

Interactive comment on “Synthetic ozone deposition and stomatal uptake at flux tower sites” by Jason A. Ducker et al.

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Clifton's comment asks for two things: (1) to clarify our calculation of stomatal and non-stomatal fractions of O₃ deposition at Harvard Forest and (2) to provide detailed accounting of the differences in methods and results between this work and that of Clifton et al. (2017). We address both items below and in changes to the manuscript.

While preparing this response, we found an error in the supplemental SynFlux csv file provided with our paper. The files did not include the recalibration of Harvard Forest water vapor flux. No results or figures in the paper were affected, but the gs and other values in that csv file were inconsistent with Figure 5, as Clifton's comment suggested. We have attached corrected csv files to this comment and will ensure that the final pub-

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lished paper contains the correct supplemental files. We apologize for this confusion, but very much appreciate that Clifton identified this issue.

In addition, we found that the Harvard Forest data archive (<http://harvardforest.fas.harvard.edu:8080/exist/apps/datasets/showData.html?id=hf004>), which provides the measured O₃ mole fraction and flux measurements used in our work, contained some bad O₃ flux values because a data filtering criterion was not applied to the posted data. Filtering out those values caused small changes (~0.05 cm/s) in the mean deposition velocity of some years. One of us (Munger) is updating the web archive. We will use the updated values in our revisions and in the response below. This change modestly affects the deposition velocity, canopy conductance, and non-stomatal fraction of deposition derived from O₃ fluxes at Harvard Forest, so we will update all affected figures.

"Ducker et al. (2018) find that their estimates of stomatal conductance suggest that the nonstomatal fraction of ozone dry deposition ranges from 4 to 32% across years at Harvard Forest and note that this is different from what Clifton et al. (2017) find (20 to 58%) (see Ducker et al. (2018) lines 318-323). Stomatal conductance estimates are critical for inferring the nonstomatal fraction of deposition as the nonstomatal conductance is calculated as a residual of the canopy conductance (inferred from ozone eddy covariance fluxes) and the stomatal conductance. Ducker et al. (2018) attribute the difference between our estimates of the year-to-year range in the nonstomatal fraction of deposition to their re-calibration of the water vapor fluxes used in the stomatal conductance estimate. First, I ask them to clarify whether their range of the nonstomatal fraction is indeed comparable with mine. Second, I use the authors' stomatal conductance estimates (given in their supplementary material) to show below that their estimates of the nonstomatal fraction of deposition are similar to those given by Clifton et al. (2017). I recommend that the authors clarify their discussion of my previous work based on this finding."

Clifton elaborates on these points below, so we respond there.

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"First, it is unclear how Ducker et al. (2018) arrive at their reported 4-32% range. When I examine their Figure 5, the figure referenced for these numbers, I can infer the 4-32% range from the error bars, which the caption says represent one standard deviation across daily estimates. If the 4-32% range represents the standard deviation across daily estimates, then these numbers are not directly comparable to mine and the text should be revised accordingly. The 20-58% given in my paper represents the range in the summertime daytime mean nonstomatal conductance across yearly values, not the spread across daily values. If Ducker et al. (2018) actually calculate the mean non-stomatal conductance for each year to obtain their range of 4-32% to compare directly with Clifton et al. (2017), then their approach needs to be more clearly documented in the manuscript."

As suggested, the 4-32% range was not calculated in the same way as Clifton et al. (2017) calculated the interannual variability in non-stomatal deposition, so the two should not be directly compared. We have redone the calculation in the same way as Clifton et al. (2017). This changed our central estimate (from 15% to 9% non-stomatal) and the interannual range. We will replace the text as follows and add supplementary text that provides additional details on the comparison of our results with Clifton et al. (2017). The proposed supplement is provided at the end of our response. The paragraph in Sect. 3.2 (line 318) will become... "A recent analysis of O₃ flux measurements at Harvard Forest suggests that non-stomatal deposition averages 40% of daytime O₃ deposition during summer months, with a range of 20-60% across years (Clifton et al., 2017). Our analysis of the same site does not support such a large role for non-stomatal deposition at this site in summer. For each year, we calculate summer daytime means of *g_s* and *g_c* by averaging the June-September values, then calculate the non-stomatal fraction of deposition (1- *g_s*/*g_c*). Averaged across years 1993-2000, we find that 8% of daytime O₃ deposition is non-stomatal during the summer, with a range of -33% to 34% across years. Negative fractions mean that stomatal conductance is large enough to explain all O₃ deposition. A large negative non-stomatal fraction (-33%) occurs in only one year (1996) and no other year is less than -11%, which is within

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uncertainty of 0% (2 std.) according to the error propagation. Despite the small or zero non-stomatal fraction found here, our results continue to support the large year-to-year variability of this fraction reported by Clifton et al. (2017). The re-calibrated latent heat flux measurements are the main reason that our results differ from prior work and Supplement S1 provides explanation. At Hyytiälä Forest. . [continued as previously]"

We have also revised Figure 5 (see Fig. 1 below) to also show the interannual mean and standard deviation for each month of the year.

"To investigate whether the re-calibration of water vapor fluxes leads to the differences in the fractions of stomatal (or nonstomatal) deposition in Ducker et al. (2018) vs. Clifton et al. (2017), I downloaded the authors' monthly mean stomatal conductance estimates given in their supplementary material. I divided their summertime (June-September) daytime mean stomatal conductance estimates at Harvard by my own estimates of canopy conductance (9am-4pm June-September for each year). My methods for inferring canopy conductance from the ozone eddy covariance flux measurements at Harvard Forest are described in Clifton et al. (2017), and are similar to those of Ducker et al. (2018). Inferring the canopy conductance depends on estimates of the resistances to turbulence and molecular diffusion. These resistances are typically relatively small during the summer daytime compared to the total resistance to deposition calculated from the ozone eddy covariance fluxes, so there should not be substantial differences between our estimates of canopy conductance. Dividing their stomatal conductance estimate by my estimate of canopy conductance suggests that summertime mean stomatal deposition varies from 50 to 100% of the total deposition during 1993- 2000. This corresponds to nonstomatal deposition varying from 0 to 50% of the total ozone dry deposition from year to year (see Figure 1 below). This is wider than the range presented by Ducker et al. (2018) (i.e., 4-32%). Given the uncertainties in the measurements and differences in our approaches especially with respect to inferring stomatal conductance, I think it is fair to say that this range is similar to the range in Clifton et al. (2017) of 20-58%. My analysis here suggests that re-calibrated

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water vapor fluxes are not the root cause of the major differences in the ranges given by Ducker et al. (2018) vs. Clifton et al. (2017). Rather, it seems more likely that the differences reflect consideration of the spread in daily variability (Ducker et al., 2018) rather than the year-to-year range (Clifton et al., 2017)."

As we said above, the supplemental files included with our paper did not include the water vapor flux correction at Harvard Forest. As the commenter found, with the uncorrected water vapor fluxes, we estimate roughly the same non-stomatal fraction of O₃ deposition as Clifton et al. (2017). However, our best estimate of stomatal conductance at Harvard Forest, using the corrected vapor fluxes, is larger than the numbers used by the commenter, so the stomatal fraction of O₃ deposition is larger as well. Again, we apologize for the confusion.

In addition, our monthly and summer averages of g_c are generally smaller than those of Clifton et al. (2017). Our hour-by-hour values of g_c should be similar, for the reasons given by the commenter, but our monthly averages weight each hourly value by its uncertainty. Large values or outliers of g_c tend to have large uncertainty. That is because $g_c = (v_d^{-1} - (r_a + r_b))^{-1}$. When v_d^{-1} and $r_a + r_b$ have similar magnitudes, these conditions cause loss of significant figures in subtraction (sometimes called catastrophic cancellation) meaning that g_c is very sensitive to measurement errors, which can produce spuriously large g_c . Error propagation diagnoses this growth of uncertainty and those values are weighted less in the monthly averages. As a result, our g_c values are somewhat smaller (0.4-0.7 cm s⁻¹ for summer averages 1993-2000) and have less interannual variability than Clifton et al. (2017; 0.5-1.2 cm s⁻¹). The lesser variability with error-weighted averages suggests that some apparent interannual variability may be a spurious result of outliers caused by random measurement error. Our weighted g_c is similar to the median g_c for each month, both of which have similar interannual variability that is less than the unweighted mean g_c . Moreover, the unweighted averages are sensitive to how aggressively outliers are discarded, while error-weighted averages are essentially unchanged even if no outliers are excluded.

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We will briefly explain the effect of error weighting in Section 2.4 (line 249) by adding, "The error-weighted averages tend to be smaller and less variable than unweighted averages because the error propagation identifies when outliers and large values have greater uncertainty. For example, the monthly values of g_c derived from observations at Harvard Forest are 0.57 ± 0.11 cm s⁻¹ with error weighting and 0.68 ± 0.17 cm s⁻¹ without."

The following paragraph, which will be added to the supplement, addresses the remaining questions about comparing our results with Clifton et al. (2017). – "Our estimate of the non-stomatal fraction of O₃ deposition at Harvard Forest (8%, range: -33 to 34%; Sect. 3.2) is smaller than was previously reported at that site (40%, range 20-60%; Clifton et al., 2017). The main reason for the different results is the re-calibration of the water vapor fluxes in this work, which is described in Sect. 2.2. Here, we show how other differences between our analysis and that of Clifton et al. (2017) affect the estimate of non-stomatal fraction of O₃ deposition at Harvard Forest. Using our gap-filled data, the annual range of the non-stomatal fraction of O₃ deposition at Harvard Forest slightly increases while the mean estimate remains the same (8%, range: -36 to 38%). With uncorrected water vapor fluxes, our estimate would be 51% (range: 32% to 63%). If we also ignore the propagated uncertainty, which varies from hour to hour, and calculate averages with equal weight (i.e. equal uncertainty) for each time interval, as Clifton et al. did, then we would estimate 53% (range: 34% to 66%). If we also use data filtering criteria from Clifton et al. (i.e. remove 3 outliers of v_d and g_s , but no filtering for precipitation and high relative humidity), then we would estimate 48% (range: 28% to 61%). Finally, if we also restrict our averages to 9am-3pm, as Clifton et al. did, instead of all daylight data, then we would estimate 45% (range: 25% to 60%). This final estimate is very close to the method and value reported by Clifton et al. (2017). The remaining small differences are probably due to Clifton et al. including 1992 in their analysis and differences in the form of the Penman-Monteith and stability functions. Since the re-calibration of water vapor fluxes (Sect. 2.2) is an improvement in this work and the main reason for our results differing from Clifton et al. (2017), our

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estimates of small non-stomatal fraction O₃ deposition at Harvard Forest appear to be most reliable estimate for this site.

References

Clifton, O. E., Fiore, A. M., Munger, J. W., Malyshev, S., Horowitz, L. W., Shevliakova, E., Paulot, F., Murray, L. T. and Griffin, K. L.: Interannual variability in ozone removal by a temperate deciduous forest, *Geophys. Res. Lett.*, 44(1), 542–552, doi:10.1002/2016GL070923, 2017.

Figure 1. Observed O₃ deposition velocity and its in-canopy components at sites with O₃ flux measurements. Lines show the multi-year mean and multi-year standard deviation calculated from the monthly averages described in Sect. 2.4. Dashed lines on the stomatal conductance panel show the stomatal fraction of total canopy conductance ($g_s g_c^{-1}$) and dashed lines on the non-stomatal conductance panel show the parameterized g_{ns} value.

Please also note the supplement to this comment:

<https://www.biogeosciences-discuss.net/bg-2018-172/bg-2018-172-AC1-supplement.zip>

Interactive comment on Biogeosciences Discuss., <https://doi.org/10.5194/bg-2018-172>, 2018.

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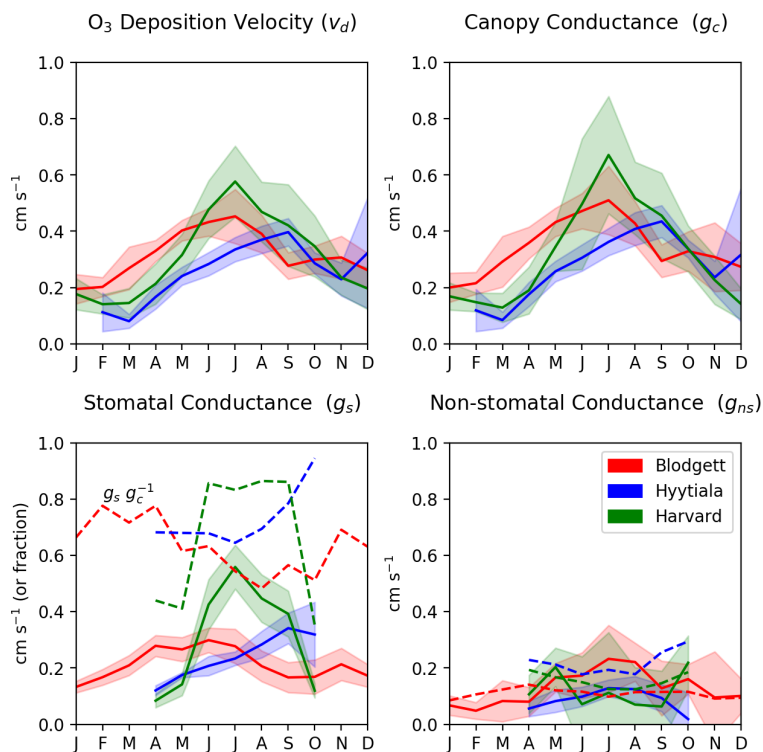


Fig. 1. Revised version of Figure #5

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