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Atmospheric Measurement Techniques Discussions

Interactive comment on "A relaxed eddy accumulation system for measuring vertical fluxes of nitrous acid" *by* X. Ren et al.

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Reply to the Review by Referee #2

We thank Referee #2 for providing us valuable review comments that have improved the manuscript. We have included the review comments followed by our responses. In the revision of this manuscript, we will highlight the changes accordingly.

Comment (1): 4110 L2: might want to indicate here that at least one of the channels (updraft or donwdraft) is going to be sampling zero air at any given time so all samples will be diluted at the analyzer. This imposes even greater conditions of sensitivity on the instrumentation. Maybe modify and move the paragraph at the end of section 2.2

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to here.

Response: As suggested, we have added here: “During field deployments, at least one of the channels (updraft or downdraft) samples zero air at any given time so all samples are diluted at the analyzer.”. For a better flow of the context, we also moved the last paragraph of Section 2.2 to here.

Comment (2): 4111 L11: what is the pH of the buffer?

Response: The pH of the buffer is 6.8. We have mentioned this here in the text.

Comment (3): 4112 L5: Can the authors indicate where the inlets were situated relative to the forest, field, road that was closest? Were the measurements made above a forest in both cases? From the same height above the forest?

Response: We have added a few sentences here to describe the inlets in the two field campaigns as: “The instrument was placed on an 18 m scaffolding tower, standing on an agricultural field. The height of the inlet was 13.5 m above the ground.” for CalNex 2010 and “The inlet was located on a scaffolding tower at a height of 13.5 m above the ground, or $\sim$5.5 m above the canopy of the surrounding pine trees.” for BEARPEX 2009.

Comment (4): 4114 L8: From this discussion it is not clear WHY Eq. (2) was used to calculate beta and then why Eq. (3) was even discussed. A few sentences here will help the reader understand the authors’ rationale for their analysis.

Response: We believe that the discussion of Eq. (3) is necessary for the comparison of the proportionality coefficient calculated from Eq. (2) with theoretical estimation. As discussed at the end of the first paragraph in Section 3.1, there are large uncertainties in Eq. (2) when the differences of Tupdraft and Tdowndraft are small, e.g. at night during near neutral atmospheric stability. To smooth the flow of the text, we have added a few sentences at the beginning of this paragraph as: “Because of the large uncertainties in the proportionality coefficient at night, theoretical estimation of the β-value has also been established. Comparison of the β-value calculated from Eq. (2) with the theoretical estimation can ensure that we can apply the daytime β-value calculated from Eq. (2) for the night time."

Comment (5): 4114 L12: The description of time lag calculation can be made clearer. Based on the diagram in Fig1, the delay time is going to vary based on the mode of sampling. Total flow rate through the inlet is going to be the sum of 3 different flows. Because at least one sampler will be sampling zero air at any given time, there are two possible total flows: 9 LPM during deadband periods or 11 LPM during sampling. Which flow rate was used to calculate the delay time? Will the 20% change in flow during deadband or any pressure effects in the inlet affect the delay time for molecules to get to the scrubber? I would have been more comfortable with a dosing experiment with a standard gas to determine this rather than straight calculation.

Response: We considered this flow difference in the data acquisition system for the two cases in terms of air motion switches. Case 1: air motion switches from deadband to either updraft or downdraft. In this case the total air flow rate through the sample line is 9 L min-1 prior to the valve switching and the delay time in the data acquisition was set to 430 ms (the time for air traveling through the sampling line). Case 2: air motion switches either from updraft to downdraft or from downdraft to updraft. In this case the total air flow rate through the sample line is 11 L min-1 prior to the valve switching and the delay time in the data acquisition was set to 350 ms (the time for air traveling through the sampling line).

We agree with the reviewer that the best approach to determine the delay time is using a dosing experiment and a fast sensor. On the other hand, because the flow in the sampling line was most likely turbulent, we do not expect a large difference between the calculated and actual delay times. Meanwhile the durations when air motion stayed in updraft or downdraft were normally more than a few seconds. Even if there is a small discrepancy between the calculated and actual delay times, there was little possibility for this discrepancy to make significant errors in the HONO flux measurement.

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We have developed a new sampler for HONO flux, in which we got rid of the sampling line. The new sampler is currently being deployed in a field study and is directly placed next to a sonic anemometer. In this case, no delay time is needed (delay time = 0) because the air motions are sampled instantaneously.

Comment (6): 4115 L15: Testing for artifacts related to retention or destruction of HONO on the sampling line is good, but were zero tests done through the end of the inlet to determine if other effects, like photolysis of particle nitrate on the tubing, were an issue? Were there any issues/interferences under high NOx conditions?

Response: The tests with a dry denuder placing at the inlet of the sampler essentially determined the effects like the photolysis of particle nitrate on the tubing. We noticed that interferences were slightly high under high NOx conditions, but the small amount of artifacts were cancelled out because only the difference of HONO concentration in updraft and downdraft samples was used for the HONO flux calculation.

Comment (7): 4118 L13: The detection limit is given here for the REA-HONO system in units different than described previously for the analyzer. Given that there is a total uncertainty for a single analyzer of about 15% and that 56% of air sampled was zero air, can the authors show that their system has sufficient PRECISION to calculate fluxes? I think this needs to be explicitly included in the manuscript since the flux is essentially the difference of two numbers.

Response: It is rather conservative for the estimated uncertainty ($\pm$15%, 2σ), which included the uncertainties in the aqueous nitrite calibration standards, the sampling flow rate, the liquid flow rate through the coil to scrub HONO, the conversion efficiency of nitrite to azo dye, and the drift of absorbance of the spectrometer. Because the HONO difference in the updraft and downdraft is used to calculated, some of the uncertainties are cancelled out (e.g., the calibration standards and the conversion efficiency of nitrite to azo dye, and the drift of absorbance of the spectrometer). So the actual accuracy of HONO flux measurement is most likely much better than $\pm$15% (more likely $\pm$5-10%

at a 2σ confidence level).

We have added a few sentences at the end of this paragraph as: "This uncertainty was estimated from the combined uncertainties in the nitrite standards (±5%), the sampling flow rate (±6%), the liquid flow rate through the coil to scrub HONO (±6%), the conversion efficiency of nitrite to azo dye (±10%), and the drift of absorbance of the spectrometer (±5%). Because the HONO difference in the updraft and downdraft is used to calculate HONO flux, some of the uncertainties are cancelled out (e.g., the calibration standards, the conversion efficiency of nitrite to azo dye, and the drift of absorbance of the spectrometer). So the actual accuracy of HONO flux measurement is more likely within ±5-10% at a 2σ confidence level."

Comment (8): Is Fig 4 from CALNEX or BEARPEX? I think CALNEX but it's not mentioned in caption. Why include both graphs in this figure? It seems that the one on the right is sufficient.

Response: This figure is for CalNex 2010. We have changed the caption as "...Data are shown for CalNex 2010 for hours between 10:00 and 18:00 ..." to reflect this addition. We believe that the plot on the left is also valuable to show diurnal variations of the two parameters and would like to retain it as it is.

Comment (9): 4109 L23: "digital"

Response: Corrected.

Comment (10): 4109 L27: How was delay time of 350 ms determined? Maybe mention it will be discussed later

Response: As suggested, we have added "...for about 350 ms (discussed in Section 3.2)..."

Comment (11): 4113 L8: Not sure what "When a non-zero threshold issued" means.

Response: To clarify this, we have modified the sentence as: "When a non-zero thresh-

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old issued (e.g., 0.5σw in this study) to determine updraft and downdraft air motions, the proportionality coefficient is. . . ”.

Comment (12): 4119 L12: “environment”

Response: Corrected.

Interactive comment on Atmos. Meas. Tech. Discuss., 4, 4105, 2011.