



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

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**Abstract:** Continental ecosystems play an important role in carbon dioxide (CO<sub>2</sub>) uptake and aerosol production, which helps to mitigate climate change. The concept of ‘CarbonSink+ potential’ enables a direct comparison of CO<sub>2</sub> uptake and local aerosol production at ecosystem scale. Following this concept, momentary net ecosystem exchange (NEE) and number concentration of negative intermediate ions at 2.0-2.3 nm ( $N_{\text{neg}}$ ) were analysed for boreal and hemi-boreal ecosystems across Finland and in Estonia.  $N_{\text{neg}}$  can tell us how effectively biogenic emissions from an ecosystem initiate the new particle formation. Four forests, three agricultural fields, an open peatland, an urban garden, and a coastal site were included focusing on summertime. We compared the NEE and  $N_{\text{neg}}$  at each site to the Hyytiälä forest as it is the dominant ecosystem type in Finland.  $N_{\text{neg}}$  was highest at the urban garden and lowest at the coastal site. The agricultural fields had higher or similar net CO<sub>2</sub> uptake rate and higher  $N_{\text{neg}}$  than all studied forests. The median net CO<sub>2</sub> uptake rate of the open peatland was only 31% of that in Hyytiälä, while the median  $N_{\text{neg}}$  was 77% of that in Hyytiälä. The median net CO<sub>2</sub> uptake rate in the urban garden was 63% of that in Hyytiälä, implying the importance of urban green areas in CO<sub>2</sub> sequestration. The coastal site was a minor CO<sub>2</sub> source. Considering the combined effect of CO<sub>2</sub> uptake and aerosol formation and the large area of forests in Finland, the forests are the most important ecosystems helping to mitigate climate warming.

## 1. Introduction

Carbon dioxide (CO<sub>2</sub>) is one of the most abundant greenhouse gases in the atmosphere and the most important cause of global warming (e.g. Jia et al., 2022). Terrestrial ecosystems have an essential role in the global CO<sub>2</sub> budget through carbon uptake from the atmosphere by photosynthesis and its consequent sequestration to various pools (Walker et al., 2021; Friedlingstein et al., 2022). Globally, the net terrestrial ecosystem uptake of CO<sub>2</sub> (i.e. the net carbon sink) is 3.1 Gt C yr<sup>-1</sup>, which accounts for 32% of CO<sub>2</sub> emissions from fossil fuel combustion (Friedlingstein et al., 2022). Terrestrial carbon sequestration, i.e., the process of storing carbon in a carbon pool (IPCC 2022), takes place in both belowground carbon storages (Walker et al., 2021; and the reference therein). Belowground storage includes soil carbon pools, while aboveground storage is primarily in biomass. As a transition between land and open ocean, the coastal environment is identified as an import carbon sink and estimated to uptake 0.4 Gt C yr<sup>-1</sup> (Regnier



et al., 2022). Large spatiotemporal variation of continental CO<sub>2</sub> uptake is assumed due to different ecosystem and land-use types, climatic conditions, and management pathways (Chang et al., 2021; Friedlingstein et al., 2022). The challenge of increasing the carbon sequestration of ecosystems has been attracting more and more attention with the global goal of reducing CO<sub>2</sub> concentrations  
55 in the atmosphere.

Apart from acting as CO<sub>2</sub> sinks, terrestrial ecosystems can influence climate by contributing to the formation of new aerosol particles (Kulmala et al., 2004; Kulmala et al., 2014; Kulmala et al., 2020; Yli-Juuti et al., 2021; Junninen et al., 2022; Petäjä et al., 2022, Rätty et al., 2023). Globally, aerosols have been reported to induce a net climate cooling effect. The best estimate of the  
60 effective radiative forcing is  $-1.06 \text{ W m}^{-2}$  (Jia et al., 2022). However, large uncertainties exist in the aerosol net radiative forcing estimation, which is tightly associated with the large spatiotemporal heterogeneity in their origin, number concentration and chemical properties.

Atmospheric new particle formation (NPF) is an important source of cloud condensation nuclei (CCN) (e.g. Gordon et al., 2017; Ren et al., 2021; Zhang et al., 2023), which significantly  
65 contributes to aerosol-cloud and aerosol-radiation interaction (Rosenfeld et al., 2014; Ezhova et al., 2018, Artaxo et al., 2022; Petäjä et al., 2022). NPF takes place frequently in many environments, such as forests, urban cities, and coastal areas (e.g. Kerminen et al., 2018; Nieminen et al., 2018; Zheng et al., 2021). It has been reported that NPF is greatly enhanced due to the emission of biogenic volatile organic compounds (BVOCs) in boreal forests and peatlands (Junninen et al.,  
70 2022; Petäjä et al., 2022). Notably, NPF events often take place regionally, extending over distances up to over 1000 kilometres (Kerminen et al., 2018). Multiple types of ecosystems may contribute to the NPF events in a region depending, for example, on the diversity of land use types. It remains unclear whether and how various ecosystems differ in their contributions to regional NPF, and what is the magnitude of such differences.

75 To overcome the challenge of analysing the role of local ecosystems in regional aerosol formation, the concept of ‘CarbonSink+ potential’ was recently established (Kulmala et al., 2024). The CarbonSink+ potential enables a direct, ecosystem-scale comparison of CO<sub>2</sub> uptake and the intensity of local intermediate ion formation (LIIF) in the atmosphere at the ecosystem scale. The LIIF can be approximated as the number concentration of negative intermediate ions in 2.0-2.3 nm  
80 size range (Tuovinen et al., 2024), to which the aerosol formation at 3-6 nm size range is proportional (Kulmala et al., 2024). The survival probability of small aerosol particles, which



describes the probability of a single particle growing to a certain size without being scavenged, is generally high for particles from 6 nm to CCN size in rural and remote environments (Kulmala et al., 2024; Stolzenburg et al., 2023). The local contributions of certain ecosystems to regional aerosol formation can thus be quantified by LIIF.

This study utilized long-term datasets of intermediate ion concentrations and CO<sub>2</sub> fluxes from various boreal and hemi-boreal ecosystems across Finland and in Estonia. In summary, four forests, one open peatland, three agricultural fields, one urban garden, and one coastal site were investigated. The negative intermediate ion concentrations and CO<sub>2</sub> fluxes for these ecosystems were compared in different seasons with a focus on the summer. Based on the CarbonSink+ potential concept (Kulmala et al., 2024), the potential of these ecosystems to mitigate climate warming regarding CO<sub>2</sub> uptake and aerosol production is discussed.

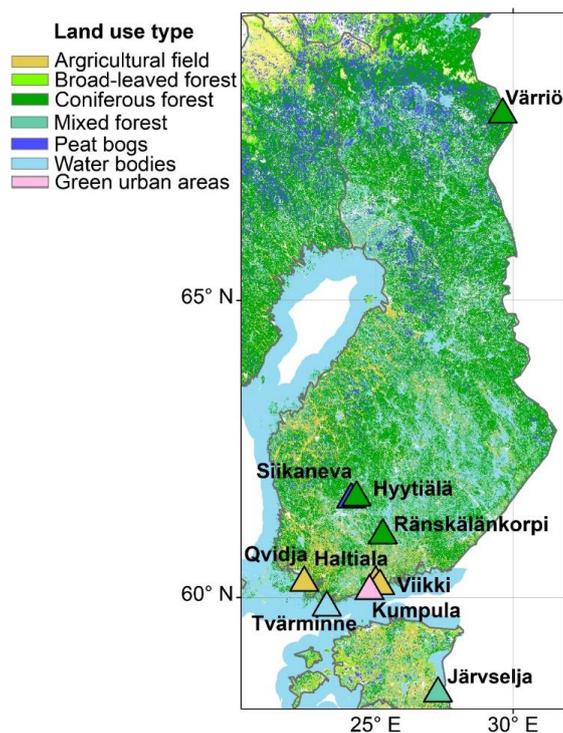
## 2. Method

### 2.1 Site description

In this study, various ecosystem types, including forests, open peatland, agricultural fields, coastal area, and an urban garden were studied (Figure 1; Table 1). All stations are utilizing the SMEAR (Station for Measuring Ecosystem-Atmosphere Relations; Hari and Kulmala, 2005) concept. The detailed location, ecosystem type, meteorological characteristics and soil type for each site are presented in Table 1. The SMEAR I in Värriö in northern Finland and SMEAR II in Hyytiälä in southern Finland are forest sites both dominated by Scots pine (Kulmala et al., 2019; Kolari et al., 2022), while the forests in Ränskälänkorpi and SMEAR Estonia at Järvelja are mixtures of coniferous and broadleaf trees (Table 1). While Hyytiälä and Värriö are upland forests, i.e., growing on mineral soil, Ränskälänkorpi is a drained-peatland forest (Laurila et al., 2021) and Järvelja has a mosaic of drained swamp, drained peat, and leached gleyic pseudo-podzols (Kangur et al., 2021; Noe et al., 2015). Two of the agricultural (SMEAR-Agri) sites, i.e. Haltiala, a cereal cropland and Viikki, a managed grassland which was renewed in 2023 with a cereal crop (Pihlatie et al., in preparation), are located in Helsinki. The third agricultural site, Qvidja, is a managed grassland located in southwest Finland (Heimsch et al., 2021). The SMEAR II site at Siikaneva is an open, pristine peatland site ~5 km southwest from the Hyytiälä forest site (Rinne et al., 2018). The SMEAR III at Kumpula, Helsinki is an urban background site and the University of Helsinki



botanical garden, and the city of Helsinki allotment garden are located in the southwest of SMEAR III station with high fraction of vegetation (Järvi et al., 2012). The coastal site is in Tvärminne Zoological Station (TZS). TZS is a 600-ha nature reserve at the Gulf of Finland entrance (northern Baltic Sea), southwest Finland (Virtasalo et al., 2023; Vähä et al., 2024). During the measurement  
115 period, the annual mean temperature for these sites ranged between 0.4 and 7.2°C, while the annual precipitation ranged between 500 and 750 mm (Table 1). SMEAR Estonia, Tvärminne, and Qvidja belong to hemi-boreal climate, while the other ecosystems are characterized by boreal climate.



120 Figure 1. Land type distribution across Finland (Copernicus Land Monitoring Service 2018) and the studied sites with their ecosystem type shown.



Table 1. Meteorological and other main characteristics of the studied sites.

Stations	Location	Selected period	Mean air temperature (°C)	Rainfall (mm/yr)	Dominant plant species	Peak LAI	Soil type	
	Hyytiälä, SMEAR II	61°51'N, 24°17'E	11/2009-12/2022	4.8	709 <sup>1</sup>	Scots pine and Norway spruce	4.6	Haplic podzol
	Värriö, SMEAR I	67°46'N, 29°35' E	3/2019-12/2022	0.4	601 <sup>2</sup>	Scots pine	3.2	Haplic podzol
Forest	Ränskälänkorpi	61°10'N, 25°16'E	4/2021-12/2022	5.4	600 <sup>3</sup>	Norway spruce, Scots pine, downy birch	----	Drained peat
	Järvelja	58°16'N, 27°16'E	10/2016-12/2020	6.8	500-750 <sup>4</sup>	Birch species, Scots pine, Norway spruce	6	Pseudo podzolic
	Haltiala, SMEAR Agri	60°16'N, 24°57'E	6/2021-12/2022	6.5	700 <sup>5</sup>	Oat	5.5	Silty clay
Agricultural fields	Qvidja	60°18'N, 22°24'E	12/2018-8/2022	7.0	679 <sup>6</sup>	Timothy, meadow fescue	6.2	Clay loam
	Viikki, SMEAR Agri	60°13'N, 25°01'E	7/2022-6/2023	6.5	792 <sup>5</sup>	Timothy (2022), Barley (2023)	5.2	Clay loam
Peatland	Siikaneva, SMEAR II	61°50'N, 24°12'E	11/2019-12/2022	5.0	710 <sup>7</sup>	Moss and sedges	0.6	Peat
Urban garden	Kumpula, SMEAR III	60°12'N, 24°58'E	5/2016-12/2022	6.3 <sup>5</sup>	731 <sup>5</sup>	Mixed	-----	-----
Coastal area	Tvärminne	59°51'N, 23°15'E	6/2022-8/2023	7.2 <sup>5</sup>	639 <sup>5</sup>	Seagrass and seaweed	-----	Sediments

<sup>1</sup>Neeffes et al. (2022); <sup>2</sup>Kulmala et al. (2019); <sup>3</sup>Laurila et al. (2021); <sup>4</sup>Noe et al. (2015); <sup>5</sup>Finnish Meteorology Institute, only data at the same calendar year of selected period and same or nearby stations as NAIS and eddy covariance measurements were applied; <sup>6</sup>Heimsch et al. (2021); <sup>7</sup>Rinne et al. (2018); ---- data not available.



## 2.2 Atmospheric measurements: intermediate ions, CO<sub>2</sub> flux, and meteorological parameters

The number concentration of ions and particles and net ecosystem exchange of CO<sub>2</sub> (NEE) were  
130 measured using a Neutral cluster and air ion spectrometer (NAIS, Airl Ltd; Mirme and Mirme,  
2013) and eddy covariance method (Aubinet et al., 1999), respectively. The meteorological data,  
e.g., air temperature, air humidity, and photosynthetic photon flux density (PPFD), were measured  
simultaneously at same heights with the eddy covariance setup. If the meteorological measurement  
at the same height was not available, it was replaced by the one from the nearest height. The types  
135 of analysers and detectors at each site are listed in Table S1.

The NAIS is capable of continuous monitoring of ion and total particle concentrations and size  
distributions over the diameter range of 0.8-42 nm. The ions can be divided into three different  
size ranges, namely small ions (also named as cluster ions) in sub-2 nm size range, intermediate  
ions (2-7 nm), and large ions (>7 nm; Tammet et al., 2014). The time resolution was set to five  
140 minutes to optimize signal-to-noise ratio (Mirme and Mirme, 2013). The data were cleaned and  
quality-checked, considering e.g. the potential interference of rainfall and snow events on the  
measurements (Manninen et al., 2016). The ion and total particle concentration were further  
averaged over half an hour.

In this study, we identified the concentration of negative intermediate ions, specifically within the  
145 range of 2.0-2.3 nm ( $N_{\text{neg}}$ ), as an indicator of the local intermediate ion formation (LIIF). It is  
important to note that the intensity of LIIF can serve as an estimate of the local contribution to the  
regional NPF (Kulmala et al., 2024). It has been observed that  $N_{\text{neg}}$  displays distinct difference  
between new particle formation and non-formation periods of intermediate ions (2-7 nm; Tuovinen  
et al., 2024), thereby making  $N_{\text{neg}}$  a reliable indicator of LIIF. Moreover, the measurement of  
150 negative intermediate ions between 2.0 and 2.3 nm by NAIS provides a relatively high degree of  
accuracy, and their footprints are constrained within the ecosystem scale (sub-1 km; Tuovinen et  
al., 2024; Kulmala et al., 2024). Moreover, the median values of  $N_{\text{neg}}$  between 00:00 and 06:00,  
i.e. outside the active hours of the ecosystem, were taken as the background concentration at each  
site. The background value of  $N_{\text{neg}}$  was calculated separately for each season. A narrower time  
155 window for background concentration compared to the one proposed by Aliaga et al. (2023),  
21:00-06:00, was applied due to the more northern site Värriö with longer day length in the  
summer in this study. We then calculated the changes of  $N_{\text{neg}}$  ( $\Delta N_{\text{neg}}$ ) by subtracting the  
background concentration in each season from  $N_{\text{neg}}$ . The diurnal variation of median  $\Delta N_{\text{neg}}$  were



presented together with  $N_{\text{neg}}$  (Section 3). The use of  $\Delta N_{\text{neg}}$  was assumed to eliminate the influence  
160 of background clustering at different sites, so that it reflects the intensity of negative intermediate  
ion production from the specific ecosystem.

The eddy covariance measurement of CO<sub>2</sub> fluxes is based on the turbulence theory, i.e. assumption  
that the turbulent flux remains relatively stable in a constant flux layer above the canopy (Lee and  
Hu, 2002), and it is equal to the covariance of vertical wind speed and ambient CO<sub>2</sub> concentration  
165 in flat and horizontally homogeneous surface (Aubinet et al., 1999). The measurement system  
requires a fast-response analyser of the CO<sub>2</sub> concentration (10 Hz) and 3-D sonic anemometer.  
The raw eddy covariance 10 Hz-data were pre-processed with standard steps, including despiking,  
detrrending, dilution correction and 2-D coordinate rotation (Aubinet et al., 1999). The fluxes were  
further lag-time adjusted and corrected for spectral loss (Aubinet et al., 1999). Either EddyUH  
170 (Mammarella et al., 2016) or EddyPro (Fratini and Mauder, 2014), or the program introduced by  
Heimsch et al. (2021) were applied for the pre-processing for one site. The processed fluxes were  
accepted only if they met the stationarity and developed turbulence criterion (Foken and Wichura,  
1996) exceeding the site-specific friction velocity thresholds (Table S1). The quality-checked CO<sub>2</sub>  
fluxes at the forest sites were further partitioned into gross primary production (GPP) and  
175 ecosystem respiration ( $R$ ) using site-specific dependence of  $R$  on the air and/or soil temperature  
and GPP on the PPFD and air and/or soil temperature (Kulmala et al., 2019).

### 2.3 Data selection criteria

In this study, the analyses were restricted to periods when both negative intermediate ion  
concentration and NEE were available (Table 1). Therefore, different time periods were applied  
180 for each of different sites. For Hyytiälä, Värriö, Järvelja, Qvidja, Siikaneva, and Kumpula sites,  
the long-term data were available for more than 3 years. At Hyytiälä, 12 years of continuous  
observations were used. For the sites with recently established atmospheric measurement,  
Tvärminne, Ränskälänkorpi, Haltiala and Viikki - data were available for approximately one to  
one and a half years. In total, 35 site-years of data were utilized in this study. As we focused on  
185 the potential of the ecosystem to uptake CO<sub>2</sub> and form intermediate ions, the inter-annual variation  
at the sites was not discussed in this study (Kulmala et al., 2019; Alekseychik et al., 2021; Heimsch  
et al., 2021).



Due to the thinning of Hyytiälä forest in the beginning of year 2020, when 30% of tree basal area was removed (Aalto et al., 2023), data from that year were discarded from the analyses to exclude  
190 the immediate thinning effect on the studied variables. In the Ränskälänkorpi forest, the western part of the site was selectively harvested (~60% of basal area removed) and the eastern part of the site was clear-cut in the spring and summer of 2021 with control site left in the middle. The NAIS equipment was located in the border between the control and clear-out. The location was ~230 m east from the eddy covariance tower located in the border between control and selective harvested  
195 sites. In this study, only data with wind blowing from the selective harvested site from the west ( $WD > 180^\circ$ ) and wind speed higher than  $2 \text{ m s}^{-1}$ , were considered. Note that carbon removed from the site in harvested tree biomass is not accounted in the measured flux of  $\text{CO}_2$ . At Kumpula site, data from the garden area, i.e.,  $180^\circ$ - $320^\circ$ , were utilized (Järvi et al., 2012).

At the agricultural sites, the management activity is relatively intense and can distinctly influence  
200 the  $\text{CO}_2$  fluxes (Heimsch et al., 2021). Note that the carbon removed in harvested crop biomass and the carbon added to the site in fertilizers do not directly contribute to the measured net flux of  $\text{CO}_2$ . For the Qvidja site, only measurements from wind direction between  $0^\circ$  and  $30^\circ$  or  $140^\circ$  and  $360^\circ$  were included to avoid interference from the nearby experimental areas. Similarly, at the Viikki site, only measurements from wind direction between  $145^\circ$  and  $245^\circ$  were included in the  
205 analysis to avoid data from other nearby fields.

The open peatland at Siikaneva is surrounded by forests. By applying a footprint model (Kljun et al., 2015), 90% of the  $\text{CO}_2$  flux footprint is within ~200 m from the measurement tower, i.e., constrained within the peatland. At the coastal Tvärminne site, the NAIS instrument trailer is on the shore, and the eddy covariance mast is on an island, ~110 m east of the shore. Only data with  
210 wind direction from  $95^\circ$  to  $165^\circ$  and from  $205^\circ$  to  $240^\circ$ , i.e., from the coastal water without being disturbed by trees on nearby islands, were included in the analysis at this site.

### 3. Results and discussion

#### 3.1 Comparison of momentary NEE in different ecosystems

The diurnal variation of NEE between the studied forests, urban garden area, agricultural fields,  
215 open peatland, and coastal site in spring (MAM) and summer (JJA) are presented in Figures 2-4.



The corresponding comparison in the autumn (SON) and winter (DJF) are presented in Figures S1-S3.

For the forest sites, the hemi-boreal Järvelja site tended to have the highest net CO<sub>2</sub> uptake rate (absolute values of NEE when it is negative) at midday (10:00-14:00) in both spring and summer.

220 The median net CO<sub>2</sub> uptake rate at midday in Järvelja forest reached 12.2 μmol m<sup>-2</sup> s<sup>-1</sup> in summer. The lowest net CO<sub>2</sub> uptake rate at midday was found in the northern Värriö site, with the median being 4.69 μmol m<sup>-2</sup> s<sup>-1</sup>. This difference may be due to the higher air temperature in the hemi-boreal Estonian site and lower temperature at Värriö (Figure S4), as the ecosystem productivity at high latitudes in Europe is typically temperature limited (Yi et al., 2010).

225 In summer, the net CO<sub>2</sub> uptake rate in the urban garden area at Kumpula was comparable with the drained peatland forest in Ränskälänkorpi. 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).



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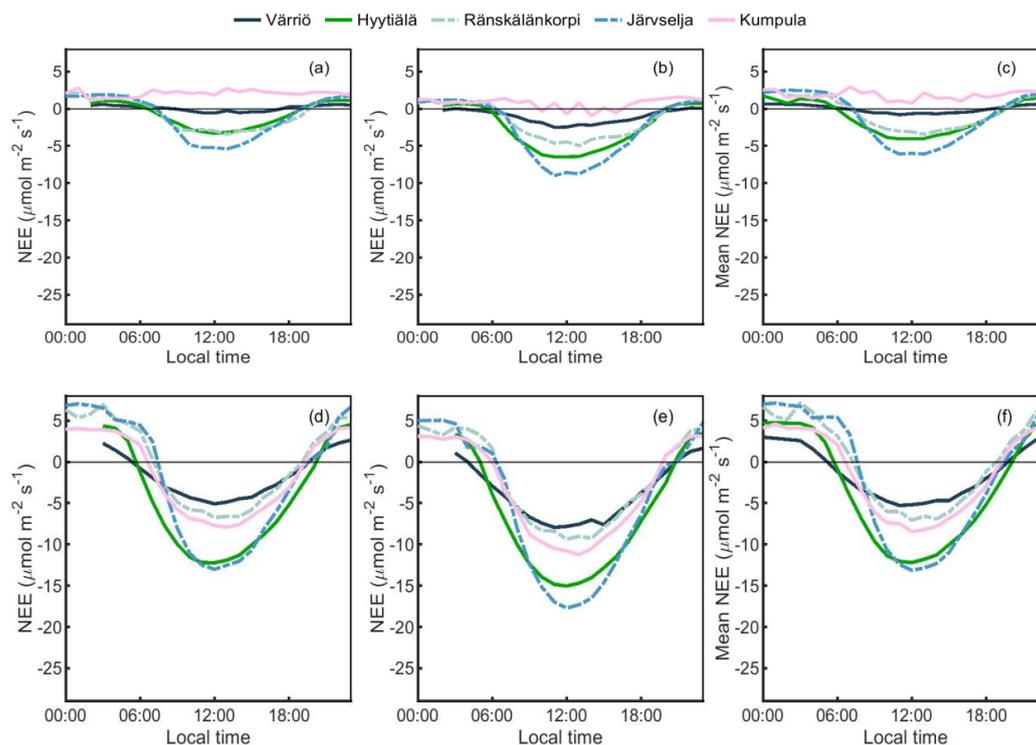


Figure 2. The 50<sup>th</sup> percentile (a), 25<sup>th</sup> percentile (b), and mean values (c) of NEE at each hour for the forest sites and urban garden in spring (MAM) and the corresponding 50<sup>th</sup> percentile, 25<sup>th</sup> percentile, and mean values in summer (JJA), (d), (e), (f), respectively.

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. Notably, in spring, the croplands in Viikki and Haltiala were net sources of CO<sub>2</sub>, while the grassland in Qvidja was a CO<sub>2</sub> sink during daytime with a similar uptake rate to the Hyytiälä forest. 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. The upper quartile of the momentary net CO<sub>2</sub> uptake, i.e., absolute values of 25<sup>th</sup> percentile NEE, was also about two times higher in Haltiala cropland than in Hyytiälä forest in summer. The midday momentary net CO<sub>2</sub> uptake rate in Viikki cropland was slightly higher than that in Hyytiälä forest, while that in Qvidja agricultural grassland was slightly lower than in Hyytiälä. It is also important to note that the harvests of plant biomass decreased local carbon



storage which was not accounted for in the measured CO<sub>2</sub> fluxes. Qvidja and Viikki agricultural sites were harvested twice in summer and the harvest in Haltiala cropland was done only at the end of the growing season, whereas the typical rotation length in managed boreal are 60-100 years in Southern Finland.

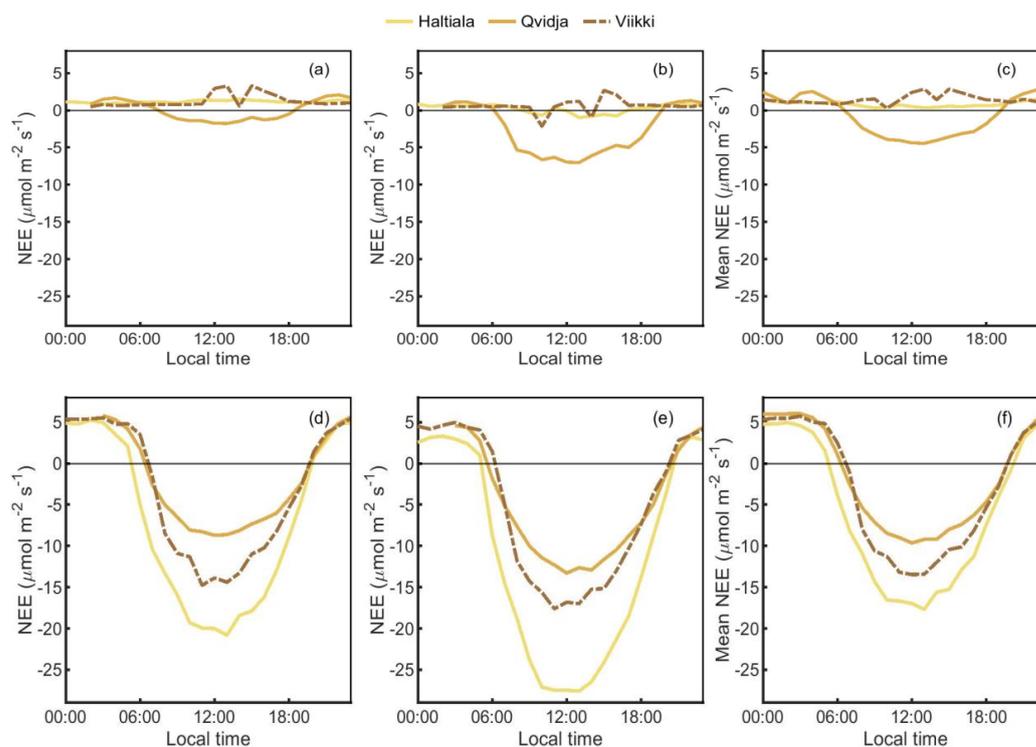


Figure 3. The 50<sup>th</sup> percentile (a), 25<sup>th</sup> percentile (b), and mean values (c) of NEE at each hour for the agricultural fields in spring (MAM) and the corresponding 50<sup>th</sup> percentile, 25<sup>th</sup> percentile, and mean values, (d), (e), (f), in summer (JJA), respectively.

The CO<sub>2</sub> uptake rate and respiration rate (nighttime CO<sub>2</sub> fluxes) in the open peatland and coastal area (Figure 4) were much lower than those in the agricultural fields and forests during spring and summer. Still, the Siikaneva peatland remained a weak net sink of CO<sub>2</sub> during daytimes in all the seasons except in winter. The midday NEE at Tvärminne were -0.26 and 0.01 μmol m<sup>-2</sup> s<sup>-1</sup> in spring and summer, respectively. Hence, net CO<sub>2</sub> uptake possibly appears in spring in this Baltic coastal area under certain conditions, i.e., when the partial pressure of CO<sub>2</sub> in the water is lower



than that in the air (Roth et al., 2023). This may be induced by phytoplankton and submerged  
 265 vegetation CO<sub>2</sub> uptake in the spring (Roth et al., 2023).

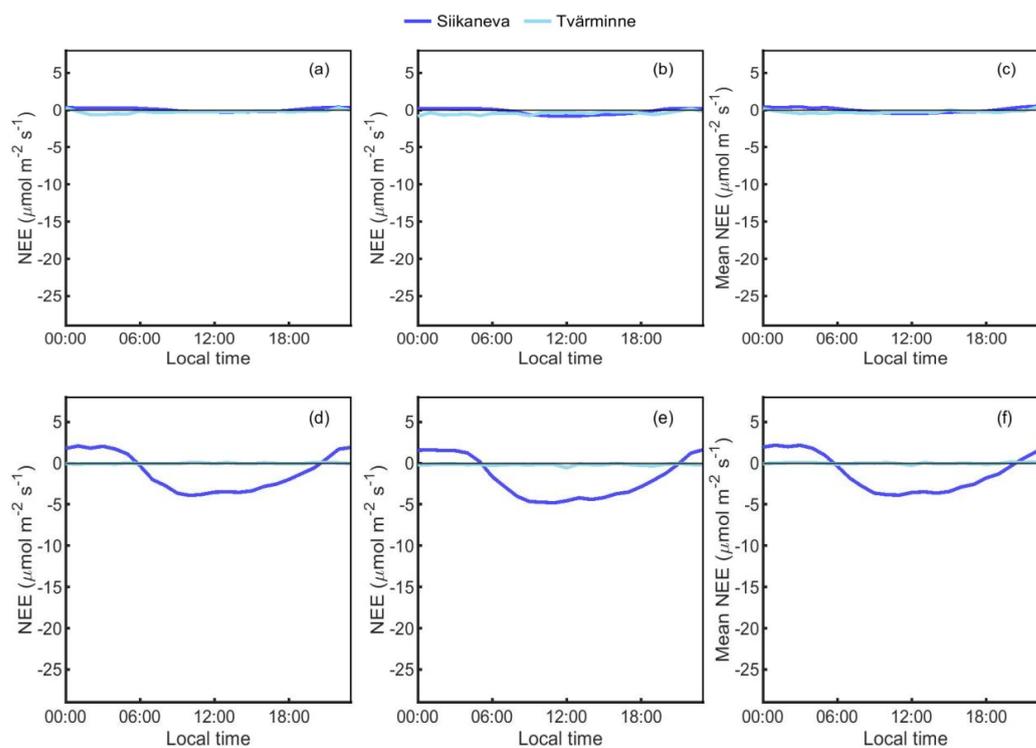
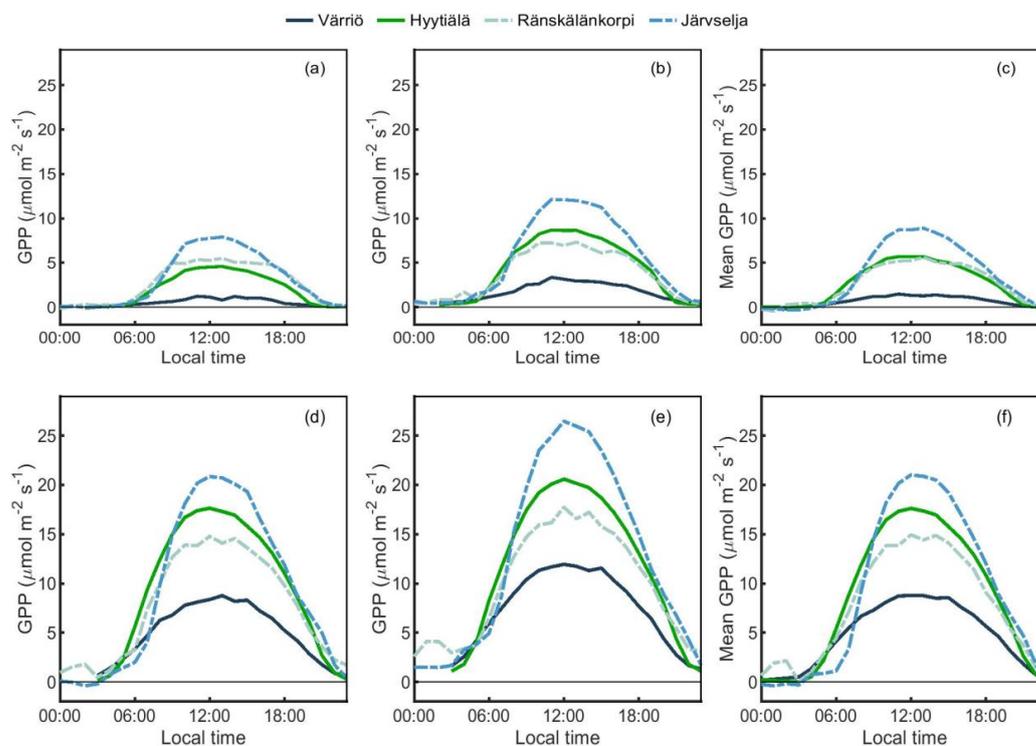


Figure 4. The 50<sup>th</sup> percentile(a), 25<sup>th</sup> percentile (b), and mean values (c) of NEE at each hour for  
 270 the peatland and coastal area in spring (MAM) and the corresponding 50<sup>th</sup> percentile, 25<sup>th</sup>  
 percentile, and mean values in summer (JJA), respectively.

Additionally, the Ränskälänkorpi and Järvelja forests turned into a CO<sub>2</sub> source 1-2 hours earlier  
 in the late afternoon of summer than the other two forests (Figure 2). Note that the soil at  
 275 Ränskälänkorpi and Järvelja is mainly drained peatland and water-logged soil (Table 1),  
 respectively, which is indicated by high organic carbon content (Laurila et al., 2021; Noe et al.,  
 2015). The higher air temperature and soil organic carbon content may drive higher respiration at  
 the two sites, which is reflected in the nighttime fluxes (Figure 2). Hence, even though the GPP at  
 Järvelja and Ränskälänkorpi in the late afternoon were close to that at Hyytiälä forest (Figure 5),



280 net emissions of CO<sub>2</sub>, i.e., positive NEE values, were observed at these two forest sites in the earlier and later hours of the day.



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Figure 5. The 50<sup>th</sup> percentile (a), 75<sup>th</sup> percentiles (b), and mean values (c) of GPP at each hour for the forest sites in spring (MAM) and the corresponding 50<sup>th</sup> percentile, 75<sup>th</sup> percentile, and mean values in summer (JJA), (d), (e), (f), respectively.

### 3.2 Comparison of negative intermediate ion concentrations across different ecosystems

290 The comparison of  $N_{\text{neg}}$  between different ecosystems in spring and summer are presented in Figures 6-8. It was assumed that negative intermediate ions at 2.0-2.3 nm can describe how efficiently the ecosystem can produce new aerosol particles (Kulmala et al., 2024; Tuovinen et al., 2024). The corresponding values of  $N_{\text{neg}}$  in autumn and winter were much lower than those in spring and summer (Figures S5-S7). The median values of  $N_{\text{neg}}$  in the daytime in spring were  
 295 higher than those in the Haltiala and Viikki croplands, Siikaneva peatland, and Kumpula urban



garden area. At the other sites, summer median values were higher. In contrast, the difference between 75<sup>th</sup> and 50<sup>th</sup> percentile of  $N_{\text{neg}}$  in spring was higher than that in summer in all the studied sites. The larger upper quartile deviation of  $N_{\text{neg}}$  in spring implied that the LFII were either more frequent or stronger in spring than in summer at all the sites (Dal Maso et al., 2005; Dada et al., 2018; Nieminen et al., 2018). For all the sites, the diurnal variation of negative intermediate ions in spring and summer was clear, i.e., a distinct peak in the daytime. In the winter, the diurnal cycle of  $N_{\text{neg}}$  was not visible in any of the studied sites (Figures S6-S8). This agrees with the observation that the global radiation and air temperature are observed to correlate positively with concentration of negative intermediate ions at 2-4 nm in the Hyytiälä boreal forest (Neeffjes et al., 2022).

305

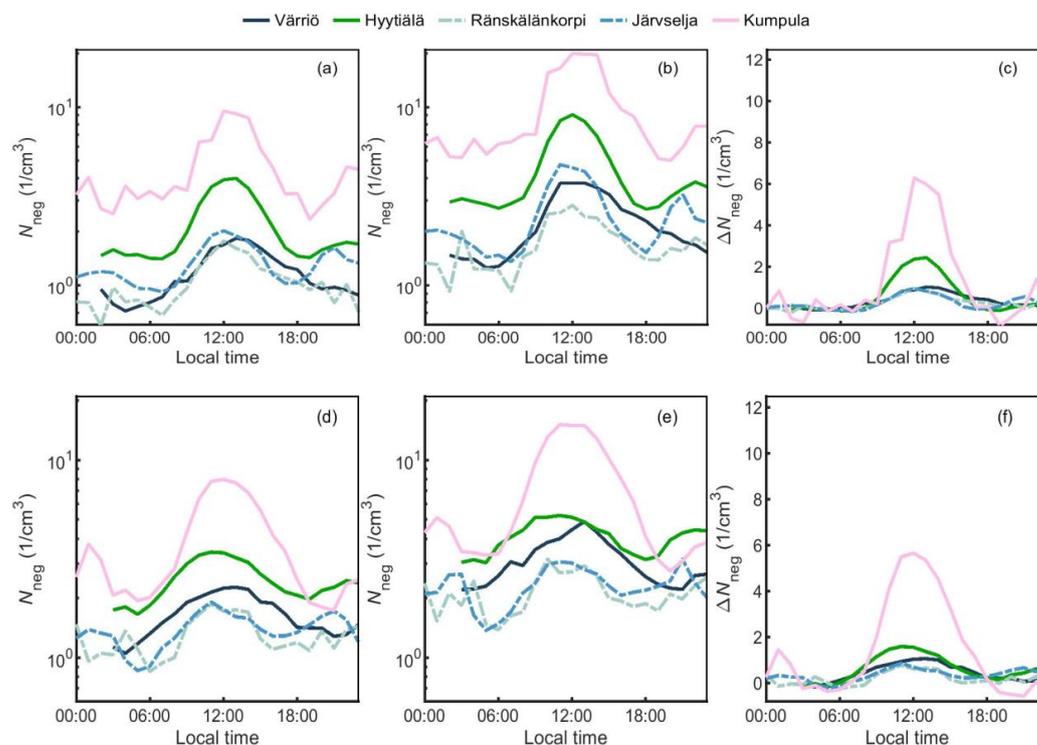
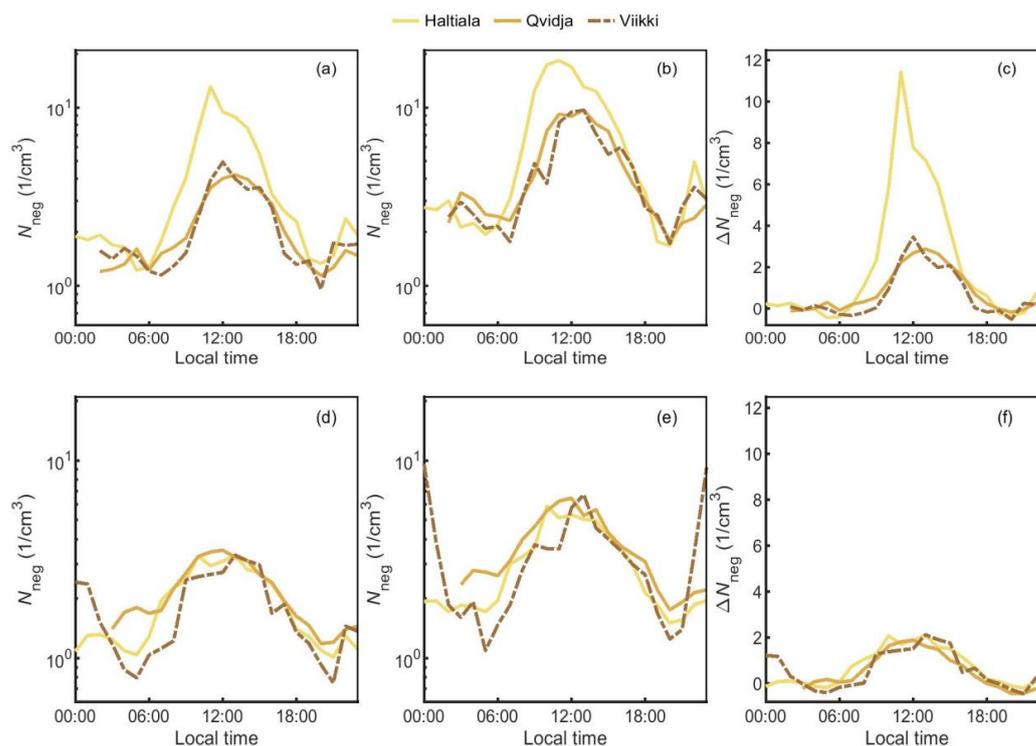


Figure 6. The 50<sup>th</sup> percentile (a) and 75<sup>th</sup> percentile (b) of negative intermediate ions ( $N_{\text{neg}}$ ) at 2.0-2.3 nm ( $N_{\text{neg}}$ ) at each hour and the daily fluctuations of  $N_{\text{neg}}$  (c) for the forests and urban garden in spring (MAM) and the corresponding 50<sup>th</sup> percentile, 75<sup>th</sup> percentile, and normalized concentration for median values in summer (JJA), (d), (e), (f), respectively.



315 Figure 7. The 50<sup>th</sup> and 75<sup>th</sup> percentile (b) of negative intermediate ions ( $N_{\text{neg}}$ ) at 2.0-2.3 nm at each hour and the daily fluctuations of  $N_{\text{neg}}$  (c) for the agricultural fields in spring (MAM) and the corresponding 50<sup>th</sup> percentile, 75<sup>th</sup> percentile and normalized concentration for median values, (d), (e), (f), in summer (JJA), respectively.



320

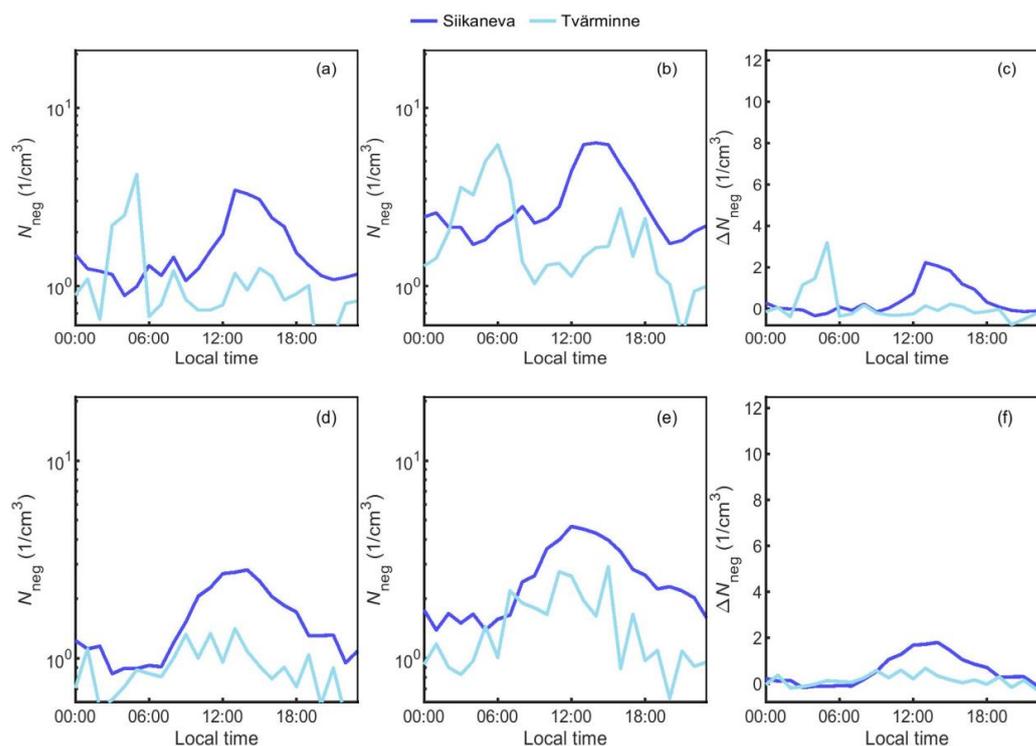


Figure 8. The 50<sup>th</sup> percentile (a) and 75<sup>th</sup> percentile (b) of negative intermediate ions ( $N_{\text{neg}}$ ) at 2.0-2.3 nm at each hour and the daily fluctuations of  $N_{\text{neg}}$  (c) for the peatland and coastal area in spring (MAM) and the corresponding 50<sup>th</sup> percentile, 75<sup>th</sup> percentile, and normalized concentration for median values in summer (JJA), (d), (e), (f), respectively.

The daily fluctuations of  $N_{\text{neg}}$  ( $\Delta N_{\text{neg}}$ ) were calculated by subtracting the background concentration from  $N_{\text{neg}}$  in each season (Section 2.2). In spring, median  $\Delta N_{\text{neg}}$  in the midday for the forests ranged between 0.8 and 2.0  $\text{cm}^{-3}$  (Table S2), with the lowest value in Järvelja and the highest value in Hyytiälä forest. The midday mean  $\Delta N_{\text{neg}}$  at the Kumpula urban garden area was 4.9  $\text{cm}^{-3}$ , which was higher than in any of the studied forests. The presence of more abundant nucleation precursors at the Kumpula urban area may facilitate the ion formation (Nieminen et al., 2018). In summer,  $\Delta N_{\text{neg}}$  decreased compared to spring at all the sites except Siikaneva peatland and Tvärminne coastal areas. Seasonal changes in the clustering precursors and their dependence on air temperature and radiation may drive the seasonal variation of  $\Delta N_{\text{neg}}$  at all the sites.



It is notable that all the agricultural sites had higher midday  $\Delta N_{\text{neg}}$  than the forest sites in spring, varying between 2.3 and 7.7  $\text{cm}^{-3}$ . The application of fertilizers in agricultural fields is known to remarkably increase the atmospheric concentration of ammonia ( $\text{NH}_3$ ) (Olin et al., 2022).  $\text{NH}_3$  can stabilize the critical clusters in the nucleation process driven by sulfuric acid ( $\text{H}_2\text{SO}_4$ ) (Kulmala et al., 2013).  $\text{H}_2\text{SO}_4$  in the air is majorly formed by oxidation of sulphur dioxide, which can be transported from a longer range than the intermediate ions. However, the frequency of NPF events was found not to increase after the fertilization in Qvidja grasslands (Dada et al., 2023). Similarly, the frequency of daytime NPF events did not correlate with agriculture activities in a cropland in France (Kammer et al., 2023). Dada et al. (2023) observed that  $\text{NH}_3$ ,  $\text{H}_2\text{SO}_4$ , and low volatile organic compounds originating from BVOC oxidation play a synergistic role in clustering in Qvidja, resulting in a higher formation rate and number concentration of particles than in Hyytiälä forest. Note that since the Haltiala and Viikki croplands are located in Helsinki, the nucleation precursors and thereby the nucleation rate may be enhanced by anthropogenic pollution in the city. The exact reasons why there were higher  $N_{\text{neg}}$  and  $\Delta N_{\text{neg}}$  at these agricultural sites require more measurement of the clustering precursors.

Furthermore, in spring and summer, the night-time  $N_{\text{neg}}$  increased again at around 20:00 for all the sites, suggesting a ubiquitous nighttime clustering in warm seasons (Mazon et al., 2016). Moreover, in summer, the 75<sup>th</sup> percentile of nighttime  $N_{\text{neg}}$  at Viikki was comparable with the daytime  $N_{\text{neg}}$ . The decreased boundary layer height (Chen et al., 2016; Neeffjes et al., 2022), especially in clear nights, may also facilitate the accumulation of formed clusters and eventually lead to the nighttime peak.

### 3.3 Potential of different ecosystems to contribute to $\text{CO}_2$ uptake and negative intermediate ion production

Since we aimed to compare the potential of ecosystems for net  $\text{CO}_2$  uptake and local production of negative intermediate ions (LIIF), the most active periods for the ecosystem plants are discussed in detail in this section, i.e., midday in summertime. The potential of the studied ecosystems for net  $\text{CO}_2$  uptake and LIIF at midday during summertime are listed in Table 2. For median values in summer,  $N_{\text{neg}}$  was found to be highest in the urban garden, followed by the agricultural fields (Figure 9). The agricultural fields generally had higher  $N_{\text{neg}}$  than the studied forests. The open peatland had lower  $N_{\text{neg}}$  than Hyytiälä forest but higher than the other forests. The  $N_{\text{neg}}$  at the



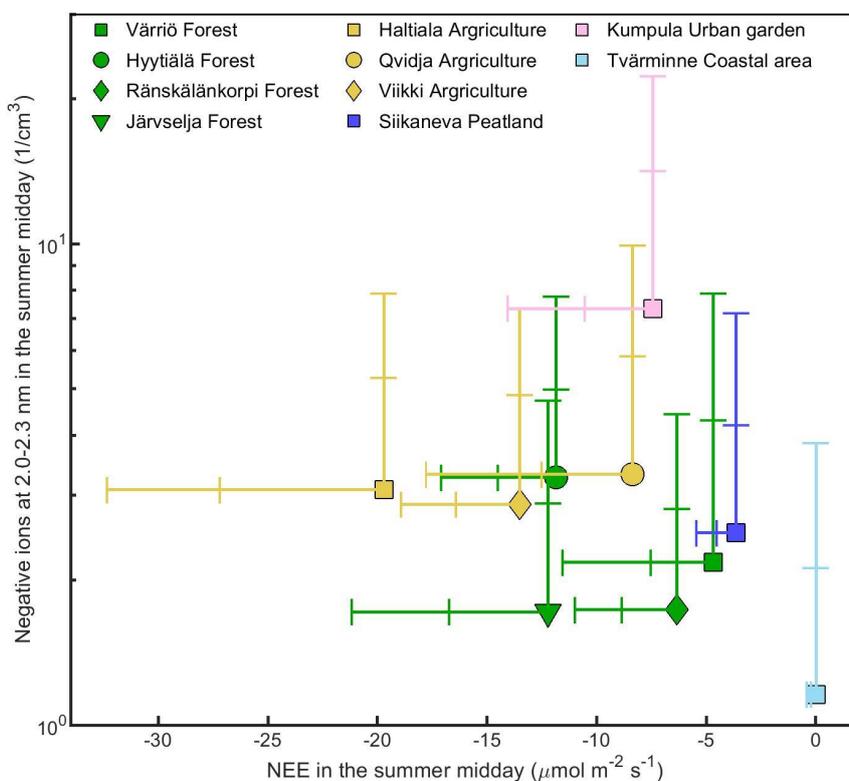
coastal area was the lowest. The momentary net CO<sub>2</sub> uptake rate at midday in summer was highest in agricultural fields, followed by the forests. The urban garden in this study displayed distinct net CO<sub>2</sub> uptake, lower than the forests and higher than the open peatland. The coastal area at midday  
370 in summer was a very weak CO<sub>2</sub> sink. In the urban garden area in Kumpula, median  $N_{\text{neg}}$  was 2.2 times of that in Hyytiälä forest, while the median NEE only reached 63% of that in Hyytiälä forest. The variation of momentary NEE and  $N_{\text{neg}}$  were distinct even between a similar type of ecosystem in a similar latitude, e.g., within forests and agricultural fields. For forests, the most southern Järvelja had the highest net CO<sub>2</sub> uptake rate, while the median  $N_{\text{neg}}$  in the midday in summer was  
375 similar to Ränskälänkorpi and 53% of that in Hyytiälä forest. Hyytiälä forest had higher  $N_{\text{neg}}$  than the other forests. For agricultural sites, the net CO<sub>2</sub> uptake rate at Qvidja and Viikki were close to that in Hyytiälä forest, while it was much higher in Haltiala croplands than in Hyytiälä forest. On the contrary, the  $N_{\text{neg}}$  were highest in Qvidja between the three agricultural sites, and median  $N_{\text{neg}}$  in the other two sites were slightly smaller than in Hyytiälä forest.

380 Another potent greenhouse gas, methane (CH<sub>4</sub>) can be emitted through microbial activities in anoxic conditions, e.g., peatlands and coastal areas (Mathijssen et al., 2022; Roth et al., 2023). Considering that CH<sub>4</sub> has a sustained-flux global warming potential 45 times of CO<sub>2</sub> over 100 years (Roth et al., 2023; and the reference therein), the net CO<sub>2</sub> equivalent emission of CH<sub>4</sub> is estimated 2.5-8.6 times of CO<sub>2</sub> uptake in Siikaneva peatland (Mathijssen et al., 2022). CH<sub>4</sub>  
385 emissions may largely compensate the CO<sub>2</sub> uptake in open and non-ditched peatlands. Similarly, the emission of CH<sub>4</sub> from coastal environment around Baltic Sea may offset 28% of the CO<sub>2</sub> sink in macroalgae-dominated coastal area (Roth et al., 2023). For ions, the summertime midday median  $N_{\text{neg}}$  at the peatland in Siikaneva was 77% of that in Hyytiälä forests (Table 2). As the open peatland is surrounded by forest within 1 km, the negative ion at 2.0-2.3 nm may be influenced by  
390 nearby forests. Also, the terpene emissions from the peatlands can initiate stronger NPF than in the Hyytiälä boreal forest (Junninen et al., 2022; Huang et al., 2024). However, these events were majorly reported to occur at late evening.

The CarbonSink+ potential, especially CO<sub>2</sub> uptake, may largely vary within agricultural fields in Finland. Agricultural fields may be highly productive in local formation of negative intermediate  
395 ions, affected by their vegetation and management practises. However, considering the much larger area of forests in Finland than that of agricultural fields (Table 2), boreal forests in Finland



in total are likely the largest contributor of climate cooling when considering the CO<sub>2</sub> uptake and local new particle formation.



400

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



404 Table 2. Comparison of NEE and negative intermediate ions at 2.0-2.3 nm size range across  
405 the hemi-boreal and boreal ecosystems at midday (10:00-14:00) in summer.

Ecosystem	Site	Area in Finland (ha)	Median $N_{neg}$ ( $1/cm^3$ )	Median $N_{neg}/median N_{neg, Hyytiälä}$	75 <sup>th</sup> percentile $N_{neg}/75^{th}$ percentile $N_{neg, Hyytiälä}$	Midday NEE ( $\mu mol m^{-2} s^{-1}$ )	Median NEE/ Median $NEE_{Hyytiälä}$	25 <sup>th</sup> percentile NEE/ 25 <sup>th</sup> percentile $NEE_{Hyytiälä}$
Forest	Hyytiälä	20.3 million <sup>a</sup>	3.27	1	1	-11.84	1	1
	Värriö		2.18	0.67	0.87	-4.69	0.4	0.52
	Järvelselja		1.72	0.53	0.58	-12.23	1.03	1.15
Drained peatland forest	Ränskälänk orpi	4.2 million <sup>a</sup>	1.74	0.53	0.57	-6.35	0.54	0.61
Agricultural field	Haltiala	2.3 million <sup>a</sup>	3.08	0.94	1.06	-19.69	1.66	1.88
	Qvidja		3.32	1.01	1.17	-8.37	0.71	0.86
	Viikki		2.88	0.88	0.97	-13.52	1.14	1.13
Open peatland	Siikaneva	0.21 million <sup>c</sup>	2.51	0.77	0.85	-3.65	0.31	0.31
Urban garden area	Kumpula	-----	7.33	2.24	2.86	-7.44	0.63	0.73
Coastal area	Tvärminne	-----	1.46	0.45	0.53	0.01	0.00	0.01

406 <sup>a</sup> Natural Resources Institute Finland 2022; <sup>b</sup> The area of oligotrophic open fens (Turunen and Valpola 2020);

407 ----- data not available

408

#### 409 4. Conclusions

410 The CarbonSink+ potential concept was established recently and provides a direct comparison  
411 of local contribution to CO<sub>2</sub> uptake and aerosol formation at ecosystem scale. The value of  
412 negative intermediate ion concentration at 2.0-2.3 nm size range ( $N_{neg}$ ) was applied as an  
413 indicator of the corresponding contribution of each ecosystem to produce new aerosol particles  
414 which, after their subsequent growth to larger sizes, are able to cool the atmosphere in a  
415 regional scale. Following this concept, net ecosystem CO<sub>2</sub> exchange fluxes (NEE) and  $N_{neg}$   
416 were analysed in ten hemi-boreal and boreal ecosystems in Finland and Estonia. The boreal



417 forest in Hyytiälä was chosen as a reference site, to which the values of NEE and  $N_{\text{neg}}$  at all  
418 other sites were all compared.

419 The results showed that the agricultural fields had similar or even higher CO<sub>2</sub> uptake potential  
420 compared to Hyytiälä forest during the summer. Note that the decreased carbon storage due to  
421 harvest in the fields was not taken into account in this study. A distinct CO<sub>2</sub> uptake in the urban  
422 garden at midday in summer was observed, lower than that in Hyytiälä forest but higher than  
423 observed in the open peatland. The coastal area considered in this study remained a very small  
424 CO<sub>2</sub> source during summertime. The differences in  $N_{\text{neg}}$  between the studied sites were not as  
425 large as those in NEE. Ubiquitous nighttime clustering was observed across the ecosystems.  
426 At midday in summer,  $N_{\text{neg}}$  was highest in the urban garden, followed by the agricultural fields.  
427 The coastal area had the lowest  $N_{\text{neg}}$ . The forest sites generally had lower  $N_{\text{neg}}$  than the  
428 agricultural sites. The  $N_{\text{neg}}$  in the open peatland was lower than Hyytiälä forest but higher than  
429 other studied forests. Note that the urban garden and agricultural sites in Helsinki might be  
430 more influenced by air pollution compared to the forests and open peatland that were  
431 background sites. Overall, considering the large area of forests in Finland and Estonia, the  
432 forests in total have the largest potential of climate cooling when considering the CO<sub>2</sub> uptake  
433 and local new particle formation.

#### 434 **Data availability**

435 Measurement data at the sites, including ions data, eddy covariance data and meteorological  
436 data, are available upon request from the corresponding author before the relevant databases  
437 are open to the public.

#### 438 **Author contributions**

439 ST, JL, and RT were responsible for the ion measurements. PS, AL, MP, AL, MK, HR, LH,  
440 AV, IM, and SN were responsible for the eddy covariance measurement and analysed the raw  
441 data. MK designed the study. PKe, AL, PKo, TN, OP, EE, TK, JB, VMK, and MK analysed  
442 the data and interpreted the results. PKe prepared the first-draft paper. All authors contributed  
443 to discussion of the results and provided input for the paper.



#### 444 **Competing interests**

445 The authors declare no competing interests.

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