Atmos. Chem. Phys. Discuss., 15, C12699–C12707, 2016
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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Airborne observation of mixing across the entrainment zone during PARADE 2011" *by* F. Berkes et al.

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Received and published: 24 February 2016

We would like to thank both anonymous referees for their valuable comments on the manuscript which led to a significant improvement of the manuscript. According to the reviewers suggestions we included an additional Figure (now Figure 6). For the specific replies below we refer to the figure numbering of the original version. Referee 2 comments are given in bold, the answers in standard font. Changes to the text are in italics.

Comments The impact of the paper could be enhanced by drawing more connections with current research. The introduction is excellent and thoroughly describes many of the foundations for this work. But it isn't completely clear

to me how this research that carefully examines a single event can be used for future studies. Will it be necessary to have the complete suite of measurements and analyses presented here to interrogate mixing? Do clouds regularly drive this sort of mixing, and can these results be expected for other locations? Is CO2 a reliable tracer for mixing between the free troposphere and PBL? Can anything be said more generally about when and where this sort of mixing can be expected?

The questions are repeated below including specific comments and are answered there.

If it is possible to make more general statements about the case study presented here, then the results may be especially helpful for understanding ozone entrainment and mixing in the US. In the western US, vertical transport is particularly important for understanding compliance with air quality regulations, since ozone standards are tightening and some regions are strongly influenced by downward ozone transport from the FT. Referencing a few of the recent papers that discuss baseline ozone in the US and the contribution from downward mixing (e.g. Jaffe, EST, 2011; Cooper et al., Science, 2015; Lin et al., JGR, 2012) will help connect this work to a large and active research area.

We agree that the linkage to the studies in the US is relevant and included the following lines at P29174, L12: *Cooper et al (2015) highlighted that surface air quality regulations in the US strongly depend on accurate knowledge of baseline ozone, which is defined as ozone transported downwind from anthropogenic and natural sources, while forecast of baseline ozone needs large knowledge of all different transport processes. This includes intercontinental transport from Asia, which is often associated with episodes of enhanced ozone concentrations over large parts of the western US that is located at relatively high altitudes (Lin et al ,2012). Moreover, downward transport of ozone rich air from the FT can enhance ozone mixing ratio in the PBL (Beck et al., 1997, McKendry et al, 2000, Jaffe, 2011).*

Many papers use CO rather than CO2 as a tracer for tropospheric air. Why is this the first use of CO2 and O3 to examine mixing (pg 29175, line 6), as both molecules are regularly measured? Justifying the use of CO2, which can be influenced by the biosphere as well as anthropogenic emissions, would be helpful. CO2 has been used as a tracer for pollution in winter, but in summer CO2's utility as a tracer is diminished because uptake and respiration introduces considerable variability. Noting the limitations for using CO2 to diagnose mixing would be helpful. CO2 variability is used here as an indicator of mixing. Quantifying this variability will help to connect these results with other studies. The mixing lines are a consequence of CO2 and O3 differences between the free troposphere and PBL, and comparing the variability to this difference may also provide a metric that could be used to quantify mixing.

We agree that we need to extend the discussion on CO2 as tracer and its limitations. On the days studied in our case study, CO2 showed stable mixing ratios of about 386 ppm with a small variability of less than 0.5 ppm in the FT. Additionally, we demonstrate that the air mass in the PBL was not affected by long range transport within the FT, which is supported by backward trajectories. Pataki et al 2005 showed that CO2 can be used as tracer when no major sink is available, which is the case for the FT. This gives confidence that CO2 can be used as a tracer to identify mixing events at this particular day. Importantly, the variability of CO2 near the surface is needed to identify potential mixing events, since this leads to the formation of a gradient at the inversion. During night time respiration and potential impact from anthropogenic emissions are expected to dominate the surface measurements for the period of our case study. During night we observe larger CO2 mixing ratios than during the day close to the surface in the PBL, which is also the case during the mixing event. The mixture consists of air masses of high CO2 from the early morning and of low CO2 from day. We agree that CO or the combination of CO2 and CO would increase the potential to identify and interpret the mixing event, but CO measurements are not available for the specific time period. We conclude that CO2 is not the perfect tracer to study mixing, especially

when biogenic update cannot be excluded, but in our particular case with the given circumstances it can be used to identify mixing events. To the authors' knowledge, it is the first study which used CO2 as tracer for mixing at the PBL top along with the tracer-tracer correlation method.

The figures are clear, but some of the legends are confusing. The text describing figures 6 and 7 mentions panels a) and b), but those labels don't appear on the figures, or appear with the date so as to be confusing. For figures 6-8, the confusing date/time label should be removed, and the date should be mentioned in the caption (the time already is). The label in figure 8 that lists the time contradicts the caption. The legend for figure 5 should change "PBLH/RLH" to "aerosol layer height". I don't understand the CO2 scale at the top of figure 5. If the times are given in local solar time, then the "morning/noon/afternoon" labels could be removed to declutter the figure.

We improved the figures 5 – 9 as suggested. The CO2 scale in figure 5 helps to link this figure with Figure 6 and is valid for the CO2 profile at 8:00 UTC, for the two remaining profiles the origin of the x-axis was shifted to time of measurement (11:30 and 13:15 UTC).

Smaller comments 1) Verb tense switches inconsistently between past and present, which leads to some confusion. I recommend that all descriptions of the field measurements from 2011 be written in the past tense and all descriptions of the current analysis be written in the present tense. For example, section 2.2.2 "The pressure was obtained. . .". Section 3.2 "PBLH decreased. . ." and "air mass composition was probed. . ." Section 3.3 "residual layer was. . ." and "CO2 was again well mixed. . .". Section 4.2 "PBLH grew to . . ." Section 5 "PBL was convectively driven during that day. . ."

We improved the grammar of the manuscript. Please see the track changes.

2) Some of the terminology is unclear. The stable layer is mentioned in the 2nd

sentence, but never again in the text, yet it shows up on the figures. The text should discuss more clearly when the term PBL is used, and when SL/RL are used. I don't understand the sentence pg 29174, line 17 that mentions the inversion layer. Which layer is this? And how does the barrier exhibit gradients?

We clarified the terminology throughout the manuscript and changed the respective sentence as follows: *However, the boundary layer inversion acts as a barrier for mixing and often exhibits strong trace gas gradients.*

3) Please note time response for all the measurements 2.2.1, which will be important if variability is quantified.

We updated the time response for CO2 and O3 within the text: *The response time of the instruments (5%/95%) was 3 s (CO2) and 9 s (O3) and the final data correction accounted for different residence times in the inlet lines.*

4) pg 29184, line 9: replace "not directly related to clouds" with a sentence that states the measurements were not obtained in clouds. Later, the importance of clouds was noted, and that the measurements are related to clouds. Changed sentence to: *Since the aircraft was allowed only to operate in cloud-free conditions, the large humidity values were only observed within the PBL.*

5) Why do the O3, CO2 and water mixing ratios change with altitude in the PBL, as shown in Fig 7? Is the potential temperature constant? Is the PBL well-mixed when there is a strong vertical gradient in mixing ratios?

We included an additional figure (now Figure 6, see response to reviewer 1). The potential temperature is constant within the PBL, which is an indication for neutral stratification. The variability of ozone, CO2 and water vapor is affected by different sources, which we cannot separate but may be related to upward mixing from the dissipating night-time stable layer. At this time in the morning the trace gases are not completely mixed. Later that day, the profiles of CO2 and O3 become more constant with altitude

(see Figure 5, after 13 UTC). We think our study is related exactly to this transition from a state of incomplete mixing (with large tracer variability) to the well mixed state during day time.

Pg 29174, line 29: replace aloft with above

Changed

Pg 29175, line 21-23: remove sentence, as the air mass influences are described better in the next paragraph

Removed

Pg 29177, line 5: use m asl instead of km to be consistent with the rest of the paper

Changed

Pg 29177, line 3: describe the profiles more specifically: spirals, and give the radius of the spirals.

We modified the sentence: *The descending flight legs were flown in spirals with a diameter of about 10 to 15 km.*

Pg 29178, line 24: replace an with a

Changed

Pg 29179, line 3: replace "in 10 min average" with "as 10 min averages"

Changed

Pg 29181, line 11: replace "High CO2 values" with "CO2"

Changed

Pg 29184, line 13-14: Remove "after discussing" and replace "we use these information" with "are used"

Changed

Pg 29185, line 5: replace sentence beginning with "In this altitude. . .." With "The EZ was sampled at this location and time".

Changed to : *The EZ was situated at this location and time.*

Pg 29187, line 3: replace "has grown" with "grew".

Changed

Pg 29190, line 1: difference rather than differences

Changed

Pg 29192, line 4: ". . .lead to variability that can be characterized by mixing lines"

Changed

Fig 1 – include Mt Kleiner Feldberg on map

Included

Fig 5 caption: replace rain bow with Rainbow

Changed

Fig 7 caption: replace "related due to the mixed air masses.." with "influenced by mixing air masses from the PBL. . ."

Changed

References:

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Lin, M., Fiore, A. M., Horowitz, L. W., Cooper, O. R., Naik, V., Holloway, J., Wyman, B. (2012). Transport of Asian ozone pollution into surface air over the western United States in spring. Journal of Geophysical Research: Atmospheres, 117(4), 1–20. http://doi.org/10.1029/2011JD016961

Pataki, D. E., B. J. Tyler, R. E. Peterson, A. P. Nair, W. J. Steenburgh, and E. R. Pardyjak (2005), Can carbon dioxide be used as a tracer of urban atmospheric transport?, J. Geophys. Res., 110, D15102, doi:10.1029/2004JD005723.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 29171, 2015.

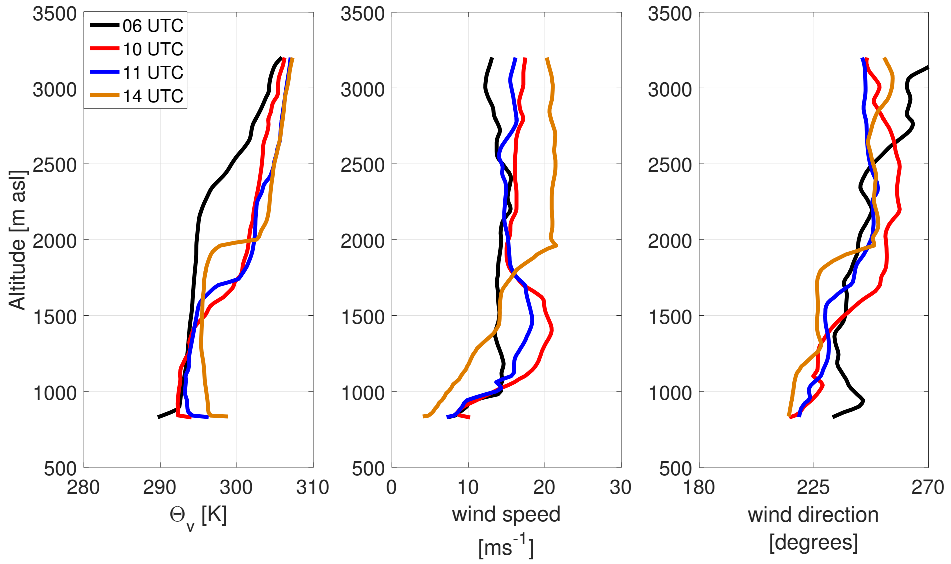

Fig. 1. Profiles of a) virtual potential temperature, b) wind speed, and c) wind direction derived from radiosondes during the day (black : 6 UTC, red: 10 UTC, blue: 11 UTC, orange: 14 UTC) on 6 September 201