# Ozone deposition impact assessments for forest canopies require accurate ozone flux partitioning on diurnal timescales

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## Abstract

We thank Anonymous Referees #1 and #2 for their valuable and constructive feedback on our manuscript. In this response letter, we carefully address each of the points that were raised. New/modified text is shown in blue.

# 1 Anonymous Referee #1

# 1.1 General comments

#### Comment 1

This paper evaluates the potential added value of a multi-layer representation of vegetation canopies with respect to the traditional big leaf approach (e.g. Wesely, 1989) for simulating ozone deposition and ozone impact metrics for forest canopies. In the Appendix, It also compares for corrected version (vapor pressure and soil moisture of dry deposition such as Zhang et al (2003)).

This is an relevant paper making an interesting comparison between the proposed MLC-CHEM scheme and W89. Although the authors mention that the use of W89 mechanism is not valid for flux-based assessments (e.g from model simulation or observations), they do not discuss how their MLC-CHEM scheme could be implemented in a practical way in chemical transport models. Since the proposed methodology is constrained by flux measurement **observations**, it cannot technically be implemented in regional transport models since it would require a spatial density of ozone flux measurements which does not exist over the domain of such models. As the authors point out: "sites with long-term ozone flux measurements are scarce especially these sites with long-term ozone flux measurements". Therefore, the authors should explain how MLC-CHEM is an added value for current air quality models providing near real-time simulations (and using modified versions of W89).

#### Response

We thank Anonymous Referee 1 for the constructive feedback on our work. The referee here raises one main issue regarding the implementation of MLC-CHEM in larger-scale models, e.g. air quality models, given that ozone flux measurements are not available for the model's domain. In the current model set-up, we indeed constrain the multi-layer model's stomatal conductance and photosynthesis parameters using the complementary information derived from eddy-covariance observations of latent heat and CO<sub>2</sub> fluxes. We use the updated estimate of stomatal conductance to simulate the ozone dry deposition, which is then compared with the observed ozone fluxes.

MLC-CHEM has already been applied previously in a global chemistry-climate model system (EMAC; Ganzeveld et al., 2002, 2010). In these studies, MLC-CHEM uses EMAC-simulated stomatal conductance, to obtain consistent simulations of evapotranspiration (and its effect on micro-meteorology and the boundary layer) and atmosphere-biosphere exchange of ozone and other trace gases. Models that can simulate  $CO_2$  mixing ratios as well as greenhouse gases (e.g. WRF-Chem) are well-suited for using MLC-CHEM as a land surface exchange parameterization. Alternatively, in case stomatal conductance or  $CO_2$  mixing ratios are unavailable from the driving model, A-g<sub>s</sub> could be applied with an offline set-up of MLC-CHEM, for example by using prescribed  $CO_2$  concentrations and other (micro)meteorological parameters for the online simulation of ozone, latent heat and  $CO_2$  fluxes.

To include this point, we add the following section to the discussion:

MLC-CHEM can be driven by diagnostic variables available from CTM output (or their driving meteorological models), favoring its implementation to represent atmosphere-biosphere fluxes of reactive compounds (Ganzeveld et al., 2002, 2010). In such a coupled setup, MLC-CHEM would use simulated stomatal conductance from the driving model to consistently represent atmosphere-biosphere exchange with the model's representation of (micro-)meteorology. An implementation of A-g<sub>s</sub> with prescribed CO<sub>2</sub> mixing ratios, calculated online or offline, can be tested if simulated stomatal conductance estimates are unavailable.

# Comment 2

Similarly, the authors also wrote: "Observational studies indicate that ozone deposition exhibits substantial temporal variability that is not reproduced by atmospheric chemistry models due to a simplified representation of 5 vegetation uptake processes in these models". The authors should elaborate a bit on that and mention that the lack of observational routine data for dry deposition is an handicap to appropriately simulate processes described by MLC-CHEM. It would be nice if clear recommendations would be given to modelers in that respect (other than "W89 is invalid").

#### Response

These suggestions are highly welcomed. In the introduction, we already included the following statement regarding ozone deposition data availability:

However, sites with long-term ozone flux measurements are scarce (Clifton et al., 2020), which limits the characterization of the seasonal to inter-annual temporal variability in the stomatal and non-stomatal components of ozone removal.

We now also add another statement later in the introduction:

Additionally, spatio-temporal controls of ozone deposition pathways remain incompletely understood (Clifton et al., 2017, 2020), in part owing to the scarcity of long-term ozone flux observations. Therefore, we here study temporal controls on stomatal and non-stomatal ozone deposition pathways, and their implications for simulations of CUO, using two multi-year ozone deposition datasets as well as a big leaf and multi-layer parameterization of land surface ozone uptake.

In order to communicate clearer messages to modellers, we remove the statement regarding the invalidity of W89 for flux-based impact assessments (see also our reply to Comment 4 by Anonymous Referee #1). Instead, we propose the following future directions to the ozone deposition modelling community, in the discussion:

To stimulate improvement of big leaf and multi-layer parameterizations, modelers may benefit from evaluations against existing long-term dry deposition observations in various ecosystems (e.g. tropical forest, temperate forest and grassland) and for contrasting environmental conditions (e.g. during droughts) to improve simulated temporal variability in different ozone sinks on diurnal to inter-annual timescales. Such an assessment is currently underway in stage 4 of the Air Quality Model Evaluation International Initiative (AQMEII4; Galmarini et al., 2021). Additionally, evaluation against in- and above-canopy ozone flux measurements (e.g Fares et al., 2014; Finco et al., 2018) can reveal information about non-stomatal sinks in these parameterizations, such as soil deposition and in-canopy chemical removal. Lastly, the application of proposed parameterizations for non-stomatal ozone sinks, such as for wet leaf uptake (Potier et al., 2015) and soil uptake (Stella et al., 2019), should be tested in 3D and single-point models of ozone deposition.

#### Comment 3

Note that W89 is rather obsolete nowadays as many authors have shown its weaknesses through the years. For example, LAI scaling to canopy was omitted in W89 and should be included. VPD weakness has been corrected by using Jarvis (1976), etc.

#### Response

This is correct. However, we have the impression that the atmospheric chemistry modelling community is not fully aware of the limitations of the original W89 scheme. In fact, the code implementation of the W89 scheme in this study is derived from the often-used community model WRF-Chem (Grell et al., 2005). Therefore, we consider it important to communicate this message. Besides, in Appendix B we show that another popular big leaf parameterization (Zhang et al., 2003), which includes a more advanced description of stomatal conductance that accounts for sunlit/shaded leafs, VPD and soil moisture stress, does not consistently outperform W89 at simulating stomatal conductance for our two study sites.

# 1.2 Specific questions/issues

# Comment 4

1) The authors wrote: "This invalidates the use of the W89 mechanism for flux-based assessments...". The authors would be careful with such general statement. Moreover, the comparison with Wesely (1989) could not be entirely appropriate since very few institutions and models use W89 as in its original form. Most of models use different corrected forms of W89 ( exchange of ozone, e.g. accounting for stomatal closure based on the vapor pressure deficit (VPD) and soil moisture, LAI scaling correction in W89,

#### Response

We partially agree with this statement. Indeed, many authors have proposed updates to the Wesely scheme, and directly after this statement we discuss important improvements regarding accounting for VPD and soil moisture. However, as discussed above, the W89 parameterization remains in use in state-of-science models (see e.g. Galmarini et al., 2021).

In the main text, we have removed this statement and instead focus on effects of an instantaneous structural bias when summed over the growing season:

We have shown that stomatal and non-stomatal sinks are not accurately reproduced using the W89 big leaf parameterization compared to observations at two forested ozone flux sites, leading to structurally biased instantaneous and growing season-cumulated (stomatal) ozone flux simulations.

## Comment 5

2) Line 115-166. W89 can be deemed representative for the representation of dry deposition in other atmospheric chemistry model. Please be careful since several models have upgraded W89 to correct for some of its weakness (include VPD deficit, wetness on foliage, corrected form LAI missing in W89, etc.). e.g. (Jarvis, 1976; Ref. Valmartin, M, Heald, CL and Arnold, SR (2014) Geophysical Research Letters, 41 (8). 2988 - 2996.etc.).

#### Response

We have removed this particular statement and revised this section as follows, giving credit to the mentioned upgrades:

The discussion below considers the implementation of the big-leaf dry deposition approach in the coupled meteorology-chemistry model WRF-Chem (Grell et al., 2005). Other big leaf parameterizations are available with improved treatment of stomatal (e.g. Emberson et al., 2000; Val Martin et al., 2014; Lin et al., 2019) and non-stomatal uptake (e.g. Zhang et al., 2003). However, the common use of Wesely's (1989) parameterization in state-of-science 3D atmospheric chemistry and transport models (see e.g. Galmarini et al., 2021) motivates the choice for this scheme.

#### Comment 6

3) Conclusion: How can MLC-CHEM be included in a regional transport model. ?. Avoidance of Wesely type parametrization might not be possible unless CO2 is better evaluated by these models (which does not seems to be the case now) and routine flux and near-real time measurements are available.

#### Response

As explained in our response to Comment 1, MLC-CHEM can readily be applied in coupled carbon cycle/atmospheric chemistry models. In models without any (accurate) representation of the carbon cycle, modellers may experiment with using stomatal conductance from the driving meteorological model (for consistency with simulations of evapotranspiration, as in Ganzeveld et al., 2002, 2010), or by prescribing temporally varying CO<sub>2</sub> mixing ratios (e.g. from CAMS data products Agustí-Panareda et al., 2014).

#### Comment 7

4) In the Appendix it would be useful and informative to also show the diurnal profile of CO2 concentration (not only CO2 fluxes). It would be also interesting to provide values of CO2 compensation point and diurnal variations of CO2 concentrations for the benefit of the reader (in Supplementary material).

#### Response

These are interesting suggestions. We chose not to include these diagnostic graphs, since a complete interpretation of these variables would require additional observational constraints on in-canopy  $CO_2$  mixing ratios and contributing processes (e.g. soil respiration), which are not available from observations. We therefore consider this analysis beyond the scope of the current manuscript. In the discussion, we now emphasize the need for better observational constraints on stomatal conductance and canopy-atmosphere  $CO_2$  exchange in the discussion:

Our results suggest that  $A_{net}$ - $g_s$  parameterizations, as applied in MLC-CHEM, simulate stomatal conductance in good agreement with observation-inferred values throughout the diurnal cycle. Such models are sensitive to parameters typically derived at leaf level that display spatio-temporal variability. Further observational constraints on these parameters, e.g. from leaf-level ecophysiological measurements, improve the representation of stomatal conductance and biosphere-atmosphere exchange (Vilà-Guerau De Arellano et al., 2020), benefitting simulations of CO<sub>2</sub> and ozone exchange as simulated by A- $g_s$  within MLC-CHEM.

# 1.3 Technical corrections

## Comment 8

- 1) Line 77: detailed below -> which are detailed below.
- 2) Typo: Line 117 uptak -> uptake
- 3) Typo: Legend of Figure 2 shaed -> shade

4) Legend of Figure 1: "and their in- and output variables" -> and their input and output variables.

5) Line 131 and 175 is a repeat of the same information.

6) Please avoid starting a section or a paragraph with the word **Figure** (section 3.1.1 and elsewhere)

7) Figure 5. "The shaded area in panel d highlights the daytime period (defined as 8-20 h LT) over which the stomatal flux is calculated". It looks that the gray (shaded) area rather corresponds to overnight time not daytime. Please review and correct.
8) Figure 8. Please specify the meaning of the numbers just below the figure ("0.28 0.23 0.31", etc.). the meaning does not appear anywhere in the document, it seems.

#### Response

Thanks for these suggestions. We have corrected technical comments 1-4, 6 and 7.

Regarding technical comment 5: lines 131 and 175 refer to the soil resistance settings in W89 and MLC-CHEM, respectively, which we set to site-inferred values from the literature rather than using the original values in the parameterization. For completeness it is important to include this information in both locations in the text, as otherwise it could suggest that we use the model's default soil uptake resistance.

Regarding technical comment 6: we have removed all occurrences in the manuscript, and have changed the structure of these paragraphs.

Regarding technical comment 8: we do include this information in the caption of Figure 8. We made the caption more specific:

Only the data points with valid observation-inferred stomatal conductance estimates are selected for this comparison, the fraction of valid data points per growing season that remains is shown at the bottom of panels a and c.

# 2 Anonymous Referee #2

# Comment 1

This work compares a well know big leaf model with a multi-layer canopy model to demonstrate that the latter best represent ozone deposition in two forest canopies. As the author mention, there are not many field measurements globally, therefore testing models vs observation remains challenging. However, for future studies the authors are invited to use data from other field studies which have been collected in the last decades, especially those collected in dry sites. Results are quite predictable in the sense that a multi-layer model constrained with observation return more realistic data especially in reproducing diurnal site. Unfortunately, without field data to constrain MLC models, it is hard to obtain realistic results and therefore global application of MCL model remains challenging. The work reads well, and shed light on the need to use multi-layer models to derive ozone fluxes, in particular CUO used as a metric for ozone damage to vegetation.

# Response

We thank Anonymous Referee #2 for the constructive evaluation of our study. Below, we address each of the issues raised. We are indeed aware of the need to apply MLC-CHEM for additional land use types and different environmental conditions, e.g. at dry forest sites and grasslands, and an effort to expand the analysis presented in this manuscript to different sites is currently being executed in the context of the project 'Air Quality Model Evaluation International Initiative Phase 4' (AQMEII4).

# Comment 2

Introduction

"...these are limited to short timescales. For these reasons, quantifying temporal variability in stomatal and non-stomatal ozone deposition solely based on observations remains challenging."

»Please define what is an appropriate timescale to derive ozone flux partitioning. In my opinion having year-long continuous measurements is timescale long enough.

#### Response

We agree that continuous observations during one year are indeed a major step forward to quantify temporal variability in stomatal and various non-stomatal removal terms. However, extending such observational data to multiple years (which ideally span a range of environmental conditions, e.g. wet/dry years, high/low ozone concentrations) would benefit studying temporal variability of the different ozone sinks and its drivers.

We propose to modify the statement as follows:

Given the scarce availability of ozone deposition observations that span at least one, and preferentially multiple years, quantifying temporal variability in stomatal and non-stomatal ozone deposition solely based on observations remains challenging.

#### Comment 3

## Site description

Other than a temperature broadleaf forest and a coniferous forest, it would have been wise to select also a dry forest in order to test models in environments with rather different climate conditions and vegetation features (i.e. the capacity to emit different VOCs). Furthermore, the effect of high VPD and drought on Gs is acritical point for models, therefore you missed such representation.

## Response

We acknowledge that conditions at dry forest sites are more difficult to reproduce given the VPD and soil moisture effects that can limit ozone uptake. As already mentioned, the MLC-CHEM evaluation will be extended to more sites, including dry forests, in the context of the model intercomparison project AQMEII4. We also added this as a suggestion for future work in the discussion, for which we refer to our response to Comment 2 by Anonymous Referee #1.

#### Comment 4

Lines 150-154: please provide a definition and/or reference for the relaxation factor of 0.5.

## Response

We have elaborated on the definition of the relaxation factor as follows:

Micro-meteorological variables are provided as input to the model, and ozone concentrations in the upper layer are nudged to observed above-canopy ozone concentrations to represent entrainment and advection. We use a weighting factor of 0.5, which implies that we force simulated above-canopy ozone mixing ratios to observed mixing ratios with a timescale of  $\pm 2$  h, based on the applied temporal resolution of 0.5 h.

## Comment 5

Line 154: please provide the formula or the reference to estimate In-canopy resistance.

#### Response

A reference to Van Pul and Jacobs (1994) has been added.

# Comment 6

Line 158: unclear the way adopted to estimate Ag. The latter parameter is crucial to estimate properly Gs. Have you tried to compare Cumulative Ag with GPP derived from EC measurements?

# Response

The procedure to derive Ag is relatively elaborate, and fully explaining this is beyond the scope of this study. Therefore, we have chosen to state that it is a function of PAR, skin temperature and internal  $CO_2$  concentration. We have resolved this unclarity by providing a reference to the original A-gs model description in Ronda et al. (2001), Appendix A:

 $A_g$  is gross assimilation, calculated as a function of photosynthetically active radiation (PAR), skin temperature, the internal CO<sub>2</sub> concentration and the soil water content (SWC). We refer the reader to Appendix A in Ronda et al. (2001) for more details on the calculation of  $A_g$ .

The suggestion to compare cumulative  $A_g$  with observation-derived GPP is interesting and worthwile, but beyond the scope of the current study that focuses on ozone deposition. However, we recognize that application of MLC-CHEM would benefit from more robust evaluation of CO<sub>2</sub> exchange within A-g<sub>s</sub> using, for example, campaign-based leaf-level ecophysiological measurements (see also our response to Comment 7 by Anonymous Referee #1).

# Comment 7

Lines 220-230: il looks like a mid-day depression in ozone deposition most likely due to partial stomatal closure is not taken into account by the MLC model.

# Response

MLC-CHEM indeed overestimates  $V_d(O_3)$  somewhat compared to the observations at Ispra, but unaccounted stomatal closure is unlikely to be the cause. MLC-CHEM does account for stomatal closure due to VPD stress, as included in A-gs (Ronda et al., 2001). Additionally, this overestimation occurs mostly in the morning (9-13 h LT), while VPD stress is particularly expected in the afternoon. Furthermore, the effect of (soil moisture) drought on stomatal conductance is small (see Discussion). We therefore attribute the morning overestimation to overestimated non-stomatal uptake (see Fig. 5c).

# Comment 8

Line 326: why do you exclude relevant chemical removal of ozone which may contribute to non-stomatal deposition? Perhaps low NOx and low BVOC emitted by the two ecosystems? Sesquitrpenoids from pines and to a minor extent monoterpenoids may react with ozone at time scales even lower than those used in the study.

# Response

In this section we describe sensitivity experiments to disentangle the contribution of different processes (leaf uptake, soil uptake, chemical removal) to the total non-stomatal ozone sink. Our strategy was to perform sensitivity simulations where these sinks were deactivated one at a time, in order to quantify their contribution to total ozone removal.

From these sensitivity experiments, we find that the contribution from chemical removal to the total non-stomatal sink is low. We acknowledge that this conclusion is uncertain, since we could have missed contributions by soil-emitted NO and BVOCs unaccounted for in the applied parameterizations (Yienger and Levy, 1995; Guenther et al., 2006, 2012), but we lack the observational constraints to study such chemical sinks. At Hyytiälä, where the highest emissions of BVOCs reacting with ozone are expected, there are indeed indications for a chemical ozone sink by BVOCs, but the mechanisms remain incompletely understood (Rannik et al., 2012). A previous (shorter-term) multi-layer modelling analysis for Hyytiälä found a rather small contribution of chemical removal to the total ozone sink at this site (Zhou et al., 2017). For decidous forests like Ispra, the lifetime of the site-emitted BVOCs is on the order of 2.5 hours or higher (Jensen et al., 2018), which is larger than a typical deposition timescale (defined as the canopy height divided by the deposition velocity) of 1-2 hours.

# Comment 9

Line 354: have you tried gap-filling so to produce CUOst for all year?

#### Response

Panels b and d in Figure 8 show a first-order estimate of observation-derived  $CUO_{st}$  derived by scaling the cumulative stomatal uptake (calculated for all valid observations) with the fraction of valid observations. This is a first-order gap-filling method, which assigns the average stomatal conductance of valid observations to all non-valid data points. While this method is first-order, we discuss its shortcomings in Section 3.4 (for example, the  $CUO_{st}$ estimate for Hyytiälä in 2005 is lower than in other years, since data availability is low in the summer months, with the highest stomatal conductance). We believe this is sufficient to highlight our main point of this paragraph: growing-season ozone uptake is higher at Ispra than at Hyytiälä, which is not immediately evident from panels a and c in Figure 8.

# Comment 10

Line 488: rather than using the term "plant's interior" I would say "plant's chloroplast" or more generally "plant's carboxylation site".

## Response

Corrected to plant's chloroplast.

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