980, l. 13: ...should be maximized. (Actually more accurately, if you sample significantly more than 1/integral scale, increasing the sampling rate won't gain you anything.)

We completely agree and modified the text accordingly.

30) p. 7980, l. 18: It's not clear to me what you mean by "total cycle length." Do you mean the sample rate for each species is 1.25 to 2 samples/s?

We meant the duty cycle length which is the sum of the dwell times of all measured species in the disjunct mode, but we now also describe how this relates to the sampling rate.

ACPD

14, C2671–C2680, 2014

31) p. 7980, l. 25-26: Do you really mean non-stationarities or do you mean horizontal heterogeneity? In l. 24 "affected by diurnal effects" is not usually a problem for aircraft measurements of a single flight segment.

We actually meant both or in other words non-stationarities related to heterogeneities. A single flight segment could take a few hours for a slow flying aircraft, but in our case this should not have been a problem for receiving contributions from diurnal effects.

32) p.7982, l. 9: ...an integrated straight stretch...What is meant by integrated? What is integrated? Do you mean continuous or contiguous? Why does it have to be straight?

We have replaced "integrated" with a different word to avoid confusion. The stretches were the straight, even and uninterrupted segments we selected for flux calculation. We think that it could be less accurate to calculate the flux on the curved paths when the aircraft is turning because of more complex behavior of horizontal wind and potentially larger advection errors.

33) p. 7982, l. 16: See earlier comment about random error in flux measurements as function of sample length.

As stated earlier, 2 km was not the flux integration length, but the resulting flux representation from the wavelet decomposition of the long stretch. We add a note to the text "meaningful spatial representation at 1-2 km resolution (note the integration length was ~2 orders of magnitude larger)".

34) p. 7982, l. 29: ...controlled by its relatively. . . . . ."racetracks" at multiple levels...(profiles is redundant)

Done.

35) p. 7983, l. 1: again, flux divergence has units of concentration/time, not percent.

The sentence was rephrased to inform the reader of the percent difference of flux

ACPD
14, C2671–C2680, 2014

at aircraft-level vs flux at the surface. "...It was found to be linear, so the scaled surface flux was in the range of 5 – 30% larger than measured at the given z/z_i ratio

36) p. 7983, l. 3: A storage term of 2-5% of what? The chemical reaction term? This seems very small to me in view of the diurnal variability of the isoprene concentration in the PBL. Was the buoyancy flux also extrapolated down to the surface similar to the isoprene flux when used to estimate w*?

The range of determined storage term calculations is based on the vertical profiling flights for the duration of one vertical profile measurement and not the entire diurnal cycle. Within this time period the change of isoprene concentration relative to the flux divergence term due to chemistry was small. The isoprene concentration was relatively constant due to the offsetting influences of increasing boundary layer depth and increasing emission source. The buoyancy flux was not extrapolated down to the surface.

37) p. 7983, Eq. (4): Where did you get this equation come from? It isn't in Horst and Weil (1992), who considered only the surface layer not the mixed layer. It seems that the authors have the wrong citation; this equation is presented in Weil and Horst, 1992: Footprint estimates for atmospheric flux measurements in the convective boundary layer. A chapter in Precipitation Scavenging and Atmospheric Surface Exchange, Vol 2, S. E. Schwartz and W. G. N. Slinn, Coords, pp. 717-728, Hemisphere Publishing Corporation, Washington, 1172 pp.

Thank you for spotting this oversight. We have now corrected the citation to Weil and Horst (1992).

38) p. 7983, l. 24: "superposition?" or "summation"?

now is "summation".

39) p.7984, l.12: do you mean "minimal roll angle"? Constant roll angle could imply a

ACPD
14, C2671–C2680, 2014

constant rate turn.

Yes. We changed the text accordingly.

40) p. 7984, l. 22: Standard usage for cross-spectrum is the real and imaginary parts of the Fourier transform, and co-spectrum for the real part, which you are using here.

We have changed cross-spectrum to co-spectrum.

41) p. 7986, l. 12: By "measurement footprint" it seems that you are no longer talking about the flux footprint that you were discussing earlier, but the concentration footprint, since the flux footprint does not extend hundreds of km upwind. This needs to be pointed out.

We point out here that we refer to the concentration footprint.

42) p. 7990, l. 25: ...for a setting where...

Done.

43) Table 1: You might also include w* or the surface virtual temperature flux in the table, so the reader can e.g. estimate the convective turnover time. Your estimate of 10 minutes (p. 7979, l. 9) means a large surface virtual temperature (i.e. buoyancy) flux.

That is a nice suggestion, and we have now included the summary statistics for convective velocity scale (w^*) to the Table 1. In order to keep the table within the size limits we decided to remove the summary statistics for relative humidity but we state in the text that the environment was becoming dryer as the temperature increased.

Once again we thank Don Lenschow for his time and providing all these extremely useful comments.

Pawel Misztal

ACPD
14, C2671–C2680, 2014