Review of the manuscript acp-2014-84

Title: “Airborne flux measurements of Biogenic Volatile Organic Compounds over California”

by P. K. Misztal et al.

General Comments

The goal of the manuscript is “to measure the distribution of isoprene flux across the oak woodland areas of California in order to test and improve the landscape-scale emission models that are used for regional air quality assessments”. The main question addressed by the research is whether measurements of biogenic volatile organic compounds (BVOCs) confirm the spatial patterning of model results. For this purpose, data from ground-based and airborne platforms are used, as well as multitude of data processing approaches. The authors conclude that horizontally varying source distributions of isoprene were successfully mapped out across dominant ecosystems in California.

The central question is interesting and important as BVOCs contribute to regional air quality through ozone production and acting as radical sinks in the source regions. The authors approximate that 50% of the total global BVOC emission is constituted of isoprene. A wide array of methods is used, including initial isoprene emission measurements using airborne eddy-covariance. Most (but not all) of the outlined methods are taken into consideration in the results and conclusions. The results are mainly of technical nature and support the conclusions. However, results and conclusions miss to quantitatively address the actual research question of testing and improving landscape-scale emission models. As such, in its current form the manuscript constitutes a collection of methods and must be considered as an incremental advance to the field.

The paper partially fits the scope of ACP in that it presents a combination of field measurements, remote sensing and modelling of biosphere-atmosphere interactions. However, in its current form the manuscript is primarily of technical and regional interest, and misses to unravel general implications for atmospheric science in a rigorous, quantitative manner. In general, the flow of the paper is logical, references are adequate and from my perspective no copy-editing is required. However, the authors tend to hypotactic sentence structures (e.g., p. 7969 l. 1) which could be broken apart to make it easier for the reader to follow. With 28 (discussion) text pages the length of the manuscript is reasonable. However, the allocation is not well balanced with 17 pages on methodology but only 7 pages on results, discussion and conclusions, half of which actually attributed to concentration (and not flux) results.

Consequently, I recommend major revisions, and see two principal ways for successful publication. (i) Publication in ACP: Addressing the stated goal through removing technical details (description of individual flight days and leg separation, FFT vs. CWT comparison, mixed boundary layer technique, length scales vs. time scales…), adding a quantitative model-observation inter-comparison and expanding on general implications (results, discussion and conclusions). (ii) Publication in more regional (BGC) or technical (AMT) oriented journal: Reformulating the stated goal to focus on regional implications (BGC) or technical questions of airborne isoprene concentration and flux measurements (AMT).

Specific Comments

p. 7966 l. 7: It does not appear that 10,000 km of flights were performed in the 8 measurement days presented in the manuscript.

p. 7966 l. 12, p. 7985 l. 15: Neither FFT nor CWT eddy-covariance (EC) approaches are "independent" of non-stationarities. While CWT does not require a stationary time series, non-stationarity principally violates reducing the Navier-Stokes equation to the 1-D problem posed by EC. Hence CWT is not necessarily more "accurate" as long as not considering all divergence terms. Can the authors explain how CWT is making their results more valid?

p. 7966 l. 13, p. 7983 l. 1, p. 7983 l. 2: Extrapolating flux measurements at 400 m above ground to the surface heavily relies on the accurate knowledge of the vertical flux divergence. In the present case, the vertical flux divergence is determined from measurements at different heights, i.e. with different source areas and assuming a linear and monotonous function with height. Are the extrapolated fluxes still significant when, in addition to the residual error in the regressions, contributions from different surfaces are considered? Maybe an inverse method like Bange et al. (2006) might be superior? Also, inference of vertical flux divergence from profile soundings is not a "direct" measurement as claimed on p. 7982 l. 29. Lastly, it is not clear at which flight levels and horizontal extent the stacked patterns were performed, which type of regression was used (considering error in variables?), and whether the regression results are actually significant.

p. 7966 l. 23: The authors relate concentrations to source regions. It must be noted that at 400 m a.g.l. flight altitude, the concentration source areas can extend several ten to hundreds of kilometres upwind (e.g., Griffis et al., 2007). Was such a source area analysis performed to substantiate the conclusions?

p. 7967 l. 5: Why did the authors chose a spatial resolution of 2 km? Principally, the flight altitude should be chosen so that the resulting blending length and flux footprint extend matches the spatial scale of surface patchiness (e.g., Mahrt, 2000; Mason, 1988; Raupach and Finnigan, 1995; Wood and Mason, 1991). Now, a matching wavelet integration interval can be chosen, which optimizes the trade-off among high spatial discretization and small random sampling errors. Also, it has been shown that individual flux estimates require aggregation to be statistically significant (Sühring and Raasch, 2013). What is the strategy of the authors to attain statistical significance?

p. 7967 l. 20: …resolution and coverage… Principally, one wants to determine a functional operator that allows scaling up and down between different observations in different reference frames. Here, it is important to note that atmospheric observations are a convolution of biogenic emission (e.g., from leaf-level measurements) but also atmospheric transport. Hence a functional operator has to explicitly consider both of these processes.

p. 7969 l. 8–18: Repetition that can be omitted. Instead, it would be helpful for the reader to provide an overview of what's to come in the next sections.

p. 7969 l. 19–23: Belongs to goal on p. 7968 l. 22–25.

p. 7970 l. 8: Surface fluxes cannot be measured at 400 m a.g.l.

p. 7970 l. 12: Tertiary levels of what?

p. 7970 l. 20: Measured temperature is not very meaningful, as it will vary with flight altitude. Better: Potential temperature. The standard deviation on l. 22: Within flight-track or among flight tracks? Also (Table 1): Relative humidity at flight altitude does not appear very meaningful, better dry mole fraction or partial pressure;

p. 7971 l. 16: Is a dynamic upwash correction applied to the wind measurements?

p. 7972 l. 3 – p. 7975 l. 2: Description of flight patterns overly detailed. Move to supplementary materials;

p. 7975 l. 9: EGU journals use metric units.

p. 7975 l. 20: Sensor models and pre-/post calibrations?

p. 7976 l. 17: …were kept constant across all flights?

p. 7977 l. 18: I am not familiar with the PTR-MS methodology. Does the sensor report dry mole fraction, or do density corrections due to temperature differences and humidity have to be applied? If so, these might be significant, as relative humidities as high as 100% are reported.

p. 7978 l. 7: w'c' is missing the overbar.

p. 7978 l. 13: …frequency of the transporting eddies…

p. 7978 l. 23: "build" and not "built".

p. 7978 l. 25 – p. 7979 l. 18: Does not contribute to manuscript objectives and can be omitted.

p. 7980 l. 3: How was the depth of the surface layer determined?

p. 7980 l. 20: The conventional method of determining EC flux is a time-domain Reynolds-decomposition (e.g., Foken, 2008). FFT requires additional pre-processing steps such as tapering etc. and hence alters that data basis.

p. 7980 l. 25: …affected by non-stationarities…

p. 7981 l. 18: …preserves the energy…?

p. 7981 l. 20 – p. 7982 l. 4: Does not contribute to manuscript objectives and can be omitted.

p. 7981 l. 24: Nordbo and Katul (2012) focus on a spectral correction method but do not specifically address long-term CO_2 fluxes from soil.

p. 7981 l. 5: Repetitive, can be shortened.

p. 7981 l. 11: Suggest clarification: …integration of a sub-segment (e.g. 2 km) or an entire flight segment (e.g., 100 km)…

p. 7982 l. 19: Why would agreement between FFT and CWT results add confidence to the flux estimates? The agreement is basically a measure of how well the stricter assumptions on FFT are fulfilled, and how modifications of the data such as de-trending and tapering affect the results.

p. 7982 l. 22: To this point, no spectral correction was mentioned in the text. Hence for a comparison among FFT and CWT it doesn't matter whether high-frequency spectral loss is present or not - it should be reflected by either method. How were high-frequency spectral corrections performed by the authors?

p. 7982 l. 24: The approach of Nordbo and Katul (2012) can correct spectral attenuation as long as not related to sensor displacement.

p. 7983 l. 2: How was the contribution of the storage term below aircraft flux sounding level determined from profile flights above this level?

p. 7983 l. 10: Footprint results are nowhere mentioned in the results/discussion, but are crucial to be considered for a model validation. What is the accuracy of the simplified approach to source area quantification? Despite omitting the use of actual along- and cross-wind probability density function, is the approach sufficiently reliable to allow distinguishing different surface sources from a flight altitude of 400 m? Is the along-wind PDF evaluated each 2 km, or is turbulence statistics calculated over an entire transect? How are footprints superimposed along transects?

p. 7983 l. 21, p. 7984 l. 7: The authors are making an effort to characterize uncertainty, but it is not clear how individual error sources are propagated. Principally, the study is lacking a thorough, quantitative uncertainty budget on a per-sample basis, from which the statistical significance of the reported 2 km results would be evident (e.g., Fig. 7, Sühring and Raasch, 2013). In addition to errors originating from instruments and turbulent sampling, such budget should quantify uncertainty resulting from time-frequency and source area analyses, parametric and structural errors in the data processing. For example, random sampling uncertainty for 2 km segments are expected to be much larger than the stated 5% (Finkelstein and Sims, 2001; Salesky et al., 2012), and no flux detection limit is provided (Billesbach, 2011).

p. 7984 l. 21: The authors preserve the global covariance through considering wavelet coefficients above the cone of influence while attempting to offset edge-effects resulting from a limited length of the time series. How is this achieved by padding with zeroes? I would imagine that cyclic boundary conditions are less prone to these edge effects?

p. 7984 l. 27: Heat flux is used as spectral reference for BVOC fluxes. This assumes that (i) the frequency response of the temperature sensing element is sufficient, (ii) temperature is measured in the free airstream and not subjection to dampening effects from housing (e.g., Rosemount), (iii) radiation error has been corrected, (iv) adiabatic heating caused by aircraft propagation has been corrected, and (v) adiabatic heating caused by the aircraft vertical movements has been corrected (potential temperature at average flight level). Have these steps been considered?

p. 7985 l. 15: ≈20 m s^{-1} is a slow flying aircraft (e.g., van den Kroonenberg et al., 2008) and <<100 m is a flight altitude close to the surface for flux measurements (e.g., Zulueta et al., 2013).

p. 7986 l. 19: How can measurements of reactive trace gas species be performed reliably after transporting an air sample through more than 500 m of tubing? I would imagine that the dark room reaction kinetics in the tubing is quite different from the ambient reaction kinetics?

p. 7986 l. 19: …Twin Otter…?

p. 7987 l. 12: What is the isoprene flux detection limit?

p. 7987 l. 15, p. 7988 l. 6, p. 7988 l. 19: How was the measured concentration mapped to the landscape? I did not see the application of a concentration footprint model. Do the authors actually distinguish between flux and concentration footprint (e.g., Schmid, 1997)?

p. 7989 l. 2: PTR-MS doesn't measure fluxes.

p. 7989 l. 8: Technical detail that can be moved to supplementary materials.

p. 7989 l. 20: Repetition from Sect. 2.7.1 that can be omitted.

p. 7990 l. 26: …emission strength…?

p. 7991 l. 1: Here it says 1–10 mg m^{-2} h^{-1}, while on p. 7990 l. 17 it says 1–15 mg m^{-2} h^{-1}. Where do the differences originate from?

p. 7991 l. 11: This is a very qualitative analysis, how does that address the manuscripts goal of testing emission models? Quantitatively relating measured emission strength to LAI and land cover type would be desirable.

p. 7991 l. 25: What are the uncertainties around these values?

p. 7992 l. 6: Repetitive and qualitative, paragraph can be omitted.

p. 7992 l. 14: What is the source for temperature and radiation information to perform the normalization? At 400 m above ground, temperature, radiation and (passive vs. active scalar) fluxes originate from very different source areas. How do you take this into account? Also, Have atmospheric corrections been applied?

p. 7992 l. 23: No reference is provided for Misztal et al. (2014).

p. 7993 l. 1: Paragraph wholly qualitative, not living up to the manuscripts goal.

p. 7993 l. 10: This is a nice summary. But what about the potential future impacts of the summarized activities?

p. 7994 l. 15: The bibliography should be as consistent as possible. For example, Lenschow (1986) appearing before Lenschow et al. (1980), or names being spelled out as Lenschow, D. one time and Lenschow D.H. the other time.

p. 8004 Fig. 2: Technical detail that can be omitted.

p. 8005 Fig. 3: Optically, the space series of instantaneous fluxes doesn't appear to match the coefficients of the wavelet cross-scalogram very well. For example, at ≈45 km and towards the end of the track at at ≈110 km.

p. 8006 Fig. 4: What about comparing the co-spectra to a reference model?

p. 8007 Fig. 5: How exactly is the difference between net flux and turbulent flux (vertical flux divergence/storage)´being calculated? The ratio appears to decrease towards the end of the flight track. How can this be significantly supported by the profile measurements?

p. 8008 Fig. 6: Not really meaningful, as source areas for the concentration measurements are different from the location of the aircraft. A spatial projection is required for that purpose.

p. 8012 Fig. 7: This is OK for an overview. However, for the claimed observation-model inter-comparison, the quantitative agreement with the MEGAN emissions in the flux footprint would have to be shown. For example, an error-in-variables regression.

p. 8012 Fig. S2: Looks nice, but doesn't really tell anything about vertical flux divergence, a term that is sought for to prove significance of the stated emissions.

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