Review of Improved isoprene emission estimates over the Finnish boreal forest using the MEGANv3.2 model

Bettineschi et al.

The authors thank the referees for their effort in reading and providing positive and constructive feedback for the improvement of this article. Below, we report in red the referees' comments and in black the authors' answers. In the authors' reply, *italic purple* is used to report text from the previous version of the manuscript, while *italic blue* is used for the updated text. All line numbers refer to the track changes version.

Answer to Anonymous Referee #1

This paper provides an improved framework for modeling isoprene emissions in the Finnish boreal forest by incorporating high-resolution, species-specific tree and emission data. The authors found that the new framework can better reproduce the measured patterns and further claimed an overall improvement in secondary organic aerosol estimates. Based on Figure 11, the performance on OA shows only a marginal improvement, so please revise this statement to be more accurate.

We thank Anonymous Referee #1 for this comment. We agree that the model performance in reproducing OA only shows a marginal improvement. We modified the text to acknowledge that, specifically, we modified the following:

L10-11: *both result in overall improvements* → *both result in overall improvements (although small)*

L421-422: *reduction in model biases for both BVOCs and OA* → *reduction in model biases for both BVOCs and OA, even though the impact on OA is small)*

In addition to these changes, in the original draft we already extensively discussed the persistence of the OA bias in both the results (L410-455) and discussion (L490-507) sections.

Major comments

(1) The authors compared the "simple" and "advanced" species distribution, noting 20% differences, but did not run the WRF-CHIMERE model using this simple specification. As this 20% differences might result in negligible differences in the CTM, I see the need to run a control simulation with this "simple speciation" to logically claim that highly detailed species distributions are essential for atmospheric chemistry modelling;

We agree with the reviewer that the differences between the EF calculated using the "simple" and "advanced" species distribution might result in negligible differences in the CTM. As we mentioned in the original manuscript, we recommend incorporating detailed species

distribution if the data are available; however, we also mentioned that the "simple" approach is a valid alternative:
For BVOCs emissions estimation, incorporating high-resolution species distribution information is highly recommended over uniform distribution for each ecotype. If this is not possible, including all the main tree species or genera (with uniform distribution) present in a given ecotype should be preferred over the simple needleleaf/broadleaf classification.
Additionally, we performed an additional sensitivity run using the "simple" species distribution to calculate isoprene emission factors and thus emissions. Figure 1 shows the average relative difference in isoprene emissions vs concentrations for each grid cell. The great ma-

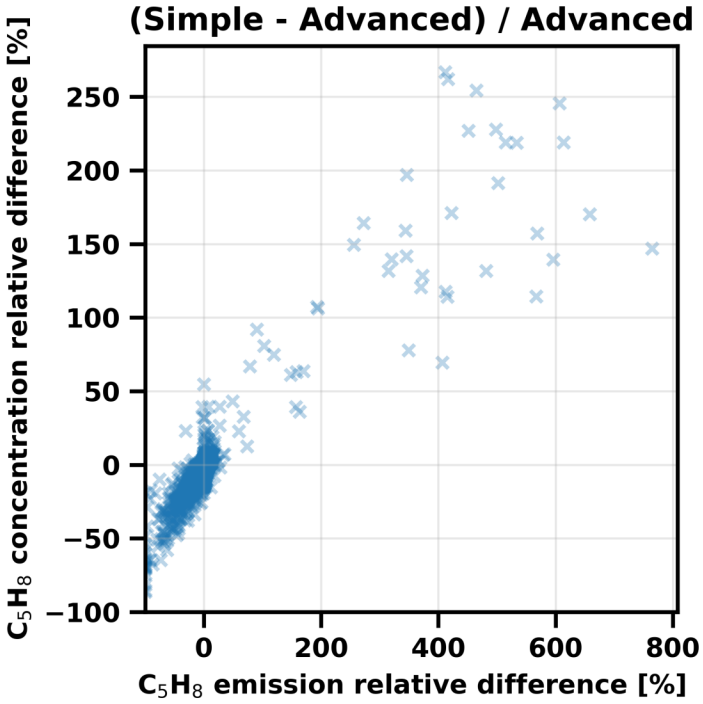

Figure 1: Average relative difference, for each grid cell in Finland, between C_5H_8 emissions and concentrations. The relative difference is calculated between the simulation using the "simple" and the "advanced" speciation.

jority of points show small average differences; however, in some grid cells, the differences in emissions and concentrations are significant. While for large-scale modeling these differences are probably not important, for high-resolution regional modelling this can affect the results.

(2) Why only focus on isoprene changes in this study, as these species are not strong isoprene emitter at all, so the changes/improvement we see here might be overturned/non-valid if the authors also account for main-emitted compounds and their atmospheric interactions;
We agree with the Referee that isoprene is not the most abundant BVOC in the boreal forest. However, we decided to focus only on isoprene for different reasons. Firstly, previous studies (Ciarelli et al., 2024; Zhao et al., 2024) have shown that isoprene emissions in the boreal for-

est have been greatly overestimated by previous MEGAN applications, while monoterpenes were reproduced fairly well. This makes isoprene a critical target for model improvement, as biases in its representation can disproportionately affect atmospheric chemistry despite its lower absolute abundance.
Secondly, isoprene plays an important role in atmospheric oxidation chemistry due to its high reactivity and its influence on OH radical budgets, ozone formation, and secondary organic aerosol (SOA) production. Even relatively small changes in isoprene emissions can therefore lead to non-linear and system-wide impacts on atmospheric composition. Focusing on isoprene thus allows us to isolate and evaluate the sensitivity of the chemical system to uncertainties in one of its most reactive BVOCs.
Thirdly, by constraining our analysis to a single compound class, we are able to more clearly attribute the observed changes to the modifications introduced in the emission, without the added complexity arising from simultaneous adjustments in multiple BVOC species and their interactions. This reduction in dimensionality is particularly important for a process-oriented evaluation.
We acknowledge that a comprehensive assessment of all major BVOCs and their interactions would be valuable. However, such an analysis is beyond the scope of the present study and would require a dedicated investigation.

(3) "EFs calculated by MEGANv3.2 represent the leaf-level EF with units of nanomoles m-2 s-1 LAI-1". I don't think this is correct. If it is leaf-area EF, it should be in units of per leaf area. In Section 2.2, the manuscript states that a "careful conversion is required" to translate leaf-level EFs from MEGANv3.2 to canopy-scale EFs for MEGANv2.1. Equation 2 is presented for this conversion, with both LAI_{std} and LAI_{max} defined as 5 m^2 m^{-2}. Consequently, the scaling factor (LAI_{std} / LAI_{max}) equals exactly 1, meaning the numerical values of the EFs remain mathematically unchanged. For me, this is not correct, and equation 1 cannot convert the unit from per leaf area to per ground area in my understanding.
We thank the reviewer for this comment. We would like to clarify that in MEGANv3.2, the EF is indeed defined at the leaf-level. The unit "nmol m^{-2} s^{-1} LAI^{-1}" reflects this, since dividing by LAI (m^2 leaf per m^2 ground) converts a ground-area flux to a leaf-area basis. Therefore, this unit is consistent with a leaf-level EF, even though it is expressed with LAI^{-1} rather than explicitly as m_{leaf}^{-2}. This has also been pointed out in previous work (Silva et al., 2020).

Secondly, this section has been completely rewritten because Eq. 1 was incorrectly reported and did not correspond to the actual conversion used in this study. We added a new section explaining in detail the difference between MEGANv2.1 and MEGANv3.2 and the correct conversion between EF. The new section (MEGAN emission model) is as follows (L163-179):
BVOC emissions are estimated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN; Guenther et al., 2006, 2020). Both MEGANv3.2 and MEGANv2.1 share the same general emission equation:

$$E = EF \cdot EA \quad (1)$$

The two versions differ in both the definition of EF and the formulation of EA. In MEGANv3.2,

the EA is defined as follows:

$$EA_{\mathrm{v3.2}} = Cce \cdot LAI \cdot \gamma_{\mathrm{P}} \cdot \gamma_{\mathrm{T}} \cdot \gamma_{\mathrm{HT}} \cdot \gamma_{\mathrm{LT}} \cdot \gamma_{\mathrm{SW}} \cdot \gamma_{\mathrm{O3}} \cdot \gamma_{\mathrm{A}} \cdot \gamma_{\mathrm{SM}} \cdot \gamma_{\mathrm{CO2}} \quad (2)$$

where Cce represents the canopy environment coefficient accounting for within-canopy light and environmental attenuation, LAI is the leaf area index, γ_{P}*,* γ_{T}*,* γ_{HT}*,* γ_{LT}*,* γ_{SW}*,* γ_{O3}*,* γ_{A}*,* γ_{SM}*, and* γ_{CO2} *represent the activity factors for downward shortwave radiation, 2 meter air temperature, high temperature, low temperature, strong wind,* CO_2 *pollution, leaf age, soil moisture, and* CO_2 *concentration, respectively. In the MEGANv2.1 version implemented within CHIMERE, the EA term is:*

$$EA_{\mathrm{v2.1}} = \gamma_{\mathrm{LAI}} \cdot \gamma_{\mathrm{P}} \cdot \gamma_{\mathrm{T}} \cdot \gamma_{\mathrm{A}} \cdot \gamma_{\mathrm{SM}} \quad (3)$$

where $\gamma_{\mathrm{LAI}} = 0.49\, LAI/\sqrt{1 + 0.2\, LAI^2}$.

Because EF in MEGANv3.2 is defined per unit LAI while in MEGANv2.1 it is not, the two EF datasets are not directly interchangeable. Equating Eqs. (2) and (3) at standard conditions and solving for $EF_{\mathrm{v2.1}}$ *gives:*

$$EF_{\mathrm{v2.1}} = EF_{\mathrm{v3.2}} \cdot \frac{LAI \cdot Cce}{\gamma_{\mathrm{LAI}}} = EF_{\mathrm{v3.2}} \cdot \frac{Cce}{0.49} \cdot \sqrt{1 + 0.2 \cdot LAI^2} \quad (4)$$

where LAI in Eq. (4) is at standard condition (i.e., 5 $m^2\ m^{-2}$*)*

Detailed comments

L110: All the simulations are performed "online". What does this mean? Do meteorological variables affect emissions at each time step?

In this case, "online" refers to the coupling between CHIMERE and WRF. Specifically, the CHIMERE size distribution is passed to WRF, which affects the meteorology. Specifically, aerosol feedback (both direct and indirect) is activated in all simulations. We added further explanation in the text as follows (L116-117):

All the simulations are performed "online", with both direct and indirect aerosol effects activated.

On top of this, meteorological variables do indeed affect emissions at each time step as standard when using the MEGANv2.1 algorithm.

L145-147: In MEGAN 2.1, EF is at the ecosystem level, but not at the gridcell level. The EF is still at the PFT level. It is important not to mix this. When used in CLM, for instance, the EFs are only used at the PFT level!

We agree with the Referee that this can be a source of confusion. We originally used the term emission factor to refer to the grid cell, as this is commonly done in chemistry-transport model (CTM) applications. However, to avoid confusion and be more precise, we updated the text to use the term emission potential (EP) when referring to the grid cell. We modified

the text as follows (L180-185):
To begin, we would like to clarify the terminology used throughout this text, as EF can refer to different concepts. When we use the general term EF, we refer specifically to the ecosystem level. To differentiate this from the EF associated with a specific tree species, we will consistently use the term "species-specific EF" when referring to the latter. Additionally, from here onwards, we use the term emission potential (EP) to refer to the "emission capacity" of the entire grid cell. The EP is used in the MEGANv2.1 algorithm to calculate actual emissions.
On top of this, we modified the whole text and figures consistently (i.e., by using EP when referring to the grid cell).

L149-150: Not sure I understand here. First of all, why only focus on isoprene emissions, as isoprene emissions are generally low or absent in these listed trees. Then, about this EF processor, what does it do with your measured EF? Before introducing the emission factor to be used in the model. It is important to describe how these measured EFs were derived/measured.
We have now clarified why we specifically focus on isoprene emissions, as detailed in comment 22. Regarding the EF processor, this code is available in the MEGANv3.2 release. It calculates the EP for each grid cell based on tree cover, ecotype (similar to the old PFT) cover, the tree species present in each ecotype, and the EF of each tree species. So the EF processor is not used to derive/measure species-specific EFs, but rather to "convert" them into EP.

Equation 6: Why integrate SRR over a fixed vertical depth of 500 meters? I assume this fixed depth will systematically bias the AME metric by underestimating daytime but overestimating nighttime. How does this fixed depth affect Fig. 12?
It is true that a fixed depth could bias the AME metric. However, integrating over a fixed vertical depth is a standard approach used in many other studies (Aliaga et al., 2021; Bettineschi et al., 2025; Hakala et al., 2022). The reason is that it provides a framework independent of uncertainties in planetary boundary layer height (PBLH) estimation, which can vary substantially across models and retrieval methods. Using a fixed depth avoids introducing additional variability and potential errors associated with diagnosing PBLH. Furthermore, Bettineschi et al., 2025 showed that using a fixed depth or a depth that varies with the PBLH (simulated by WRF) does not result in substantial differences in the resulting metrics, suggesting that the sensitivity of AME to the choice of integration depth is limited. Consequently, the main features and interpretation of Fig. 12 remain robust despite the potential bias in the AME.

L296-297: Where does the isoprene come from in the night?
At Hyytiälä in summer, true nighttime is very short. There are only a few hours without sunlight, while solar radiation is present for most of the 24-hour period (often 18–20 hours including twilight). As a result, isoprene emissions go to zero only in few hours, because light levels remain sufficient for low but non-zero production into the late evening and early morning. We modified the text to clarify this, as follows (L340-343):
A cluster of points, however, shows underestimations greater than a factor of ten; these correspond mostly to nighttime (when in Hyytiälä light levels during short summer nights allow low but non-zero isoprene emissions).

Fig. 11 the almost unchanged patterns when compared with the observed OA. Back to my question again: why improve only isoprene emissions in this manuscript?

As outlined in our response to major comment #2, the primary objective of this study is to improve the representation of isoprene emissions in a region (the Finnish boreal forest) where large uncertainties have been identified, rather than to reproduce OA concentrations as accurately as possible.

Accordingly, the limited changes in OA shown in Fig. 11 are consistent with our targeted approach. By focusing on isoprene alone, we can isolate its specific contribution and demonstrate that its impact on OA is relatively small in this environment. Including all BVOCs would introduce additional non-linear interactions, making it more difficult to disentangle the role of individual compounds and to clearly attribute the effects of the emission updates. However, we agree with the referee that a comprehensive treatment of all BVOCs would be valuable, but this is beyond the scope of the present work.

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Answer to Referee #2

M. Bettineschi and co-authors present a well-thought and valuable study addressing the model estimation of isoprene emissions and concentrations over Finland. Several previous studies have hinted at a significant overestimation of isoprene fluxes calculated using the MEGAN algorithm. Here, high-resolution distributions of the major tree species in Finland are used in MEGANv3.2 to provide improved emission estimates. These emissions are used in a regional model to calculate the concentrations of BVOCs and organic aerosols (OA) over Finland and neighboring areas. The isoprene fluxes and BVOC and OA concentrations are evaluated against in situ and flux data over Finland. Improved agreement for the isoprene fluxes and concentrations is obtained when the updated distributions are used, in conjunction with best estimates of the emission factors, as well as with a canopy correction to the wind speed and diffusion coefficients within the canopy. The study demonstrates quite well the importance of using local information on tree distributions in biogenic emissions models. The canopy correction also proves to be very valuable. The results for OA abundances are comparatively disappointing. The analysis suggests an underestimation of secondary OA formation from the oxidation of monoterpenes. An improved model representation of SOA formation is one of the main recommendations for future studies.
We thank the Referee 2 for this comment on our manuscript.

Major comments

1) The MEGANv3.2 algorithm is not properly described. A reference (Guenther et al. 2020) is provided, but it is a book chapter (conference paper) and does not fully describe the MEGANv3.2 algorithm. There is some confusion regarding the units and definition of the emission factors - providing equations with properly defined variables would make it easier for the reader to understand the algorithmic differences between the versions 2.1 and 3.2 of MEGAN. Also, a conversion is required for the reported emission factors of the main tree

species, since the literature generally gives EF per gram of dry matter, instead of per square meter. I'm also not sure whether the standard conditions of MEGAN (either 2.1 or 3.2) are identical to the standard conditions of the reported emission factors. These aspects should be clarified.

We thank the reviewer for this comment. This section has been completely rewritten because Eq. 1 was incorrectly reported and did not correspond to the actual conversion used in this study. We added a new section explaining in detail the difference between MEGANv2.1 and MEGANv3.2 and the correct conversion between EF. The new section (MEGAN emission model) is as follows (L163-179):

BVOC emissions are estimated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN; Guenther et al., 2006, 2020). Both MEGANv3.2 and MEGANv2.1 share the same general emission equation:

$$E = EF \cdot EA \tag{1}$$

The two versions differ in both the definition of EF and the formulation of EA. In MEGANv3.2, the EA is defined as follows:

$$EA_{\mathrm{v3.2}} = Cce \cdot LAI \cdot \gamma_{\mathrm{P}} \cdot \gamma_{\mathrm{T}} \cdot \gamma_{\mathrm{HT}} \cdot \gamma_{\mathrm{LT}} \cdot \gamma_{\mathrm{SW}} \cdot \gamma_{\mathrm{O3}} \cdot \gamma_{\mathrm{A}} \cdot \gamma_{\mathrm{SM}} \cdot \gamma_{\mathrm{CO2}} \tag{2}$$

where Cce represents the canopy environment coefficient accounting for within-canopy light and environmental attenuation, LAI is the leaf area index, γ_{P}, γ_{T}, γ_{HT}, γ_{LT}, γ_{SW}, γ_{O3}, γ_{A}, γ_{SM}, and γ_{CO2} represent the activity factors for downward shortwave radiation, 2 meter air temperature, high temperature, low temperature, strong wind, CO_2 pollution, leaf age, soil moisture, and CO_2 concentration, respectively. In the MEGANv2.1 version implemented within CHIMERE, the EA term is:

$$EA_{\mathrm{v2.1}} = \gamma_{\mathrm{LAI}} \cdot \gamma_{\mathrm{P}} \cdot \gamma_{\mathrm{T}} \cdot \gamma_{\mathrm{A}} \cdot \gamma_{\mathrm{SM}} \tag{3}$$

where $\gamma_{\mathrm{LAI}} = 0.49\, LAI / \sqrt{1 + 0.2\, LAI^2}$.

Because EF in MEGANv3.2 is defined per unit LAI while in MEGANv2.1 it is not, the two EF datasets are not directly interchangeable. Equating Eqs. (2) and (3) at standard conditions and solving for $EF_{\mathrm{v2.1}}$ gives:

$$EF_{\mathrm{v2.1}} = EF_{\mathrm{v3.2}} \cdot \frac{LAI \cdot Cce}{\gamma_{\mathrm{LAI}}} = EF_{\mathrm{v3.2}} \cdot \frac{Cce}{0.49} \cdot \sqrt{1 + 0.2 \cdot LAI^2} \tag{4}$$

where LAI in Eq. (4) is at standard condition (i.e., 5 m^2 m^{-2})

2) One important aspect of the study is overlooked: the chemical mechanism of isoprene oxidation in WRF-CHIMERE and in the model studies (Ciarelli et al., 2024; Cholakian et al., 2022) cited in the introduction to back the claim of isoprene overestimation against in situ data. The oxidation mechanisms in these studies (including this one) are obsolete, and lack OH-recycling mechanisms (epoxide formation, peroxy radical isomerisation, etc.) that are

now widely accepted to occur (see e.g. Wennberg et al., https://doi.org/10.1021/acs.chemrev.7b00439; Novelli et al., https://doi.org/10.5194/acp-20-3333-2020). The absence of such reactions might lead to isoprene overestimation in source regions, especially at remote locations such as the boreal forest. The manuscript should be amended to acknowledge this limitation, and if possible, to provide a crude estimate of the possible impact. More caution is advised when discussing the causes of model overestimation of isoprene levels. Among the recommendations of the paper, the implementation of a more appropriate chemical mechanism is advisable.

We thank the Referee for raising this important point. We agree that the representation of isoprene oxidation chemistry is a key factor when interpreting model–measurement discrepancies, ans that the current configuration of WRF-CHIMERE, as well as the mechanisms used in the studies cited, relies on a simplified and now outdated description of isoprene oxidation. We have revised the manuscript to explicitly acknowledge this limitation. The text now clarifies that part of the discrepancy may arise from incomplete chemical mechanisms, in addition to uncertainties in emissions factors. Specifically, the text was modified as follows (L123-124):

The SAPRC-07A chemical mechanism scheme (Carter, 2010) is used for gas-phase chemistry. This mechanism does not explicitly account for recently established low-NOx pathways such as peroxy radical isomerization and epoxide (e.g., IEPOX) formation, which enhance OH recycling and accelerate isoprene oxidation (Wennberg et al., 2018).

L508-513:

Additionally, our results confirm earlier findings that isoprene reduction can increase OA production under certain conditions, due to reduced scavenging of OH radicals and enhanced oxidation of monoterpenes. However, it is important to acknowledge that this result might be biased by the chemical mechanism used in this study. In particular, missing OH-recycling pathways in isoprene oxidation can reduce modelled OH levels and slow down isoprene loss, contributing to positive concentration biases under low-NOx conditions (Wennberg et al., 2018). This effect is expected to be relevant in boreal environments and should be considered when interpreting model–measurement discrepancies.

3) Another important aspect is the large variability in the reported isoprene emission factors for Norway spruce (see Table 2 in Hakola et al., 2023). This variability implies an uncertainty that might exceed the differences found here between different emission estimation methods. I think that this point requires a further examination and discussion of the available data.

We agree that the range of emission factors reported in the literature is substantial and may introduce an uncertainty comparable to, or larger than, the differences arising from the emission estimation methods compared in this study. We have revised the manuscript to explicitly acknowledge and discuss this source of uncertainty in the context of our results. In the revised discussion, we now emphasize that variability in species-specific EF represents a key limitation in emission modelling. L219-234:

It is important to acknowledge that for Norway spruce, reported isoprene EFs span a very wide range (0–7140 ng $g_{(dw)}^{-1}$ h^{-1}), introducing a notable source of uncertainty. This variability reflects differences in measurement approaches, environmental conditions, and site-specific factors, and represents a limitation in the current emission estimates. Consequently, the choice of a representative value (e.g., median) may influence the resulting emissions.

Minor comments

l. 31 Isoprene reacts also with O3. The relative contribution of OH, O3 and NO3 to the total isoprene sink has been estimated using models (https://doi.org/10.5194/acp-19-9613-2019, https://doi.org/10.5194/gmd-12-2307-2019).
We agree and modified the text as follows (L30-35):
Isoprene is removed from the atmosphere primarily through its reaction with the OH radical, resulting in a lifetime of about 1.4 hours under typical OH levels (2×10^6 molecule cm^{-3}). On a global scale, this pathway accounts for approximately 85% of the total isoprene sink, while oxidation by O_3 and NO_3 accounts for about 9% and 5%, respectively (Müller et al., 2019). Although isoprene can react efficiently with the NO_3 radical, the absence of night-time isoprene emissions makes this pathway overall less important (Atkinson, 2000; Seinfeld and Pandis, 2016).

l. 45 I would add that these differences highlight the complexity and uncertainties of SOA formation, as well as the importance of accurate estimation.
We agree and modified the text as follows (L48-50):
These differences highlight the complexity and uncertainties of SOA formation and thus the importance of accurately estimating BVOC emissions to simulate SOA formation mechanisms correctly in global and regional models.

l. 51 Harrison et al 2013 is not the best reference for this aspect.
We agree and changed the reference using the following studies: Barkley et al., 2013; Palmer et al., 2006

l. 65-71 I could not find any mention of isoprene overestimation over boreal forests in Zhao et al. (2024); in fact, their model was found in good agreement with measurements along the Trans-Siberian.
While it is true that they do not mention isoprene overestimation directly, Zhao et al., 2024 is cited for the results shown in Figure 2(f). The Figure shows that the model predicts isoprene to dominate across the great majority of their domain. However, the same figure also shows that observations report monoterpenes as the dominant BVOC in most locations.

l. 116-117 What are these reactions rates? Please clarify.
The reaction rate refers to the second-order rate constants for the oxidation "aging" of organics by OH radicals. We revised the text to improve the clarity as follows (L128-134):
The aging of anthropogenic secondary organic aerosol (ASOA) and biogenic secondary organic aerosol (BSOA) is parametrized as oxidation by OH radicals using effective second-order rate constants of 1×10^{-11} and 4×10^{-12} cm^3 $molecule^{-1}$ s^{-1}, respectively (Murphy and Pandis, 2009; Cholakian et al., 2018; Bergström et al., 2012; Ciarelli et al., 2024). These rate constants represent lumped chemical aging processes that convert organic gases into more oxidized, lower-volatility species.

Figure 1 legend "the shading represents the percentage of the ceell covered by forest": I cannot see this on the plot.

The forest cover is represented by transparency, with higher forest cover corresponding to lower transparency (i.e., a darker appearance). We modified the caption, substituting the word "shading" with "transparency".

l. 141 In the Supplement, it would be useful to see a plot of typical vertical profiles of the correction factors to the diffusion coefficient and windspeed.
Thank you for this suggestion; however, in our implementation, the canopy correction following Cholakian et al., 2022 is applied only to the first model layer, where both the diffusion coefficient and horizontal wind speed are multiplied by stability-dependent correction factors. As a result, no vertical profile of correction factors is explicitly represented in the model. We have clarified this point in the text to avoid confusion (L156-158):
In the final sensitivity simulation built upon the second, we added a canopy correction to the first model layer, based on the approach introduced by Cholakian et al. (2022), to account for the effects of forest canopy on dispersion (further details are provided in the supplementary material)

Figure 2. Please add color bars to each subplot. It would be interesting to compare the subplot d) with the EF from the unmodified MEGANv3.2 model.
Figure 2 is intended as a schematic illustration rather than a quantitative map, and therefore, we decided not to add any color scale. Its purpose is to highlight the different steps required to calculate the new EFs. For this reason, we have not added color bars to the subplots.

Eq. (1) Define LAIstd and LAImax.
This part of the text has been removed and completely rewritten.

l. 297. The temperature underestimation being of the order of 1 degree at night, it probably cannot explain the large flux underestimation.
Thank you for this comment. It is true that in relative terms the underestimation is large (more than an order of magnitude); however, it is important to note that these fluxes are very small in absolute terms during nighttime conditions, when isoprene emissions are inherently low. As a result, even modest absolute differences in temperature can be the cause of the underestimation during nighttime. For this reason we decided to leave the statement as is.

l. 325-330 The calculated effect of isoprene on OH might be unrealistic.
We thank the Referee for this comment. We acknowledge this limitation arising from the chemical scheme used in this study, as discussed in response to Major Comment 2. Please refer there for further details.

Fig. 10. I am not convinced that the normalized concentration plot makes much sense. The subplots a and c are sufficient for the discussion.
We removed the subplots b and d and modified the text accordingly.

l. 393-397 If typical pyrogenic compounds were measured at the site (e.g. CO or CH3CN), filtering of the BVOC dataset based on this filter could help removing the effect of fires.

We thank the reviewer for this comment. To assess the potential influence of fire emissions on the observed OA concentrations, we computed the Pearson correlation between the measured OA and CO concentrations (JJA 2017–2019, Hyytiälä SMEAR II station). As shown in Figure 1, a correlation is present, but it is low (r = 0.30, R^2 = 0.09), indicating that only 9% of the variance in observed OA can be explained by CO variability. This confirms that fire influence is not a dominant driver of OA at this site during the study period, but does not exclude an episodic and partial contribution, consistent with what is stated in the manuscript. We therefore consider the existing text to be appropriate and have not modified it.

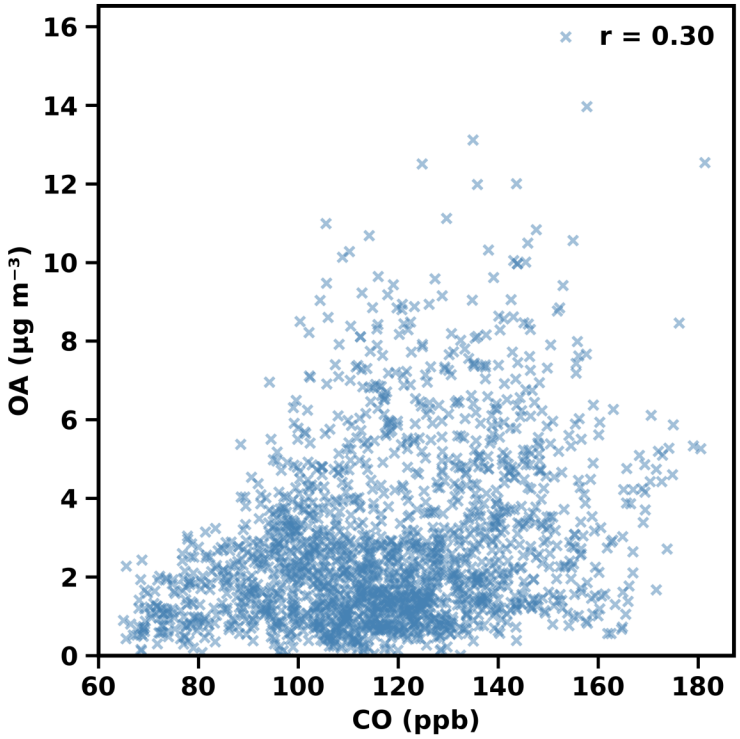

Figure 1: Scatter plot of the observed CO vs OA concentration at Hyytiälä during summer.

l. 460 This OH reduction is dependent on the chemical mechanism, which is not a strong feature of the model used here.
We thank the Referee for this comment. We acknowledge this limitation arising from the chemical scheme used in this study; a detailed discussion is provided in the discussion of Major comment 2. Please refer there for further details.

Sections 4 and 5 of the paper are somewhat long, with some repetition. I suggest revising, and possibly merging, these two sections.
We thank the referee for this suggestion. We have revised Sections 5 to reduce repetition and improve the distinction between discussion and conclusions. In particular, the Conclusions section has been shortened and streamlined to focus on the study's principal findings and

implications. However, we prefer to keep the sections separate, as the Discussion provides interpretation and recommendations, while the Conclusions summarize the main outcomes of the work.

Technical/language corrections

l. 6 "reduction in bias": not clear. With respect to what?
As stated in the original draft: *reduction in bias for both isoprene emissions fluxes and concentrations compared to previous versions of MEGAN*

l. 8 insert "the" before "vertical", and delete the word "concentrations"
We deleted the word "concentrations", but did not add the word "the" before "vertical". The new sentence is:
We further evaluate a canopy correction model to account for the effects of forest canopy on vertical and horizontal transport of biogenic volatile organic compounds (BVOCs).

l. 9 replace "additionally" by "further"
Done.

l. 10 delete the commas
Done.

l. 11 "transportation" $\longrightarrow$ "transport"
Done.

l. 11 and 14: avoid repetition
Modified as follows:
The enhanced representation of isoprene emissions and the effects of canopy on dispersion processes both result in overall improvements (although small) in SOA formation and transport. Our findings highlight the importance of moving beyond broad vegetation categories and incorporating detailed tree species distributions in emission factor calculations, demonstrating that ecosystem-specific adjustments are essential for realistic modelling of biogenic emissions and their impacts on atmospheric chemistry.

l. 16 and 17: delete "the" before "atmospheric"
Done.

l. 19 insert a comma after "globally"
Done.

l. 22 delete "the" before "biogenic"
Done.

l. 24 "among which"

Done.

l. 25 mention the formula of monoterpenes (C10H16)
Done.

l. 35 "a different range of ..." : many words, little meaning. Maybe replace by "lower-volatility compounds, which might partition to the aerosol phase..."
Done.

l. 35 Replace the comma by a new sentence.
Done.

l. 41 replace "bigger" by "higher"
Done.

l. 42 "the oxidant"
Fixed.

l. 52 delete "it"
Done.

l. 55 delete the second "emissions"
Done.

l. 56 "... factors accounting for..."
Fixed.

l. 58 The unique EF value is not just challenging, it is impossible. I think the idea is clear from the first part of the sentence.
We believe this sentence is needed to highlight the problem.

l. 59 "... in the same PFT coexist..." weird, rephrase. Maybe "the same PFT includes..."
Fixed.

l. 90 "BVOCs driven aerosol dynamics" weird wording, please rephrase.
Done.

l. 109 "the parent domain has". Same mistake on l. 110.
Fixed.

l. 158 "bigger" $\longrightarrow$ "larger"
Fixed.

l. 193 "can be chosen"
This part of text was removed and completely rewritten.

l. 258-259 The correction is not described in the methods section but in the Supplement.
Fixed.

l. 309 high-emission area
Fixed.

l. 323 "when compared to..."
Done.

l. 429 "Further improvements for future work" weird, please rephrase.
Done.

l. 456 "The absence, or too simplistic representation, ..."
Fixed.

l. 457 "in underestimated BSOA production"
Fixed.

l. 479 "shows a significant improvement"
Fixed.

References

Barkley, M. P., Smedt, I. D., Van Roozendael, M., Kurosu, T. P., Chance, K., Arneth, A., Hagberg, D., Guenther, A., Paulot, F., Marais, E., et al. (2013). Top-down isoprene emissions over tropical south america inferred from sciamachy and omi formaldehyde columns. *Journal of Geophysical Research: Atmospheres*, *118*(12), 6849–6868. https://doi.org/10.1002/jgrd.50552

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and its main drivers at northern high latitudes. *Atmospheric Chemistry and Physics*, *24*(10), 6105–6121. https://doi.org/10.5194/acp-24-6105-2024

Review of Improved isoprene emission estimates over the Finnish boreal forest using the MEGANv3.2 model

Bettineschi et al.

The authors thank the referees for their effort in reading and providing positive and constructive feedback for the improvement of this article. Below, we report in red the referees' comments and in black the authors' answers. In the authors' reply, *italic purple* is used to report text from the previous version of the manuscript, while *italic blue* is used for the updated text. All line numbers refer to the track changes version.

Answer to Anonymous Referee #3

The manuscript by Bettineschi et al. tests the effects of using tree species-specific emission factors in the calculation of isoprene emissions by the bottom-up model MEGAN and the resulting simulated concentrations of BVOCs and OA at different various heights above the ground. It also examines the possible causes of model underestimates in non-isoprene BVOC (especially monoterpenes) and OA concentrations such as lateral boundary conditions, missing emissions from wetlands and fires, and the limited representation of SOA formation chemistry. To achieve these, they apply multiple models including MEGAN v2.1 and v3.2, WRF-CHIMERE, and FLEXPART. Based on their results, the authors show the importance of accurate EFs for bottom-up BVOC emission estimate (like in MEGAN), canopy correction, other emission sources and chemistry updates. Overall, the manuscript presents an interesting study of the effect of isoprene emission updates, and is well written and relatively easy to read (despite with some difficulty in methodological descriptions). It fits the scope and requirement of the journal. Below are some suggestions to clarify the methodology and strengthen the findings of the manuscript.
We thank Anonymous Referee #3 for this comment.

Major comments

The authors have done a thorough investigation of the effect of EFs, with a caveat that the effects of activity factors are absent. I understand the focus of this study is EF, but since the activity factors could also substantially affect the resulting emissions, it would be valuable to examine the (potential imperfectness in) activity factors and discuss their potential effects on emissions. The simulated temperature and wind speed is examined already at one site, and it would be useful to look at other factors (such as solar radiation) and at other locations.
We thank the reviewer for this thoughtful comment. We agree that activity factors (AFs) can

substantially affect BVOC emissions and that their potential bias deserves discussion. We have therefore extended the meteorological evaluation to two additional sites, Kumpula and Värriö (see Figure 1), in addition to the original Hyytiälä evaluation, providing a broader spatial assessment of the model's ability to reproduce the temperature conditions that drive the AFs. Regarding wind speed, we note that it does not affect the AF formulation used in this study and was therefore not included in this evaluation. Solar radiation would have been a valuable additional variable to assess, as it is a key driver of light-dependent AFs; however, this diagnostic was not saved in the model output due to storage limitations and cannot be retrieved at this stage. We believe the extended temperature evaluation at three

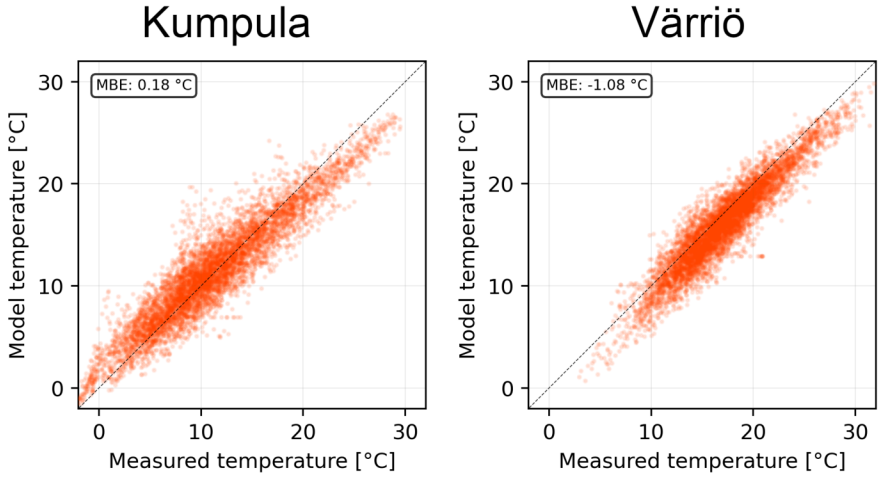

Figure 1: Observed vs modelled hourly 2-meter temperature in Kumpula (Helsinki) and Värriö.

sites provides a reasonable evaluation of the meteorological drivers of the AFs within the constraints of the available output, and we have added the Figure to the supplementary material.

I understand the ground-based measurements may not be available, but some comparison with existing assimilated meteorological datasets might still be useful to get a rough estimate the potential uncertainty in these activity factors and their impacts on calculated emissions. Given the limited number of ground measurement sites, it would be useful to include some evaluation of modeled VOC concentrations based on satellite measurements of VOCs such as HCHO, CHOCHO, and isoprene. Those data are often freely available. This evaluation would provide a more comprehensive evaluation of the uncertainty and your improvements in BVOC emissions, to complement your current results from ground sites.
We thank the reviewer for this suggestion. Following this recommendation, we compared the simulated HCHO columns against satellite-derived HCHO columns from the ESA Climate Change Initiative TROPOMI/Sentinel-5P (De Smedt et al., 2025) retrieval over Finland for the study period (we were not able to find isoprene satellite data over Finland). The results show that both the Baseline and MEG3-UPD simulations reproduce the observed spatial distribution and the probability density function of HCHO columns reasonably well (see figures below), with the MEG3-UPD simulation showing a slight improvement in the peak

of the distribution.

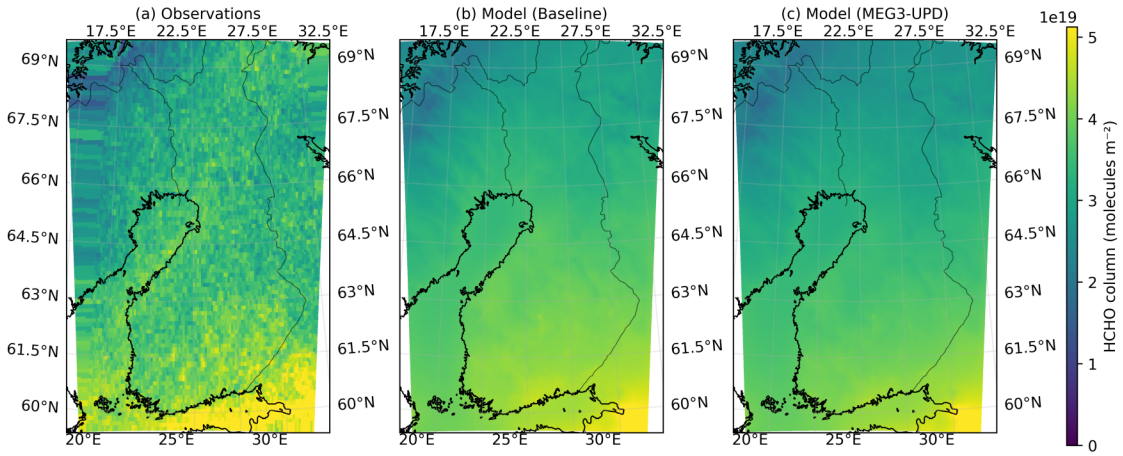

Figure 2: Average HCHO column during the summer months of 2018 and 2019 (satellite data from this source were not available during 2017).

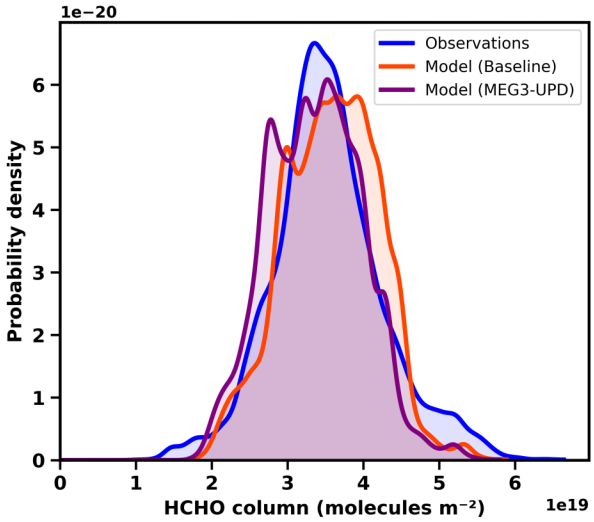

Figure 3: Probability density distribution of the HCHO column over the whole domain.

However, we have chosen not to include this analysis in the manuscript. HCHO is produced from a wide range of precursors beyond isoprene, including other BVOCs, methane oxidation, and anthropogenic emissions, and its column abundance is further modulated by transport, photochemistry, and retrieval uncertainties. Given these confounding factors, drawing robust conclusions about isoprene emission accuracy from HCHO columns alone would require a more dedicated attribution analysis that is beyond the scope of this study. We therefore consider that adding this comparison could give the impression of a stronger constraint on

BVOC emissions than is actually supported, and we prefer to keep the evaluation focused on the ground-based measurements where the interpretation is more direct. We have not modified the manuscript on this point.

Although the manuscript is overall clearly written, the methodological descriptions related to the use of SRR and SRC could be further clarified. What is the exact meaning and formula for SRR? What is the meaning of "air mass arrival time" in Eq. 3? Does SRC consider the effect of the characteristic lifetime of OA (spent from the emissions of its precursors to the deposition of OA)? If so, how? What is the meaning of omega (I guess it means some sort of spatiotemporal domain, but it should be clearly defined)?

We thank the reviewer for this comment. We address each point in turn and revised the manuscript accordingly.

Definition for SRR. The $\mathrm{SRR}(t, h; x, y, \tau)$ is the residence time (in seconds) that particles released at arrival time τ spent in the grid cell (x, y) at height h and time t during the 72-hour back-trajectory. It is a standard FLEXPART backward-mode output, computed internally by the model as the fraction of particles that resided in a given grid cell multiplied by the output time step.

Meaning of air mass arrival time τ. The variable τ refers to the time at which the particle ensemble is released from Hyytiälä (i.e. the measurement time at the receptor site). In other words, τ indexes each individual hourly release event.

Does SRC account for the characteristic lifetime of OA? The SRC as formulated here does not explicitly account for the chemical lifetime of OA or its precursors. The δ indicator function only checks whether an air mass passed through a given grid cell at any point during the 72-hour back-trajectory window, regardless of when during that window the passage occurred. The OA concentration assigned is the value measured (or modelled) at the receptor at time τ. The implicit assumption is therefore that OA measured at Hyytiälä reflects the integrated history of the air mass over the preceding 72 hours, without applying an explicit aging or decay correction. This is a known simplification of the SRC approach and is consistent with its use as a source-region diagnostic rather than a quantitative source apportionment tool.

Definition of Ω. Ω denotes the full four-dimensional spatiotemporal domain of the FLEXPART simulation, spanning the horizontal grid (x, y), the vertical levels (h), and the back-trajectory integration time (t) over the 72 hours preceding each release τ.

We modified the text to improve clarity as follows:

L248-249:

The output of FLEXPART in backward mode is the Source-Receptor Relationship (SRR), expressed in units of seconds, representing the residence time of particles in each grid cell, i.e., the time spent by particles released at the receptor in a given grid cell during the backward simulation.

L253-255:

Given a simulation domain Ω, defined as the four-dimensional spatiotemporal domain of the FLEXPART simulation including time (t), height (h), longitude (x), and latitude (y) over the 72-hour back-trajectory, and an air mass arrival time (τ), defined as the time of particle release at the receptor (i.e., the measurement time at Hyytiälä)

L264-265:

The SRC formulation does not explicitly account for the chemical lifetime of OA, and it is used here as a source-region diagnostic rather than a quantitative source apportionment tool.

In addition, the difference between leaf-level EF and canopy-level EF could be better described. What is the physical rationale of the conversion in Eq. 1? Eq. 1 suggests the converted EF to have exactly the same unit as the EF before the conversion, which appears to be contradictory to the difference between leaf-level EF (per LAI basis) and canopy-level EF (not per LAI basis) shown in Line 182-183. What are the meanings of LAI_{max} and LAI_{std}, and where are their specific values from?

We thank the reviewer for this comment. This section has been completely rewritten because Eq. 1 was incorrectly reported and did not correspond to the actual conversion used in this study. We added a new section explaining in detail the difference between MEGANv2.1 and MEGANv3.2 and the correct conversion between EF. The new section (MEGAN emission model) is as follows (L163-179):

BVOC emissions are estimated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN; Guenther et al., 2006, 2020). Both MEGANv3.2 and MEGANv2.1 share the same general emission equation:

$$E = EF \cdot EA \tag{1}$$

The two versions differ in both the definition of EF and the formulation of EA. In MEGANv3.2, the EA is defined as follows:

$$EA_{\mathrm{v3.2}} = Cce \cdot LAI \cdot \gamma_{\mathrm{P}} \cdot \gamma_{\mathrm{T}} \cdot \gamma_{\mathrm{HT}} \cdot \gamma_{\mathrm{LT}} \cdot \gamma_{\mathrm{SW}} \cdot \gamma_{\mathrm{O3}} \cdot \gamma_{\mathrm{A}} \cdot \gamma_{\mathrm{SM}} \cdot \gamma_{\mathrm{CO2}} \tag{2}$$

where Cce represents the canopy environment coefficient accounting for within-canopy light and environmental attenuation, LAI is the leaf area index, γ_{P}, γ_{T}, γ_{HT}, γ_{LT}, γ_{SW}, γ_{O3}, γ_{A}, γ_{SM}, and γ_{CO2} represent the activity factors for downward shortwave radiation, 2 meter air temperature, high temperature, low temperature, strong wind, CO_2 pollution, leaf age, soil moisture, and CO_2 concentration, respectively. In the MEGANv2.1 version implemented within CHIMERE, the EA term is:

$$EA_{\mathrm{v2.1}} = \gamma_{\mathrm{LAI}} \cdot \gamma_{\mathrm{P}} \cdot \gamma_{\mathrm{T}} \cdot \gamma_{\mathrm{A}} \cdot \gamma_{\mathrm{SM}} \tag{3}$$

where $\gamma_{\mathrm{LAI}} = 0.49\, LAI / \sqrt{1 + 0.2\, LAI^2}$.

Because EF in MEGANv3.2 is defined per unit LAI while in MEGANv2.1 it is not, the two EF datasets are not directly interchangeable. Equating Eqs. (2) and (3) at standard conditions and solving for $EF_{\mathrm{v2.1}}$ gives:

$$EF_{\mathrm{v2.1}} = EF_{\mathrm{v3.2}} \cdot \frac{LAI \cdot Cce}{\gamma_{\mathrm{LAI}}} = EF_{\mathrm{v3.2}} \cdot \frac{Cce}{0.49} \cdot \sqrt{1 + 0.2 \cdot LAI^2} \tag{4}$$

where LAI in Eq. (4) is at standard condition (i.e., 5 m^2 m^{-2})

Furthermore, different sets of experiments are shown for isoprene emissions and WRF-CHIMERE simulations, and it is hard to link the emission-focused experiments to WRF-focused experiments. To help understand these experiments, it would be good to add a table (or expanding Table 1) to describe both emission-focused experiments and WRF-focused experiments side by side.
We expanded Table 1 to include which simulation is emissions-focused and which is meteorology-focused. The new table is as follows:

Table 1: Summary of the modification between each simulation. The "Emission factor" column refers to which version of MEGAN has been used to calculate the EF. The "Tree cover + speciation" column indicates whether updated tree cover and species distribution have been taken into account or not. The "Canopy correction" column shows which simulation includes the canopy correction.

Simulation name	*Emission factor*	*Tree cover + speciation*	*Canopy correction*	*Focus*
Baseline	*MEGAN2.1*	*Not included*	*Not included*	-
MEG3	*MEGAN3.2*	*Not included*	*Not included*	*Emissions*
MEG3-UPD	*MEGAN3.2*	*Included*	*Not included*	*Emissions*
MEG3-UPD-CC	*MEGAN3.2*	*Included*	*Included*	*Meteorology*

Minor comments

The boundary layer mixing scheme, along with convection scheme and the fact that the canopy process is not well represented, could be added in Method (i.e., at the beginning of Sect. 2.1), to improve the understanding of results, particularly on the vertical profiles.
We added the following part to the text (L117-121):
Simulations were performed using the Rapid Radiative Transfer Model scheme (Mlawer et al., 1997), the Thompson aerosol-aware microphysics scheme (Hong et al., 2004), the Monin–Obukhov surface-layer scheme (Janjic, 2003), and the Noah land surface model for land surface physics (Chen and Dudhia, 2001). The boundary-layer option was the Mellor–Yamada–Janjic turbulent kinetic energy scheme (Janjic, 1994). Convection was parameterized using the Kain–Fritsch scheme (Kain and Fritsch, 1993). WRF simulations were performed on 46 vertical eta levels.

Line 174-175: "This simplified version was tested because detailed information on tree species distribution is often unavailable in many regions." – it would be good to show the extent of such limitation (for example, in the SI).
Here we do not refers to regions over Finland, where detailed tree species distribution information are available everywhere, but rather in regions around the world. We modified the text to clarify this as follows:
This simplified version was tested because detailed information on tree species distribution is often unavailable in many regions around the world.

Line 179-181: "Specifically, for Norway spruce we used the average value of all the EFs available, while for Scots pine and Silver birch we assigned the minimum value present in MEGANv3.2, as for both species extremely small EFs are reported in Finland." – add references.
We added the following reference: Lindfors and Laurila, 2000.

Figure 4: Change "+ Trees emission factor" to "+ Tree emission factor updates"
Done.

Figure 10: To improve readability, it would be better to reduce the number of colors, for example, use red color dashed line for CC Day.
We appreciate the suggestion. However, reducing the number of colors and relying on line styles (e.g., dashed lines) would decrease readability, especially when profiles overlap. Using color as the primary distinction allows clearer separation of cases, so we retained the current scheme.

Line 423-425: "Because the standard MEGANv3.2 configuration includes species information only for the contiguous United States, this limitation introduces considerable uncertainty in global applications." – This information could be already mentioned in the introduction or method section.
This information is already included in the introduction (L79-81):
However, currently, many ecotypes in the standard version of MEGANv3.2 do not contain actual tree species but only needleleaf/broadleaf divisions, which are ecotype-specific, meaning that needleleaf/broadleaf in different ecotypes are considered as different "species" with different species-specific EFs.

Line 456-457: "The absence, or more detailed representation, of LVOCs, ELVOCs, and ULVOCs in the VBS scheme used in this study can therefore lead to two main issues" – Grammar error.
Fixed.

References

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