Reviewer 1 comments and response

The manuscript by Walker et al. presents results from a study investigating atmospheric deposition of reactive nitrogen to a deciduous forest at the USDA Forest Service Coweeta Hydrologic Laboratory in the southern Appalachian Mountains. The authors use several well-established measurement methods to differentiate between oxidized and reduced as well as organic and inorganic compounds found in wet and dry deposition. Finally, they apply a bi-directional resistance-based model driven with the observed measurements of Nr air concentrations, micrometeorology, canopy structure, and biogeochemical parameters to present the full reactive nitrogen budget for the site.

While the character of the paper is a report-style compilation of results from a multitude of methods rather than following a clear scientific question, the authors do a great job in thoroughly describing the complexity of reactive nitrogen field investigations and long-term observation. Though continuous eddy-covariance observations are not included, the study represents the state-of-the-art in Nr monitoring and data interpretation. I particularly appreciate the inclusion of field investigations of the ammonia emission potential of green and senescent leaves as well as from litter, which is crucial for model parameterization and rarely conducted. The results are put into a broader context and discussed with regard to air quality regulations in the past, e.g. reduction in oxidized N is now clearly visible. Method uncertainties are sufficiently considered and presented.

The text is very well written and easy to follow. Figures are clear and easy to grasp. The supplemental material is useful and the selection of graphs and tables that were put into this section is good. This is the most comprehensive single-site study I am aware of and definitely deserves publication.

I only have a few, rather minor, points that should be considered before final presentation in the BG journal:

Response: We sincerely thank the reviewer for their comments and questions. We have addressed each in detail below.

Comment: With regard to Section 2.2.7, how exactly were the NH3 data from hourly measurements used to impose diurnal variability on the biweekly data to be used as hourly input for the model? It is stated in line 393 that continuous NH3 concentrations were only measured during the last two intensives (in spring and summer, I guess?). The diurnal variability is known to be driven by temperature, humidity, light availability, phenology, etc., how was the amplitude of the variability from these two campaigns transferred to the other – probably much cooler – seasons?

Response: The reviewer is correct that continuous hourly measurements were only conducted during the last two intensives. To account for diurnal variability in the NH_3 air concentration, the diel concentration pattern determined during the spring and summer intensives (Supplemental Figure S7) was imposed on the bi-weekly AMoN NH_3 concentration. First, the hourly profile of NH_3 concentrations was normalized by the corresponding overall mean

concentration to produce a normalized mean diel concentration profile. This profile was then applied to each biweekly AMoN air concentration, temporally scaling the NH_3 concentration by time of day while maintaining the measured biweekly AMoN concentration. In this way, the hourly time series derived for the entire year from AMoN measurements displays the diel variability observed during the spring and summer.

The reviewer is correct, the amplitude of the diel concentration profile would be expected to change throughout the year in response to a number of factors, e.g., temperature, local NH_3 emissions, boundary layer dynamics, biogeochemistry. Our approach does not incorporate the seasonality that may be observed in the diel profile for winter and fall. However, while the amplitude of the diel cycle may differ from our observations during these seasons, the seasonality of the air concentration on a longer averaging period (i.e., two weeks) is reflected in the AMoN concentrations. An alternative approach that could be employed in the future would be to derive diel profiles from a chemical transport model at shorter than seasonal time scales, perhaps monthly, to scale the AMoN measurement to the hourly time scale for flux modeling.

Comment: The method section is very informative, but quite long. I'm wondering whether it would make sense to put all detailed descriptions from 2.2.1 up to 2.2.5 into the supplement, just adding a few sentences to 2.2 what has been done and referring to the respective part in the Supplemental Material. It's not a must, but would significantly reduce length and better highlight the findings given the potential readership of people who work in conservation and are likely more interested in the results and their interpretation than in every technical detail of the methodology.

Response: We appreciate the reviewer's comment and admittedly it was difficult to decide how much detail on methods to include in the primary text as opposed to supplemental. Ultimately, we felt that the methods themselves, and particularly the combination thereof to assess the deposition budget, were worth describing in the main text along with the results.

Other:

Comment: Introduction: I suggest adding information on measurement period, length, etc.

Response: This information will be added.

Comment: Line 41-42: "many areas" and "some regions", please specify where, e.g. near hotspots of animal husbandry, chemical industry, etc.

Response: Additional detail will be added.

Comment: Line 153: Is 8 m the correct height? What was the reason for this height?

Response: Yes, 8 m is correct. This was the height of a permanent tower immediately adjacent to the shelter housing the TD-PC-CL instrument. The tower was used opportunistically and 8 m happened to be the maximum height. Reviewer 2 also asked about the choice of measurement heights and corresponding treatment of measurements at different heights for flux modeling. As

discussed in the response to reviewer 2, we made no correction for differing measurement heights and will clarify that point and limitation in the revised text.

Comment: Line 170: What was the selection criteria for the two respective heights?

Response: In this case, the heights were chosen to maximize the separation between sampling boxes to maximize the concentration gradient. 34 m is just above the top of the canopy and 37.5 m and 43.5 m were the total height of the tower in 2015 and 2016, respectively. Ultimately, the MARGA data were not used for gradient flux calculations because colocation experiments to remove systematic bias between the sample boxes were not successful. As described in the primary text, gradient fluxes were instead calculating using measurements from the time-integrated denuder measurements.

Comment: Line 178: "to the analytical box for analysis Ion Chromatography (IC)", is there a word missing?

Response: Yes, this will be corrected.

Comment: Line 270: Check for consistency in unit notation: "g^{-1} tissue" vs. "kg $tissue^{-1}$"

Response: Thank you for pointing out this inconsistency. We will make the correction here and check throughout.

Comment: Line 312: Delete "is R_a" after "z_0"

Response: OK

Comment: Line 449: Is RH defined before?

Response: RH will be defined at line 236 in the revised text.

Comment: Line 505-506: Do two decimal places reflect the measurement accuracy?

Response: Two decimal places are appropriate for the corresponding detection limits (0.018 – 0.038 ppb).

Comment: Line 605-606: 61.4% wet plus 38.7% dry deposition equals 100.1%, check rounding.

Response: Thank you for point this out. Rounding will be checked here and throughout.

Comment: Line 617: Can a bit more explanation given why stomatal fluxes are so low compared to cuticular fluxes?

Response: We will add more detail to the explanation of the low stomatal fluxes compared to the cuticular flux. First, the stomatal resistance is generally larger than the cuticular resistance even during the summer when the stomatal resistance reaches a minimum. Second, the gradients

that drive the leaf-level stomatal (F_s) and cuticular (F_{cut}) fluxes are defined as X_s-X_l and X_{cut}-X_l, respectively. X_s and X_{cut} are the stomatal and cuticular compensation points, respectively, and X_l is the concentration of NH_3 above the leaf. In the current model formulation X_{cut} is zero and X_s is non-zero and a function of the stomatal emission potential and temperature. Thus, the concentration gradient is always larger for the cuticular versus the stomatal pathway. Together the larger stomatal resistance and smaller concentration gradient (X_s-X_l) result in $F_s < F_{cut}$.

Comment: Line 708: Why would the aerodynamic resistance become zero at steep forested slopes? R_a is turbulence and wind speed driven, so why would it approach zero?

Response: Over flat homogeneous terrain, vertical exchange between the atmosphere and vegetation is driven by vertical turbulent diffusion. In the traditional resistance analogy, the resistance to turbulent transfer is referred to as the aerodynamic resistance (R_a). Hicks (2008) describes an extreme example in which horizontal flow approaching a steep slope penetrates the canopy. In this example, the transfer of material (deposition) to the canopy elements becomes dominated by horizontal advection and filtration rather than vertical diffusion. Thus, this situation can be described as analogous to R_a tending to zero, i.e., no aerodynamic resistance to transfer.

References

Hicks, B.B., 2008. On estimating dry deposition rates in complex terrain. Journal of Applied Meteorology and Climatology, 47, 1651 – 1658.