Potential of carbon uptake and local aerosol production in boreal and hemi-boreal ecosystems across Finland and in Estonia

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This manuscript explained and researched the  $CO_2$  uptake and local aerosol production at different types of stations in Finland. Authors analysed each NEE and  $N_{neg}$  characteristics at each station and compare between them. Introduction is very persuasive and interesting why this study is necessary. However, the way to describe the result of experiments is not kind to the readers. Result was simple and it seems like lack of discussion. I also wonder whether the conclusion gives the answer to the question in the Introduction section. Hopefully authors read the manuscript carefully with reader's view and revise/describe them explicitly.

We are grateful for insightful comments and suggestions provided by the reviewer. All comments are addressed below and have significantly improved our manuscript.

We would like to make a note that we have corrected the measurement period at Haltiala croplands to '06/2021-10/2022' from '01/2022-12/2022'. All available data for both NAIS and CO<sub>2</sub> fluxes at Haltiala cropland are from 2021/06-2022/10 so far. We modified the text and the code following the corrected date range. After revision, the midday CO<sub>2</sub> uptake rate and  $N_{neg}$  in summer in Haltiala cropland decreased from 19.69 µmol m<sup>-2</sup> s<sup>-1</sup> and 3.08 # cm<sup>-3</sup> to 10.42 µmol m<sup>-2</sup> s<sup>-1</sup> and 2.66 # cm<sup>-3</sup>, respectively. The related figures and tables were all revised. However, the main conclusions remained without changes. Detailed responses to the comments can be found below.

#### General comments

 Title mentioned 'boreal and hemi-boreal ecosystem' but all manuscripts are not linked to boreal and hemi-boreal region. Authors did not explain which site is belonging to boreal or hemi-boreal. According to the category in Table 1, there are forest, agricultural, peatland, urban garden and coastal area. It was hard to match to the title. Authors should consider how to make this manuscript well match to the title and more interesting.

Reply: We have now added references assigning climate zones to these sites, in line 117 "F-JAR, C-TVA, and A-QVI belong to hemi-boreal ecosystems, while the other ecosystems are boreal (Mäki et al., 2022)". In Table 1, the climate zone of each station is added.

2. Authors did not make station ID and mentioned the station name directly (Table 1). But readers are not familiar with Finish and hard to follow up what type of station with only station's names. Hope authors reconsider the way to explain station' name.

<u>Reply: We now have added the station ID for each site and revised the name throughout</u> the manuscript. We hope this is clear now.

<u>Sites</u> (Site ID)		Locatio <u>n</u>	<u>Selecte</u> <u>d</u> period	<u>Mean air</u> <u>temperat</u> <u>ure (°C)</u>	<u>Rainfall</u> (mm/yr )	Dominant plant species	<u>Peak</u> LAI	<u>Climate</u> Zone
<u>Forest</u>	<u>Hyytiälä,</u> <u>SMEAR</u> <u>II (F-</u> <u>HYY)</u>	<u>61°51'</u> <u>N,</u> <u>24°17'</u> <u>E</u> (7°46)	<u>11/200</u> <u>9-</u> <u>12/202</u> <u>2</u>	<u>4.8</u>	<u>709<sup>1</sup></u>	Scots pine and Norway spruce	<u>4.6</u>	<u>Boreal</u>
	<u>Värriö,</u> <u>SMEAR</u> <u>I</u> (F-VAR)	<u>67°46'</u> <u>N,</u> <u>29°35'</u> <u>E</u>	<u>3/2019-</u> <u>12/202</u> <u>2</u>	<u>0.4</u>	<u>601<sup>2</sup></u>	Scots pine	<u>3.2</u>	Boreal
	<u>Ränskälä</u> <u>nkorpi</u> (F-RAN)	<u>61°10'</u> <u>N,</u> <u>25°16'</u> <u>E</u>	<u>4/2021-</u> <u>12/202</u> <u>2</u>	<u>5.4</u>	<u>600<sup>3</sup></u>	<u>Norway</u> <u>spruce,</u> <u>Scots</u> <u>pine,</u> <u>downy</u> <u>birch</u>		<u>Boreal</u>
	<u>Järvselja,</u> <u>SMEAR</u> <u>Estonia</u> (F-JAR)	<u>58°16'</u> <u>N,</u> <u>27°16'</u> <u>E</u>	$\frac{\frac{10}{201}}{\frac{6}{202}}$	<u>6.8</u>	$\frac{500}{750^4}$	<u>Birch</u> <u>species,</u> <u>Scots</u> <u>pine,</u> <u>Norway</u> <u>spruce</u>	<u>6</u>	<u>Hemi-</u> boreal
<u>Agricultural</u> <u>fields</u>	<u>Haltiala,</u> <u>SMEAR</u> <u>Agri</u> (A-HAL)	<u>60°16'</u> <u>N,</u> <u>24°57'</u> <u>E</u>	<u>6/2021-</u> <u>10/202</u> <u>2</u>	<u>6.5</u>	<u>700<sup>5</sup></u>	<u>Oat</u>	<u>5.5</u>	<u>Boreal</u>
	<u>Qvidja</u> (A-QVI)	<u>60°18'</u> <u>N,</u> <u>22°24'</u> <u>E</u>	<u>12/201</u> <u>8-</u> <u>8/2022</u>	<u>7.0</u>	<u>679<sup>6</sup></u>	<u>Timothy,</u> <u>meadow</u> <u>fescue</u>	<u>6.2</u>	<u>Hemi-</u> boreal
	<u>Viikki,</u> <u>SMEAR</u> <u>Agri</u> (A-VII)	<u>60°13'</u> <u>N,</u> <u>25°01'</u> <u>E</u>	<u>7/2022-</u> <u>6/2023</u>	<u>6.5</u>	<u>792<sup>5</sup></u>	<u>Timothy</u> (2022), <u>Barley</u> (2023)	<u>5.2</u>	Boreal
Peatland	<u>Siikanev</u> <u>a.</u> <u>SMEAR</u> <u>II</u> (P-SII)	<u>61°50'</u> <u>N,</u> <u>24°12'</u> <u>E</u>	<u>11/201</u> <u>9-</u> <u>12/202</u> <u>2</u>	<u>5.0</u>	<u>710<sup>7</sup></u>	Moss and sedges	<u>0.6</u>	<u>Boreal</u>
<u>Urban</u> garden	<u>Kumpula</u> <u>SMĒAR</u> III (G- <u>KUM)</u>	<u>60°12'</u> <u>N,</u> <u>24°58'</u> <u>E</u>	<u>5/2016-</u> <u>12/202</u> <u>2</u>	<u>6.3</u> <sup>5</sup>	<u>731<sup>5</sup></u>	Mixed		<u>Boreal</u>

Coastal area	<u>Tvärmin</u> <u>ne (C-</u> <u>TVA)</u>	<u>59°51'</u> <u>N,</u> <u>23°15'</u> <u>E</u>	<u>6/2022-</u> <u>8/2023</u>	<u>7.2<sup>5</sup></u>	<u>639<sup>5</sup></u>	<u>Seagrass</u> <u>and</u> <u>seaweed</u>		<u>Hemi-</u> boreal
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3. In many paragraphs, authors did not suggest table/figure # but also values. For example, *L367-369: The momentary net CO<sub>2</sub> uptake rate at midday in summer was highest in agricultural fields, followed by the forests. The urban garden in this study displayed distinct net CO<sub>2</sub> uptake, lower than the forests and higher than the open peatland. Next to the highest, it needs value. Lower than or higher than, authors can suggest certain values. Without certain values, all explanations such as "lower than, higher than, highest, similar" are very boring and insufficient to deliver what authors would like to say.* 

Reply: We have now revised the descriptions throughout the manuscript and added exact values for the differences, such as in lines 371-378: "The agricultural fields exhibited a 47% higher  $N_{\text{neg}}$  compared to the studied forests. In contrast, the open peatland (P-SII) had a 23% lower  $N_{\text{neg}}$  than F-HYY but 15-46% higher  $N_{\text{neg}}$  than the other forests. The  $N_{\text{neg}}$  at the coastal area was the lowest. The momentary net CO<sub>2</sub> uptake rate at midday in summer was highest in agricultural fields, followed by the forests. The urban garden in this study displayed distinct net CO<sub>2</sub> uptake, 37% lower than the forests and ~2 times that in the open peatland. The coastal area at midday in summer was a very weak CO<sub>2</sub> sink. In the urban garden area in Kumpula, median  $N_{\text{neg}}$  was double of that in Hyytiälä forest, while the median NEE only reached 63% of that in Hyytiälä forest". For general descriptions, such as in Line 271, "For median values in summer,  $N_{\text{neg}}$  was found to be the highest in the urban garden, followed by the agricultural fields (Figure 9)", we would like to follow the present style to keep the manuscript concise.

# 4. Hope authors can describe the sufficient reason after the result, for example, in section 3.3. This is scientific paper not a report.

Reply: In this study, we aimed to present the different potential of  $CO_2$  uptake and aerosol production across different ecosystems; hence we reported directly measured  $CO_2$  fluxes and local aerosol production, indicated by the negative ions at 2-2.3 nm. We focus on the comparison of potentials and have now briefly investigated the reasons that could cause the difference in NEE and  $N_{neg}$  between the ecosystems. We have revised Figure 9 and added a corresponding discussion in two paragraphs in Section 3.3 (Line 396-426):

"Multiple factors can cause the difference in NEE and  $N_{neg}$  across the sites despite the similar seasonal and diurnal variation patterns. The CO<sub>2</sub> uptake rate at midday in summer increased with an increasing air temperature in both studied forests and agricultural fields (Figure 9). Moreover, the CO<sub>2</sub> uptake rate at midday in summer increased with LAI across the studied forest ecosystems (Table 1 and Figure S9). As F-RAN was selectively harvested (Section 2.3), the leaf area was decreased, which can result in a lower CO<sub>2</sub> uptake rate than other forests under similar air temperature and PPFD. Additionally, the peat soil at F-JAR and F-RAN can induce higher respiration (Figure 2). Hence, even though the LAI and air temperature at F-JAR were 23% and 10% higher than that in F-HYY, respectively, the NEE at F-JAR was only 4% lower than that at F-HYY. In the agricultural fields, the LAI and air temperature were comparable or higher than that in the forests, which may explain the high momentary  $CO_2$  uptake rate at summer midday in the agricultural fields.

In the case of  $N_{\text{neg}}$ , the precursor of aerosol production largely influences  $N_{\text{neg}}$ . The trends of N<sub>neg</sub> varying with air temperature and radiation were not evident (Figures 9 and S9). H<sub>2</sub>SO<sub>4</sub> formation can drive the nucleation process and is influenced by the sulphur dioxide concentration and radiation. As the garden area and agricultural fields in this study are located in or nearby cities, the SO<sub>2</sub> concentration there may be enhanced due to the anthropogenic pollution and its long-range transport. Also, the terpene emissions can initiate NPF, which has been observed in P-SII and led to stronger NPF there than that in F-HYY (Junninen et al., 2022; Huang et al., 2024). However, these events were reported to occur mostly in the late evening. Different plant species can emit different types of BVOCs (Guenther et al, 2012), e.g., monoterpenes are found dominant in coniferous forests and isoprene dominant in broadleaf forests. The oxidation products of monoterpenes can enhance aerosol formation and growth (Rose et al., 2018), while isoprene has been reported to inhibit new particle formation (Kiendler-Scharr et al., 2009). As birch species are mixed with coniferous species in F-JAR, the possibly higher isoprene emission than in the other three predominantly coniferous forests may partially explain the lower  $N_{\text{neg}}$  in F-JAR. Moreover, the enhanced NH<sub>3</sub> in agricultural fields can play a synergistic role with both H<sub>2</sub>SO<sub>4</sub> and low volatile organic compounds in clustering (Dada et al., 2023), which may explain the generally high N<sub>neg</sub> in the three studied agricultural fields."

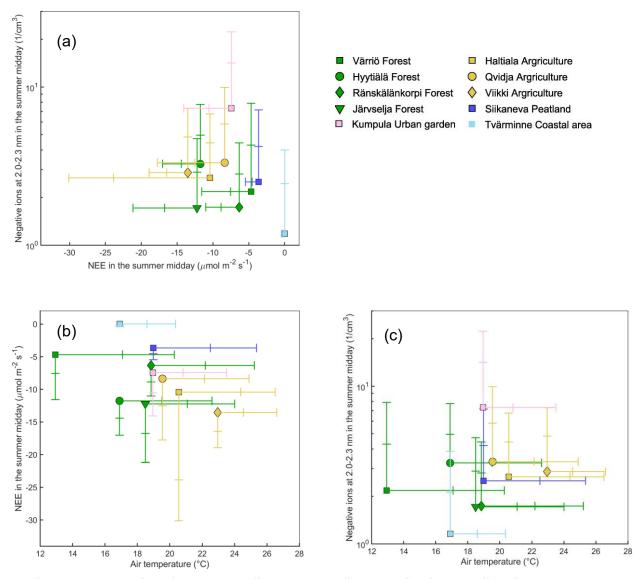
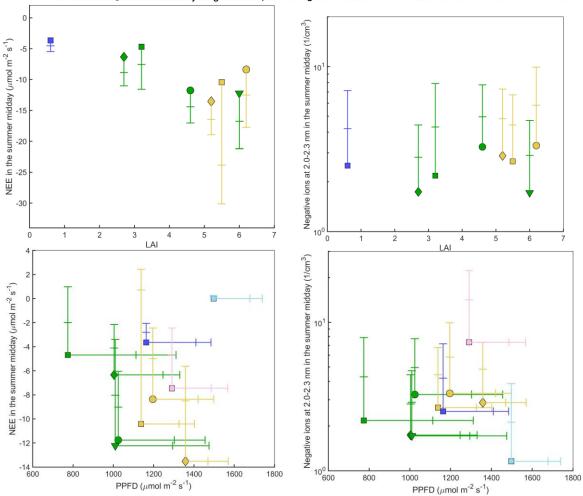


Figure 9. Comparison between median NEE, median negative intermediate ions at 2.0-2.3 nm, and median air temperature at midday in summer between the sites. The error bars are 10<sup>th</sup> and 25<sup>th</sup> percentile for NEE, 75<sup>th</sup> and 90<sup>th</sup> percentile for the negative intermediate ions, and 75<sup>th</sup> and 90<sup>th</sup> percentile for the air temperature at each site.



Värriö Forest Hyytiälä Forest Ränskälänkorpi Forest Värriö Forest Kumpula Urban garden
Haltiala Argriculture Quvidja Argriculture Viikki Argriculture Siikaneva Peatland Tvärminne Coastal area

Figure S9. Comparison between median NEE, median negative intermediate ions at 2.0-2.3 nm, leaf area index, and median photosynthetic photo flux density (PPFD) at midday in summer between the sites. The error bars are 10<sup>th</sup> and 25<sup>th</sup> percentile for NEE, 75<sup>th</sup> and 90<sup>th</sup> percentile for the negative intermediate ions, and 75<sup>th</sup> and 90<sup>th</sup> percentile for PPFD at each site.

#### Minor comments

1. L58: Räty et al., 2023 was not on the reference list.

Reply: It is now added to the list.

2. L86: long-term datasets. What's the definition of 'long-term' here? Only long-term datasets are from Hyytiälä station and others are less than a decade.

<u>Reply: It is revised to "This study utilized 1 to 10 year-long datasets of intermediate ion</u> <u>concentrations."</u> **3**. Table 1: Between the category (such as Forest and Agricultural) it would be good to have a line.

Reply: The lines are added now.

4. Table 1: Hope station name can be reconsider. Especially Hyytiälä station is 'reference station', used everywhere but in the forest category. Author can name stations with their characteristic.

Reply: The site ID is now added (see response to the major comment 2) and used throughout the manuscript. We took Hyytiälä forest as a reference station as it is one typical ecosystem type in Finland and has the longest record of data and endures relatively little anthropogenic interference. Hence, we still kept the Hyytiälä site in forest category.

5. Table 1: Location. Hope author can add height of each station.

Reply: We added this information in Table S1. We have added in line 145 "The inlets for all the NAIS in the studied sites are 1-2 m high above the ground." and in line 170 "The fluxes were measured above the ecosystem canopies and below 30 m. The detailed measurement height for each site is listed in Table S1.

L133: at same heights. Please add values.

Reply: The height information is included in Table S1. As the measurement heights (1-70 m) vary between the stations, dependent on the ecosystem canopy height, we referred to Table S1 in the text.

6. L137: *Diameter range*. Does it mean particle size?

Reply: Yes, it describes the range of particle size in diameter.

7. L140: *The data were cleaned*. What does it mean? Does it mean 'filtered' or 'selected'?

Reply: This is now deleted as it is the same meaning of 'quality-checked'. "The data were quality-checked, considering e.g. the potential interference of rainfall and snow events on the measurements (Manninen et al., 2016)."

8. L142: *total particle concentration*. Is it mass concentrations or number concentrations or others?

<u>Reply: It is a number concentration. It is revised as "The ion and total particle number concentration" in the text.</u>

9. L151: *their footprints are constrained within the ecosystem scale*. Don't authors think that it can be differed according to the height of station?

Reply: Indeed, the footprints of both eddy covariance and  $N_{\text{neg}}$  are impacted by the height of the station. This sentence is revised as "their footprints are constrained within the ecosystem scale when measured at a height between 1 and 70 m (Section 2.2 and Table S1)." The forest canopy is below 30 m and even shorter for agricultural fields and peatlands. The footprint for eddy covariance (Kljun et al., 2015) and  $N_{\text{neg}}$  (Tuovinen et al., 2024) are within the studied ecosystems.

# **10**. L153: *outside the active hours of the ecosystem, were taken as the background concentration at each site.* This sentence is conflict against L352-357.

Reply: The ecosystem at nighttime is assumed to be relatively inactive with no photosynthesis and low BVOC emission. The nighttime clustering explained in L352-357 is more likely a consequence of meteorological conditions associated with atmospheric chemical reactions. To clarify the difference between drivers for daytime and nighttime clustering, the sentence is revised as "However, these negative ions clustered at nighttime are likely unable to grow >3 nm in diameter (Mazon et al., 2016)."

# 11. L194: *tower*. Hope to add the tower height.

Reply: The tower height can differ largely with the measurement station. Here we added the measurement height in case of confusion. We revised the text as "The NAIS equipment was positioned in the border between the control and clear-out, ~230 m east from the eddy covariance tower (measurement height of 29 m)".

## 12. L203: *interference*. What kind of interference?

Reply: There is an experimental area (for different cutting heights) between 30° and 140° of the eddy covariance mast in the agricultural grassland. The different management in the experimental area may influence the measured CO<sub>2</sub> uptake and  $N_{\text{neg.}}$ . This sentence was revised to avoid confusion "For the Qvidja site, NAIS and eddy covariance data from wind directions between 30° and 140° were discarded due to another experimental plot located in that part of the field (Heimsch et al., 2021)".

## 13. L205: *fields*. What is the possible source to affect on data?

Reply: Revised as "Similarly, at the Viikki site, only measurements from wind direction between 145° and 245° were included in the analysis to avoid data from other nearby fields with different vegetation and management activities." Vegetation and management activities, such as fertilization, may impact the CO<sub>2</sub> uptake, emission of BVOCs and NH<sub>3</sub> concentration.

14. L226: In the other seasons, the urban garden area was a net source of CO<sub>2</sub> most of the time, similar to the results previously reported for the years 2006-2010 from the same site (Järvi et al., 2012). Most of section did not explain the reason. Why is that?

Reply: This part is now revised (L230): "There are residential buildings and traffic within the eddy covariance measurement footprint in G-KUM. The CO<sub>2</sub> emissions from the residential buildings, traffic and bare soil outweighed photosynthetic uptake of CO<sub>2</sub> except during summer daytime".

15. Figure 2-4: Why did authors add 50<sup>th</sup> and 25<sup>th</sup> graph? It would better to add median with standard deviation to see the variation of the values since experimental periods are different at each station.

Reply: The 50th percentile (median) of the NEE described the average status of  $CO_2$  fluxes. For the 25th percentile of  $CO_2$  fluxes, they corresponded to the conditions when the ecosystem is very active. In the case of  $N_{neg}$ , 75<sup>th</sup> percentile can be a sign of new particle formation event (Aliaga et al., 2023). We want a uniform presentation of NEE and  $N_{neg}$  in the manuscript. Also, as we aimed to emphasize the  $CO_2$  uptake and local aerosol production potentials, we prefer keeping the original way of presenting the data.

16. L237: In the case of agricultural fields in summer (Figure 3), the Haltiala site had higher momentary net CO<sub>2</sub> uptake than the other two agricultural sites. Why don't authors add specific value for 'higher than' precisely?

Reply: It is revised to "In the case of agricultural fields in summer (Figure 3), the A-HAL and A-VII croplands had 2-5  $\mu$ mol m<sup>-2</sup> s<sup>-1</sup> (for midday median values) higher momentary net CO<sub>2</sub> uptake rate than the other agricultural grassland." The similar issue, i.e., the specific values for "higher than" or "lower than", in the whole manuscript have been revised.

17. L239: *Qvidja was a CO<sub>2</sub> sink during daytime with a similar uptake rate to the Hyytiälä forest.* Add specific value and figure #.

<u>Reply: This line is revised as "A-QVI was a CO<sub>2</sub> sink during daytime with a comparable uptake rate to the F-HYY (ranging between 0 and 4  $\mu$ mol m<sup>-2</sup> s<sup>-1</sup>)."</u>

18. L240: The different plant species (Table 1) and management activities between the agricultural fields likely caused the differences in their seasonal  $CO_2$  fluxes. This is very confusing to readers. In table 1, authors mentioned of plants type but not management activities. I confirmed that it was described in the section 2.1, it would be good to add table 1.

Reply: We now added the description of management activities in the three agricultural fields in Section 2.3 (line 143) "A-QVI was harvested in June and August, A-VII was harvested twice in August during the reported period, and A-HAL was harvested once around the end of August during the measurement periods. The sowing (over-seeding for A-QVI and only in 2022) and first fertilization in the year usually takes place at the end of spring." And following line 240 "The different plant species (Table 1) and management activities between the agricultural fields likely caused the differences in

their seasonal CO<sub>2</sub> fluxes. During the measurement period, perennial plants were grown in A-QVI, while the growth of the annual crops in A-HAL and A-VII relied on the sowing and fertilization date, normally at the end of spring. This may explain the springtime CO<sub>2</sub> emission in A-HAL and A-VII. In the summer, the A-HAL and A-VII was harvested only in August, while A-QVI was harvested in June and August separately, which may explain the higher CO<sub>2</sub> uptake rate in A-HAL and A-VII."

**19**. L294: *The median values of*  $N_{neg}$  *in the daytime in spring were higher than those in the Haltiala and Viikki croplands, Siikaneva peatland, and Kumpula urban garden area.* I cannot understand what this sentence means.

Reply: Revised as follows "The daytime median values of  $N_{neg}$  were higher in spring than that in summer at A-HAL and A-VII, P-SII, and G-KUM. At the other sites, the median values in summer were higher than those in spring".

20. L296: *summer median values were higher*. Please finalize the sentence. Higher than what?

Reply: Revised "At the other sites, the median values in summer were higher than those in spring".

21. L338: The application of fertilizers in agricultural fields is known to remarkably increase the atmospheric concentration of ammonia (NH<sub>3</sub>). This sentence seems to be generalized to all stations. Please add specific type of regions.

Reply: It is revised to "The application of fertilizers is known to remarkably increase the atmospheric concentration of ammonia (NH<sub>3</sub>) in agricultural fields, e.g., observed in A-QVI (Olin et al., 2022)".

22. Figure 9: I cannot understand the meaning of bar in Figure 9.

Reply: Line 413 for the figure caption is revised "Figure 9. Comparison between the median NEE, median negative intermediate ions at 2.0-2.3 nm, and median air temperature at midday in summer between the sites. The error bars are the 10th and 25th percentiles for NEE, the 75th and 90th percentiles for the negative intermediate ions, and the 75th and 90th percentiles for the air temperature at each site". The main scope is to present the potential of different ecosystems influencing the CO<sub>2</sub> uptake and local aerosol production. The 25<sup>th</sup> and 75<sup>th</sup> percentile for NEE and  $N_{neg}$  were presented, respectively, to show the higher range of ecosystem CO<sub>2</sub> uptake rate and local aerosol production.

23. Table 2: There is no explanation of 'c' (superscript) and there is no 'b' in the table.

Reply: The 'c' is revised to be 'b' in Table 2.

24. L409: Summary and conclusions or only summary?

Reply: It is "Conclusions"

25. L416: What are 10 stations belonging to hemi-boreal station? Why did not author define 'hemi-boreal' and 'boreal' in the manuscript?

Reply: This has been addressed above.

### 26. L421-423: Why didn't authors add the reasons?

Reply: This line is revised as "A distinct  $CO_2$  uptake in the urban garden at midday in summer was observed, due to the strong photosynthesis of vegetation inside. The uptake rate was 37% lower than that in F-HYY but ~2 times of that observed in the open peatland". We also simply explained why agricultural fields presented generally high  $CO_2$  uptake and aerosol production in Line 490 "The results showed that the agricultural fields had similar or even 15% higher  $CO_2$  uptake potentials compared to the boreal Hyytiälä forest (F-HYY) during the summer, possibly due to the high leaf area index and air temperature in the agricultural fields." and line 500 "In agricultural fields, the synergetic role of NH<sub>3</sub>, H<sub>2</sub>SO<sub>4</sub>, and low volatile organic compounds originating from BVOC oxidation may play a synergistic role in clustering and induce a high  $N_{neg}$  comparing with other ecosystem types.".

27. L431: What's the difference between reference station and background station? Please add the definition.

Reply: The background site in this context means the sites that receive little human interference. Here we actually did not need the concept of background sites, and the text is revised as "Note that the urban garden and agricultural sites in Helsinki might be more influenced by air pollution compared to the forests and open peatland that received little anthropogenic interference and pollution".

#### Reference

Kljun, N., Calanca, P., Rotach, M. W., and Schmid, H. P.: A simple two-dimensional parameterisation for Flux Footprint Prediction (FFP), Geoscientific Model Development, 8, 3695-3713, <u>https://doi.org/https://doi.org/10.5194/gmd-8-3695-2015</u>, 2015.

Tuovinen, S., Lampilahti, J., Kerminen, V.-M., and Kulmala, M.: Intermediate ions as indicator for local new particle formation, Aerosol Research, 2, 93-105, <u>https://doi.org/10.5194/ar-2-93-2024</u>, 2024.