Atmos. Meas. Tech. Discuss., 4, C1346–C1356, 2011
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Atmospheric Measurement Techniques Discussions

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.

F. Bocquet et al.

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Received and published: 10 August 2011

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

Reviewer 2 Comment:

Is the air really homogeneous in all directions? Is there an impact larger than the measured ozone gradients due to high NOx in camp influenced air? Can the camp influenced air be removed from the data by filtering on the wind direction?

Author Response:

Summit is located at the center of the Greenland ice field (http://www.summitcamp.org/). In all directions the terrain is flat with very gently downward slope. This makes this location an ideal flux experimental site. Air can get contaminated from camp operations, however. In order to minimize this effect, the flux site was located ∼250 m upwind (south) of the station, i.e., in the 'Clean Air Sector'. Approximately 90% of winds come from the clean air footprint. Data were nonetheless filtered by wind direction, and data from the 200 to 70 degree sector were eliminated. We have developed this data filtering using multiple atmospheric tracers, including NOx, which is very sensitive to camp emissions, and have found it to be a very robust method for assuring that measurements at the flux site represent clean, background air conditions.

Reviewer 2 Comment:

The authors should clarify why they have decided to do the flux calculations in these units (equations 1 & 2). It is my understanding that in most models the diffusion of gases is treated in terms of mixing ratio to ensure that there are not errors in the calculated diffusion rates. This is because differences in the air density (at a given temperature and pressure) result in different concentrations, but not different mixing ratios. In a situation without deposition towards the surface there still may be a vertical gradient in the concentration due to differences in the air density. Shouldn't calculations of the vertical flux take this into account?

Author Response:

As air was sampled from two inlets to two instruments that were collocated, and both instruments correct for pressure and cell temperature, there was no need to correct for changes in concentration due to density differences at the inlet heights. Results are presented as surface exchange velocity, ve, in analogy to deposition velocity, vd. Also, 1/vd = Rs, the surface resistance, is the variable commonly used in models.

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Reviewer 2 Comment:

The authors should comment on the conditions at Summit and if they are close to near-stable and near-unstable to understand if equation 3 applies.

Author Response:

Equation 3 applies to all stability conditions, which is useful as there is always some deviation from ideal neutral stability. The effect of the correction and the uncertainty in the flux calculation increases with the degree of deviation from neutral stability. Therefore, the stability function was only applied for a of Richardson Number values of -0.1 < Ri < +0.1, and data outside of this window was rejected from further analyses. The data in Table 1 show that a significant fraction of the measurements fell outside of this range. Figs S-3, S-6a,b, and Table S-1 in Helmig et al., 2009 show the quantitative distribution of the Richardson number and stability function for Summit conditions.

Reviewer 2 Comment:

More detail on the ozone losses discussed on P1030 should be included. Was this measured only in the lab, but not in the field? Was this a systematic, reproducible error that has been corrected for based on the tube length?

Author Response:

As already mentioned in the reply to reviewer #1, the 2% value was an upper limit, determined in laboratory experiments prior to the shipping of equipment to the field. Since this experiment we have conducted another study at South Pole, where we sampled ambient air through 135 m-long PFA tubing, $\sim$ 4 times as long as the tubing used in the Summit experiment. During the South Pole experiment the ozone loss rate in the line was monitored over 9 days, and found to be 1.8 +/- 0.8 % (1 ïĄş) (Helmig et al., 2008). We thoroughly compared the ozone data from our flux gradient measurements with the data from the NOAA surface ozone monitoring at Summit, and found the data to generally agree to within 2-3%. We did not see a need to correct for this relatively

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small bias, as in our case the differences in ozone between two measurement heights was the critical variable that was determined. And any biases in the ozone loss rate between lines or instruments were corrected by the regular same height inter-comparison measurements that were performed at the site.

Reviewer 2 Comment:

In Section 5, there is also an uncertainty in using these equations at all. How does this compare with the error from the detailed Monte Carlo simulations?

Author Response:

It is not clear to us what this reviewer question refers to.

Reviewer 2 Comment:

The authors should comment further on Figure 9 and discuss more the features and their possible meaning in the text.

Author Response:

An in-depth discussion of these data, a comparison with previous literature, and the implications of these findings on the polar ozone budget has been presented by Helmig et al. 2009, and is outside the scope of this manuscript, as here the focus is on the technical aspects of this experiment.

Reviewer 2 Comment:

Technical corrections: Abstract: The authors should add the month and year of the measurements to the abstract. It is also a bit confusing to talk about the seasonal dependence, when the measurement period is only four months. It would be clearer to simply say summer vs. spring.

Author Response:

Month and year of measurements have been included in the revised manuscript ab-

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stract, and the wording was changed from ‘seasonal’ to ‘spring versus summer’. Reviewer 2 Comment:

General: I recommend using the same units for deposition velocity as well as surface resistance throughout the paper: either cm or m as the distance unit.

Author Response:

Ozone exchange velocity is reported in units of cm s-1, which is the most common unit used in the literature.

Reviewer 2 Comment:

P1023 L5: The sentence that starts here is out of place, this paragraph should be rewarded to make it more clear that ozone behavior and chemical interactions in the snow pack are coupled. It should also be noted that by measuring very accurately the ozone deposition, you are capturing the net effect of some of the influence of the snow on boundary layer air.

Author Response:

The revised manuscript now starts with a new paragraph. It reads:

“Ozone fluxes over snow are linked to chemical interactions in the snowpack. Ozone in the polar snowpack has been found to undergo depletion, which follows both the diurnal and seasonal cycle in solar radiation (Helmig et al., 2007b). These observations suggest that ozone surface fluxes should vary similarly with time of day and season. Accurate measurements of ozone surface exchanges would provide a means for evaluating the net effect of ozone snow photochemical processes on boundary layer ozone. Previous ozone flux measurements over polar snow have not been able to demonstrate this behavior, likely due to the lack of measurement sensitivity and required long-term observations.”

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P1024 L1: A summary sentence describing why you discuss these measurements in such detail is needed.

Author Response:

The following sentences were added:

“Previous ozone flux measurements over snow have been conducted using an array of different techniques. Few of these studies provided detailed characterization of the sensitivity of the flux measurement. Galbally and Allisoin (1972) were the . . . . . .“

Reviewer 2 Comment:

P1024 L19-23: This should be reworded to be clearer. I suggest adding some additional references to these statements.

Author Response:

The text has been reworded to:

Despite this relatively low surface uptake rate, ozone deposition to polar snow is an important ozone sink because other chemical processes that determine the ozone budget are weak compared to other environments (Helmig et al., 2007a).

Reviewer 2 Comment:

P1024 L28: Add numbers, what is the total signal and what is a typical gradient?

Author Response:

The revised sentence now reads:

The challenge of flux gradient measurements is to conduct highly accurate and precise measurement of gradients that are small (on the order of tenths of ppbv for ozone, as well as tenths of oC and m s-1 for temperature and wind speed, resp.) in relation to the absolute magnitude signal of the measured variable.

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Reviewer 2 Comment:

P1026 L21: at Summit, compared to other Arctic sites.

Author Response:

Corrected sentence now reads:

"Surface ozone levels at Summit are remarkably high compared to other Arctic sites, typically falling . . .."

Reviewer 2 Comment:

P1026 L22-25: The authors should consider adding other references, there is an on-going debate as to what causes high ozone at Summit as well as what differentiates the ozone seasonal cycle at Summit from other Arctic sites. Some other papers in the literature should be referenced.

Author Response:

Two recent publications (Helmig et al., 2007b, c) investigate the surface ozone behavior at Summit and are cited for reference.

Reviewer 2 Comment:

P1029 L9: Add a citation to "most of the literature demonstrated".

Author Response:

The text has been updated with two references:

"As most of the literature have demonstrated (e.g., Oke, 1987; Stull, 1988),. . ."

Reviewer 2 Comment:

Figure 3: Is this representative of the other inter-comparison graphs?

Author Response:

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Yes, this is a typical example for the precision of the measurement. However, the absolute values of the disagreement between monitors showed some considerable variation, as shown in Figure 4.

Reviewer 2 Comment:

Section 5: More detail on the range of values tested should be included. What temperature range, what wind speed gradient range, what range of sensor accuracy, etc?

Author Response:

The section describing the Monto Carlo simulation has been revised and is now providing this particular information:

"The relative contribution of individual input variables to the uncertainty of the flux determination was assessed by the Monte Carlo simulation approach. The uncertainty was calculated for the 30-min average ozone exchange measurement results obtained from both the {2-0.75 m} and {10-2 m} gradient measurements. Thirty-min averages and estimated uncertainties were used for each pair of the air temperature gradient, wind speed gradient, ozone mixing ratio gradient, measurement height gradient, and ambient pressure. For temperature, wind speed and ozone mixing ratio, measurement uncertainty estimates were derived from the ensemble of inter-comparison experiments while for ambient pressure and heights, uncertainty estimates were derived from estimates of sensor accuracy and measurement precision, respectively. Utilized uncertainty values (see further discussion below) were 0.05oC for the temperature measurement, 0.05 m s-1 for the wind speed measurement, 0.1 ppbv for the ozone determination, 0.5 hPa for the pressure measurement, and 5 cm for the measurement height. Furthermore, the ranges of the input variables that were tested by Monte Carlo simulations were -40 to -10°C for temperature, 1 to 10 m s-1 for wind speed, and 35 to 65 ppbv for ozone. A total of 5000 Monte Carlo simulations were run for each calculated 30-min ozone exchange velocity, providing an uncertainty value for each 30-min ozone exchange velocity measurement. The Monte Carlo sensitivity analysis also in-

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vestigated how the distribution of the output variable (ve(O3)) varied with changes in only one input variable (while all other input variables are held constant at their 30-min value). This analysis, called the nominal-range analysis, was used to determine which input variable(s) had the largest influence on the output uncertainty."

Reviewer 2 Comment:

Figure 5: In the current form this figure is confusing, is this during an inter-comparison experiment? The figure caption should be more detailed and clearly state the height of the inlets. If the change in temperature is associated with error, then this should also be stated in the figure caption. Given the sudden onset of the temperature disagreement, it would be interesting to include the specific algorithm for correction and if possible the sensitivity of calculated fluxes to using it.

Author Response:

Figure caption has been revised to:

Example of wind speed (absolute) and temperature gradient data measured during an inter-comparison experiment (i.e., all sensors were kept at the same measurement height). Shown are the raw data from the three sensors prior to being subject to correction functions to account for sensor discrepancies.

Reviewer 2 Comment:

General: The figure captions all need to be updated to be more clear and contain more information. Because DOY is not easily translated to month and day, the authors should at least include the date range in the figure caption. All of the figures with gradient and/or flux data should include a definition of positive vs. negative so it is clear if there is deposition or emission. Extra information not referred to in the text (for example the text on Figure 7) should be removed from the figure.

Author Response:

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Figure captions have been updated and other changes have been made as suggested.

Reviewer 2 Comment:

Figure 8: This figure should have two panels, one for the 10-2 m data and the other for the 2-0.75 m data.

Author Response:

As noted in our response to reviewer #1 above, red and blue colors are now used for the two data series for better differentiation.

Reviewer 2 Comment:

General: I assume during this time the inlets were not always at the stated heights, can the authors include a table of day of year vs inlet height in the electronic supplement. Is there any difference in the behavior when the 0.75 m inlet was very close to the surface?

Author Response:

The measurement heights changed during the course of the experiment due to the snow accumulation around the tower. The rise in the surface was monitored continuously with an ultrasonic distance sensor (Model SR50, Campbell Sci., Logan, Utah) that was mounted on the tower. In addition, sensor heights were manually determined four times during the course of the experiment. From March 30 – August 14, instrument heights decreased by $\sim$23 cm; changes in height were accounted for in the calculation of fluxes. We did not decipher any systematic changes in the data due to the change in inlet heights. We do not see the need to present a table displaying the changes in heights as it would not add pertinent information to the discussion.

Other note: We have clarified Table 1 by revising the Table caption to:

"Percentage of data that were removed by ten successive quality control filters. Each row lists the additional percentage of data that were eliminated from the data remaining

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after applying all preceding filters."

Row 9 in this table is further detailed as:

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9 Boundary layer height (PBL)

PBL < 100m for {10–2m} 2.6 1.0 3.4

PBL < 20m for {2–0.75m} 0.0 0.0 0.0

References

Helmig D., Ganzeveld L., Butler T., and Oltmans S.J. (2007a) The role of ozone atmosphere-snow gas exchange on polar, boundary-layer tropospheric ozone – a review and sensitivity analysis. Atmos. Chem. Phys. 7, 15–30, 2007.

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Helmig D., Johnson B., Oltmans S.J., Neff W., Eisele F., Davis D.D. (2008) Elevated ozone in the boundary-layer at South Pole. Atmos. Environ. 42, 2788-2803.

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