Answer to the comments of Prof. David Fitzjarrald

We are grateful to Prof. Fitzjarrald for the detailed review and constructive criticism, which helps to improve the clarity of the manuscript. Please find below the replies to the specific comments and an account of the modifications to be implemented.

The review starts with an account of different points about the manuscript's quality, and Reviewer's assessment related to these points.

"+" = agree; "-" = disagree; "0" = neutral.

1. Does the paper address relevant scientific questions within the scope of ACP? +
2. Does the paper present novel concepts, ideas, tools, or data? -
3. Are substantial conclusions reached? 0
4. Are the scientific methods and assumptions valid and clearly outlined? +
5. Are the results sufficient to support the interpretations and conclusions? 0
6. Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)? +
7. Do the authors give proper credit to related work and clearly indicate their own new/ original contribution? +
8. Does the title clearly reflect the contents of the paper? +
9. Does the abstract provide a concise and complete summary? +
10. Is the overall presentation well structured and clear? +
11. Is the language fluent and precise? 0
12. Are mathematical formulae, symbols, abbreviations, and units correctly defined and used? +
13. Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated? +
14. Are the number and quality of references appropriate? 0
15. Is the amount and quality of supplementary material appropriate? 0

While almost all of the above items are marked neutral or positive, the Reviewer finds lack of novelty in the paper. We feel it is important to clarify this point and strengthen it in the manuscript. While the method (Lagrangian modelling) and application of Lagrangian modelling to calculate footprint functions of inert and non-inert gases are well-known, **it is the first time this method has been used for local aerosol formation**. This manuscript combines and develops the ideas from several groups: footprint calculation and boundary layer meteorology (Rannik, Rinne, Vesala), aerosol dynamics (Kerminen, Kulmala, Tuovinen), forest ecology (Lintunen, Ke) and atmospheric chemistry (Garmash, Peräkylä, Kulmala). In subsections 2.1-2.2, we explain the process of aerosol formation and discuss when and how Lagrangian models can be used for local aerosol formation. Furthermore, **we introduce source contribution function, eq. (1), which has not been used before**. Therefore, we argue that the manuscript does present novel concepts and ideas.

We also want to emphasize that footprint-based approach is not an accidental choice. Regional growth of aerosol particles within air masses was linked to temperature and forest emissions using back trajectories (e.g., Tunved et al 2006, Petäjä et al 2021); therefore, Lagrangian footprint functions, also employing trajectories of air parcels but considering forest structure influence on turbulence, was a natural choice for local formation of small particles.

1.They argue that a Lagrangian model applicable to neutral conditions for boreal forest and adjacent agricultural fields is adequate to identify how finely one can divide a landscape for the purpose of assigning source areas for growing small aerosols. They do not make clear what they mean by considering a neutral atmospheric surface layer. I write this because they do refer to what is clearly a morning convective boundary layer—definitely not neutral in terms of surface sensible heat flux. At another place, they discuss a "shallow stable" layer but such caveats about the actual atmospheric surface layer appear only toward the end of the text.

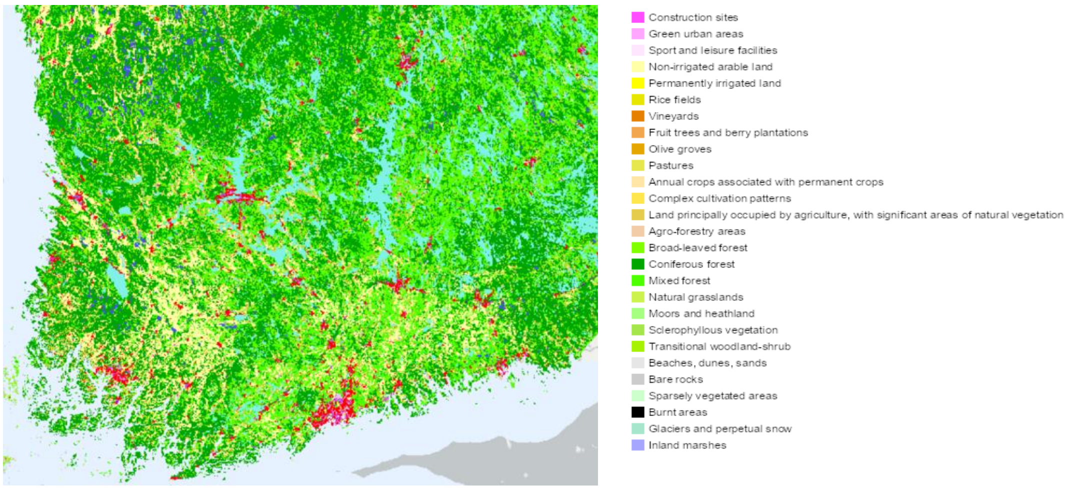
The aims of this study were twofold: 1) to outline the processes contributing to local aerosol formation in forests and agricultural fields and suggest a method for determining the relevant horizontal scales, and 2) to estimate how different horizontal scales of local aerosol formation can be in a closed-canopy forest versus open vegetation areas. The main reason for choosing neutral stratification conditions was to use the simplest possible setup.

The reviewer is correct that convective unstable boundary layer could be a good choice for daytime aerosol formation, and shallow stable layer for nighttime. However, for realistic modelling of stability impacts on turbulence fields both within and above canopy, that serve as inputs for Lagrangian stochastic dispersion modelling, a more complex model considering both radiative and turbulence transfer would be needed. Therefore, a simplified model with neutral stratification assumption was used in this manuscript. We have modified the second aim to read:

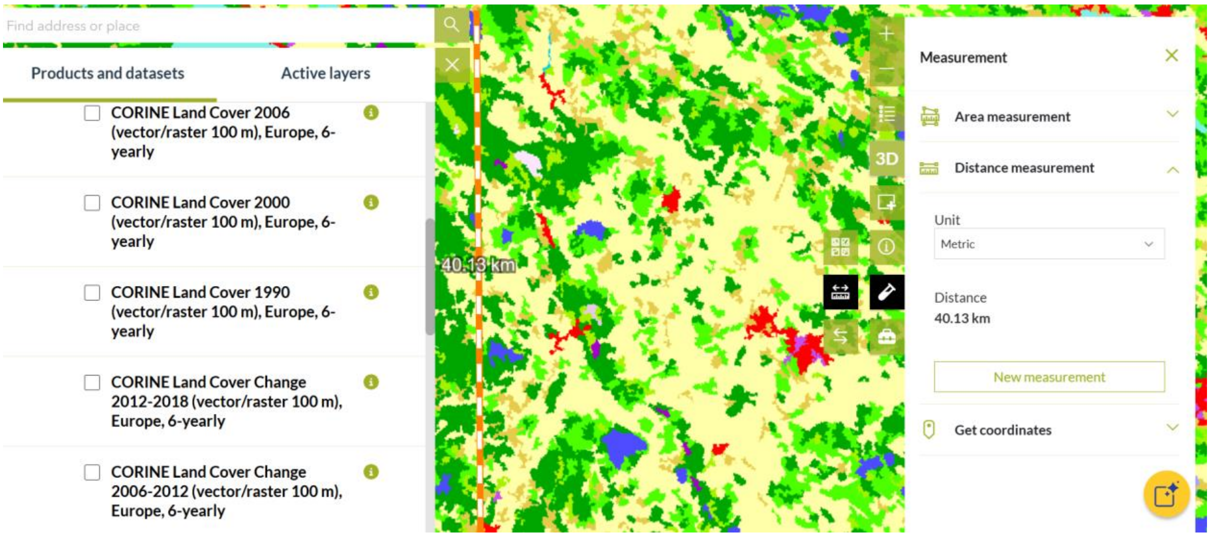
'To quantify horizontal spatial scales corresponding to the growth of ions from 1.7 to 2 nm size linked to a local ecosystem via VOC emissions **for a simple case of neutral stratification'**

An important issue may be discussion of the forest canopy structure. This is not a small point, since the emergence of aerosols from a forest canopy and into the lower atmosphere depends a lot on the architecture of the canopy and the mixing state of the atmospheric boundary layer. Complementing this there needs to be information about the 'typical' area of the patches that constitute the landscape mosaic of forest, wetland, and agricultural field. Do not leave this only to references. This paper is strewn with references, but the reader shouldn't be sent scurrying to other texts to find this basic information essential to this paper.

We are grateful to the reviewer for raising this important point. Forest canopy structure including leaf area density profile and LAI is addressed later. Below one can see the map obtained from EGU CORINE cover 2018 data set (vector/raster 100 m) and focused on southern Finland, where both our reference sites (Viikki and Hyytiälä) are located (https://land.copernicus.eu/en/map-viewer?dataset=0407d497d3c44bcd93ce8fd5bf78596a, accessed 25.05.2026).

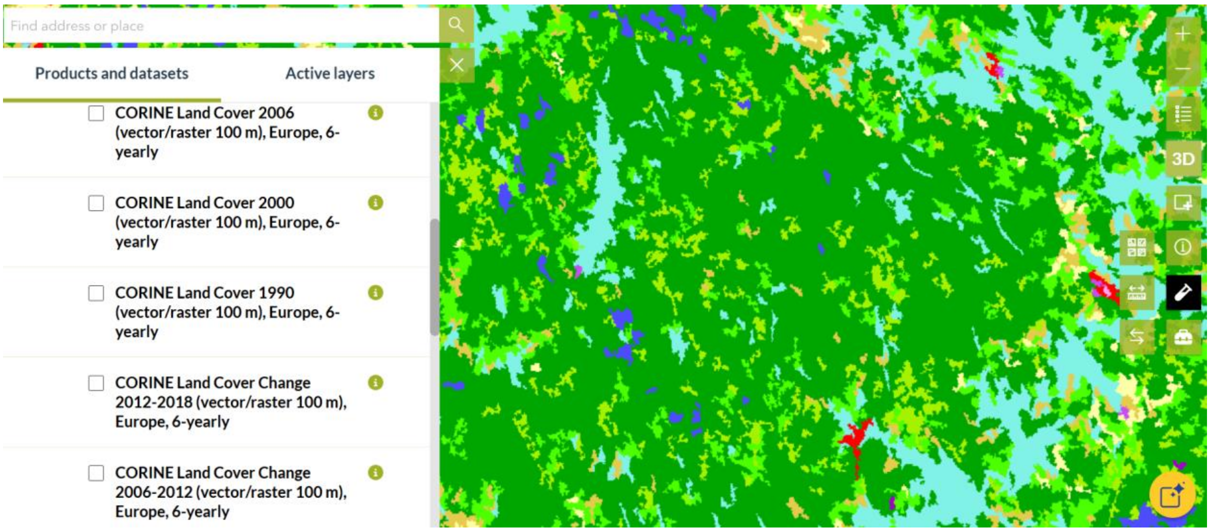


The prevailing colors in the figure are green (forests: dark – coniferous, two shades of light green – mixed or broad-leaved), yellow (non-irrigated arable land), light-blue (lakes and rivers), red – urban and in the upper left-hand corner one can notice blue patches (wetland). One can clearly see that forests are prevailing in Finland, whereas in the south-western part, more agricultural land can be noticed.



This is the map zoomed on agricultural land-dominated part. One can see there are also darker yellow-brown patches which are lands principally occupied with agriculture but with significant natural vegetation. Even in yellow-dominated part, there are small inclusions of forest and wetland. As the total vertical scale of the zoomed map is 40 km, one can estimate that most of the yellow patches would have characteristic scales of 1.5-2 km but they are elongated in some directions up to ca 10 km.

Zooming on a forest-dominated part of the same size, one can notice that larger patches can be found of ca 5x5 km size, and there are inclusions of small lakes and wetland. In addition, also forests are inhomogeneous, in this area dominated by coniferous but with inclusions of mixed/broadleaved forest areas. The scales of patches of homogeneous coniferous forest could be estimated ca 2-3 km.



So all in all, based on this not very precise analysis, the sizes of patches with different land use in southern Finland are typically quite small, roughly on the order of 1-10 km, with a larger fraction of the patches with smaller scales. We have modified the sentence about landscape mosaic in the manuscript to read:

‘A typical boreal landscape, e.g. in Finland, is a mosaic of agricultural fields, wetlands and especially forests representing patches with scales roughly on the order of 1-10 km, skewed to have a larger fraction of the patches at the lower end of this range’.

We agree with Prof. Fitzjarrald that local aerosol production depends strongly on both structure of vegetation and the state of boundary layer, but it depends also on vegetation and soil emissions, i.e. plant species and soils, and presence of oxidants. To address the issue, we have established recently about 30 new observational sites in Finland and more in Denmark, targeting forests of different age/structure/species/soil, and agricultural land with different management practices and different cultures, focusing on aerosol measurements. The goal is to better understand the effect of different land use on aerosol production and its connection to the meteorological conditions.

The authors need to convince this reader that that the proximity of the origin of the aerosol is essential to understand the consequent density of larger particles. Presumably, this cohort is to provide a basis for growth to larger sizes that may be important cloud condensation nuclei or act to diffuse incident solar radiation, which some believe to be the source of a ‘diffuse fertilization effect’ on subsequent carbon uptake (e.g., Durand et la., 2026), though this idea has a competing hypothesis related to shallow clouds and shading (e.g., Kivalov and Fitzjarrald, 2019). In either case the role of aerosols is essential to either diffuse enhancement or cloud formation.

While this cohort is to provide the basis for the growth to larger sizes, and the link between local and regional aerosol particle formation does exist, this link is not 1:1 but rather R = 0.6, as the reviewer has noticed in his other comments. The growth of aerosol population through small sizes can be interrupted because of, e.g., lack of condensing vapours to grow the particles to larger sizes or large amount of pre-existing aerosol in the air which acts as a sink for small particles. Previous studies showed that the bottleneck size for newly born aerosol particles to survive is about 2.5-5 nm (Aliaga et al, 2023, Garmash et al, 2024). Large number of particles in this size bin is clearly associated with strong regional new particle formation events in boreal forests (Aliaga et al 2023, Lampilahti et al, 2025). Simple correlation does not work also because different growth rates make the peaks of concentration in these different size ranges occur with varying time delays. To overcome these difficulties, similar to these abovementioned manuscripts, we introduce a metric, which assesses the difference between the concentration peaks and low background values separately for each day, . We have decided to add in the manuscript figures (Fig. A1 in the end of this document), which show a positive statistically significant correlation between 2-2.3 nm ions and 2.5-5 nm aerosol particles at four sites including forests and agricultural fields, where we have longer data sets (two co-authors to be added due to new data included, J. Lampilahti and M. Lampimäki). We have added the Methods related to the derivation of these figures as Appendix. The correlations differ especially between spring and other seasons (besides the most northern Värriö forest, where conditions similar to spring at other sites are observed in June), with more particles growing in spring. Given the seasonal difference between sources and sinks, the positive association is expected but for the reasons indicated above, it involves other processes to be considered, and moderate correlations should not be surprising. Another study, confirming correlations between formation rates of ions at 2.0-2.3 nm and aerosol particles at 3-5 nm based on Hyytiälä and Beijing data is under review in Environmental Science and Technology (Tuovinen et al., under review).

As for diffuse fertilization effect, separating aerosol from cloudy conditions, we can see the increasing photosynthesis with aerosol load in the absence of clouds too. However, recent analysis where machine learning was used to identify patterns in the data showed that at a certain level of diffuse radiation associated with onset of patchy cumulus clouds, there is a disruption in the monotonic increase of gross primary production at the ecosystem level with diffuse radiation, looking more like a jump increase. It could probably be due to another mechanism contributing, which Prof. Fitzjarrald mentions. We will have the reference in mind for the future.

To reiterate: distinguishing the landscape among agricultural fields, wetlands and boreal forest is important to the premise of the paper. To understand better the importance of one land use type compared to another the authors should give some examples of the relative fraction of each across the landscape. Quantify the spectrum of land cover areas that your 'mosaic' exhibits. How big are your 'patches' in the areas of Finland where they work? I've worked in the boreal forest in northern Quebec, for example, and the relatively sparse tree density might have a distinctly different 'aerosol signature' than where the current focus is. (See photo below).

Specify how to describe the mosaic of land cover types in the regions on which they focus.

a. *Why* is it important to distinguish between 'local' and 'regional' spatial scales? The authors aim to improve over crude estimates of the 'aerosol footprint'. Can the reader conclude that there might be a linear combination of the constituent landscape mosaic features to assess the overall population of reactive aerosols that emerge?

b. *Where* is their definition of these scales, 'local', 'regional'?

c. *What* are the characteristic scales in the region around where their example measurement are made. This needs to be specified in this article. Perhaps somewhere in the many references this information is given, but simply referring to '*boreal forest*' and '*agricultural use'* is insufficient.

d Line 30: "Indeed, the concentration of ions in the 2-2.3 nm range demonstrates a reasonable correlation (R = 0.61) with the concentration of 3-6 nm particles …" Perhaps explaining 36% of the variance (r^2!) is 'reasonable' to the authors, but this reader is skeptical.

All in all, to address the reviewer's comments about the link between local and regional NPF, we have produced a table below to be added in Introduction in the revised manuscript.

Table 1. The difference between local and regional aerosol formation.

Variable	Regional aerosol formation	Local aerosol formation
Time and spatial scales	100-500 km, ~10 h	50 – 5000 m, <30 min
Size of aerosol particles produced	climatically relevant size, cloud condensation nuclei (>50 nm)	>2.0 nm
Air layers involved	air mass scale, the whole boundary layer is important, forest/agricultural land simply as a rough surface	'footprint' scale, inside-forest turbulence and canopy effect on turbulence are important, atmospheric surface layer+ lower boundary layer
Plant emission contribution	dominating vegetation in the region is important, well-mixed VOCs and vapours in the air mass	contributions from an ecosystem and its different components
Compounds contributing to aerosol growth	sulfuric acid, ammonia, oxidants +ELVOCs, HOMs + semi-volatile vapours	sulfuric acid, ammonia, oxidants +ELVOCs, HOMs
Meteorology	synoptic scale processes	boundary layer and inside-canopy stratification

3-5 nm aerosol size is 'mesoscale' in this case, with particle growth to this size at the time scale of ~1 h, and all the complex effects from the interactions between the ecosystems, changes in roughness, and day and night transformations of boundary layer take place and define aerosol dynamics.

While the table and answers above address most of the comments made by the reviewer, the comment dealing with linear combination (point *a*) remains. It is certainly not that simple. Too small land use patches, strong wind conditions, well-mixed convective boundary layer contributing to increasing vertical scales and dilution, low concentrations of oxidants and weak VOC emissions in the atmosphere will all work against this simplification. Atmospheric chemistry and meteorology, together with size of patches and vegetation density will ultimately define when this is a valid assumption. Putting it simple: closed-canopy forest, strong VOC emissions, abundance of oxidants in the atmosphere and light winds or stable stratification will contribute to localized aerosol formation while open vegetation, weak emissions, small amount of oxidants and strong winds will do the opposite.

Please justify why the whole effort is done for relatively light wind conditions in a *neutral stability* atmospheric boundary layer, something that rarely occurs in nature. Is this approach common in this field of has this been done because it is complicated to implement the Lagrangian model for conditions with stable layers present? *Please* include some more extensive explanation why this approach is adequate to support the arguments made. That is, how can such an assumption be defended, even as we suspect that the neutral model case may be standard starting point for Lagrangian model efforts.

Neutral stability was chosen indeed because it is the simplest possible model setup for the forest, this statement we now added in the manuscript. The wind speed with friction velocity of 0.4 m/s is a typical average value in May-September at Hyytiälä forest (e.g., Supplementary Fig. S2 to Launiainen et al, 2022).

Neutral observation: The plethora of abbreviations (e.g., SA, for sulphuric acid) saves space, but it sends any reader whose specialization is somewhat distant from that of the authors scrambling back and forth through the text. I see no clean alternative to the authors' approach.

We are grateful to the reviewer for pointing this out and changed the abbreviation SA to H_2SO_4 throughout the text. We have also chosen to use full words instead of abbreviations in Subsection 2.1, containing many abbreviations used in atmospheric chemistry.

In the Introduction, the authors put forward wave after wave of generalizations, supported by many references, about the time scale for aerosol new particle growth from ≈1 nm to ≈1.7 nm. This reader would welcome more information about the significance of this process. A few sentences would relieve the reader from mystery, presumably because of the importance to climate alteration by the presence of larger particles. It wouldn't be too difficult to lay this out more clearly. In the Introduction CCN are mentioned—are these particles likely to be cloud condensation nuclei?

In the range outlined by the reviewer, 1.0-1.7 nm, atmospheric molecules form clusters. Atmospheric clustering is a continuous process, so there is always a distribution of aerosol particles in sub2-nm range. However, the growth to larger sizes occurs only occasionally and first as 'activation' of the particles from approximately 1.7 nm by condensing vapours, which in boreal environment include highly oxidized organic molecules. At the same time, growing particles are also lost to pre-existing aerosol population, condensation sink. Previous studies showed that survival through 3-5 nm size range is associated with further growth to larger sizes. After 50 nm, the particles can be considered to reach climatically relevant size, meaning that they can serve as cloud condensation nuclei. The description of the process of aerosol growth has been added in the Introduction.

The importance of the physical environment is shunted to the side much too quickly. This makes one wonder how the authors can be so confident in dealing with 'neutral stability', a situation that occurs quite rarely near the surface over the diurnal cycle. It would be grand if the authors would include a modest amount of text to explain why neutral stability was chosen. (As noted earlier, I suspect that this was done to facilitate using the Lagrangian model of the footprint function. If so, say so.) To an aging student of the turbulent boundary layer, the presence of the forest canopy brings in not only a 'displacement height' for momentum absorption, but also brings a possibility of an stable inversion at canopy top, both day and night, though these would be of quite different character. On top of this, there is the roughness sublayer above the forest.

Yes, the reviewer is correct that neutral stratification was chosen because we need turbulence statistics for the Lagrangian stochastic particle dispersion model. For inside canopy turbulence statistics we used 2nd order closure model by Massman and Weil (1999) (reference provided in Appendix B), which is limited with neutral stratification only. The neutral stability aspect has been addressed in the previous comments and added in the manuscript. Note however, that in the manuscript, we address both below-canopy and above-canopy processes, and the turbulence model accounts for the presence of canopy, which indeed has a profound effect on the source contribution functions, with above-canopy scales being much longer than below-canopy.

Specific comments

1. Line 6. "Here, we consider forest and agricultural ecosystems, and distinguish situations in which aerosol production is relatively slow and vertically distributed within the well-mixed boundary layer and when it can occur quickly close to the surface." What 'well-mixed boundary layer' is this? Is it the convective boundary layer during the day, perhaps up to 1 km in thickness? Is it the free convection surface layer, up to 50 m above the surface or a 'roughness sublayer' above canopy top?

It is the first, convective boundary layer or simply boundary layer of about 1 km high. We added this clarification in the manuscript.

2. Lines 20-22: Again the authors invoke for turbulent fluxes are important to describe evapotranspiration, cloud formation and the like, but these are convective boundary layer issues, with appreciable surface heat flux.

This is a general introduction to the topic about forest influence on boundary layer clouds.

3. Line 34: Is r2 ≈ 0.4 a 'reasonable correlation' in this line of work? Just explain, thanks.

We have explained in the previous comments why we consider this a reasonable correlation. The explanation is added also in the manuscript.

4. Line 70. "We limit the current study to a neutral atmosphere, representing growing season conditions with moderate winds and cloudiness." In what world does a neutral atmosphere represent growing season conditions? This sounds like the authors are trying to deal with a forest canopy beneath a convective boundary layer, which has a nearly adiabatic lapse rate—some might call that 'neutral', but the whole layer is motivated by mixing via buoyant eddies. This surely alters the footpring, Langrangian or otherwise.

Atmospheric stability does alter the concentration footprint, which is well illustrated in the paper by Kljun et al. (2002). At the same wind speed, concentration footprint has a maximum closer to measurement point under convective turbulence conditions and this maximum is not pronounced at all under stable conditions. The curve corresponding to neutral stratification is between those two. However, the source contribution function, as compared to inert scalar concentration footprint, depends also strongly on the growth rate of aerosol, which is determined by the presence of VOCs and oxidants. It can be large under stable conditions and make the local scale of aerosol formation smaller than under convective conditions.

Neutral curve is the case between the two. However, all these footprint functions do not converge, and in that sense they will not have as strong influence as aerosol distribution or below-canopy change in turbulence. It is wind speed, growing part of sub-2 nm distribution and growth rate that define the scale of aerosol formation.

5. Line 186. Please explain more clearly why growth from 1.7 nm to 2nm is so important.

This has been explained in the previous comments and we added this paragraph in the Introduction as well.

6. Line 234. The forest height is 20 m. Do aerosol particles pass out of the canopy going against no stable layer?

Aerosol particles can certainly pass out of the canopy.

7 Line 270: "Oppositely, for the measurement heights above the canopy, even at 1 m above canopy top, the peak is much less pronounce…"d due to developed turbulence above the vegetation in neutral stratification…"Why do the authors insist on neutral stratification?

We understand this is a simplification, but we had a model in our usage for neutral stratification.

8 Lines 310, 327: Please repair the text: "Fig. ??"!'

Thank you for noticing, it should be Fig. 8. This is now resolved.

9. Line 362. Here is the first time the authors own up: "The effects of meteorology should be considered further in future studies. An especially striking example is the decoupling in the evenings when temperature quickly drops, …" Please include this honesty also earlier in the paper, in the Introduction.

The importance of atmospheric stability for the spatial scale of local aerosol formation is mentioned in the manuscript already in the Introduction, in L 55-56:

'Meteorological conditions, such as atmospheric stability and wind speed (and direction, in case of inhomogeneous land cover) will also be important'.

Now we state in the end of Introduction that the study is limited to neutral atmosphere for simplicity. Furthermore, L 142-156 in Subsection 2.1 also include considerations of atmospheric stability.

10. Line 365: "If there are on-going emissions of VOCs from any nearby sources, dilution of these components is inefficient in a shallow stable boundary layer… " We have to wait until near the end of the paper to be reminded that the analysis is severely hampered by not taking into account the stable conditions, which can exist not only just above the canopy at night, but also within a closed canopy during the daytime. This confusion could be removed by discussing earlier in the paper what is the canopy density vertical profile (leaf and branch).

We agree with the reviewer that atmospheric processes, including dispersion, are strongly affected by atmospheric stability. However, in this study the source areas of small particle production are of interest, and it will have even a stronger impacting factor which is the aerosol growth rates. To study this impact, we have used a model that limits as to neutral atmospheric stability. We acknowledge that further investigations would benefit from including the effect of atmospheric stability. The density vertical profile is proportional to leaf area density in Hyytiälä forest, and included in the Appendix.

11. Line 420 equation. Please define Q. Or did I miss something?

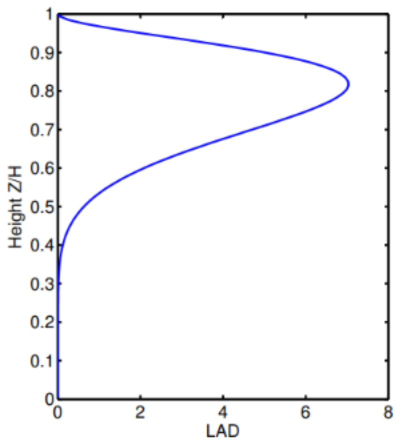
Thank you for noticing: Q is the source of extremely low volatile organic vapours (ELVOC), we added this in the manuscript.

12. Line 443. Choosing a 1km convective boundary layer over the agricultural field and a 2km one over the forest is another thing that depends on the size of the patches. If the forest is a small patch, for example, the boundary layer won't show that great a change. Please comment in the text.

For the estimates here, we have assumed homogeneous surface cover, so effect of small patches is not considered. This is added now in previous L 443.

13. Do you have dense, or a closed boreal forest canopy? How does the 'Lagrangian footprint' vary depending on the canopy closure?

Hyytiälä forest is a closed-canopy forest of intermediate to low leaf density. We simulated LAI of 2.5, with LAD profile shown in the figure. According to fig. S2 in Launiainen et al, 2022, LAI = 2.5 corresponds to pine/spruce, which are dominant species in Hyytiälä (Kolari et al, 2022, tree density data).



We added information about closed canopy in Subsection 2.3, in Model calculations setup, and LAD profile and LAI in Appendix B. We have not considered the effect of the canopy closure in this paper, but it can be expected that open-canopy forest is a case between open vegetation and closed-canopy forest. Thus, for the same wind/stratification conditions, the 'footprint' area will increase from the case of the closed canopy.

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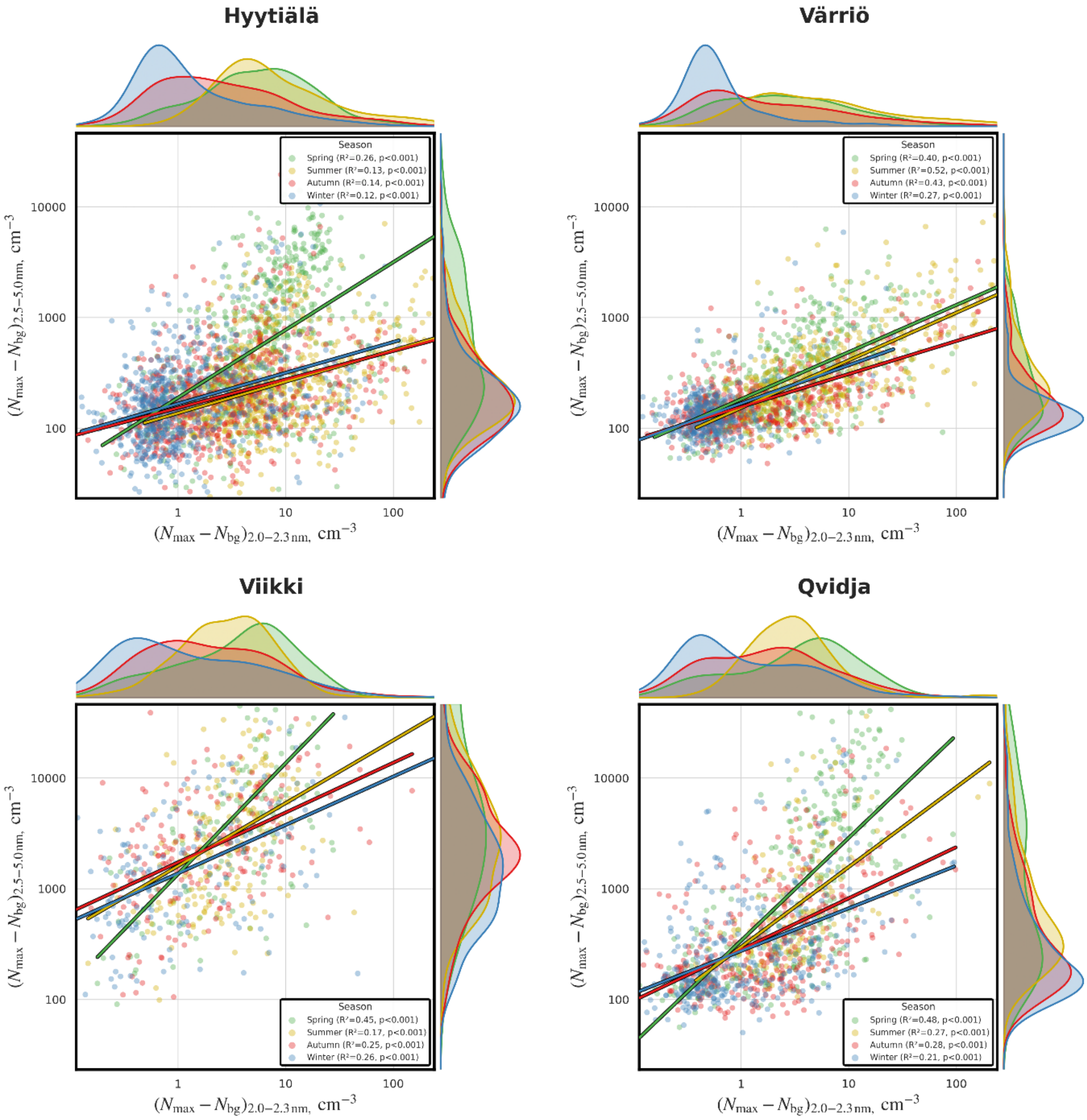

Fig. A1. Correlations between the peaks in number concentration relative to their respective background values, daily time scale: x-axis 2-2.3 nm ions, y-axis 3-5 nm aerosol particles.