

Interactive comment on “Evaluation of the flux gradient technique for measurement of ozone surface fluxes over snowpack at Summit, Greenland” by F. Bocquet et al.

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Received and published: 10 August 2011

We appreciate the insightful comments from both reviewers. Below are our responses to their comments and the corrections to the manuscript.

Reviewer 1 Comment:

General Comment: Since instrumental precision is of such paramount importance in determining the small ozone exchange velocities (or concentration differences with height), I am curious as to why the investigators chose to use three separate analyzers to measure ozone at the three heights as opposed to a single analyzer that

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would switch between levels. I am not suggesting that there is anything wrong with the current work, but if the precision of a single analyzer is 0.02-0.03 ppbv (pg. 1040, for averaging thirty 1-min measurements), then if you only spent 10 minutes on each level with a single analyzer (during a single 30 min. flux period), one would expect the ozone gradients to be good to 0.04-0.05 ppb (dividing by square root of 10 instead of 30, pg. 1040). This is nearly a factor of two better than the reported \sim 0.1 ppbv overall uncertainty reported on pg. 1040 when the differing instrument offsets were factored in. One would have to cycle through the levels relatively quickly (e.g., 1 or 2 min/level) to obtain a representative average concentration at each level and would likely still have to bring the inlets to a common height to test for the possibility of differing losses in the individual lines.

Author Response:

This is a valid suggestion. As a matter of fact, we debated at length at the startup of the project which of these two options would be the better one to pursue. There are a number of pluses and minuses to both of these approaches. An advantage of the two-instrument approach is that one will get close to 100% time coverage for gradient measurements, twice as much as for a switched inlet measurement. No switching manifold is needed, which technically simplifies the measurement, makes it more robust, and eliminates potential chemical ozone losses and analytical biases from the sampling manifold. The three-instrument approach gave us a set of three gradients, which provided a valuable opportunity for quality control of the measurement and data analysis protocol (however, in the course of this experiment we found out that at times fluxes within the measurement height interval were not constant under conditions encountered at Summit). This approach gave us one level of redundancy, allowing continuation of measurements if one of the instruments was to fail.

As rightly stated, working with several instruments requires the correction for the calibration offset of the analyzers. While our protocol accommodated that need quite well, an unexpected and major complication arose from instruments drifting differently over

time, and during several occasions experiencing sudden step changes in response (Manuscript Figure 4).

Working with one analyzer and switching between inlets eliminates the need for accuracy correction, which is a major advantage when wanting to achieve a high accuracy in the gradient determination. One analyzer will spend only $\frac{1}{2}$ of the time sampling from a given measurement height, and consequently only half as many data points are recorded at each height as with the two-analyzer configuration. As additional time is needed to purge the instrument and plumbing components every time the sample is switched to a new inlet height, data from the transition periods need to be eliminated, resulting in further reduction of useable data. We were particularly concerned about the analyzer equilibration time to the changing water vapor levels that may be in air pulled from different heights. Another disadvantage is that changes in ambient concentration with time occurring between the switching intervals could falsely be interpreted as a vertical concentration gradient. Since the experiments described in this paper we have done further ozone flux gradient measurements, and in several of these studies we have used the 'switched gradient' approach. We are still evaluating these results but intend to present a quantitative comparison of both techniques soon.

Reviewer 1 Comment:

Specific Comments: pg 1023, lines 6-12. It may be worth mentioning that although it is much easier to measure the ozone gradients within the snowpack, it is exceedingly difficult to quantitatively ascertain the flux from these types of measurements due to the difficulty in estimating gas diffusion within a constantly changing snowpack, as well as steep gradients in solar irradiance and other chemical constituents within the snowpack that may play a role in the ozone loss. (there are also sampling issues within the interstitial snow)

Author Response:

Yes, this is 100% correct. We have added a statement reflecting this in the revised

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manuscript version.

Reviewer 1 Comment:

Pg. 1024, line 24. I am not sure that there is much distinction between the "gradient method" and the "gradient profile method". Another name is typically the "aerodynamic profile method" (note that Section 3.1 is titled: Aerodynamic gradient method).

Author Response:

Since we represented the flux in terms of the vertical gradient, we consider it to be the gradient method. 'Aerodynamic' does not really add anything, so we will delete this term in order to stay consistent throughout the paper.

Reviewer 1 Comment:

Pg 1027, line 20. Is showing the ideal gas law really necessary? Merely stating that you use it along with ambient temperature and pressure measurements to convert mixing ratio to density should be sufficient.

Author Response:

We have eliminated the equation and instead given the reviewer's explanation in the text.

Reviewer 1 Comment:

Pg 1030, line 5. The authors state that ozone losses through all of the system components was less than 2%, but this is nearly 1 ppbv (at 50 ppbv ambient concentration) which is large compared to the gradients you are measuring. The field tests described later where all instruments sample from the same height are more instructive as they not only test for instrument offset, but also for bias (or losses) that differ from inlet to inlet.

Author Response:

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We did several tests, and generally the ozone loss rate through one of these long lines was not much different compared to a shorter line. A loss of 2% is a rather conservative, upper value. As rightly stated by the reviewer, this concern was one of the motivations why we chose to do the frequent inter-comparison measurements.

Reviewer 1 Comment:

Pg. 1031, lines 23-25. The sentence concerning “temperature fluctuations from vertical temperature gradients: : : as well as from sensible heat flux: : :” is a bit redundant. If you have vertical temperature gradients, this implies a sensible heat flux (and vice versa). Since you are looking at mean quantities (and not fluctuations from that mean), I do not think that fast temperature fluctuations are a concern here. On a further note, how do you know that the temperature fluctuations have been equilibrated? For non-conducting sample line material, Leuning and Judd (Glob. Change Biol., 1996, 2, 241) suggest that it can take several hundred meters of tubing to equilibrate temperature fluctuations.

Author Response:

We have deleted “sensible heat flux” from this sentence.

Thanks to the reviewer for pointing out the article by Leuning and Judd, which we had not seen before. We never monitored the temperature of the sample inside the UV absorption monitor at high time resolution and therefore do not have a concrete data record to further investigate this question. Please note that during most times temperature gradients between the inlet heights were on the order of tenths of oC. Given the length of the sampling tubing, the thermal mass of the monitor and the number of ports, filters, and valves that the sample passes through, we still assume that air from the three inlet heights is at very similar temperature inside the analyzer. Secondly, the TEI monitors measure the optical cell temperature and use this measurement for correcting the ozone mixing ratio. Therefore we do not expect any bias from temperature effects in the sample air.

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Reviewer 1 Comment:

Pg. 1034, line24. Should be the “von Karman constant” (not “van”, this occurs several other places in the manuscript).

Author Response:

This has been corrected in our revised manuscript.

Reviewer 1 Comment:

Pg. 1035, paragraph beginning on line 26. What was the lower wind speed threshold used for filtering the data? It seems that there may be a subset of data where the lowest height would be below this threshold, but a gradient flux could still be obtained from the two highest inlets. Although less reliable (since no corroborating lower gradient), these could still be useful flux data.

Author Response:

The wind speed threshold used for filtering data was 0.25 m s⁻¹. This information has been added to line 14 of P 1034. This value was selected because it corresponds to the manufacturer’s threshold value for the cup anemometers we used. Gradient data were evaluated individually for each measurement interval. Data were retained when both cup anemometers of a gradient interval recorded average wind speeds of > 0.25 m s⁻¹ over a 30-min measurement period.

Reviewer 1 Comment:

Pg 1037, lines 15-18. The corrections applied do assume that you are always operating within the linear range of the instrument. For example, if there are nonlinearities in the wind speed response at low wind speeds, they may not be apparent in a side-by-side intercomparison; however, they may result in a bias when applied to gradients where one is measuring two different wind speeds at two heights.

Author Response:

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We plotted the wind speed recordings from the 3 instruments during inter-comparison periods against each other. We then chose one of them to be the 'good/true' reference instrument, and determined a first order function that would fit the second sensor data to the good instrument. Deviations between sensors were less than 2% (slope values of 1.02 and intercept of 0.07 and 0.08 m s⁻¹.) The correction functions were then used to correct data obtained during the regular flux gradient measurements.

Reviewer 1 Comment:

Pg. 1041, lines 21-27. This behavior is somewhat typical for all gradient flux measurements. As there is more turbulent mixing – the gradients become more difficult to measure. The observation that the highest fluxes also have the largest uncertainty suggests that the flux may be dependent upon turbulent mixing. A plot of the flux vs. friction velocity for periods of similar irradiance might be useful to discern this. This would not be unexpected given the prior evidence for pressure pumping in the upper layers of the snowpack.

Author Response:

We followed this suggestion and prepared graphs plotting the ozone v_e against u^* , broken up in 100 W m⁻² radiation bins. For all but the lowest (0-100 W m⁻²) bin, there was a positive correlation between the flux and u^* . This result supports the hypothesis that the ozone surface flux increases with wind speed and radiation. We intend to apply this analysis to a more extensive data set that has since been obtained at Summit during a follow-up experiment and to present these interesting findings with an in-depth discussion in a future publication.

Reviewer 1 Comment:

Pg 1042, Ozone deposition results. Where there any systematic difference between fluxes determined from the bottom two levels relative to those determined using the top two inlets? The authors should discuss this as this can serve to prove that the

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measurements were made within the “constant flux layer” or indicate possible biases in the measurements.

Author Response:

The measurement of flux divergences, i.e. changes in ozone flux with height was one of the goals at the beginning of the proposal with the intention to decipher chemical ozone production or loss from atmospheric reactions. This was one of the reasons why we had multiple flux measurement heights on the tower. From the collected data we learned that the 10 - 2 m height interval was frequently outside the lowest 10% of the mixed boundary layer (surface layer). Under those conditions the data from this interval were not suitable for flux calculations (see page 1035, lines 12-25). As shown in the manuscript, the absolute magnitude of the flux was very low, and the relative uncertainty of flux results was high. Consequently, the differences in flux results from two height intervals rarely deviated more than the combined uncertainty in each measurement. We continue working on this question, and have conducted several new experiments with multiple height ozone eddy covariance measurements over snow. These data are still being analyzed and we intend to present a comparison of these two different flux measurement approaches in another upcoming publication.

Reviewer 1 Comment:

Figure 5. Please clarify or describe what the legends represent in this figure. It is not clear what WS-10, WS-2, WS-30, Grad_10_2a, etc. stand for relative to what is described in the text.

Author Response:

The new Figure caption will read:

“Fig. 5. Example of results from the inter-comparison measurement during DOY 89-91 (March 29-31) (upper graph) and DOY 89 (March 29) (lower graph), where the three instruments were brought to the same height and run side by side. The upper graph

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shows ~1 day of wind speed data from the three cup anemometers prior to being subjected to correction functions. WS_10, WS_2, and WS_30 refer to the instruments that were operated at the 10 m, 2 m and 0.3 m height, respectively. Deviations in data between the sensors were on average < 2%, almost indiscernible in this plot. The bottom graph shows the raw, uncorrected data from the intercomparison of the temperature sensors. Gradients were determined directly. For example, 'Grad_10-2' stands for the gradient measurement between the 10- and 2-m measurement height. Deviations from zero are due to instrument noise and measurement artifact (see discussion in section 6.1)."

We have also reproduced this figure in color to better differentiate the data series shown in these graphs.

Reviewer 1 Comment:

Figure 8. Since many of these points overlay one another – it would help clarify if you used a different color for one of the data sets.

Author Response:

We have followed this suggestion and are now using red and blue color for the two data series.

Interactive comment on Atmos. Meas. Tech. Discuss., 4, 1021, 2011.