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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A model-based analysis of foliar NO_x deposition" *by* Erin R. Delaria and Ronald C. Cohen

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The authors present a modeling study of NOx deposition to forest foliage using a 1-D column model consisting of 8 vertically-stacked boxes extending to the top of the PBL. Each box uses the continuity equation incorporating all production and loss processes occurring within and above a forest canopy. They apply this model at two contrasting forest ecosystems in the USA: a high-altitude Ponderosa pine forest in California and a northern deciduous mixed forest in Michigan from which they have a comprehensive suite of measurements from intensive field campaigns. They consider two commonly-used parameterisations of the stomatal deposition of ozone to model NOx deposition. They conduct a number of sensitivity studies to explore the processes that have the largest effect on modeled deposition velocities and quantify the subsequent impact on

fluxes and budgets of NOx, O3 and other key atmospheric gas-phase species within and above the canopy.

Loss rates of reactive gas-phase species, such as NOx, through deposition is a question of critical importance to our ability to model the complex interactions between NOx-HOx-Ox in the lower troposphere. While much effort has been put into improving parameterisations of O3 dry deposition, and particularly stomatal fluxes of O3, very little attention has been given to other gases. Yet the nitrogen cycle is fundamental to ecosystem functioning and health, the capacity of the terrestrial carbon sink and atmospheric composition (air quality and climate).

This study is highly timely and in my opinion excitingly novel and important. I commend the authors for their thorough and insightful approach to this global issue.

I have a couple of general concerns and a few specific comments that the authors should address prior to publication.

General:

1. While I appreciate that NO-NO2 cycling is rapid, non-linear and highly complex, I would like to authors to be explicit in precisely which molecule they are considering the dry deposition of. They rather inconsistently refer to deposition of NO2 and of NOx. Presumably they are assuming that NO deposition is negligible and hence deposition of NO2 can be used as a proxy of NOx deposition. If so, this should be explicitly stated early in the manuscript and a single term used from that point on.

2. The study purports to use two field sites, Blodgett Forest (BEARPEX campaign 2009) and University of Michigan Biological Station (UMBS campaign 2012). However, the authors almost exclusively focus their model description, parameterizations, sensitivity tests, results and discussions on Blodgett with scant details given to the results from UMBS other than to corroborate (or highlight differences) those from Blodgett. The authors should either reduce their analysis to a single site or, preferably, give simi-

lar attention to UMBS. The differences between model outcomes for the two sites is, to my mind, of real importance to enable the modeling and measurements communities to understand the processes that require further elucidation.

3. While the authors explicitly quantify the differences in NOx concentrations and fluxes between the two deposition schemes and between the perturbed parameter sensitivity tests, they do not similarly evaluate the relative performances against observations, relying instead on qualitative, descriptive differences. The results would be far stronger if this aspect of the model outcome were better explored and presented.

Specific:

Introduction

Throughout: All of the deposition models and studies presented here are specifically focused on the dry deposition of O3. The authors need to build a stronger argument that NO2 deposition should be assumed to follow the same process. In particular, in the case of O3, there still remain questions around the relative contributions of stomatal vs cuticular fluxes to the total leaf conductance. Most O3 deposition calculations assume that mesophyllic conductance is zero, is there evidence that this is the case for NO2.

p2, L19 (and elsewhere): VPD is a convenient proxy for leaf water potential as it can be calculated from routinely measured meteorological variables but it is often not a good metric to use under conditions of drought.

p2, L20: make clear that “season” and “seasonality” refers to plant phenology

p3, L4-5 (and elsewhere): Technically, the DO3SE model estimates stomatal conductance for use in deposition schemes to calculate deposition velocities and hence O3 fluxes.

p3, L12: Could the authors explicitly state some of these “other molecules”

p3, L15-17: YES!!! This should be emphasised!

2 Model description

p3, L21: A value of 100m for the PBL height during the peak growth season (summer) seems low, particularly for Blodgett. Under clear skies and high insolation I would expect to see values of 1500-2000m. Is their value based on observations at the two sites? If so, please provide references; if not please justify.

p3, 21: “Gaussian”

p3, L30: Δh is surely the height / depth of the box. I assume that the model has a horizontal scale of 1m2 or 1cm2, but please clarify this.

p4, L1-19: This paragraph (which should really be split in two for BEARPEX and UMBS) is not a description of the model, rather the two field sites and should have a separate section.

p4, L7: I am surprised that UMBS was modelled here without a separate understory, see e.g. Bryan et al (2015) Atmos Environ.

p4, L20-21: Make clear here that this is simply following Beer’s Law.

p4, L30: What are tau and TL in this context?

p4, L30 (and Table 1): Where is the value of u* taken from and why is it a constant value?

p5, L15: Please explain why the rate constants require adjustable parameters to make them site-specific. Are the authors assuming segregation? recycling?

p5, L21: Where are the basal emission rates taken from? Are they average values for deciduous and evergreen mid-latitude forests, site-specific, dominant-species specific?

p5, L22: Deposition should be described in a separate section. In fact, given it is the main focus of the study, it should be the first.

p5, L26-p6, L5: This is the Baldocchi parameterisation of total resistance. Why have

the authors not used the subsequent Gao et al (1993) update?

p6, L5-7: If all processes are correctly included and paratemerized there should be no need to use a compensation point; this is merely a formulation that is used when the production and loss terms are not fully represented in a model.

p6, L13: The authors have not defined SR

p6, L14: Eqn 12 is essentially the Jarvis (1976) parameterisation of stomatal conductance. It has been modified since, with additional adjustment factors. It forms the basis of the DO3SE model, but really the DO3SE model is about the damage and therefore incorporates an additional modifying factor fo3 to the Jarvis expression for gs.

p7, L6 & L8: VOC or BVOC?

p7, L16: Please expand on how fluxes are calculated within this model.

p7, L17: How is the PAN formation / NOx removal incorporated? It is not clear if or how these processes are included in the authors' considerations of chemical production and loss, lifetime calculations and OPE.

3 Sensitivity to parameterizations:

As previously noted, this section appears only to consider Blodgett Forest (unless all parameters were the same at both sites, which other parts of the manuscript suggest was not the case)

p7, L22-23: How were these values of total deposition velocity chosen?

p8, L10: Why have the authors chosen a value of 2 for tau/TL; Wolfe and Thornton (2011) used a value of 4 for this site when developing the CAFE model.

p8, L10: "resulting in a canopy residence time of 152s" at both sites? Or just Blodgett?

p8, L22: Please explain why Rb and specifically lw has a larger impact on species with high rates of leaf deposition.

p9, L14: I realise this is taken from a previous study but it is not clear why UMBS should be modeled using parameters for a European beech species when it is dominated by aspen.

p9, L19-p10, L4: Please quantify the model-obs fit rather than providing simply a qualitative overview.

p9, L25: Please explicitly state what is meant by NOx enhancement. I think it is the difference between in-canopy and above canopy concentrations. But these will differ between levels in the canopy and PBL

p10, L6: Wesely

p10, L26-28: How do these deposition velocities compare with observations? In L10, the authors state that values of 1.4, 0.77 and 1 are used in global models. Do the author have site-specific measurements on which they have based their choice of 0.3 and 1.4 as upper and lower bounds?

p11, L1: The authors are comparing 2 sites with a range of differences so I'm not sure they can claim "regional" differences. Surely it's more to do with different forest types, different soils, different meteorology, . . . Please could the authors be a little more specific.

P11, L2: I have a problem with the use of "wet" and "dry" in this context as deposition itself is referred to as wet or dry. Perhaps the authors can find an alternative way to describe wet and dry environments (I couldn't think of an obvious alternative I'm afraid)

p11, L4-7: Do these values of SWP and RH match long-term observations?

p11, L20-25: It would be good to see a more considered discussion of the results and the reasons (i.e the processes) behind the similarities and differences between the sites.

5 Discussion

p12, L6-7: Suggest the authors extend their view beyond the USA. Surely their findings are GLOBALLY applicable?

p12, L9: CLM includes a specific parameterization of stomatal conductance and is the land surface model for both regional and global models of chemistry-climate (see Lombardozzi et al, various). Models with a full land surface module already calculate stomatal conductance and plant physiology so have no need to incorporate either the Wesely or Emberson approaches for estimating gs.

p12, L21-22: Following on from the above point, this point about the relative simplicity of the Emberson approach should be made explicitly clear from the outset by the authors.

p13, L2: How is OPE defined? As molecule of O3 produced per "molecule" of NOx lost?

p13, L8: PBL

p13, L20-p14, L10: Parameterized for BEARPEX again?

p13, L26: Is PBL height fixed?

p14, L1 and L3: The plots of observed NOx concentrations for both sites suggest they are $\sim\leq$1ppb so why have the authors explored up to 100 ppb here?

6 Conclusions

p14, L25: missing closing parenthesis.

p14, L30-31: It's also imperative to accurately measure gs in a way that reflects differences between leaf-level and canopy-scale gs.

p14, L31-32: DO3SE is NOT a deposition model; it is a model of stomatal conductance that can be used in a deposition scheme so effectively it also uses the resistance in series approach.

p15, L14: Why is this important? What does this miss? Do we know that is wrong?

p15, L8: think GLOBAL!

p15, L8-9: Please could the authors be more specific in their recommendations? Precisely what do they mean by explore? More measurements? More modeling? And specifically of what, when and where?

Figures and Tables

p24, Fig 1: I am surprised that the authors have chosen only to vary PBL for the top two layers in the active mixed layer. I would expect the lower 2 of these layers to similarly evolve over the course of the day but with lower amplitude.

p24, Fig 1: Right-hand labels on plot say "remnant" and caption says "residual". Would personally use the latter.

p25, Fig 2: Not sure that this figure (or Fig S9) add anything to the paper. The authors have given the lat-long coordinates for both sites so readers could look for them on a map and they do not refer more than in passing to the figure from the text.

p26, Fig 3(d): Clarify what is meant by NOx enhancement in this context.

p27, Fig 4: A panel showing a time series of NOx would be helpful for direct comparison between the two sites.

p32, Fig 9: PAN? Daylight hours or 24-hour average?

Overall, I feel this is work of global significance that I hope will start to focus the attention of the atmospheric chemistry community on the importance of deposition in general and the fate of reactive nitrogen species in particular.

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