

Investigating evidence of enhanced aerosol formation and growth due to autumnal moth larvae feeding on mountain birch at SMEAR I in northern Finland

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Laboratory studies have shown that herbivory-induced biogenic volatile organic compound (BVOC) emissions might enhance aerosol formation and growth. To increase understanding of the atmospheric relevance of this enhancement, we analyzed 25 years of data from SMEAR I (Station for Measuring Ecosystem-Atmosphere Relations) in northern Finland, where autumnal moth (*Epirrita autumnata*) larvae are prominent defoliators of mountain birch. We did not find a direct correlation between the autumnal moth density and aerosol processes, nor between the total number concentration and temperature, and hence the basal BVOC emissions. Instead, there is some evidence that the total particle concentration is elevated even for a few years after the infestation due to delayed defense response of mountain birch. The low total biomass of the trees concomitantly with low autumnal moth densities during most of the years of our study, may have impacted our results, hindering the enhancement of aerosol processes.

Introduction

In northern Fennoscandia, periodic defoliation of mountain birch (*Betula pubescens* var. *pumila*; former spp. *czerepanovii*) and subsequent mortality of trees have received a lot of attention throughout time (Tenow 1972, Lehtonen and Heikkinen 1995, Tenow and Bylund 2000, Jepsen *et al.* 2008, Saravesi *et al.* 2015). The cause for the defoliation is due to insect herbivory, of which Autumnal moth (*Epirrita*

autumnata, Lepidoptera: Geometridae) larvae are among the most abundant defoliators. As plants are immobile, they have developed several defense mechanisms, including emissions of biogenic volatile organic compounds (BVOC), to diminish stress and further damage (Dudareva *et al.* 2006, Mäntylä *et al.* 2008, Holopainen and Gershenson 2010, Peñuelas and Staudt 2010, Mithöfer and Boland 2012, Yli-Pirilä *et al.* 2016). In the atmosphere, BVOCs are oxidized (Atkinson and Arey 2003, Mogensen *et*

al. 2015, Hellén et al. 2018), and some of the oxidation products may contribute to the formation of secondary organic aerosol (SOA) and further growth of the particles (Laaksonen et al. 2008, Hallquist et al. 2009, Riipinen et al. 2012, Donahue et al. 2013, Schobesberger et al. 2013, Ehn et al. 2014, Kulmala et al. 2014a, Riccobono et al. 2014). By scattering and absorbing solar radiation, aerosol particles can modulate the radiative budget of the Earth (IPCC 2013). Additionally, alterations in cloudiness and cloud properties, due to changes in cloud condensation nuclei (CCN) concentrations, influence the radiative conditions (Twomey 1977, Albrecht 1989, Twomey 1991, Haywood and Boucher 2000, Kerminen et al. 2005, Kerminen et al. 2012, IPCC 2013, Gryspeerdt et al. 2014, Rosenfeld et al. 2014), which can further enhance the photosynthetic activity of the forest ecosystem (Gu et al. 1999, Gu et al. 2002, Kulmala et al. 2014b, Ezhova et al. 2018).

Plant stresses are categorized into abiotic stresses and biotic stresses. Abiotic stresses are caused by external circumstances, such as drought, heat wave and air pollution. Meanwhile, biotic stresses are caused by living species, such as herbivores and pathogens. Apart from phenological purposes, BVOCs mainly function as stress relief and as a defense mechanism (Holopainen and Gershenson 2010). In the presence of herbivory, the emitted BVOCs can be toxic or used as a repellent against the attacking herbivores. In addition, they can also function as infochemicals: warning neighboring plants or attracting predators and parasitoids of the herbivores — thereby minimizing plant damage (Dudareva et al. 2006, Mäntylä et al. 2008, Dicke and Baldwin 2010, Holopainen and Gershenson 2010). Plant stresses can increase the emissions of constitutively-emitted BVOCs and alter their relative share, or they can set off a production of new, stress-induced compounds (Holopainen 2004, Vickers et al. 2009, Fineschi and Loreto 2012). The emission rates of the compounds depend on plant species and environmental conditions, in addition to stress types and their severity (Vuorinen et al. 2007, Dicke and Baldwin 2010, Holopainen and Gershenson 2010, Loreto and Schnitzler 2010, Mithöfer and Boland 2012, Yli-Pirilä et al. 2016, Faiola and Taipale 2020).

Many recent studies have already investigated the herbivory-induced stress emissions from vegetation and their importance in aerosol processes in laboratory conditions (Joutsensaari et al. 2005, Hamilton et al. 2009, Mentel et al. 2013, Faiola et al. 2015, Joutsensaari et al. 2015, Yli-Pirilä et al. 2016, Faiola et al. 2018, Faiola et al. 2019). The emissions have been proposed to have higher SOA-forming potential, leading to increased mass yield of particles (Mentel et al. 2013, Joutsensaari et al. 2015, Yli-Pirilä et al. 2016), though opposing results have also been found (Faiola et al. 2018, Faiola et al. 2019).

Yli-Pirilä et al. (2016) studied SOA formation from BVOC emissions of mountain birch infested by autumnal moths in a laboratory, and concluded that the formed SOA mass load was up to 150-fold compared to SOA mass load formed from control tree emissions. They repeated the study using silver birch (*Betula pendula*), in which case the mass load was 285- and 8260-fold in control and laboratory trees, respectively.

In Joutsensaari et al. (2015), several approaches to study the same topic with different tree species were used. Both field (Scots pine (*Pinus sylvestris*) infested by European pine sawfly (*Neodiprion sertifer*) and laboratory experiments (Scots pine and Norway spruce (*Picea abies*) infested by pine weevils (*Hylobius abietis*)) showed 10- to 50-fold increases in monoterpene and sesquiterpene emissions. In the laboratory experiments, the increase in SOA mass from the emissions of infested trees was 200- to 1000-fold in comparison to the SOA mass from control tree emissions. Joutsensaari et al. (2015) also performed a global-scale model simulation of aerosol mass increase in a scenario where the monoterpene emissions were 10-fold higher and 10% of the boreal forest area were infested with herbivore species. The results revealed local increases of up to 480% in aerosol mass and 45% in CCN concentrations. When investigating satellite observations, they found a 2-fold increase in aerosol optical depth over a pine forest in Canada during a bark beetle outbreak.

Zhao et al. (2017) studied CCN number concentration in a chamber experiment using BVOC emissions from typical boreal forest trees:

Scots pine, Norway spruce and silver birch. They showed that the CCN concentration was increased by 47% due to aphid infestation. When the trees were exposed to both aphids and heat stress, a 93% increase in the CCN number concentration was observed (Zhao *et al.* 2017).

In chamber experiments by Mentel *et al.* (2013), the mixture of stress-induced BVOCs comprising sesquiterpenes, methyl salicylate and C₁₇-BVOCs led to 17–33% SOA yields compared to 4–6% SOA yields from mainly monoterpenes containing BVOC mixtures. In their experiments, green leaf volatiles (GLV) tended to suppress SOA formation. The authors hypothesized that this could be due to OH scavenging in their chamber by GLVs, which has previously been shown for isoprene (Kiendler-Scharr *et al.* 2009, Kiendler-Scharr *et al.* 2012, McFiggans *et al.* 2019).

Bergström *et al.* (2014) used the results of the chamber measurements by Mentel *et al.* (2013), and studied the effects of BVOC emissions on aerosol load in northern Europe using a modeling approach. According to their estimations, less than 20% of organic aerosol (< 2.5 µm in diameter) was due to the background BVOC emissions, whereas stress emissions, first approximated only by sesquiterpene emissions, caused 20–40% of the total organic aerosol. When stress emissions were approximated both by sesquiterpene and methyl salicylate emissions, the relative fraction was 50–80% of the organic aerosol.

Faiola *et al.* (2018; 2019) observed Scots pine trees infested by pine weevils and aphids, emitted larger amounts of sesquiterpenes compared to healthy trees in laboratory experiments. In Faiola *et al.* (2019), the collected BVOC samples were exposed to ozone, in which case SOA mass yield decreased from 9.7–14.6% to 6.9–10.4% from infested trees. Exposure to OH did not affect the SOA mass yield. They hypothesized that the reduction in the ozone exposure was caused by the emissions of acyclic sesquiterpenes that produced higher volatility oxidation products in dark ozonolysis. Faiola *et al.* (2018) used different volatile blends altered by stress treatments of plants. They concluded that the SOA mass yield from healthy plant emissions was limited by sesquiterpene emissions, whereas the mass yield from the emissions of

infested trees was limited by the specific blend of monoterpenes. Hence, though the infestation increased the volatile emissions from the trees, and therefore provided more condensable vapors, the emission blend was not favorable in terms of SOA formation.

Both constitutive and stress-induced emissions have been estimated to increase in the future (Peñuelas and Llusà 2003, Yuan *et al.* 2009, Peñuelas and Staudt 2010). The constitutive emissions are modulated by the rising air temperature, making the forest more active and prolonging the growing season, but it may also cause changes in tree species and density of the forest, and move the treeline towards higher latitudes and altitudes (Chmielewski and Rötzer 2001, Koca *et al.* 2006, Tømmervik *et al.* 2009, Hari *et al.* 2013, Jepsen *et al.* 2013, Valolahti *et al.* 2015, Tang *et al.* 2018, Ghirardo *et al.* 2020). As for stress emissions, many herbivore species have shown an increasing population density because the living conditions have become more optimal, although also opposite cases exist (Lehmann *et al.* 2020). Additionally, some studies have shown amplified increase in BVOC emissions in case of simultaneous warming and herbivore infestation (Kivimäenpää *et al.* 2016, Zhao *et al.* 2017, Li *et al.* 2019).

All these changes are particularly important at high latitudes where the impacts of climate change are stronger than the global average (Law and Stohl 2007, IPCC 2013). Moreover, for the boreal region, a climate feedback mechanism linking enhanced photosynthesis and BVOC emissions due to the elevated CO₂ and temperature levels to increase in aerosol load and cloudiness, leading to cooling of the climate, has been proposed (Paasonen *et al.* 2013, Kulmala *et al.* 2014b, Ezhova *et al.* 2018, Scott *et al.* 2018, Sporre *et al.* 2019). In that area, particulate mass comprises significantly of oxidized organic compounds (Jimenez *et al.* 2009, Heikkinen *et al.* 2020). Hence, BVOC emissions have an increasingly important role in the boundary layer chemistry and physics in the boreal region.

Autumnal moth is distributed over the northern parts of the Holarctic region, restricting approximately to the 45° N latitude (Tenow 1972). In Finland, the moth is commonly found where deciduous trees and shrubs grow, but

mass outbreaks occur only in Lapland (Tenow 1972, Tanhuanpää *et al.* 1999, Ruohomäki *et al.* 2000), where the main host plant is mountain birch. In these areas, autumnal moth population density has a statistically significant 9–11 years periodic cycle with or without severe outbreak levels (Haukioja *et al.* 1988). Autumnal moth is obligatorily univoltine, meaning that it can have only one generation per year (Ruohomäki *et al.* 2000). Females lay eggs on e.g., the branches of host plants where the eggs overwinter. In spring, the larvae hatch from the eggs concurrently with bud burst and start feeding (Kaitaniemi *et al.* 1997, Kaitaniemi and Ruohomäki 1999, Fält-Nardmann *et al.* 2016). Bud burst usually takes place in early June in Finnish Lapland. When growing, larvae go through five development stages, instars within 4–7 weeks, after which they descend to the ground to pupate (Haukioja *et al.* 1988, Kaitaniemi and Ruohomäki 1999). The adult moths fly from mid-August to late September to breed (Kaitaniemi and Ruohomäki 1999).

In early summer, when the larvae are small, they consume only little foliage. Later, the level of defoliation increases as the larvae grow. Usually, when larval density is low, the total consumption of foliage is also low, and the level of defoliation remains less than 15% (Bylund 1995). During those years, trees can recover well, contrary to the years of total foliage loss when even mortality of the trees has been observed (Lehtonen and Heikkinen 1995, Tenow and Bylund 2000, Karlsson *et al.* 2005, Tenow *et al.* 2005). The emission rates of BVOCs depend on the level of defoliation, and thus the number of larvae and their size (Yli-Pirilä *et al.* 2016), although reduced leaf biomass due to feeding suppresses also BVOC emissions. Therefore, during mass outbreak years, the BVOC emissions may be even less than the constitutive emissions from stress-free mountain birch.

When exposed to autumnal moth larvae, mountain birch have shown to induce production of many compounds that are typically emitted by plants under herbivore attack (Yli-Pirilä *et al.* 2016, Faiola and Taipale 2020). These compounds include several terpenes (e.g. (E)- β -ocimene (monoterpene, MT), linalool (MT), β -bourbonene (sesquiterpene,

SQT) and homoterpene (E)-4,8-dimethyl-1,3,7-nonatriene), GLVs (e.g. (E)-2-hexenal and (Z)-3-hexen-1-ol), and methyl salicylate (Mäntylä *et al.* 2008, Yli-Pirilä *et al.* 2016). GLVs are especially related to the mechanical damage caused by larval feeding (Joutsensaari *et al.* 2015, Yli-Pirilä *et al.* 2016).

Haapanala *et al.* (2009) studied BVOC emissions of mountain birch in field conditions during two different summers. During the first summer, they found that the most abundant sesquiterpenes were α -farnesene and β -caryophyllene, and during the second summer, β -caryophyllene dominated the emissions whereas α -farnesene was not observed at all. Moreover, the total emissions of sesquiterpenes were less than 1% of the emissions during the previous year. They hypothesized that the autumnal moth mass outbreak two years prior might have still affected the emissions during the first year. Due to the delayed-induced defense response of mountain birch (Kaitaniemi *et al.* 1998, Ruuhola *et al.* 2007), BVOC emissions may also be elevated for several years after the autumnal moth infestation.

Studies on downy birch (*Betula pubescens*), which mountain birch is a subspecies of, and dwarf birch (*Betula nana*), which have common genomes with mountain birch, show increased emissions of similar compounds as has been found to be emitted by stressed mountain birch (Hakola *et al.* 2001, Li *et al.* 2019). Hakola *et al.* (2001) stressed downy birch mechanically, and Li *et al.* (2019) measured emissions of dwarf birch when they were exposed to autumnal moth larvae and methyl jasmonate, which is a plant hormone that mimics herbivore attack. When exposing dwarf birch to methyl jasmonate, the total emissions of terpenoids was 13-fold, and when exposing additionally to 2°C warming, the increase in total terpenoid emissions was 46-fold (Li *et al.* 2019). In autumnal moth infestation, dwarf birch emitted GLVs and methyl salicylate, and monoterpene and homoterpene emissions were 4-fold compared to intact trees (Li *et al.* 2019). In contrast to other studies, Li *et al.* (2019) did not find increased emissions of sesquiterpenes (Haapanala *et al.* 2009, Yli-Pirilä *et al.* 2016). The experiment was repeated with methyl jasmonate in warmer (21.5°C

vs. 15°C) and brighter (120 $\mu\text{mol m}^{-2} \text{s}^{-1}$ vs. 50 $\mu\text{mol m}^{-2} \text{s}^{-1}$) conditions, in which case the response of sesquiterpene emission was similar to monoterpene and homoterpene emissions (Li *et al.* 2019).

Despite the increasing interest towards the effects of biotic stress-induced plant volatiles on SOA formation and growth of the particles, the atmospherically relevant importance of the processes has never been investigated in field conditions. If the enhancement is as strong in the field as laboratory studies have proposed, the stress emissions should be considered also e.g., in climate models, especially when predicting the changes caused by global warming.

In this work, we analyze 25 years of field data at SMEAR I research station in Värriö, Finland. Data include autumnal moth densities, different aerosol and meteorological variables and trace gases. We focus on the question of herbivory by autumnal moths as one of the most significant biotic stresses in northern Fennoscandia, and mountain birch make up the major share of plant biomass in their distribution area, having a significant role in the northern ecosystem (Lehtonen and Heikkinen 1995, Tenow and Bylund 2000, Wielgolaski 2005, Jepsen *et al.* 2008, Tømmervik *et al.* 2009, Jepsen *et al.* 2013). The remote location of our study site ensures low background concentrations of anthropogenic pollutants, and the varying population density of autumnal moths promote the investigation of stress-free as well as stressed conditions with varying intensity.

The motivation for this study is to fill the gaps in understanding the atmospheric relevance of the phenomenon, particularly at high latitudes where the effects of climate change are more pronounced compared to other regions. Consequently, it is important to understand comprehensively the causes of BVOC emissions and how they affect atmospheric processes.

Material and methods

In this section, we first introduce the measurement site, the methods to estimate the population density of autumnal moths, and the data set of the employed atmospheric measurements. We

also describe the data handling needed in the analysis.

Measurement site

The SMEAR I (Station for Measuring Ecosystem-Atmosphere Relations) measurement site is located in the Värriö Strict Nature Reserve (Värriön luonnonpuisto) in eastern Finnish Lapland (67°44' N, 29°37' E, 390 m a.s.l.) (Hari *et al.* 1994). The area lies 130 km north from the Arctic Circle and 6 km from the Finnish–Russian border. There is no permanent residence nearby and anthropogenic influence in the area is minor. However, occasional events of high concentrations of atmospheric trace gases and aerosol particles occur when the wind advects them from the industrial area in the Kola peninsula, Russia (east–northeast from the station) (Pirjola *et al.* 1998, Kyrö *et al.* 2014). The average temperature in January (the coldest month) during the climatological normal period (1981–2010) was -11.4°C , and in July (the warmest month) was 13.1°C . The region is free from snow approximately from the end of May to mid-October. The sun does not set between ca. 27 May and 15 July.

Collection of autumnal moths

The population density of autumnal moths in Värriö was estimated with three different collection methods: time-unit specified collection by hands, sweeping with a net, and catching with light traps. The two former methods directly count the number of larvae, whereas light traps attract adult moths. Consequently, the first two methods reflect the density of autumnal moths locally, while adults fly across wider areas. Each collection method is described in more detail.

The time-unit specific method (hereafter "larval index"), has been conducted once per year since 1987 near the station and timed when the larvae had molted to the fourth instar. The larval index is used to calculate a relative index of larval density per 10 min of search from the leaves of mountain birch (Fig. 1a and b). The number of found larvae were summed to obtain the relative index value. The method is explained in more detail in Klemola *et al.* (2016).



Fig. 1. (a) View of a mountain birch forest defoliated by autumnal moth larvae at the time of full-grown leaves on 01 July 2005 at the foot of Nuvvus-Ailigas fjeld in Utsjoki, Finland. (b) Autumnal moth larvae feeding on mountain birch leaves. (c) Collection of caterpillars from bilberry bushes was conducted by sweeping 100 times with a net. (d) Jalas model light trap set-up used in Värriö. The light-attracted moths are directed to the catch which is emptied every morning. Figures courtesy of Tero Klemola (a, b), Veli Pohjonen (b) and Esko Karvinen (d)

Sweeping was conducted approximately every fifth day from June to September. The collection was done by sweeping 100 times with a net (Fig. 1c) both from mountain birch branches and bilberries (*Vaccinium myrtillus*) on the northern slope of the fjeld (Värriötunturi) near the station (<https://www.helsinki.fi/en/research-stations/varrio-subarctic-research-station/research/long-term-research-series>, last accessed 27 Oct. 2020). The spot where the sweeping was conducted was selected at random each time. All the caught caterpillars were counted and stored in ethanol-filled tubes. The species were not separated, but the most abundant one by far in Värriö was the autumnal moth (Hunter *et al.* 2014). In

this study, we used data from 2006 onwards, excluding the year 2012.

In most parts of the analysis, we used sweeping data averaged over June and July, because the number of days when sweeping was conducted varied between the years. We used data only from June and July due to the timing of the feeding of autumnal moth larvae (see Introduction section). Additionally, we used data where the number of caught larvae was interpolated linearly between the two consecutive collections. Interpolation helped to extend the analysis of daily values to days when the sweeping was not originally conducted. The interpolation was performed year-by-year between the first day when sweeping was

conducted and 31 July. In the analysis, the sweeping data are presented as a sum of larvae found from mountain birch branches and bilberries.

Jalas-model light traps (see Table 1 for a detailed list of instruments used in the study) were used to attract and capture nocturnal moths (Fig. 1d; Leinonen *et al.* 1998). Each trap had a 500-W light bulb that was switched on every night between 20:00 and 08:00 (UTC+3), approximately from mid-May to mid-October (Hunter *et al.* 2014). Every morning the traps were emptied and the collected moths were stored in a freezer. In autumn, the samples were counted and identified to species (Hunter *et al.* 2014). From 1987 to 2012, eleven light traps were used, but was reduced to two thereafter. Due to the reduction in the number of light traps over the course of the study, the number of collected moths was normalized by the number of occupied light traps. The light trap method collects adult moths, but the larval feeding causes the defoliation earlier in the summer. Hence, we calculated averages of the data from August and September and used that as an indicator of abundance of autumnal moth larvae in June and July.

Atmospheric measurements

Aerosol total number concentration (N_{tot}) was obtained in two ways: 1) from direct ambient

air measurements with a condensation particle counter; and 2) by integrating the concentrations in separate size classes from number size distribution data. CPC measurements at the station have been continuously running since 1991. The aerosol number size distribution has been measured with a Differential Mobility Particle Sizer (DMPS) since 1997 (Aalto *et al.* 2001). Before 2003, the lowest cut-off size of the DMPS was 8 nm and thereafter it was 3 nm. In 2005, the uppermost cut-off size was extended from 500 nm to 850 nm. In the analysis, we additionally used concentration of nucleation mode and CCN-sized particles, which were also derived from DMPS measurements. Due to the changing cut-off size, the concentration of nucleation mode particles was calculated between 8 nm and 25 nm (N_{8-25}). The concentration of CCN-sized particles was calculated between 70 nm and 500 nm (N_{70-500}).

Two variables derived from aerosol number concentration were also used in the analysis: 1) the growth rate (GR), which describes how rapidly newly-formed particles can grow to larger sizes (Kulmala *et al.* 2012); and 2) the formation rate (J), which quantifies the strength of a particle formation event (Kulmala *et al.* 2012). The GR and J were always calculated for particles with a diameter in the range from 8–25 nm due to the changing cut-off diameters. The formation rate is then denoted as J_g . The

Table 1. List of instruments used in this study.

Measurement	Years measured	Model; Manufacturer
Jalas-model light trap	1992–2016	Custom-built; University of Oulu, Finland
Condensation particle counter	1991–2006	3760; TSI Inc., USA
Condensation particle counter	2007–2015	3022; TSI Inc., USA
Condensation particle counter	2016–	A20; Airmodus Ltd., Finland
Differential Mobility Particle Sizer	1997–2016	Custom-built; University of Helsinki, Finland
Pulsed Fluorescence analyzer (SO ₂)	1991–1996	TEI 43S; Thermo Fisher Scientific, USA
Pulsed Fluorescence analyzer (SO ₂)	1996–2003	TEI 43 CTL; Thermo Fisher Scientific, USA
Pulsed Fluorescence analyzer (SO ₂)	2004–2007	API 100AS; AMP-Cherokee, USA
Pulsed Fluorescence analyzer (SO ₂)	2008–2010	TEI 43 i-TLE; Thermo Fisher Scientific, USA
Pulsed Fluorescence analyzer (SO ₂)	2011–	TEI 43 CTL; Thermo Fisher Scientific, USA
Air temperature	1991–	Pt100
Relative humidity	1993–2004	HMI32 Humicap; Vaisala, Finland
Relative humidity	2005–	MP106A; Rotronic, Switzerland
UV-B	1996–	Model 501A; Solar Light, USA
Pyranometer (global radiation)	1991–2007	Reemann TP-3; Astrodata, Estonia
Pyranometer (global radiation)	2008–	Middleton SK08; Middleton Solar, Australia

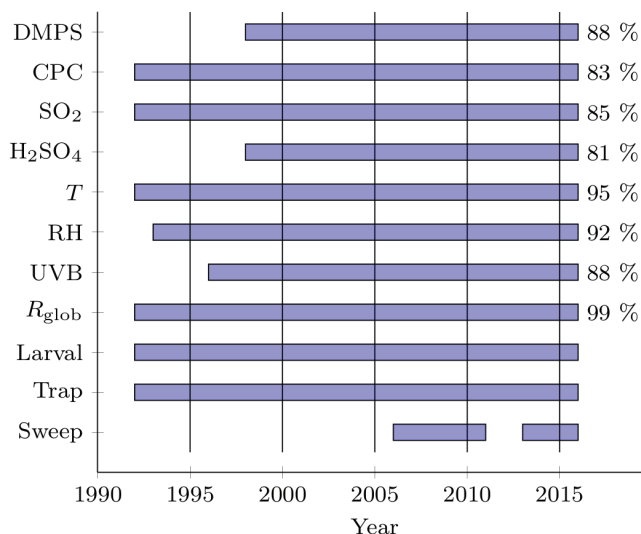


Fig. 2. Timeline of ongoing measurements used in this study. The percentage value shows the data availability in June and July from the time period when the measurement was ongoing. Availability was calculated from 30 min averages. Larval, trap and sweep are abbreviations for larval index, light trap and sweeping data, respectively.

used new particle formation (NPF) event classification analysis follows the procedure proposed by Dal Maso *et al.* (2005).

Sulfur dioxide (SO₂) has been measured with a pulsed fluorescence analyzer since 1991 at SMEAR I. Sulfuric acid (H₂SO₄) concentration was calculated from a proxy developed by Petäjä *et al.* (2009; Eq. (3)). Other measurements used were air temperature, relative humidity, UV-B and global radiation (Hari *et al.* 1994).

We used 30-min averages of quality-checked data. In order to focus on the time of larval feeding, we only analysed measured data during June and July between 1992 and 2016 or from beginning of the year when the measurements were launched at SMEAR I (Fig. 2).

Data analysis

Sulfuric acid, which is an oxidation product of sulfur dioxide, is known to contribute to NPF (Donahue *et al.* 2013, Schobesberger *et al.* 2013, Kulmala *et al.* 2014a). Thus, it was important to minimize its effect when analysing the data. Our interest lies on days when the total particle concentration was high but sulfur dioxide was low. Hence, when mentioned, we filtered all the used data sets according to total particle and sulfur dioxide concentrations. The day was included in the analysis if the 09:00–17:00 (UTC+3)

median total particle concentration was higher than or equal to 500 cm⁻³ and sulfur dioxide concentration was less than or equal to 0.7 ppb. We selected the magnitude of suitable filtering conditions based on the results reported by Pirjola *et al.* (1998) and Kyrö *et al.* (2014). As there were gaps in total particle and sulfur dioxide concentration data, also data from those periods were treated as to qualify the filtering conditions, to increase the number of data points in the analysis.

When examining the correlation between the variables, we calculated linear least squares fits to the data points. We used the Pearson's correlation coefficient (r) as an estimate of dependency, and the reported p -values to describe how certain the relations were. We used a 5% limit to determine whether the correlation was statistically significant. The slopes and the number of data points per figure are listed for clarification. Due to the relatively low number of data points, we plotted the 95% confidence intervals to the figures. The confidence intervals show the limits between which the actual linear fit lies with 95% confidence.

A two-sample Kolmogorov–Smirnov test was additionally used for examining the differences in the two sample vectors. It tests the hypothesis that the two samples come from the same distribution by comparing the cumulative distribution functions of the vectors. If the null

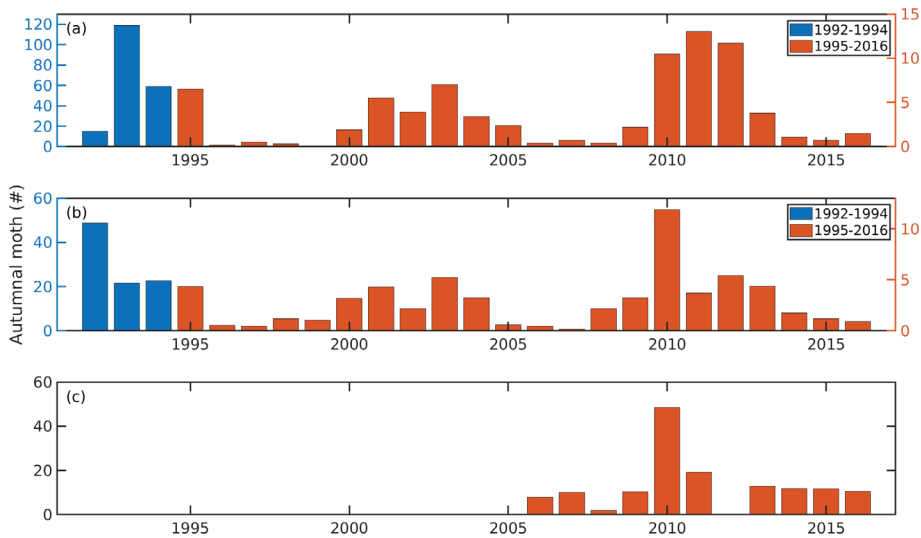


Fig. 3. The number of autumnal moths from 1992–2016 collected with three methods: (a) larval index, (b) light trap and (c) sweeping. Light trap and sweeping data are presented as averages of the original data. Sweeping data are presented as sum of larvae found from mountain birches and bilberry bushes. In (a) and (b), the y-axis is different for the first three years (1992–1994, left y-axis) than for the rest of the years (1995–2016, right y-axis).

hypothesis could not be rejected at the 5% significance level, we could assume that there were no significant differences in the data between the two sample vectors.

DMPS number size distribution analysis

We investigated the difference in median aerosol number size distribution between the four years of the highest and lowest autumnal moth densities. The statistical testing of two time series with two variables (particle diameter and concentration) is not trivial. Hence, the testing was conducted by first taking daily medians of the aerosol number concentration from all June and July days between 2003 and 2016. The range covered all the years when the lower particle diameter limit of the DMPS system was 3 nm. The daily median of the number concentration was taken to simplify the analysis. We then selected four years with the highest autumnal moth densities and calculated a median over those June and July daily median number concentrations. The procedure was repeated analogously for the four years with the lowest moth densities. Then, to

gain a distribution of median values to which we could compare the medians from the highest and lowest autumnal moth density years, we selected four random years from the range of years and calculated analogously the median over the June and July daily median number concentrations of those four random years. Altogether, this procedure was repeated as many times as there were different combinations of sets of four years ($\binom{4}{4} = 1001$ different combinations), and thus the distribution of medians was reached. If the median number concentration from the low or high moth density years were more extreme than the 5th or 95th percentiles of the distribution of medians, we could conclude that they differ statistically significantly from the medians compared with the years with an average autumnal moth density.

Results and discussion

First, we investigated autumnal moth population density dynamics in Värriö, followed by analyses of its relation to NPF event frequency, and formation and growth rates of aerosol par-

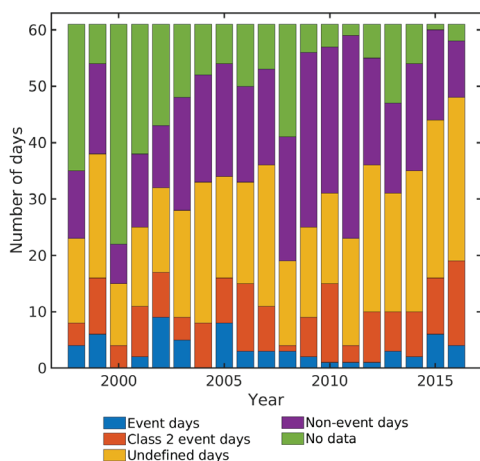


Fig. 4. NPF event classification in June and July from 1998–2016.

ticles. The second part of the analysis is focused on aerosol number size distribution and number concentration. We also studied the effect of several atmospheric variables on aerosol load, and finally compared our findings with earlier studies.

Moth population density during the study period

We began the analysis by studying population dynamics of autumnal moths (Fig. 3). Larval index and light trap data (Fig. 3a and b) are presented so that the *y*-axis is different for the first three years (1992–1994) than the rest (1995–2016), due to the much higher moth densities during the former years. The light trap and sweeping data (Fig. 3b and c) are reported here as averages of the original data (see Collection of autumnal moths section). Larval index is presented as such since it has only one value per year.

The larval index and light trap data both especially indicated cyclic population density dynamics (Fig. 3). The same pattern was also seen in sweeping data, although the available time series was shorter. Moreover, the estimates of the population density with different methods showed approximately a similar pattern. The timing of the peak densities was also simi-

lar in northern Norway and continental Lapland (Klemola *et al.* 2006, Jepsen *et al.* 2013, Klemola *et al.* 2016).

According to Klemola *et al.* (2016), moth outbreak takes place when the larval index exceeds 100 larvae per 10 min of search. In our study (from 1992 to 2016), there was only one year (1993) when an outbreak level was reached. Unfortunately, the data coverage from atmospheric measurements were limited in 1993. Therefore, the year 2003 and from 2010–2012 were treated as years with high moth density; meanwhile the years 1998, 1999, 2007, 2008 and 2016 were indicated as low moth density years. At some parts of the analysis, when data availability allowed, the years from 1992–1994 were included as years with high moth density. Additionally, we examined the possible delayed-induced defense response of mountain birch. The delay could induce BVOC emissions to remain high for several years after infestation. Therefore, we studied the three years following the autumnal moth density peaks (i.e. years 2004–2006 and 2013–2015, and years 1995–1997), when the data availability allowed, as potential years of delayed defense response.

Reduction of leaf biomass occurs quickly during severe moth outbreaks and it further decreases the emissions of BVOCs. It may also prevent the enhancement or lead to a decrease in SOA production and growth. With the current data set we were not able to estimate the level of defoliation and therefore we had to ignore the reduction of biomass in this analysis. However, as the autumnal moth densities remained low during most of the measurement period, the level of defoliation was also likely low.

NPF event frequency analysis

We investigated the frequency of NPF events in June and July (Fig. 4). The fraction of event and non-event days varied, but examined closely, they did not follow the periodic pattern seen with autumnal moth data (Fig. 3). The correlation was analyzed further by plotting the annual percentage of event and non-event days from June and July against the larval index data, and against the averaged number of autumnal moths caught

with light traps and sweeping (Fig. 5). When calculating the percentage of event and non-event days, the number of days with missing data were subtracted from the total number of days in June and July, to eliminate the effect of data gaps on the percentage value of event or non-event days. Only class 1 NPF events were included to this analysis. We initially wished to include class 2 type of NPF events into the analysis (figure not shown), but the inclusion of class 2 events revealed a lower correlation coefficient ($r < 0.29$) and higher p -values ($0.23 < p < 0.65$).

According to the assumption that larval feeding modifies SOA formation via emitted BVOCs, the number of event days should correlate positively with the number of autumnal moths as long as the amount of biomass is not decreased significantly. Our results showed, however, a moderate negative correlation between the number of event days and autumnal moths (r ranging from -0.42 to -0.6), though the p -values exceeded slightly beyond the 5% significance level (Fig. 5). This could at least partly be explained by the small number of data points. The data from non-event days were more scat-

tered and showed a weak, non-significant positive correlation. The inverse relation of event and non-event days to the number of autumnal moths is caused by the natural exclusionary behavior of the number of event and non-event days. Although autumnal moth population densities remained rather low between 1998 and 2016 and even during the peak years, and thus the level of defoliation likely did not suppress BVOC emissions (Karlsson *et al.* 2005, Yli-Pirilä *et al.* 2016). Additionally, we could not find a correlation between the number of NPF events and population density of autumnal moths.

To test the possible delayed-induced defense response of mountain birch, we shifted autumnal moth data 1–3 years forward (figure not shown). In this case, the correlation coefficients between the number of event days and autumnal moth density became less negative or slightly positive ($r < 0.24$). Moreover, the correlation between the number of non-event days and moth density turned out to be less positive or negative ($r > -0.38$). However, the data were scattered, and the p -values exceeded the 5% significance level ($0.07 < p < 0.85$).

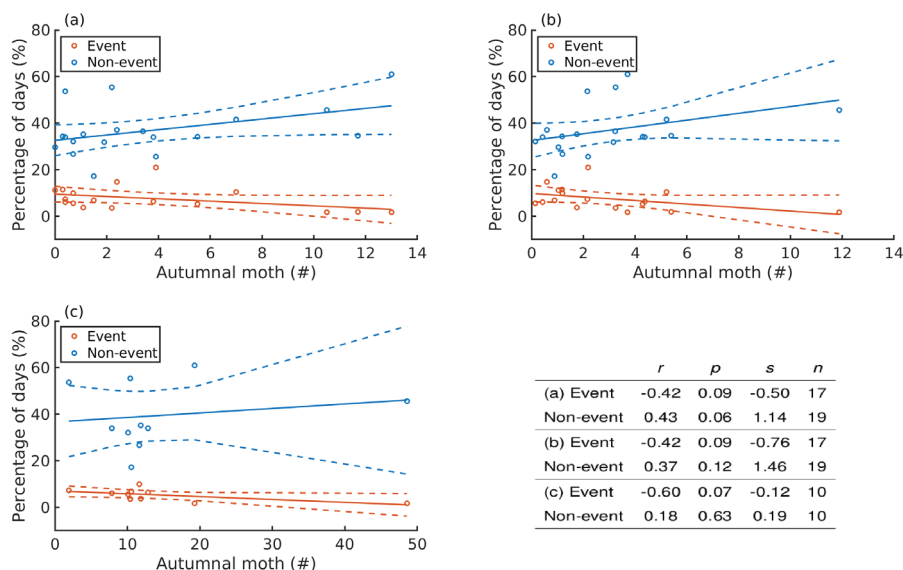


Fig. 5. Percentage of NPF event (red) and non-event (blue) days in June and July as a function of (a) larval index, and yearly averaged (b) light trap and (c) sweeping data. The number of days when DMPS data were missing have been excluded from the analysis. Solid lines are the linear least squares fits to the data points, and dashed lines show the 95% confidence intervals. Slopes of the fits (s), correlation coefficients (r), p -values and number of data points (n) are presented in the table for each subplot.

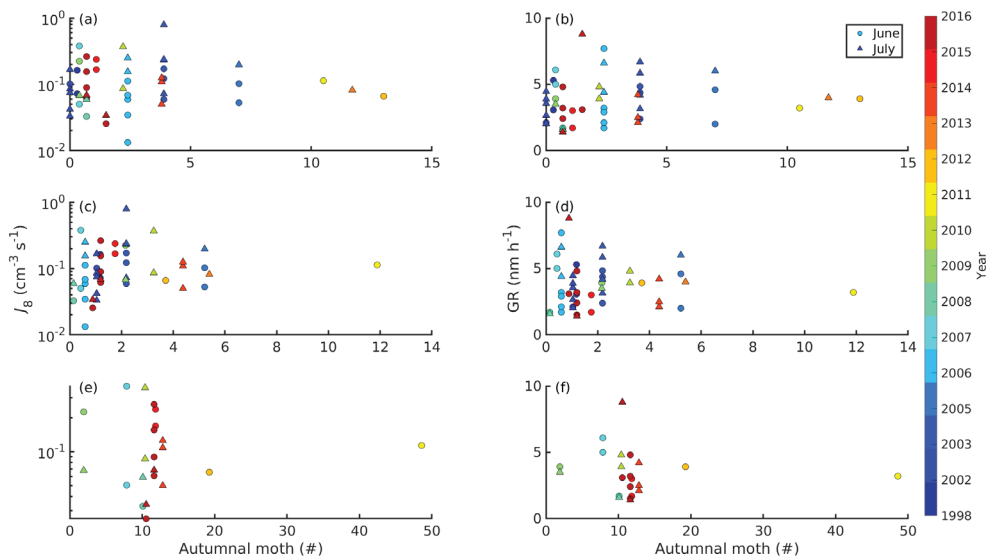


Fig. 6. The calculated (a) J_8 and (b) GR as a function of larval index data, (c) J_8 and (d) GR as a function of light trap data, and (e) J_8 and (f) GR as a function of sweeping data. Averaged light trap and sweeping data are used. The data were restricted to days when the total particle concentration was high and the SO_2 concentration was low. Coloring refers to the year and marker shape to the month when the observations were made.

Formation and growth rates

Although there seemed to be no correlation between autumnal moth population density and the number of NPF events, the autumnal moth feeding-induced BVOC emissions could affect the growth of the newly formed particles rather than the initial nucleation (Riipinen *et al.* 2012, Donahue *et al.* 2013, Ehn *et al.* 2014, Peräkylä *et al.* 2014). Thus, the behavior of formation and growth rates of particles between 8 nm and 25 nm were examined with respect to the number of autumnal moths (Fig. 6). We used larval index and averaged light trap and averaged sweeping data to estimate the autumnal moth density during the measurement period. The data were filtered to consider only days when the total particle concentration was high and the sulfur dioxide concentration was low. It should be noted that there were years without or with only a few data points.

Our results showed that the data points were scattered and we could not find dependency between the number of autumnal moths and the formation nor growth rates (Fig. 6). We also applied a two-sample Kolmogorov–Smirnov test for high (2003, 2010, 2011 and 2013) and low (1998, 1999, 2007, 2008 and 2016) autumnal

moth density years, as well as years after the population density peaks (2004–2006, 2013–2015). The results indicated that the data were from same distribution, and thus there were not significant differences in the formation or growth rates between the years. Additionally, the formation or growth rates were not consistently lower in June compared with July when the BVOC emissions are expected to be higher due to the larger size of larvae and therefore a larger consumption of foliage (Haukioja 2003, Lempa *et al.* 2004).

To investigate the effect of a delayed defense response, we shifted the autumnal moth density data 1–3 years forward (figure not shown). In the case of formation and growth rates, we could not find any response in the results to the shift as seen previously with the number of event and non-event days.

DMPS number size distribution

The four years with the highest and lowest autumnal moth densities were compared with each other by plotting the median DMPS number size distribution from June and July of high (2003, 2010–2012) and low (2006–2008 and 2016)

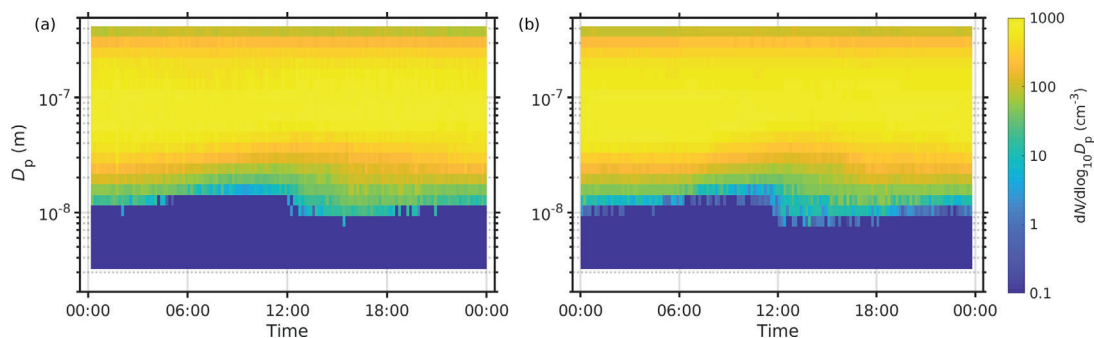


Fig. 7. The median diurnal DMPS number size distribution from (a) years with low moth density (2006–2008 and 2016) and (b) years with high moth density (2003, 2010–2012).

autumnal moth density years (Fig. 7). For this analysis, we opted to include data of the smallest particles (down to 3 nm). Therefore, we only considered data from 2003 onwards when the DMPS' lowest cut-off size measured down to 3 nm. Due to this, the year 2006 was also included among the years of low autumnal moth density in order to have an equal number of high and low moth densities. The differences were minor and statistical examination was needed.

To simplify the statistical analysis, we calculated first daily medians of the aerosol number concentration data from June and July days between 2003 and 2016, and then medians of those over the high and low moth density years, and finally over all different combinations of sets of four years to get a distribution of medians during the years with average moth densities (see DMPS number size distributions analysis). We fitted a lognormal distribution to the median values, and calculated 5th, 25th, 75th, and 95th percentiles (Fig. S1 in Supplementary Information). Then, we could compare the median values during the high and low autumnal moth years with the medians during average moth densities.

When reducing the number size distribution to a single median value over four years from June and July data, the diurnal variation information was lost. Yet, the value represents the overall aerosol particle concentration that is also affected by NPF. If the daily median number concentrations had been compared directly, the meteorologically-driven autocorrelation of subsequent days should have been taken into account. Also, it should be noted that the data were not normally distributed, and hence many statistical

tests could not be used. When dividing the data to random combinations of sets of four years, the years could be assumed to be independent, and the autocorrelation could be neglected. Hence, the procedure simplified the analysis considerably, and the statistical significance of the differences between the lowest and highest autumnal moth density years with respect to other years with average moth densities could roughly be estimated with this method.

Assuming that the aerosol number concentration should be higher during the years of high autumnal moth densities compared with low and normal autumnal moth density years, the median value from high autumnal moth density years should stand out from the distribution. Instead, both the medians from the highest and lowest moth density years lie within 25% and 75% percentiles, indicating that there is not a significant difference in aerosol number size distribution between low, normal and high autumnal moth density years (Fig. S1 in Supplementary Information).

We additionally performed a two-sample Kolmogorov–Smirnov test for the daily median number concentration data from low and high moth density years. The results indicated that the distributions of data from both high and low moth years were similar and there were no significant differences in the data.

Aerosol number concentration

As the years 1992–1994 had considerably higher number of autumnal moths than other years, we

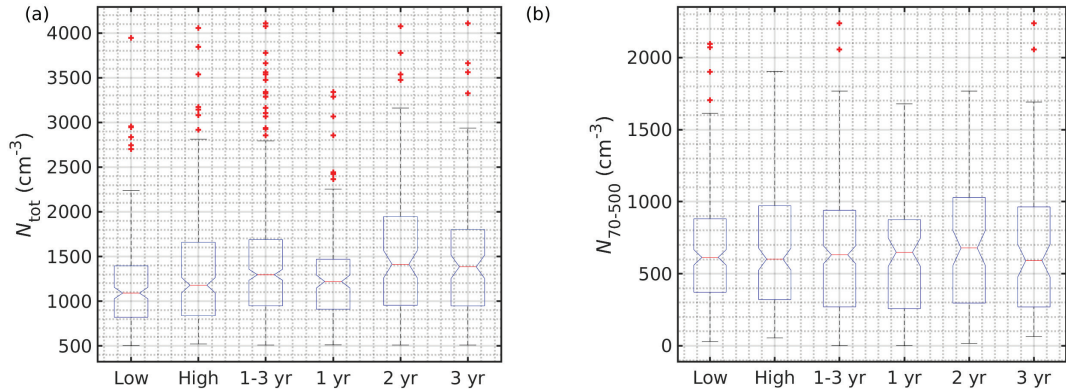


Fig. 8. Notched box plots of median daytime (09:00–17:00) (a) total particle concentration (measured with CPC) and (b) concentration of CCN-sized particles. The data has been separated between the years of high autumnal moth density ((a): 1992–1994, 2003, 2010–2012; (b): 2003, 2010–2012), low moth density ((a) and (b): 1998, 1999, 2007, 2008 and 2016), and 1–3 years after peak densities ((a): 1995–1997, 2004–2006 and 2013–2015; (b): 2004–2006 and 2013–2015). Additionally, the last one has also been divided such that "1 yr" contains data from the first years after the peak densities ((a): 1995, 2004 and 2013; (b): 2004 and 2013), "2 yr" from the second years after the peak densities ((a): 1996, 2005 and 2014; (b): 2005 and 2014), and "3 yr" from the third years after the peak densities ((a): 1997, 2006 and 2015; (b): 2006 and 2015). The data have been filtered to consider only days with high total particle and low SO_2 concentration. The red horizontal lines represent the median values, and edges of the blue boxes are the 25th and 75th percentiles. Black dashed lines represent whiskers extending to 1.5 times the interquartile range, and outliers are plotted individually using a red "+" symbol.

investigated the median daytime (09:00–17:00) total particle number concentration annually to see whether the number concentration from 1992–1994 differed from other years (Fig. S2 in Supplementary Information). We used the median of June and July total particle number concentration data from CPC measurements, and added larval index data to indicate the autumnal moth density dynamics. We applied the total particle and sulfur dioxide concentration filtering to the total particle number concentration data.

Total particle concentration in 1992 was lower compared with other years (Fig. S2 in Supplementary Information). Unfortunately, there were only few data points, so we cannot conclude that the highest autumnal moth density years would be associated with the lowest total particle concentrations. Moreover, there seems to be no direct relation between the total particle concentration and the number of autumnal moths. We also tested the potential impact due to delayed-induced defense response on aerosol processes by applying a built-in function "finddelay" of MATLAB (R2019b, MathWorks Inc, USA). The function searches for correlation between two variables, in this case, autumnal

moth density and total particle concentration. However, no significant correlation was found.

We also separated the aerosol number concentration data between the years of high (1992–1994, 2003 and 2010–2012) and low (1998, 1999, 2007, 2008 and 2016) autumnal moth densities, and years after the peak densities (1995–1997, 2004–2006 and 2013–2015) (Fig. 8a). The years of potential delayed defense response were additionally divided and studied by the number of years after the peak densities: first year after the peak densities (1995, 2004 and 2013); second year (1996, 2005 and 2014); and third year (1997, 2006 and 2015). The total particle concentration data were from CPC measurements. The introduced total particle and sulfur dioxide concentration filters were applied to the data.

The notches overlap between high and low autumnal moth years and between high and data from 1–3 years after the peak densities, indicating that the medians do not differ statistically significantly at 5% significance level. Differences can, however, be found between the data from low autumnal moth density years and 1–3 years after the peak densities. We additionally applied

the two-sample Kolmogorov–Smirnov test to the data. Contrary to the results of the box plot, the results of the test indicate that the data in high and low moth density years, as well as between low moth density years and 1–3 years after peak densities, are from different distributions.

We performed a similar analysis for the concentrations of CCN-sized particles (Fig. 8b) and nucleation mode particles (8–25 nm, figure not shown), with the only exception that the data begins from the year 1998. Neither the results of the box plots nor the two-sample Kolmogorov–Smirnov tests indicated any differences between the data. Several other combinations were also performed using more and less years, filtering the data using sulfuric acid interval, or using no filtering at all. Additionally, number concentration data were replaced with the formation and growth rates. The same filtering procedures were applied to these data sets. Nonetheless, all the figures gave similar results: the median values did not differ statistically significantly at the 5% significance level and neither the two-sample Kolmogorov–Smirnov test indicated differences between years of high or low autumnal moth data or data after the peak years. Thereby, according to our findings, it is possible that there is an enhancement in the aerosol total number concentration during and after the peak density years, and on NPF frequency after the peak densities but these effects can not be explicitly disentangled.

To emphasize the effect of autumnal moths to total particle concentration, several types of normalizations were additionally tested. The analyses included daytime median total particle concentration normalized by corresponding sulfuric acid concentration, UV-B radiation, the product of UV-B radiation, temperature and sulfuric acid concentration, and also the formula of the linear fit of the total particle concentration as a function of UV-B radiation. Besides total particle concentration, also concentration of nucleation mode particles was investigated. Different time averages and event–non-event day separation were additionally implied to the normalized data. In all of the cases, the variables did not correlate linearly ($r < 0.25$). This analysis did not indicate that herbivory by autumnal moths would enhance aerosol load in field conditions.

An example figure of this investigation is given in Appendix (Fig. S3 in Supplementary Information).

Effect of atmospheric variables on aerosol particles

The relations between daytime (09:00–17:00) median total particle concentration and corresponding meteorological variables or trace gas concentrations were examined with respect to the number of autumnal moths estimated with larval index (Fig. S4 in Supplementary Information). The data had been filtered according to the total particle and sulfur dioxide concentrations. Here, we used total particle concentration from direct ambient aerosol particle measurements, i.e., CPC data, to extend the analysis to begin from 1992. Each subplot does not have the same amount of data, due to different launching times of the measurements, because we opted to use as much data as was available (Fig. 2). The available years and data coverages are listed in Table S1 (in Supplementary Information). Coloring in the figures represents the number of autumnal moths, and the gray dots show the original, non-filtered data. The correlation coefficients and p -values of the original data are presented in Table S2 (in Supplementary Information).

None of the presented variables showed a strong linear correlation with total particle concentration ($r < 0.29$), and the differences were small between filtered and non-filtered data (Fig. S4 in Supplementary Information). Not even temperature, and hence the basal BVOC emissions correlated with the number of aerosol particles (Guenther *et al.* 1995). This non-correlation also extended to sulfur dioxide and sulfuric acid concentrations, which is what we would not have expected due to the role of sulfuric acid in NPF (Donahue *et al.* 2013, Schobesberger *et al.* 2013, Ehn *et al.* 2014). Moreover, the number of autumnal moths (coloring) did not show any relation to the measured variables. The procedure was repeated using averaged light trap and averaged sweeping data as estimates of the number of autumnal moths. The results of those figures were, however, similar as

shown here (Fig. S4 in Supplementary Information), and hence they are not presented.

We also studied whether the pre-existing particles, acting as condensation sink (CS) to precursor gases, suppress aerosol particle formation and growth from BVOCs caused by larval feeding. We plotted daytime (09:00–17:00) median total number concentration and concentration of nucleation mode particles as a function of the number of autumnal moths from all summer days (Fig. S5a and b in Supplementary Information) and including only NPF event days (Fig. S5c and d in Supplementary Information). The color of the markers represents the corresponding CS. The non-filtered data are shown in gray and data filtered by total particle and sulfur dioxide concentration are represented with the color (Fig. S5 in Supplementary Information). Notice the differences in the scaling of y-axes.

During event days, CS is low (Fig. S5c and d in Supplementary Information), which is usually the case (Dada *et al.* 2017). However, the concentration of nucleation mode particles tends to be lower when CS is higher (Fig. S5b in Supplementary Information). This is mostly caused by the very low concentration of nucleation mode particles on non-event days but indicates also that there still might be days when nucleation mode particles are not formed due to condensing of vapors on pre-existing particles.

According to our results, CS is not, however, affected by the number of autumnal moths (Fig. S5a and b in Supplementary Information). The differences between CS data during high (2003, 2010–2012) and low (1998, 1999, 2007, 2008 and 2016) autumnal moth density years as well as years after the peak densities (2004–2006 and 2013–2015) were also tested with the two-sample Kolmogorov–Smirnov test. The test was repeated both for the data from all days that used the filtering conditions and for data from NPF event days. The results of the tests indicated that the data were from the same distributions, and hence, there were no significant differences in CS between the years. Here, we used the larval index as an estimate of the number of autumnal moths (Fig. S5 in Supplementary Information), but light trap and sweeping data led to similar results and are therefore not shown.

General discussion

We studied regional NPF events that typically have a spatial scale of more than 100 km in Värriö (Hussein *et al.* 2009). According to Klemola *et al.* (2006), the spatial synchrony of autumnal moths can reach several hundreds of kilometers in Fennoscandia, though there might be 1–2 years of lag between the peak years. The large spatial scale of regional events and local differences in peak years of autumnal moth population density might explain the low correlation between the number of NPF events and autumnal moth population density (Fig. 5). However, we also studied the distributions of disrupted or fluctuating class 2 types of NPF events and undefined days (Fig. 4), as well as aerosol number concentration data (Figs. 7 and 8), which are also affected by possible aerosol bursts and other local processes.

Tunved *et al.* (2006) concluded that BVOCs contribute to the aerosol load in northern Finland. Our analysis did not, however, show correlation between ambient temperature and aerosol number concentration (Fig. S4c in Supplementary Information), and thus, the basal temperature-driven BVOC emissions (Guenther *et al.* 1995). In the study by Kivimäenpää *et al.* (2016), the emission rates of some monoterpenes of Scots pine were lower on the following spring after herbivory but the temperature dependency of the emission rates was stronger compared with the intact trees. As we were not able to detect any temperature or stress-driven changes in aerosol load, we hypothesize that the total leaf biomass of mountain birch in the area was maybe too small. Mountain birch grow as bushes or up to 10 m high trees (Fig. 1a), and the size of their leaves is usually smaller than the leaves of downy birch (Wielgolaski 2005). In Dahlberg *et al.* (2004) and Heiskanen (2006) the estimated average leaf area index of a mountain birch forest from field measurements was $0.17 \text{ m}^2 \text{ m}^{-2}$ and $0.76 \text{ m}^2 \text{ m}^{-2}$, respectively. Hence, the estimated average biomass in subarctic region is considerably smaller than that of the North European boreal forests (Stenberg *et al.* 2008, Aalto *et al.* 2014).

Results by Yli-Pirilä *et al.* (2016) additionally support our hypothesis that the biomass of

mountain birch was too low to cause detectable changes in aerosol population in field conditions. The produced SOA mass in their experiment was relatively low ($< 1.3 \mu\text{g m}^{-3}$), whereas experiments in other studies have reached SOA masses from $5.9 \mu\text{g m}^{-3}$ (Joutsensaari *et al.* 2015) to $54.6 \mu\text{g m}^{-3}$ (Faiola *et al.* 2019) with Scots pine. Furthermore, the maximum increment in SOA mass load due to the herbivore infestation in the experiment by Yli-Pirilä *et al.* (2016) was 150-fold, though the lowest increment was only 3-fold, showing a large variability in the results.

Earlier studies have shown that even though trees recover from defoliation, the effects may be seen for several years as decreased number of leaves or leaf mass, reducing thereby the total biomass of the trees. In a study by Piper *et al.* (2015) the completely defoliated winter-deciduous trees (*Nothofagus pumilio*) had smaller leaf area per branch for two years and the number of leaves and leaf mass per branch for one year after the defoliation when compared to undamaged trees. Tenow and Bylund (2000) reported that in northern Sweden, 80–90% of leaf-carrying shoots of mountain birch died after complete defoliation by autumnal moths in 1956. They noticed that on the following summer the damaged trees produced long shoots with higher-than-average number of leaves. However, the number of the leaf-carrying shoots was low and increased only gradually after the initial rapid recovery. Tenow and Bylund (2000) estimated that it would take at least 70 years for the damaged trees to reach the assumed size of a mature mountain birch tree.

Besides the low biomass of mountain birch, the low autumnal moth population densities also during our measurement period might affect our results. According to Bylund (1995), the degree of defoliation is usually less than 15% in Fennoscandia, and even during the highest population density years in the beginning of the 1990s, the degree of defoliation was mostly less than 50% (Ruohomäki *et al.* 2000). Although mild stress is usually the most efficient in inducing BVOC emissions (Niinemets 2010), the small biomass combined with a low degree of defoliation might lead to a case where the induction of stress-related BVOCs is not intense enough to enhance aerosol processes. Measure-

ments of organic compounds from the study site would give valuable information on whether the local stresses and phenological states could be detected and separated. Unfortunately, such measurements were not available from the station during our study period.

There are also other biotic stresses at the measurement site that might affect our analysis. Winter moth (*Operophtera brumata*) has recently reached cyclic outbreak dynamics in the northern Fennoscandia (Hagen *et al.* 2007, Jepsen *et al.* 2008, Klemola *et al.* 2016). The outbreak cycle is similar to that of autumnal moths, but the peak densities occur 1–3 years after those of autumnal moths (Jepsen *et al.* 2013, Klemola *et al.* 2016). Yet, the population density of winter moth in boreal Finnish Lapland is minor compared to that of autumnal moths (Hunter *et al.* 2014). Semi-domestic reindeer also graze on mountain birch in the summer. However, earlier studies have shown that even though grazing might reduce the number of mountain birch saplings, the abundance of large trees was not affected or was even higher due to increased regeneration rate of mountain birch as a response to grazing (Lehtonen and Heikkinen 1995, Bråthen and Oksanen 2001, Kumpula *et al.* 2011). Needle rust by fungal infection is additionally a common biotic stress in the area. However, needle rust occurs mainly on Norway spruce during late summer or autumn, and thus, the stress is not simultaneous with the one caused by the relevant herbivory of our current study (Tillman-Sutela *et al.* 2004).

As a consequence of climate change, many changes are predicted to occur in northern ecosystems where the warming is much faster compared to the global average (Law and Stohl 2007, IPCC 2013, Ruosteenoja *et al.* 2016). The rising air temperature and CO_2 concentration are beneficial for the trees, enabling more efficient photosynthesis, longer growing seasons and expansion of the geographical distribution both to higher latitudes and altitudes (Chmielewski and Rötzer 2001, Koca *et al.* 2006, Tømmervik *et al.* 2009, Hari *et al.* 2013, Jepsen *et al.* 2013, Valolahti *et al.* 2015, Tang *et al.* 2018, Ghirardo *et al.* 2020). Also, the abundance of many herbivore species has been predicted to increase and spread to regions where they did not succeed earlier

(Lehmann *et al.* 2020). This is affected by the spreading distribution of host plants, but also due to the rise in winter minimum temperature as, for example, the overwintering eggs of autumnal moths die if the ambient temperature decreases below -36°C (Tenow and Nilssen 1990, Virtanen *et al.* 1998, Bale *et al.* 2002, Hagen *et al.* 2007, Jepsen *et al.* 2008, Ammunét *et al.* 2012, Jepsen *et al.* 2013, Hunter *et al.* 2014). Even though the basal BVOC emissions are exponentially temperature dependent (Guenther *et al.* 1995), recent studies have shown that herbivore attack accompanied with warming, can lead to amplified BVOC emission rates (Kivimäenpää *et al.* 2016, Zhao *et al.* 2017, Li *et al.* 2019). Hence, the importance of stress-induced emissions of BVOCs to atmospheric processes are crucial to understand.

Conclusions

The aim of this study was to investigate whether the larvae of autumnal moths feeding on mountain birch have an atmospherically relevant effect on formation and growth of aerosol particles. Our study is the first to evaluate the enhancement of secondary aerosol production due to herbivory in field conditions. We utilized 25 years of field data from the SMEAR I station in eastern Finnish Lapland. At the study site, autumnal moths showed variable population density. Considering that the site is remote with only minor human impact, and that autumnal moths are by far the most abundant defoliator in the area, the site was ideal for the present study.

Despite the results of laboratory studies, we could not find a clear indication that aerosol processes would be enhanced due to stress-induced BVOC emissions. However, stress emissions may potentially increase total particle concentration for several years after the infestation, though the results found in this study were not conclusive. Temperature, and hence the basal BVOC emissions, did not either have a clear effect on the formation and growth of particles. The leaf biomass of mountain birch may be too small to be able to cause detectable changes, especially when combined with low autumnal moth population densities during our measurement period.

Unexpectedly, neither sulfur dioxide nor sulfuric acid concentrations, which is known to contribute to formation of new aerosol particles, showed a strong correlation with the number of atmospheric aerosol particles.

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Supplementary Information: The supplementary information related to this article is available online at: <http://www.borenav.net/BER/archive/pdfs/ber25/ber25-121-143-supplement.pdf>.

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