Review of paper acp-2012-137: Ozone deposition into a boreal forest over a decade of observations: evaluating deposition partitioning and driving variables by Rannik et al.

The paper describes a nice analysis of the long-term O_3 deposition flux record measured over the boreal forest of Finland at the SMEAR observation site. The availability of this measurement record on O_3 deposition complemented with many relevant micrometeorological measurements such as radiation but also soil moisture allows for an in-depth analysis of the drivers of deposition. This is done in the presented study focusing on the stomatal versus the non-stomatal component relying on the use of a multi-layer modelling system that considers the contribution by stomatal uptake and simplified terpene-ozone chemistry. This paper fits in well of the scope of this special iLEAPS ACP-BG issue. The paper is well written, although its needs according to me textual editing (see some comments found below). Consequently, I support the publication of this article in this special issue after the following comments/suggestions have been properly addressed.

Line 23: "Several papers addressing the ozone deposition over pine forests (Goldstein et al., 2004; Holzinger et al., 2005, 2006) revealed the role of oxidation products of biogenic emissions in ozone removal". You are stating here that the oxidation products of BVOC emissions are explaining the removal of ozone but checking again for example the Holzinger et al., 2005 reference it is actually been postulated that the oxidation products are indicating the role of BVOC-ozonolysis reactions.

Section 2.3.2 Bulk canopy conductances;

Reading through this section in which the analysis of the observations in terms of the stomatal and non-stomatal components is being explained the question is raised on how the aerodynamic and quasi-laminar transfer contributions have been considered in the analysis. This seems to be one of the major flaws of the presented study (or you have a good reason to leave it out and simply didn't include it in the ms).
You calculate the stomatal and non-stomatal conductances from the measured flux but this conductances then include also the (inverse of) the aerodynamic- and quasi-laminar resistance terms. I am aware that these terms are generally less important in the total transfer pathway but can become relevant especially at days with efficient stomatal (or non-stomatal) uptake. Consequently, this has be modified in the revised version of your ms.

In the section on the ozone chemical modelling is becomes clear after some reading that you apparently use here also a vertical layering system in which you calculate the concentrations and fluxes using an iterative system. I haven't read any information up to this stage on the resolution of your vertical layering. What is the selected thickness of the vertical layers of your multi-layer uptake and chemical destruction model system?

Page 12725, line 22; "In order to solve concentration and flux profiles simultaneously for ozone and B-caryophyllene, **an** iterative method was used with **the** measured ozone concentration at the canopy top **being used as** the boundary condition. For vertical integration **a** numerically stable forward Eulerian scheme was applied (also with iterative correction at each level) and zero-caryophyllene flux assumption at

upper boundary of the simulation domain was superimposed"

(this comment might reflect a difference of "taste" on UK writing styles. It addresses the fact that according to my opinion the overall document should be carefully checked for having left out too many times "the" and "a/an" terms).

What is meant with an iterative correction at each level?

Line 29: "Since no sensitivity of the results **to the assumption on** atmospheric boundary layer (ABL) height was observed from the model runs the ABL height was set for simplicity to 300 m". This remark also raises the question of your selected vertical layering system that apparently goes all the way up to 300m or ?

Page 12726: "In 2006 **there has been a relative** long **break in the** measurement **record**", how long was it, some months??

Line 7; "It is important to note that **the** flux is" (after this I stopped adding the terms "the or a/an", this should be done by the authors/text editors at the Copernicus office)

Page 12727, line 13:14, you mention that the analysis is limited to daytime conditions, which is understandable if you want to focus on the stomatal versus the non-stomatal uptake (and assuming that there is complete nighttime stomatal closure). But what is your justification for only using data with sufficient turbulence? Under conditions with relative small turbulence the chemical timescale might get even closure to the turbulent timescale implying an even more important role of chemistry in O_3 deposition.

Page 12728: line 14. In contrast to what your extensively expressed previously you actually include here in the analysis nocturnal O_3 deposition. I think this is very interesting and like to see this included in the analysis but you should modify the previous statements saying that your analysis "focusses on daytime deposition"

Page 12729; "Further, variation of total and non-stomatal conductance as a function of relative humidity (RH), temperature and photo-synthetically active radiation (PAR) was studied". In the sentence before you state that the non-stomatal component also depends on turbulence, which is obvious thinking about the role of turbulence in mixing in O_3 into the canopy space where it reacts with emitted terpenes but where the turbulence might also play an important role in efficiently transporting the O_3 all the way down to the soil surface where deposition can be another important contributor to the non-stomatal uptake term. Why do you limit your analysis to the role of PAR and RH?

And then read the following sentence: "Friction velocity did not exhibit clear impact on O3 deposition (not shown).". Still it is interesting to consider why there is apparently no dependence on u* since you would expect such a dependence.

Page 12730: "At night the conductance inferred from water flux measurements G^TO_3 was used as a surrogate to estimate stomatal deposition during dry periods.". Maybe I missed something but this is not completely clear to me. Why using the nighttime conductance to estimate stomatal deposition during dry periods? Are you referring

here to nighttime stomatal deposition (which I thought you were assuming that this is not happening having said that you focused on daytime data which apparently is not the case seeing also this section 3.2.2 on nighttime deposition)

Page 12731: line 19 "Comparison of the measurements and predictions enables to conclude that (i) **the** environmental variables **that have been included in the presented analysis** were able to predict ~~similar variation with~~ **to large extent the** observed **variability** during the course of the year, i.e. the seasonal variation with 98% of the variance **being** explained but (ii) failed to explain pattern in inter-annual differences (only 8% of the variance explained) such as elevated GO3 during the weeks 33–35 in 2005, systematically lower GO3 during the weeks 35–39 in 2008 (Fig. 8a)." This sentence is not well readable and since it is an important finding it should be revised to make the statement very clear (already put some suggestions for textual change)

In section 3.4 you analyze the role of terpene emissions in explaining to some extent the non-stomatal O_3 deposition. Previously you shortly mentioned the role of soil NOx and was wondering if you actually considered to include this in your multi-layer model calculation to assess the contribution by the NO-O_3. On the other hand, then your model can so easily become so complex and then you would need an explicit multi-layer exchanges model that considers the full suite of canopy interactions.
What are anyhow the NO emission fluxes at the site and how much could they contribute to O_3 deposition associated with NO titration?

Page 12732; "It is noteworthy that for O3 the vertical divergence of the turbulent flux cannot be disti**n**guished from stomatal sink term". This is very interesting finding on the role of the chemistry in the explaining ozone deposition for this site. Since not all readers might be informed about the issue of flux divergences associated with chemistry and why this expresses it might be worthwhile to consider to explains this in a little more detail. Anyhow, so this finding indicates that the flux divergence due to chemistry cannot be distinguished from the stomatal uptake implying that chemistry overall plays a minor role. Still we have seen quite some important role of non-stomatal uptake which apparently then is also not very strongly determined by in-canopy chemistry but more likely by a suite of other processes (soil uptake, thermal decomposition, etc.)

Page 12734: "but failed to explain major inter-annual differences". I think this is a really interesting finding. You would expect that some extent these inter-annual differences would have to do with controlling parameter that respond on relative long timescales (seasons/years). The potential parameters that operate at these timescales are soil moisture status and phenology but somehow these have been considered in your analyses. It raises the question if these have been properly included in your analyses which appears now to be done through the adjustments of your stomatal model scaling parameters. What other long-term parameter variability explains O_3 deposition?

You raise the issue on the role of soil NO but this source is simply too small to have a significant impact on O_3 deposition. In addition the isoprene-O_3 reaction is also to slow (compared to OH-isoprene) to have a significant impact.

Page 12735; the discussion on the role of BVOCs in explaining the O_3 deposition is too much repetition of what was already presented in the introduction.
Page 12736: "The multivariate analysis did not reveal dependence of O3 total and non-stomatal conductances on turbulence intensity. Turbulent transport time is directly related to turbulence intensity and the ratio of this time-scale to chemical reaction-rate time-scale should affect the non-stomatal deposition". This comment also raises the question about the calculations in-canopy turbulence in your study. You indicated that this relies on the use of the K-theory which we know generally fails in reproducing observed turbulence properties inside the roughness layer and the canopy. So, your estimates of the turbulent timescales might be not very optimal. But so you would anticipate them to be even shorter likely further reducing the role of chemistry in O_3 deposition. You can have a look on the sensitivity to some of these canopy exchange features to the representation of canopy turbulence but then with a focus on BVOC exchanges in a paper by Bryant et al. (acpd-2012-137) also currently in the discussion stage of ACPD.

Furthermore, analysis of the role of chemistry, which is limited in your analysis to the role of caryopyllene, might need to be modified including a whole suite of very reactive compounds (represented in a generic way) recognizing the fact that observations of OH reactivity at the SMEAR site suggest much more reactive compounds being present than the ones that we can currently consider in such model analysis based on observed BVOC emissions (Nölscher et al., ACP special issue on HUMPPA/COPEC campaign)

Page 12737; "It was concluded that chemical reactions with monoterpenes, or other removal mechanisms such as surface reactions, play a role as ozone non-stomatal sink inside canopy". I think that your concluding sentence (and some previous remarks) need to some re-writing. This conclusion is not very convincing. It is also because you wonder based on the discussion how the monoterpenes can be important as an non-stomatal sink whereas these much less reactive compared to the sesquiterpenes and these do not appear to play a role in ozone deposition. Is it then at the end the fact that the monoterpene emissions are substantially larger?? But the role of monoterpenes in O_3 deposition depends on the chemical timescale relative to the deposition timescale and this latter is generally short compared to the monoterpene-O_3 chemical timescale.
Finally, I would actually suggest to include also here a statement about the rather surprising missing role of turbulence in the non-stomatal uptake but also this finding that apparently some still unknown mechanisms might be involved in determining inter-annual variability.