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Interactive comment on "Simulating ozone dry deposition at a boreal forest with a multi-layer canopy deposition model" *by* Putian Zhou et al.

Anonymous Referee #1

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This paper presents an analysis of ozone deposition to a Scots pine forest based on a 1D multi-layer chemical transport model. The study includes an implementation of a resistance scheme for ozone deposition into an existing model, simulations for a one-month period and comparisons with measurement data. The results highlight the importance of non-stomatal sinks, both within the forest canopy and at the soil interface, and the role of surface wetness in mediating these fluxes. The topic of the study is highly relevant, as it becoming increasingly evident that ozone deposition to vegetation is controlled by interplay of processes that are difficult to describe by traditional 'big-leaf' models. In addition, the high-quality long-term data available provide an improved opportunity for development of more advanced surface exchange models.

The paper is fundamentally sound, although there are some issues that need further consideration. The measurement data from the SMEAR II site employed in the study

are excellent, and the SOSAA model serves as an advanced platform for studying surface exchange processes. For the most part, the paper is clearly written and based on sufficient analysis. I would recommend publishing the paper after a major revision that pays adequate attention to the following comments.

Major comments

The model description is incomplete:

(1) The authors state that they have implemented a multi-layer dry deposition model into SOSAA, which is a 1D chemical transport model. SOSAA is described in Section 2.3.1, which lists different modules and references but does not explain the model types or physical principles. The key elements of SOSAA relevant to the present study, especially turbulent mixing and the derivation of eddy diffusivity, should be described in more detail.

(2) Due to the incomplete model description, it is not obvious for the reader that the vertical mixing of O3 is calculated similarly to that of any other compound in SOSAA (Eqs. 5 and 6), and that the "multi-layer O3 deposition model" actually consists of the few resistance terms shown in Fig. 1 (of which not all are effective). As SOSAA has previously been used for simulating the exchange of reactive compounds and latent heat within a forest canopy, obviously it must include some sort of description of the surface exchange processes corresponding to stomatal uptake at least. This relationship should be explained, especially for the stomatal resistance of both overstory and understory vegetation.

(3) The presentation of input data should be clearer and specify the data actually used for the deposition calculations, i.e. which data are taken from measurements and what is derived within SOSAA. This is important, as a large part of the paper is dedicated to testing the modelled meteorological variables. For example, a comparison with observations is presented for u*, but it is not explained how the modelled profile is obtained or how it is utilised in the model.

(4) The description of O3 parameterisation (Section 2.3.2) presents parameter values with no justification or citation, or refers to model results that are not shown:

(4.1) r_soil is modified from a default value based on model simulations, which are not discussed. These simulations should be shown and would serve as a useful sensitivity test.

(4.2) r_ac is set to a very small arbitrary value. What is the point of including a resistance of 1 s m-1 in series with a resistance of 600 s m-1?

(4.3) r_b depends on molecular diffusivity and wind speed (or friction velocity, p.9). Please present the formula or an exact reference.

(4.4) r_stm is calculated from evapotranspiration rate in SOSAA. How?

(4.5) r_mes, r_cut and r_ws have constant values. Where do these come from?

(4.6) f_wet is a function of RH, for which the authors cite a grey literature report that does not even include original data on canopy wetness. Isn't there anything more substantial available?

There are some issues with the resistance scheme:

(5) An ineffective aerodynamic resistance term (r_ac) is included in series with the soil resistance (cf. Comment 4.2 above). However, a much more important term, namely the near-soil boundary layer resistance, is ignored in the model.

(6) The leaf surface resistance has a general formulation (Eq. 3) so as to represent both needle-shaped and broad leaves. This is accomplished by a scaling factor (alpha = 0.5) introduced to account for one-sided stomatal exchange on leaves. As the non-stomatal exchange takes place on both sides of such a leaf, with separate boundary layers, this scaling does not result in a correct formula for deposition on two-sided leaves. The authors describe alpha as a correction factor, so it may represent an approximation. However, this approximation should be justified.

Analysis of results could be improved:

(7) The authors demonstrate that the model performs well in high humidity conditions but fails during the night-time when RH is low. As wet needle surfaces require additional parameterisations (RH dependent resistance, wet surface fraction), it is surprising that the authors do no first try to develop a parameterisation that performs well in dry conditions, i.e. in a much simpler case. Instead, they refer to simulation and measurement problems due to weak turbulence, but do not explain how these would depend on RH. If the measurement uncertainties increase with weakening turbulence and affect the model validation (and the u* screening does not help), then this could be easily tested. As the soil resistance is given as a plausible explanation for the mismatch, it would also seem logical to test if a better fit can be obtained by varying this resistance (see Comment 4.1. above).

(8) Only ozone fluxes are considered in the analysis. As the modelled flux depends on the modelled concentration, which is affected by various processes and has a systematic diurnal cycle, it is difficult to assess how well the deposition processes are modelled by comparing fluxes alone. Perhaps you could have a look at the flux/concentration ratio (commonly called deposition velocity)?

(9) As the ozone fluxes measured at SMEAR II have been analysed in a large number of previous studies, including two different multi-layer models (Rannik et al., 2012; Launiainen et al., 2013), I would expect to see a more systematic comparison of these results. From the process modelling point of view, it would be useful to discuss how and why the modelled results differ between the three multi-layer models.

(10) The discussion of chemical removal (Sect. 3.7) is based on the reactivity estimates obtained from the literature. According to the model description, the SOSAA model employed here includes a detailed chemistry module, which I assume was used in the present simulations. Why are these calculations not utilised for estimating the importance of in-canopy chemistry?

The language should be improved:

(11) Even though I indicated in my access review that a linguistic revision is necessary, there are still numerous errors, some of which impair presentation. A few examples are given in the detailed comments below.

Minor comments

P1/L23: "under current knowledge of air chemistry" is obvious so can be removed.

P2/L24: Any more recent studies?

P2/L31-: A reference is needed for "the boreal forest emits a large portion of BVOCs". The examples discussed are for California.

P3/L9-12: Unclear logic. It is not only removal processes that are relevant. The introduction of eddy-covariance measurements to the discussion seems awkward. Please reformulate.

P3/L14: Wesely (1989) describes a single model, which is based on the big-leaf approach. So this sentence ("Among these models. . .") makes little sense.

P3/L14-15, "usually": Very often big-leaf models are used as inferential models.

P3/L15: Zhang et al. (2002) deal with deposition parameterisations rather than large-scale modelling.

P3/L18: Altimir et al. (2006) do not employ a multi-layer model.

P3/L23: A paper from 2000 is hardly suitable for evaluating recent models.

P3/L24, "process-based": Unclear which processes are referred to here. The implementation consists of a largely empirical resistance parameterisation.

P3/L33: Unclear which challenges are referred to here.

P4/L23: What is meant by "the same below"?

P4/L27-28: Why was the ozone flux calculated with data from a different anemometer than for other fluxes?

P4/L31: Did you correct the O3 flux data for high-frequency losses? How large were the corrections?

P5/L28: What does "partly constrained" mean?

P6/L11: I would recommend against using the term "deposition velocity" for layer-specific conductances.

P6/L19-20: What does "the unit is the same. . ." mean?

P6/L27-28: Unclear language; please rephrase.

P7/ Eq.5: This is a strange combination of partial derivatives and finite differences. Please present the equation in a mathematically consistent form. You also need to assume constant air density here. It would be more appropriate to present the 'flux' as mass flux density (g m-2 s-1).

P7/Eq.5: How did you solve for [O3]. If it is a common procedure within SOSAA, perhaps you could explain it in Sect. 2.3.1.

P8/L1-2: How did you do the forcing? Fig. 2b does not explain this.

P9/Table 1: u* is not limited to the canopy top.

P9/L13, P10/L1, P11/L7, "was calculated": How? These should be moved to the methods description.

P10/Fig. 2b: Gap-filling of data is not described in the paper. Why was it performed? For which variables?

P10/L8 (also elsewhere): These data are measured well above the canopy, so why are they referred to as "canopy top".

P11/Fig. 3, P13/Fig. 5: Are these data screened for low turbulence?

P12/L11-12: How do the low humidity conditions affect turbulent mixing, making this difficult to simulate?

P13/Table 2, P15/Fig. 7: Why is the R2 of the full data set higher than the R2 of any of the four subsets?

P14/L6: Can you estimate how much the correlation was affected by random uncertainty?

P15/Fig. 7: The caption is difficult to read.

P16/L11: The notation related to the cumulative flux is not obvious.

P16/L14: No stomatal contribution is indicated for the understory vegetation in Figure 9.

P16/L14-P17/L4: Unclear presentation. Does "uptake on leaf surfaces" refer to the flux or the cumulative flux (accumulated from the bottom)?

P17/Fig. 9: What explains the stomatal uptake during the night-time?

P17/L11: Please quantify the "limited O3 uptake", as it is obvious that small surface area corresponds to small uptake.

P17/L12-13: These percentage contributions only refer to the mean values of the four data sets, so discussion of variation may be misleading here.

P17/P13: This may be explained by Launiainen et al. (2013), but the meaning of the "sub-canopy layer" is unclear. Does it include some other vegetation surfaces in addition to the understory vegetation and soil?

P17/L14-17: The contributions cited from Launiainen et al. (2013) do not add up to 100%; why?

P18/L1: How was the soil resistance determined in the first place?

P18/L3-5: I do not see how this conclusion about EC measurements results from the

data presented here.

P19/L8-9: You should explain how these percentages were obtained.

P19/L24: No data on BVOC removal are presented in this study.

P20/L14: Poor presentation; please rephrase.

P20/L19-20: I do not see how these different flux partitionings would indicate "the difficulty of simulating and measuring O3 deposition at night".

Technical comments

P1/L18,L19: Incorrect grammar.

P2/L1-3: Unnecessary material for the abstract.

P3/L27-28: "manuscript in preparation" is not a useful reference.

P5/L10: Incorrect grammar.

P8/L8: Incorrect grammar.

P8/L13: Repetition from the introduction.

P9/L1 (also elsewhere): replace "showed" by "shows".

P12/L9,L13: Incorrect grammar.

P17/L17: Typo.

P20/L9: Incorrect grammar.

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