BOREAL ENVIRONMENT RESEARCH 29: 1–16
ISSN 1797-2469 (online)
Helsinki 5 January 2024

A novel concept for assessing the potential of different boreal ecosystems to mitigate climate change (CarbonSink+ Potential)

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Received 30 Apr. 2023, final version received 3 Aug. 2023, accepted 15 Sep. 2023

Kulmala M., Ke P., Lintunen A., Peräkylä O., Lohtander A., Tuovinen S., Lampilahti J., Kolari P., Schiestl-Aalto P., Kokkonen T., Nieminen T., Dada L., Ylivinkka I., Petäjä T., Bäck J., Lohila A., Heimsch L., Ezhova E. & Kerminen V.-M. 2024: A novel concept for assessing the potential of different boreal ecosystems to mitigate climate change (CarbonSink+ Potential). *Boreal Env. Res.* 29: 1–16.

In a changing climate, it is critical to reduce emissions of carbon dioxide and other greenhouse gases. At the same time, we need to remove carbon dioxide from the atmosphere. In addition, it is important to make other radiative forcing components, such as those due to changes in atmospheric aerosol loadings, clouds and albedo, to counteract the warming effects of greenhouse gases. An important way to reduce warming is the removal of CO_2 from atmosphere by ecosystems, which act as carbon sinks and storages. However, ecosystems influence also other radiative forcing components, yet the full potential of different ecosystems to mitigate climate warming is challenging to compare. Here we propose a novel concept (CarbonSink+ Potential) to compare ecosystems in terms of their carbon uptake and aerosol production capacity. In our approach, we utilize the regional aerosol formation measured at the SMEAR II station in Hyytiälä, southern Finland, together with locally measured negative ion concentrations at various ecosystems within the region (forest, peatland and grassland). The local ion concentrations are measured in the size range of 2.0–2.3 nm that indicates aerosol formation within a source area of roughly sim-

Editor in charge of this article: Antti Arola

ilar size as that of carbon sink measurements. The results show that, among the studied boreal ecosystems and per surface area, the pristine peatland has the lowest aerosol production and carbon sink, so this ecosystem is likely to have the smallest potential to contribute to climatic cooling (per land area). Forest (on mineral soil) has the highest carbon sink and grassland (on mineral soil) has the highest potential for aerosol production. This means, for example, that the relative contribution of grassland to climate mitigation is more important than when considering only the carbon sink.

Background

In order to meet the interlinked environmental grand challenges, such as climate change, biodiversity loss, air pollution, and sustainable supply and use of natural resources, an integrated multi-disciplinary approach is needed (e.g. Kulmala 2018, Kulmala *et al.* 2021). To demonstrate the effectiveness of the integrated approach, we have recently developed the CarbonSink+ concept (Kulmala *et al.* 2020), which incorporates different radiative forcing effects related to ecosystem-atmosphere interactions and provides a comprehensive impact assessment. CarbonSink+ is based on the analysis of continental biosphere-aerosol-cloud-climate (COBACC) feedback loop, tested with long-term field observations (Kulmala *et al.* 2004, 2014).

Forests cool the climate system by acting as a sink for carbon dioxide (CO_2) and source of atmospheric aerosol particles, whereas forests have relatively low surface albedo that tends to have a warming effect (Kurtén *et al.* 2003, Kalliokoski *et al.* 2020). Using the boreal forest environment (SMEAR II station; Hari and Kulmala 2005) as an example, it was found that aerosol particles produced by forests contribute to the regional cooling via aerosol-radiation and aerosol-cloud interactions (Sporre *et al.* 2019, Yli-Juuti *et al.* 2021, Petäjä *et al.* 2022). Furthermore, we estimated that the forest CO_2 uptake was enhanced by 10–50% in a boreal environment due to the combined effects of CO_2 fertilization and aerosol-induced diffuse radiation enhancement on photosynthesis (Kulmala *et al.* 2020, Launiainen *et al.* 2022). We further estimated that with afforestation or reforestation, i.e. replacing open areas with forests in a boreal environment, the radiative cooling due to forest aerosols cancels most of the radiative warming due to the decreased surface albedo. These two forcing components have, however, relatively large uncertainty ranges, resulting in large uncertainties in the overall effect of CarbonSink+. In the future, it is crucial to investigate and to quantify CarbonSink+ in different environments globally.

Radiative effects via aerosol-cloud interactions are usually the most important pathway by which secondary aerosol particles produced by terrestrial ecosystems cool the climate (e.g. Makkonen *et al.* 2012, Carslaw *et al.* 2013, Rap *et al.* 2013, Sporre *et al.* 2019). The most essential aerosol-related quantity in this respect is the number concentration of aerosol particles able to act as cloud condensation nuclei (CCN). In the boundary layer and especially over the continents, an important and often the dominant source of CCN is atmospheric new particle formation, NPF (Merikanto *et al.* 2009, Kerminen *et al.* 2012, Gordon *et al.* 2017, Peng *et al.* 2014, Ren *et al.* 2021, Kulmala *et al.* 2023). The growth of newly formed particles to CCN varies from a few hours in polluted environments to more than a day at remote sites (Ren *et al.* 2021, Petäjä *et al.* 2022, Räty *et al.* 2023). As a result, atmospheric CCN production is always a regional process involving emissions of aerosol precursors over spatial scales of tens to hundreds of kilometers, typically covering a number of different ecosystems and ecosystem types.

While the CarbonSink+ concept makes it possible to compare the relative roles of carbon uptake, atmospheric NPF and surface albedo changes in mitigating the climate in a specific environment using a single metric (Kulmala *et al.* 2020), it does not allow estimating the corresponding roles of different ecosystem types within the considered environment. The main reason for this is that some of the ecosystem-specific processes, such as the initial stages of atmospheric NPF, cause a measurable climate-relevant signal only at regional scales (CCN formation associated with NPF), whereas the climatic effects of e.g. greenhouse gases can be estimated for each ecosystem from direct measurements. The main purpose of this paper is to introduce a means to compare between different ecosystems in this respect. We name the new concept as "CarbonSink+ Potential", and illustrate briefly how to apply it to two climatically important components in a boreal forest environment: regional CCN production and carbon sink. The "CarbonSink+ Potential" concept can be extended to other climatically important components, such as surface albedo changes and other greenhouse gases, in a straightforward manner, so these components will not be considered in this work. We further note that a full estimation of the climate mitigation potential requires combining the two concepts, CarbonSink+ and CarbonSink+ Potential, which will be a topic of future investigations.

Theoretical approach

At SMEAR II and various other locations, we are measuring carbon sink as net ecosystem exchange of CO_2 (NEE), which can be partitioned into two components, the gross primary production (GPP) and respiration (Lintunen *et al.* 2023). Here, we use negative NEE as a measure of carbon sink. Besides carbon sink, we also measure the other components of CarbonSink+. Here we focus on the climate impacts of aerosols and CO_2 and exclude the surface albedo effect and the effects of other greenhouse gases.

The radiative effects of aerosol particles are regional with large source areas (e.g. Henze *et al.* 2012, Persad *et al.* 2022), and therefore aerosol radiative effects can be considered relatively similar over the whole southern and central Finland (Spracklen *et al.* 2008, Lihavainen *et al.* 2009). Although the contribution of NPF to the pool of large atmospheric aerosol particles responsible for the radiative effect is a regional phenomenon, the initial steps of NPF are local processes dictated by different formation rates of atmospheric clusters and tiny particles over different types of ecosystems. As a result, by comparing the different local formation rates and quantities of atmospheric clusters in different ecosystems to the local source around SMEAR II, we can estimate their relative contributions to the regional radiative effects of aerosol particles.

The initial stage of atmospheric NPF, called clustering, may depend on a combination of: 1) vapors typically present in the regional atmosphere also outside urban areas, such as sulfuric acid originating from the atmospheric oxidation of sulfur dioxide; 2) vapors with highly localized emissions, such as ammonia or amines; and 3) vapors having typically a notable variability over both local and regional scales, such as many low-volatile organic compounds originating from the atmospheric oxidation of biogenic volatile organic compounds (e.g. Lehtipalo *et al.* 2018, Beck *et al.* 2021, Cai *et al.* 2021, He *et al.* 2021, Yan *et al.* 2021). As a result, while atmospheric NPF tends to be observed as a regional phenomenon in continental boundary layers (e.g. Kerminen *et al.* 2018), the strength of clustering is expected to have a high spatial variability, and especially so between ecosystems with different emission rates of the precursors that determine cluster formation rates. This suggests that ecosystems with higher (lower) than average clustering rates provide a disproportionally larger (smaller) contributions to the total CCN concentration in the regional atmosphere that these ecosystems belong to. How do we determine such contributions?

Let us take the Fennoscandian part of the boreal forest region as an example case. It has been shown that aerosol particles originating from biogenic emissions from this environment dominate the regional CCN budget outside the late autumn and winter periods (Tunved *et al.* 2006, Petäjä *et al.* 2022, Kulmala *et al.* 2023). At the same time, measurements indicate highly variable clustering rates between different ecosystems (for example, between forests, peatlands and agricultural grasslands considered in this paper), as well as over relatively small spatial scales within a given environment (Svenningsson *et al.* 2008, Lampilahti *et al.* 2021, Junninen *et al.* 2022, Olin *et al.* 2022). In

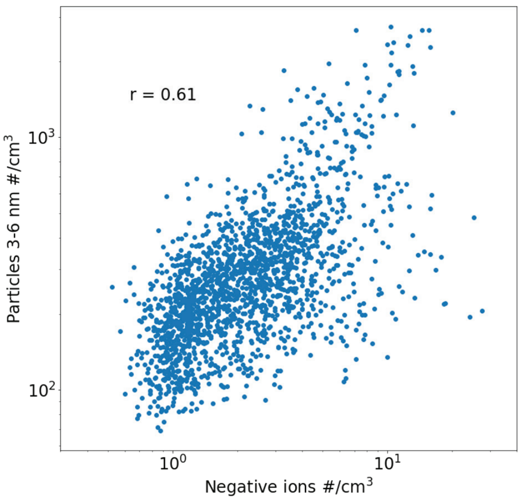

Fig 1. Daily average (08:00-16:00) total concentrations of 3-6 nm particles as a function of 2.0–2.3 nm negative ion cluster concentration measured in Hyytiälä.

order to estimate the relative contributions of an ecosystem type i to the CCN budget over this region ($f_{\mathrm{CCN,i}}$), we need to take into account the fractional area of such ecosystems in the region, the average number concentration of growing nanoparticles produced by this ecosystem into the regional atmosphere (N_i), and the survival probability (SP_i) of the newly formed particles during their growth to CCN:

$$f_{\mathrm{CCN,i}} = A_i/A_{\mathrm{tot}} \times N_i/N \times \mathrm{SP}_i/\mathrm{SP}\ . \qquad (1)$$

Here, A_i is the total area of the ecosystems of type i in this region, A_{tot} is the whole area of the region, N is the regionally averaged number concentration of growing nanoparticles and SP is their survival probability. The challenge, as discussed below, is to simultaneously estimate the last two multiplying factors in the product in Eq. 1 when using atmospheric observations.

Table 1. Particle survival probabilities during their growth from 5 nm to 100 nm calculated using the formula derived by Kerminen and Kulmala (2002).

GR (nm/h)	CS (s^{-1})		
	10^{-4}	10^{-3}	10^{-2}
2	0.98	0.80	0.11
4	0.99	0.90	0.34
8	0.99	0.95	0.58

In order to estimate N_i, i.e. to capture the contribution of local NPF, or clustering, to the number concentration of the growing nanoparticles, we can rely on ion measurements. The concentration of small air ions is a good indicator of atmospheric NPF, and a useful tool to classify individual NPF events (Leino *et al.* 2016, Dada *et al.* 2018). Our recent analysis further demonstrates that NPF is most sensitive to ion concentrations in a single size bin of 2.0–2.3 nm of the neutral cluster and air ion spectrometer (NAIS) instrument (Tuovinen *et al.* 2023). These ions are small enough to represent local NPF intensity, in addition to which there appears to be a tight connection between the concentrations of these ions and 3–6 nm aerosol particles when averaged over daytime (08:00–16:00; Fig. 1). Based on Fig. 1, it seems reasonable to assume that a daily-average N_i due to local NPF is proportional to the corresponding concentration of 2.0–2.3 nm ions measured in this ecosystem.

While solid theoretical frameworks for estimating the survival probabilities of growing clusters and larger particles exist (McMurry *et al.* 1979, Kerminen and Kulmala 2002, Lehtinen *et al.* 2007, Pierce and Adams 2007, Kuang *et al.* 2009, Westervelt *et al.* 2013), survival of clusters and particles remains poorly understood in the diameter range of 2–5 nm, especially in polluted environments (e.g. Kulmala *et al.* 2017, Tuovinen *et al.* 2022). Under non-precipitating conditions, survival of growing particles and clusters up to CCN sizes depends essentially on the ratio between their growth rate (GR), and their coagulation sink (e.g. Westervelt *et al.* 2013), the latter quantity being directly proportional to the pre-existing particle loading that is often described using condensation sink, CS (e.g. Kulmala *et al.* 2012). The sensitivity of the survival probability (SP) to the ratio GR/CS, or its variability, increases rapidly with a decreasing particle size (e.g. Kulmala *et al.* 2017, Tuovinen *et al.* 2022). Taking into account this sensitivity, together with our poor understanding on the survival probability in the sub-5 nm size range, we should avoid applying this concept to sizes smaller than about 5 nm in Eq. 1.

Let us then investigate the survival of particles during their growth from 5 nm to CCN. Table 1 shows that for this size range, SP is considerably lower than unity only when low values of GR (~2 nm/hour) occur simultaneously with values of CS approaching 10^{-2} s^{-1}. Based on long term observations, such a combination of relatively high CS and relatively low GR appears possible yet rare in a boreal forest environment (Dal Maso *et al.* 2007, Neefjes *et al.* 2022, Kulmala *et al.* 2023). Concerning the variabilities of these quantities, CS is expected to have a relatively smooth spatial variability over remote areas affected little by primary particle sources. Observations between different sites (Dal Maso *et al.* 2007), or during and between different months at the SMEAR II station (Dada *et al.* 2017, Neefjes *et al.* 2022), indicate that CS varies mostly within a factor of 2–3 in the Fennoscandian part of the boreal forest region, with the most typical values being slightly above 0.001 s^{-1}. Compared with strength of NPF, i.e. new particle formation rates, the variability of GR tends to be relatively small between different environments, as well as within the boreal forest region (Dal Maso *et al.* 2007, Dada *et al.* 2017, Kerminen *et al.* 2018, Nieminen *et al.* 2018, Kulmala *et al.* 2022a, 2023). Thus, we conclude that it is reasonable to approximate $SP_i/SP \approx 1$ in the Fennoscandian part of the boreal environment.

Long-term measurements in Hyytiälä can be thought to represent the average CCN formation potential of the whole Fennoscandian part of the boreal environment. After combining the information given above, we suggest the following approximation to Eq. 1:

$$f_{\mathrm{CCN,i}} = A_i/A_{\mathrm{tot}} \times N_{\mathrm{ion,i}}/N_{\mathrm{ion}}\,, \qquad (2)$$

where $N_{\mathrm{ion,i}}$ is the daily-average concentration of 2.0–2.3 nm ions measured in the ecosystem type i and N_{ion} is the corresponding ion concentration measured at SMEAR II. The main error source in Eq. 2 is the representativeness of the ion concentration to describe the strength at which local NPF produces growing particles to sizes > 3 nm (Fig. 1). The survival of these particles to CCN sizes is expected to be influenced much less by local emissions (i.e $SP_i/SP \approx 1$), as the bulk growth to CCN occurs anyway in the regional atmosphere.

Materials and methods

Sites

Hyytiälä (the SMEAR II station) is a medium fertile boreal Scots pine dominated upland forest site in southern Finland (61.84°N, 24.29°E, 178 m a.s.l.) (Hari and Kulmala 2005, Kolari *et al.* 2022). The mean annual temperature of the site is +4.1°C and annual precipitation 690 mm (the means for the years 1991–2020, Jokinen *et al.* 2021). The stand was established in 1962, and the mean tree height is 19.7 m. Most of the vegetation is evergreen, i.e. annual variation in leaf area index (LAI) is small.

Siikaneva is a pristine, open oligotrophic fen site located 5 km west from Hyytiälä (61.83°N, 24.19°E, 167 m a.s.l.). Its vegetation consists of *Spaghnum* moss cover, sedges and dwarf shrubs (Rinne et al. 2018). Most of the vegetation is deciduous, i.e. there is strong seasonality in LAI (Alekseychik *et al.* 2017).

Qvidja is an agricultural grassland site on mineral soil located in an archipelago in Parainen, South-West Finland (60.29°N, 22.39°E; 5 m a.s.l.). The mean annual air temperature and precipitation at this area are 5.4°C and 679 mm, respectively (1981–2010, Pirinen *et al.* 2012). The soil type at the site is clay loam, and the dominant plant species at the target area are timothy (*Phleum pratense*), meadow fescue (*Festuca pratensis*) and white clover (*Trifolium repens*). The site is managed by fertilizing twice per year and grass cutting or yield harvest 2–3 times per year. 220 and 178 kg ha^{-1} of mineral fertilizers were applied in June 2019 and April 2021, respectively, whereas 4560, 4610, 19 380, and 15 330 kg ha^{-1} of organic fertilizers were applied in 2018, 2019, 2020, and 2022, respectively. In August 2021 and 2022, there were short grazing periods. For a more detailed description of the site and management in 2018 and 2019, see Heimsch et al. (2021).

Värriö is a northern boreal Scots pine dominated forest site in northeastern Lapland (67.75°N, 29.61°E, 370 m a.s.l). It is characterized with low temperatures, short active season, and a nutrient poor, rocky soil (Hari *et al.* 1994, Kulmala *et al.* 2019). The mean annual temperature is +0.1°C and annual precipitation is

Fig 2. Map of observational sites.

607 mm (the means for the years 1991–2020, Jokinen *et al.* 2021). The mean age of trees is 80 years and their mean height is 9 m. Most of the vegetation is evergreen, i.e. annual variation in LAI is small. The location of the above sites could be seen in Fig. 2.

Eddy covariance measurements to observe carbon sink

In this study, the NEE (net ecosystem exchange) was measured with eddy covariance technique (Aubinet *et al.* 1999) at all the sites. The eddy covariance monitoring system for CO_2 at the four sites is similar, composed of a three-dimensional ultrasonic anemometer (USA-1/uSonic-3 Scientific, METEK GmbH, Germany or R2/HS-50, Gill Instruments, UK) and an enclosed infrared absorption CO_2/H_2O gas analyzer (LI-6262, LI-7200, or LI-7000, LI-COR Biosciences) mounted on a tower above the canopy. Before April 2018, the CO_2 fluxes at Hyytiälä were measured at 24.3 m height and then the measurement height was changed to 27 m as the forest canopy height was increasing. The CO_2 fluxes were measured at 16.6 m, 3.0 m, and 2.3 m for Värriö, Siikaneva and Qvidja, respectively.

The raw 10 Hz data were processed through standard steps, including de-spiking, dilution correction, 2-D coordinate rotation, lag-time adjustment, and correction for frequency response. For SMEAR sites, including Hyytiälä, Värriö and Siikaneva, the EddyUH software (Mammarella et al. 2016) was used to process the raw data into 30-min average NEE. Detailed instrument setup and raw data processing at each site can be found in Launiainen *et al.* (2022), Kulmala *et al.* (2019), Rinne *et al.* (2018) and Heimsch *et al.* (2021) for Hyytiälä, Värriö, Siikaneva and Qvidja, respectively.

The eddy covariance measurements at Hyytiälä, Värriö, Siikaneva and Qvidja started in April 1996, July 2012, January 2005 and May 2018, respectively. The NEE data with common measurement periods with NAIS (see next section) were used in this study, i.e. June 2014–December 2021, February 2019–December 2021, October 2019–December 2021, and November 2018–August 2022 for Hyytiälä, Värriö, Siikaneva and Qvidja, respectively.

The half-hour averaged CO_2 fluxes were corrected for the change in CO_2 storage below the measurement height at Hyytiälä and Värriö. The fluxes were rejected if they were measured in non-stationary conditions (Vickers and Mahrt 1997), or if friction velocity was below a site-specific threshold (0.38–0.4 m/s for Hyytiälä and Värriö, and 0.1–0.2 m s^{-1} for Siikaneva and Qvidja). For Hyytiälä, considering the effects of thinning conducted in March 2020 (see Aalto *et al.* 2023), the data from March 2020 until the end of the year were rejected to exclude the immediate harvest impact to the carbon fluxes. For Qvidja, the flux data were accepted only when the wind direction was between 0–30° or 140–360° to avoid interference from the nearby experiment sites located in the east.

The CO_2 fluxes were partitioned into ecosystem respiration and gross primary production (GPP). Empirical site-specific temperature responses of ecosystem respiration were established with the moving time window (Kulmala *et al.* 2019, Kolari *et al.* 2009). The soil and air temperature were applied in the respiration parameterization for the forested sites, while only soil temperature was used for the peatland and only air temperature for the grassland (Kulmala *et al.* 2019, Heimsch *et al.* 2021). When the flux quality and turbulence criteria were met, the GPP was calculated as the difference between modelled respiration and measured NEE.

Aerosol production measurements

Aerosol production was estimated based on ion number concentrations measured by a neutral cluster and air ion spectrometer (NAIS, Airel Ltd.; Mirme and Mirme 2013). The NAIS is capable of measuring ion and total particle size distributions in the diameter ranges of 0.8–42 nm and ~2–42 nm, respectively (e.g. Wagner *et al.* 2016). After data cleaning and quality check, the potential interference of rainfall and snow events on negative ions measurements was taken into account (Manninen et al., 2016). When the air relative humidity exceeded 70% and the error messages, i.e., fluctuating sampling flows, were reported from NAIS, the ion and particle data were discarded.

Tuovinen et al. (2023) have recently shown that the negative ion concentration from a channel with a diameter range of 2.0–2.3 nm is the best indicator for particle formation within a source area of similar magnitude to that of NEE, typically within a kilometer or less from the measurement site. The exact values of these source areas vary e.g. with local wind characteristics and, in case of ions, also their growth rate (Tuovinen et al. 2023). We used concentrations from this single channel to characterize the local particle production rate by the different areas. The quantity N_i in Eq. 2 is then the concentration of negative ions within the diameter range of 2.0–2.3 nm measured at area *i* and *N* is the corresponding value for SMEAR II during the same time window.

In addition to the negative ion concentrations, total particle concentrations in the size range 3–6 nm were used to illustrate the relationship of the ion concentrations with that of larger total particles. Particles in the size range 3–6 nm were acquired from the NAIS measuring in total mode, from the negative polarity.

Results and discussion

NEE and aerosol production at different ecosystems

First, we analyzed daily behavior of the measured NEE and negative ions having similar source areas. Spring (March, April and May, Figs. 3 and 5) and summer (June, July, August) (Figs. 4 and 6) seasons were selected for this purpose because NPF tends to be most frequent during spring and photosynthesis is most pronounced during summer (Neefjes et al. 2022). We present the data as 10th, 25th, 50th, 75th and 90th percentiles calculated of half-hourly means separately for spring and summer months (note that negative values of NEE correspond to a net carbon uptake).

During daytime, the median (i.e. 50th percentile) and mean values of NEE were negative at all the studied sites in both spring and summer, indicating carbon sink (Figs. 3 and 4 and S1–S2 in the Supplementary Information). In Hyytiälä, even the 90th percentiles were negative at daytime in the summer. The same is true for Siikaneva and Qvidja, but not in Värriö where the 90th percentile line is positive during daytime, indicating that Värriö is a carbon source more than 10th of the time. Note that the climate impacts of other greenhouse gases (GHG), such as methane that is emitted in significant amounts especially from peatland sites (e.g. Rinne et al. 2018) are not accounted for in this analysis. A difference in the carbon flux between southern and northern Finnish forests is clearly seen: the 10th and 90th NEE percentiles were distinctly higher in Hyytiälä than in Värriö. The corresponding GPP values are given in the Supplementary Information.

The median concentrations of small ions showed clear maxima slightly before the noon in all the sites and percentile categories in both spring and summer (Figs. 5 and 6), although

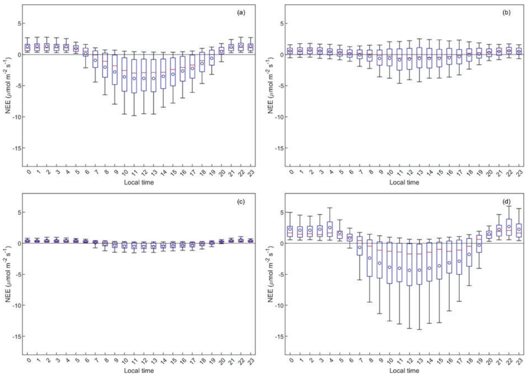

Fig 3. Hourly percentiles of NEE data during spring (MAM) for sites: (**a**) Hyytiälä; (**b**) Värriö; (**c**) Siikaneva; and (**d**) Qvidja. The data is non-gap-filled. The red center lines, the blue box and the black whiskers denote the 50th, 25th and 75th, 10th and 90th percentiles, respectively. The blue circles are the mean values.

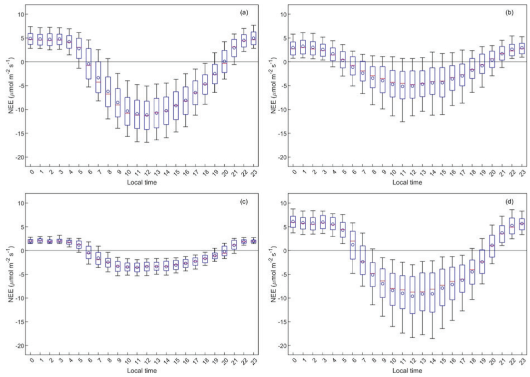

Fig 4. Hourly percentiles of NEE during summer (JJA) for sites: (**a**) Hyytiälä; (**b**) Värriö; (**c**) Siikaneva; and (**d**) Qvidja. The data is non-gap filled. The red center lines, the blue box and the black whiskers denote the 50th, 25th and 75th, 10th and 90th percentiles, respectively. The blue circles are the mean values.

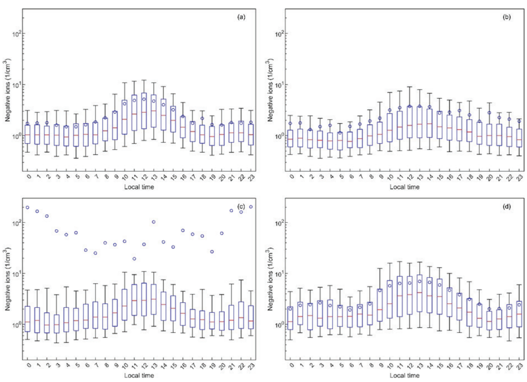

Fig 5. Hourly percentiles of negative ion concentrations (2.0–2.3 nm) during spring (MAM) for sites: (**a**) Hyytiälä; (**b**) Värriö; (**c**) Siikaneva; and (**d**) Qvidja. The red center lines, the blue box and the black whiskers denote the 50th, 25th and 75th, 10th and 90th percentiles, respectively. The blue circles are the mean values.

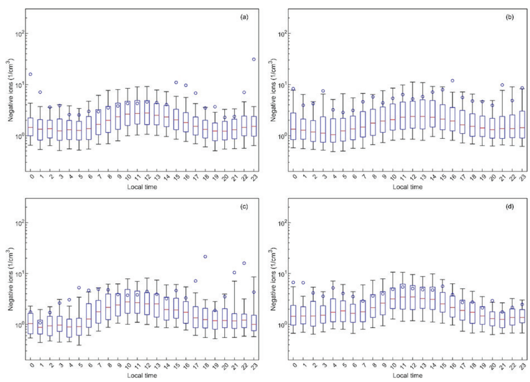

Fig 6. Hourly percentiles of negative ion concentrations (2.0–2.3 nm) during summer (JJA) for sites: (**a**) Hyytiälä; (**b**) Värriö; (**c**) Siikaneva; and (**d**) Qvidja. The red center lines, the blue box and the black whiskers denote the 50th, 25th and 75th, 10th and 90th percentiles, respectively. The blue circles are the mean values.

enhanced ion concentrations were apparent during most of the daytime. Even the 10th percentiles of the hourly data showed clear increases in ion concentrations, supporting our recent finding of quiet NPF (Kulmala *et al.* 2022b) which represents NPF taking place on days traditionally not classified as NPF event days. In general, concentrations of small ions were higher in spring than in summer. However, the concentration difference between 10th and 90th percentiles were more than factor of 20 in all the sites (Figs. 5 and 6). During the times with strong new particle formation (90th percentile), the values from Qvidja were the highest compared with the other ecosystems.

CarbonSink+ Potential

To find out the relative contribution of various ecosystems to CarbonSink+, we normalize the half-hour NEE and ion concentration values measured at each ecosystem with those measured in Hyytiälä. Figures 7 and 8 show these NEE

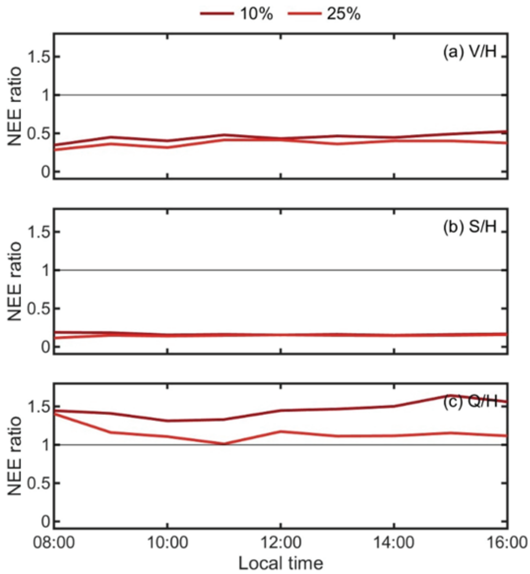

Fig 7. The diurnal variation of the ratio of NEE percentiles between different sites and Hyytiälä (SMEAR II) during spring (MAM) for: (**a**) Värriö/Hyytiälä; (**b**) Siikaneva/Hyytiälä; and (**c**) Qvidja/Hyytiälä.

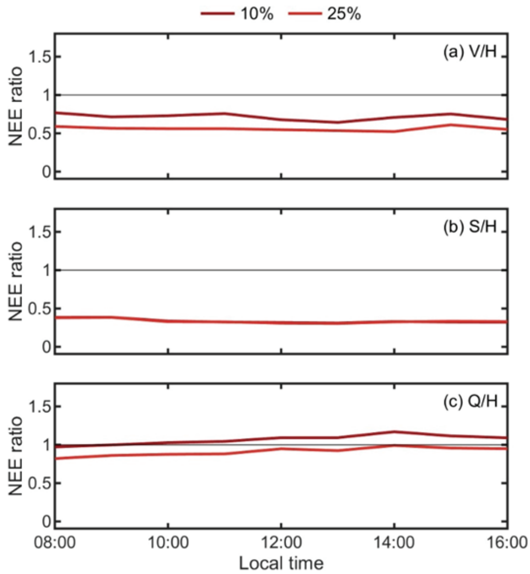

Fig 8. The diurnal variation of the ratio of NEE percentiles between different sites and Hyytiälä in the summer (JJA) for: (**a**) Värriö/Hyytiälä; (**b**) Siikaneva/Hyytiälä; and (**c**) Qvidja/Hyytiälä.

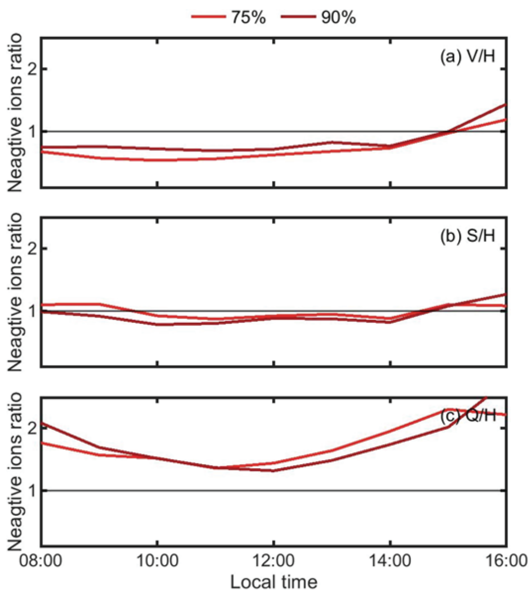

Fig 9. The diurnal variation of the ion (2.16 nm) concentration ratio between different sites and Hyytiälä (SMEAR II) during springtime (MAM) for: (**a**) Värriö/Hyytiälä; (**b**) Siikaneva/Hyytiälä; and (**c**) Qvidja/Hyytiälä.

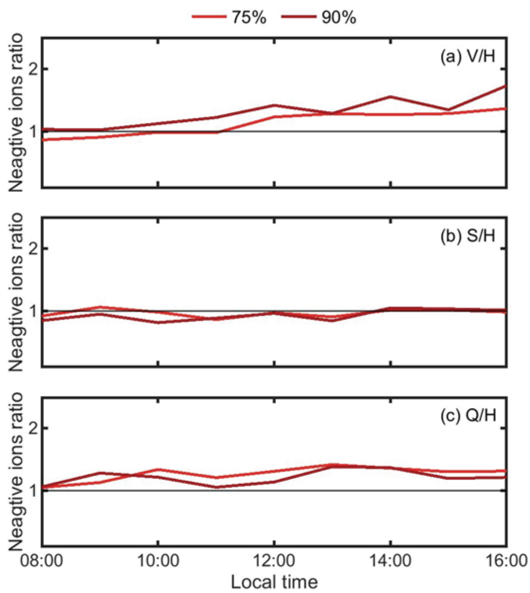

Fig 10. The diurnal variation of ratio of ion concentration percentiles during summertime (JJA) for: (**a**) Värriö/Hyytiälä; (**b**) Siikaneva/Hyytiälä; and (**c**) Qvidja/Hyytiälä.

ratios in spring and summer, respectively, while Figs. 9 and 10 show the corresponding ion concentration ratios. The 10th and 25th NEE percentiles were the lowest (corresponding to the highest carbon sink) in Qvidja during spring. During summer, the NEE values in Hyytiälä and Qvidja were rather similar (10% percentile higher in Qvidja and 25th higher in Hyytiälä). The aerosol production in Qvidja was higher than that in Hyytiälä during both spring and summer (see also Dada *et al.* 2023), indicating particularly strong contribution from agricultural grassland to regional NPF during days with strong NPF (90th percentiles).

Next, we utilize the calculated percentiles to calculate CarbonSink+ Potential. Because aerosol radiative effects are expected to be relatively similar over the southern and central Finland, but lower over the northern Finland (e.g. Spracklen *et al.* 2008, Lihavainen *et al.* 2009), we exclude Värriö site from this analysis. To calculate CarbonSink+ Potential, different seasons or emission percentiles can in principle be considered depending on the purpose of the ecosystem comparison. Here we are interested in comparing the maximum emission potential of the different ecosystems and thus focus on the 75th percentile of aerosol production and 25th of NEE (both corresponding to 75th percentile in absolute values) in the summer season. Thus, the results of this study can only be interpreted from the perspective of comparing the momentary climate mitigation potential of the studied sites, not their annual net effect on climate. During summer when the carbon sinks are reasonably high (25th percentile), NEE at midday (10:00–14:00) in southern Finland are the lowest in Hyytiälä (corresponding to the highest carbon sink), followed by Qvidja and Siikaneva (Table 2). The summertime 75th percentile values for midday negative ions are the highest in Qvidja, followed by Hyytiälä and Siikaneva (Table 2). When normalizing the values against those in Hyytiälä (Table 3), Siikaneva is a notably smaller carbon sink than other environments, whereas ion concentrations are clearly highest in Qvidja.

The relative surface areas in Finland are 0.40 for boreal forest on mineral soil (including areas with more than 1 m^3 ha^{-1} annual wood growth, Natural Resources Institute Finland 2017–2021), 0.06 for agricultural land on mineral soil (including arable land and land set aside. Natural Resources Institute Finland 2022) and 0.04 for open peatland (Natural Resources Institute Finland 2017–2021). Using the 75% percentiles for ions from Table 3 in Eq. 2, we get:

$$f_{\mathrm{CCN,i}} = 0.40 \times 1 \text{ (Hyytiälä)} = 0.40 \,, \quad (3)$$

$$f_{\mathrm{CCN,i}} = 0.06 \times 1.29 \text{ (Qvidja)} = 0.08 \,, \quad (4)$$

$$f_{\mathrm{CCN,i}} = 0.04 \times 0.93 \text{ (Siikaneva)} = 0.04 \,. \quad (5)$$

With the reservation that our sites are representative for all productive forests, cultivated agricultural lands and open peatland in the region, Eqs 3–5, together with Table 3, suggest that the potential contribution to CCN is the highest from the forest land, whereas per unit land area the agricultural grassland is the most effective source of CCN.

The contribution of agricultural grassland (Qvidja) to NPF and subsequently to CCN production is the highest (per surface area), so its contribution to climate mitigation is expected to be larger than if one only took into account the carbon sink of each ecosystem. On the other hand, compared with forests or agricultural lands, the contribution of peatland to climate mitigation is expected to be significantly smaller in both

Table 2. Comparison of mean half-hourly 25 percentile of NEE and 75 percentile of negative ion (2.0–2.3 nm) concentrations at midday (10:00–14:00) during the summer in southern Finland.

Ecosystem	NEE (μmol m^{-2} s^{-1})			Negative ions (cm^{-3})		
	50%	25%	10%	50%	75%	90%
Hyytiälä	–10.80	–13.70	–16.30	2.5	4.2	7.7
Qvidja	–8.38	–12.60	–17.60	3.1	5.4	9.3
Siikaneva	–3.42	–4.30	–5.21	2.3	3.9	6.9

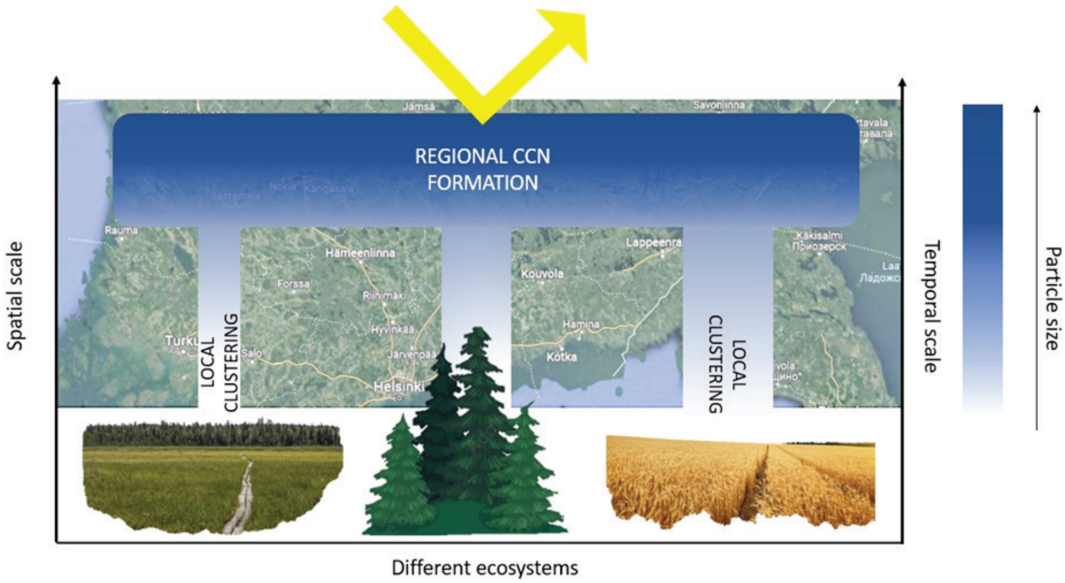

Fig 11. Schematic figure to illustrate how local clustering from different ecosystems contributes to CCN production and regional aerosol load. The width of the vertical bars denoting local clustering indicates the relative potential of local aerosol clustering in the studied ecosystems.

aerosol production and carbon sink point of view. However, it needs to be noted that in this paper, only the ecosystem-atmosphere exchange of CO_2 (i.e., NEE) was considered. As the studied grassland is under agricultural use and is fertilized and harvested several times during the growing season, the actual carbon balance of the site, defined as a sum of NEE and harvest + fertilization, is greatly affected. Figure 11 illustrates local contribution to regional aerosol and CCN load.

Conclusions

Regional environment typically consists of a number of different ecosystems, each having different contributions to the factors (e.g. carbon sink, aerosol effects, albedo and other GHG) influencing radiative forcing. Here we provide a means to estimate how the individual influencing factors (here aerosol effects and carbon uptake) vary between different ecosystems and how to take this into account in a regional environment. We call this approach CarbonSink+ Potential.

Regarding the aerosol effects, the approach is based on atmospheric clustering that has a relatively local source area comparable to those of NEE and GPP measurements (see Tuovinen *et al.* 2023). We argue that concentration of small ions (2.0–2.3 nm) is a good indicator of the strength of local NPF, subsequent formation of 3–6 nm particles, and eventually production of

Table 3. Normalized ratios at the studied ecosystems, i.e. values at site *X* divided by those obtained in Hyytiälä values at midday (10:00–14:00) during the summer.

Ecosystem	NEE (μmol m^{-2} s^{-1})			Negative ions (cm^{-3})		
	50%	25%	10%	50%	75%	90%
Hyytiälä	1	1	1	1	1	1
Qvidja	0.78	0.92	1.08	1.24	1.29	1.21
Siikaneva	0.32	0.31	0.32	0.92	0.93	0.90

CCN into the regional atmosphere across southern Finland. Here, we utilize this knowledge for ecosystem comparisons within a same region.

Since we have long-term, comprehensive observations of the environment and atmosphere from the SMEAR II station in Hyytiälä, we can compare all other sites within 500 km from this station to the data from there, and by that way estimate the contributions of these other sites to the regional CCN production using ecosystem-specific ion measurements.

With our approach, we can see that per land area, pristine peatland (based on Siikaneva results) seems to have the lowest potential for both carbon sink and CCN production, whereas agricultural grassland (based on Qvidja results) seems to have the highest momentary potential in producing CCN. The method presented here enables comparison between different ecosystems, particularly their capacity for acting as carbon sink (NEE) and producing CCN into the atmosphere.

Our previous paper (Kulmala *et al.* 2020) addressed how to compare the radiative effects due to the carbon uptake, aerosol radiative effects and surface albedo changes in a specific environment (Carbon sink+, Kulmala *et al.* 2020). The full comparison between the climate mitigation potential of different ecosystems at regional scale (using a simple metric) requires combining both of these approaches, Carbon sink+ and Carbon sink+ Potential.

Acknowledgements: We acknowledge the following projects: ACCC Flagship funded by the Academy of Finland grant number 337549 (UH) and 337552 (FMI), Academy professorship funded by the Academy of Finland (grant no. 302958), Academy of Finland projects no. 1325656, 311932, 334792, 316114, 325647, 325681, 347782, 324259, the Strategic Research Council (SRC) at the Academy of Finland (#352431), "Quantifying carbon sink, CarbonSink+ and their interaction with air quality" INAR project funded by Jane and Aatos Erkko Foundation, "Gigacity" project funded by Wihuri foundation, grant VN/28414/2021 by the Ministry of Agriculture and Forestry of Finland, European Union (Grant no. 101059888, Climb-Forest and Grant no. 871128, eLTER PLUS), European Research Council (ERC) project ATM-GTP Contract No. 742206, and European Union via Non-CO_2 Forcers and their Climate, Weather, Air Quality and Health Impacts (FOCI). We acknowledge Magdalena Okuljar and Jonathan Duplissy for managing the observations at Qvidja. University of Helsinki support via ACTRIS-HY is acknowledged. Support of the technical and scientific staff in Hyytiälä, Värriö and Qvidja are acknowledged.

Supplementary Information: The supplementary information related to this article is available online at: http://www.borenv.net/BER/archive/pdfs/ber29/ber29-001-016-supplement.pdf

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