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INSIGHTS INTO CARBON-BASED TERRESTRIAL CLIMATE FEEDBACK MECHANISMS

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Academic dissertation

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Important abbreviations and nomenclature

AOD	Aerosol optical depth
BVOC	Biogenic volatile organic compound
CCN	Cloud condensation nuclei
CO_2	Carbon dioxide
CPC	Condensation particle counter
CS	Condensation sink
DMA	Differential mobility analyzer
DMPS	Differential mobility particle sizer
GPP	Gross primary production
GR	Growth rate
GRE	Global radiation enhancement
H_2SO_4	Sulfuric acid
J	Formation rate
LAI	Leaf area index
LUE	Light use efficiency
NO_3	Nitrate radical
NPF	New particle formation
O_3	Ozone
OH	Hydroxyl radical
PAR	Photosynthetically active radiation
PTR-MS	Proton transfer reaction mass spectrometer
PW	Precipitable water
$R_{\mathrm{g,meas}}$	Measured global radiation
$R_{\mathrm{g,mod}}$	Modeled global radiation
$R_{\mathrm{d,meas}}$	Measured diffuse radiation
$R_{\mathrm{d,mod}}$	Modeled diffuse radiation
RH	Relative humidity
sCI	Stabilized Criegee intermediate
SMEAR	Station for measuring ecosystem–atmosphere relations
SO_2	Sulfur dioxide
SOA	Secondary organic aerosol
SZA	Solar zenith angle
T	Temperature
VOC	Volatile organic compound

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Abstract

Feedback mechanisms between atmosphere and biosphere have a potential to mitigate climate change. The connection arises from the activation of photosynthesis due to rising CO_2 concentration and air temperature, leading to enhanced emissions of biogenic volatile organic compounds (BVOC). In the atmosphere, BVOCs can contribute to formation of stabilized Criegee intermediates which, by oxidizing sulfur dioxide, can produce sulfuric acid. Sulfuric acid and oxidized BVOCs are key components for formation and growth of aerosol particles, which can act as seeds for cloud droplets. Cloud droplet number concentration, and hence the properties of clouds depend on the concentration of aerosol seeds. Moreover, scattering of solar radiation by aerosol particles and clouds can further intensify photosynthesis, since diffuse radiation penetrates more evenly inside the canopy, but also mitigate global warming, because the amount of incoming solar radiation is reduced.

In this thesis processes behind formation and growth of aerosol particles were analyzed by determining the relevance of biotic stress induced emissions of BVOCs on atmospheric aerosol load, and deriving a proxy for sulfuric acid with revised source and sink terms. Additionally, the interactions between aerosol particles, clouds, and photosynthesis were studied by developing an algorithm for cloud type classification, and quantifying the effect of aerosol particles and clouds on radiation and photosynthesis.

The main findings of the thesis are: 1) Though clear correlation between aerosol load and intensity of biotic stress could not be observed in field data from Finnish Lapland, we found indications of delayed induced stress effect, leading to elevated aerosol load for several years after the most intense stress periods. 2) Inclusion of a term describing oxidation of sulfur dioxide by stabilized Criegee intermediates in sulfuric acid proxies enables the estimation of night and wintertime sulfuric acid concentration. Dimer formation term is important in polluted environments where molecular clusters are formed rapidly. 3) The overall effectiveness of the cloud algorithm to distinguish different cloud types was nearly 70 %. The dominating cloud types in the measurement site in southern Finland are low level patchy and uniform clouds. 4) In the boreal region, the ecosystem scale photosynthesis can be up to 30 % larger in the presence of clouds compared to clear-sky and clean atmospheric conditions. The maximal enhancement in photosynthesis occurs when the fraction of diffuse radiation from global radiation is 0.4–0.5.

The thesis covers several topics on aerosol–cloud–radiation interactions, shedding light on versatile and complex interactions between the atmosphere and boreal forest ecosystem. Furthermore, it provides tools for upcoming researches on atmospheric physics and chemistry.

Keywords: aerosol–cloud–radiation interactions, feedback loop, sulfuric acid, boreal forest, biotic stress, volatile organic compounds, cloud type

Contents

List of publications

This thesis consists of an introductory review, followed by four peer-reviewed research articles. In the introductory part, these papers are cited according to their roman numerals. **Papers I, III–IV** are reproduced under the Creative commons Attribution License, while **Paper II** is reproduced with kind permission of Boreal Environment Research Publishing Board.

I Ezhova, E., **Ylivinkka, I.**, Kuusk, J., Komsaare, K., Vana, M., Krasnova, A., Noe, S., Arshinov, M., Belan, B., Park, S.-B., Lavrič, J. V., Heimann, M., Petäjä, T., Vesala, T., Mammarella, I., Kolari, P., Bäck, J., Rannik, Ü., Kerminen, V.-M., and Kulmala, M. (2018). Direct effect of aerosols on solar radiation and gross primary production in boreal and hemiboreal forests. *Atmospheric Chemistry and Physics*, 18(24):17863–17881.

II **Ylivinkka I.**, Itämies J., Klemola T., Ruohomäki K., Kulmala M., and Taipale D. (2020). Investigating evidence of enhanced aerosol formation and growth due to autumnal moth larvae feeding on mountain birch at SMEAR I in northern Finland. *Boreal Environment Research*, 25:121–143.

III Dada, L., **Ylivinkka, I.**, Baalbaki, R., Li, C., Guo, Y., Yan, C., Yao, L., Sarnela, N., Jokinen, T., Daellenbach, K. R., Yin, R., Deng, C., Chu, B., Nieminen, T., Wang, Y., Lin, Z., Thakur, R. C., Kontkanen, J., Stolzenburg, D., Sipilä, M., Hussein, T., Paasonen, P., Bianchi, F., Salma, I., Weidinger, T., Pikridas, M., Sciare, J., Jiang, J., Liu, Y., Petäjä, T., Kerminen, V.-M., and Kulmala, M. (2020). Sources and sinks driving sulfuric acid concentrations in contrasting environments: implications on proxy calculations. *Atmospheric Chemistry and Physics*, 20(20):11747–11766.

IV **Ylivinkka, I.**, Kaupinmäki, S., Virman, M., Peltola, M., Taipale, D., Petäjä, T., Kerminen, V.-M., Kulmala, M., and Ezhova, E. (2020). Clouds over Hyytiälä, Finland: an algorithm to classify clouds based on solar radiation and cloud base height measurements. *Atmospheric Measurement Techniques*, 13(10):5595–5619.

1 Introduction

Climate change driven by human activities during the last decades is nowadays a widely researched and discussed theme. Past studies have already provided us with good knowledge on greenhouse gas emissions and how they contribute to global warming, but the impacts of climate change are still rather poorly understood (IPCC, 2021). This stems from the complex interactions between atmosphere, biosphere, and hydrosphere. They form a united system where any shift from the balance can create a long chain of changes in numerous other variables, both amplifying or dampening the original disturbance. Therefore, the attempts of predicting the future conditions require holistic research and understanding of the interlinkages.

Kulmala et al. (2013) proposed a feedback mechanism between the terrestrial ecosystem and atmosphere illustrated in Fig. 1a: the increasing atmospheric CO_2 concentration, and subsequent temperature rise, fertilize photosynthesis (Morison and Lawlor, 1999; Fernández-Martínez et al., 2017; Zhang et al., 2017; Liu et al., 2019; Tharammal et al., 2019), altering and promoting also the emissions of reactive gases from vegetation (Fuentes et al., 2000; Peñuelas and Staudt, 2010). These gases, called biogenic volatile organic compounds (BVOC), are oxidized rapidly in the atmosphere (Atkinson and Arey, 2003; Mogensen et al., 2011, 2015), forming vapors that may participate in aerosol formation and growth processes (e.g. Riipinen et al., 2011, 2012; Ehn et al., 2014; Peräkylä et al., 2014; Riccobono et al., 2014; Patoulias et al., 2015). Depending on the chemical and physical properties of the formed particles, they can act as cloud condensation nuclei (CCN), i.e. surfaces on top of which water can condense, forming cloud droplets (Merikanto et al., 2009; Kerminen et al., 2012; Scott et al., 2014; Riipinen et al., 2015).

The number concentration of CCN affects the properties of forming clouds; in the areas of high CCN concentration, water is distributed over a large number of nuclei, making the formed cloud droplets small. Clouds with small cloud droplets are optically brighter and have a longer lifetime compared to clouds with large cloud droplets, because small droplets have a higher surface area and it takes longer for them to reach the size of a rain drop. Hence, clouds with small cloud droplets as well as optically bright particles are efficient in scattering solar radiation back to space, and thereby in mitigating the warming by greenhouse gases (Rosenfeld et al., 2014; Bellouin et al., 2020; IPCC, 2021). Moreover, scattering increases the fraction of diffuse radiation

from global radiation on the surface of the Earth, which further enhances the photosynthesis of forest ecosystems, since diffuse radiation can penetrate more evenly inside the canopy (**Paper I**; Gu et al., 1999, 2002, 2003; Niyogi et al., 2004; Mercado et al., 2009; Cheng et al., 2015, 2016). Thus, photosynthesis, enhanced by the increased CO_2 concentration, is enhanced also by the presence of organic aerosol particles and clouds. Figure 1a presents the original climate feedback loop proposed by Kulmala et al. (2013) whereas Fig. 1b compiles ecosystem–atmosphere interactions more holistically, highlighting additionally the interactions touched by this thesis.

The emissions of BVOCs are modified, in addition to photosynthetic activity and temperature, by plant stresses, such as drought, herbivory, and fungal infections (e.g. Holopainen and Gershenzon, 2010; Peñuelas and Staudt, 2010). Stresses can increase or decrease the emission rates of constitutively emitted BVOCs, or induce emissions of stress-related compounds (Dudareva et al., 2006; Fineschi and Loreto, 2012; Faiola and Taipale, 2020). Co-occurrence of stresses can have variant effects on the emissions of BVOCs compared to single stresses, complicating the estimations of the emission profiles (Dicke et al., 2009; Niinemets, 2010b). Climate change brings about alterations in the environmental conditions, and therefore also the frequency and intensity of many stresses are predicted to increase in the future, affecting thus the emissions of BVOCs (Yuan et al., 2009; Peñuelas and Staudt, 2010; Holopainen et al., 2018).

The relevance of stress-induced emissions on atmospheric processes are poorly known (Faiola and Taipale, 2020). In many laboratory (Joutsensaari et al., 2005, 2015; Hamilton et al., 2009; Mentel et al., 2013; Yli-Pirilä et al., 2016; Zhao et al., 2017; Ylisirniö et al., 2020) and modeling studies (Berg et al., 2013; Bergström et al., 2014; Taipale et al., 2021) the herbivory-induced emissions have led to higher aerosol load compared to the aerosol load from the emissions of control trees, although also opposing results exist (Faiola et al., 2018, 2019). Moreover, in some studies herbivory combined with warming produced even larger increases in the emissions of BVOCs (Kivimäenpää et al., 2016; Zhao et al., 2017; Li et al., 2019). The lack of field measurements demands urgently to study the importance of stress-induced emissions of BVOCs on aerosol formation and growth processes, and the subsequent impact on terrestrial climate feedback mechanisms via formation and growth of aerosol particles (**Paper II**; Faiola and Taipale, 2020; Yli-Juuti et al., 2021; Artaxo et al., 2022).

Atmospheric new particle formation (NPF) typically requires sulfuric acid (Nieminen et al., 2014; Vakkari et al., 2015; Lehtipalo et al., 2018; Yao et al., 2018). Therefore,

knowing its atmospheric concentration is of importance also in analyses of feedback mechanisms involving aerosol particles and clouds. However, long-term sulfuric acid concentration measurements are sparse due to its low atmospheric concentration and only relatively recent development in measurement techniques (Junninen et al., 2010; Mikkonen et al., 2011). To compensate the knowledge gap, several proxies have been developed to estimate the ambient sulfuric acid concentration (Petäjä et al., 2009; Mikkonen et al., 2011; Lu et al., 2019). More recent studies have shown that biogenic or anthropogenic emissions of VOCs can modify the ambient sulfuric acid concentration via formation of stabilized Criegee intermediates (sCI) (Mauldin III et al., 2012; Boy et al., 2013; Percival et al., 2013; Sipilä et al., 2014), and that sulfuric acid dimer formation is likely the first step of NPF (Almeida et al., 2013; Yao et al., 2018; Yan et al., 2021). These new sources and sinks were not accounted for in the previous sulfuric acid proxies, and thus a revision of current proxies is required for accurate sulfuric acid concentration estimates, particularly since sulfuric acid concentration is connected to the predicted changes in the emissions of BVOCs by climate change, stresses, and feedback mechanisms due to formation of Criegee intermediates (**Paper III**; Rose et al., 2021; Yang et al., 2021).

Aerosol–cloud–radiation interactions are associated with large uncertainties due to the high variability in properties of aerosol particles (most importantly size, concentration, and chemical composition) and clouds (type, height, location, and cloud droplet size and phase), as well as in spatial distribution and time scales of different processes (IPCC, 2021; Artaxo et al., 2022). Hence, in order to improve climate change predictions, comprehensive understanding of the underlying processes is required: what is the concentration, composition, and spatial variation of aerosol particles (**Papers II**–**III**), how well do the particles and clouds scatter radiation (**Papers I**; **IV**), and what is the response of boundary layer processes, including photosynthesis, to the changes in irradiance (**Paper I**). This thesis aims to gain further insights into the individual steps of feedback mechanisms, highlighted with green color in Fig. 1b, to quantify aerosol and cloud driven changes on photosynthesis, and to provide new tools for research on the topic. More specifically, the main objectives of this thesis are to

1. evaluate the impact of biotic stress on aerosol processes using field observations from a pristine environment (**Paper II**).

2. improve the current sulfuric acid proxies by considering the influence of stabilized Criegee intermediates and sulfuric acid dimer formation (**Paper III**).

3. formulate an algorithm for cloud type identification, and to provide information of cloud type fractions in southern Finland (**Paper IV**).

4. separate aerosol and cloud effects on radiation, and to quantify how changes in radiation affect photosynthesis in boreal forests (**Paper I**).

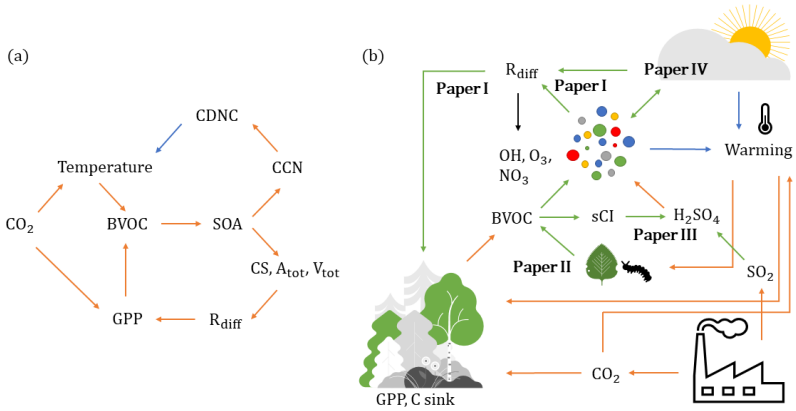

Figure 1: (a) Illustration of terrestrial feedback loop as presented in Kulmala et al. (2013). BVOC = biogenic volatile organic compound, SOA = secondary organic aerosol, CCN = cloud condensation nuclei, CDNC = cloud droplet number concentration, CS = condensation sink, A_{tot} and V_{tot} = aerosol total surface area and volume concentrations, R_{diff} = fraction of diffuse radiation from global radiation, and GPP = gross primary production. (b) Ecosystem–atmosphere interactions presented more comprehensively. sCI = stabilized Criegee intermediate. Orange and green arrows indicate increase, blue arrows decrease, and black arrows variant effects. Green arrows highlight the interactions studied in this thesis.

2 Scientific background

Forests cover 31 % of the Earth's land area, and about 27 % of that is covered by boreal forests, ranging approximately between 50°N and 70°N (FAO and UNEP, 2020). Forests mitigate global warming by acting as carbon sink, but also through climate feedbacks involving aerosol formation and growth from BVOC oxidation products (Spracklen et al., 2008; Kulmala et al., 2020). Although VOCs are emitted to the atmosphere also from anthropogenic sources, biogenic emissions dominate the total emissions globally by one order of magnitude (Guenther et al., 2012). Globally, the most extensively emitted BVOC is isoprene (C_5H_8, 50 % of emissions), which is commonly produced by tropical forests, whereas the emissions from coniferous boreal forests are dominated by groups of monoterpenes ($C_{10}H_{16}$, 15 %) and sesquiterpenes ($C_{15}H_{24}$, 3 %) (Rinne et al., 2009; Guenther et al., 2012; Rantala et al., 2015; Artaxo et al., 2022).

2.1 Photosynthesis and BVOC emissions

In photosynthesis plants fix carbon dioxide (CO_2) from the atmosphere in the presence of light. Photosynthesis is strengthened by the increasing ambient CO_2 concentration but also due to warming climate, which boosts the enzyme-controlled reactions (Morison and Lawlor, 1999; Fernández-Martínez et al., 2017; Zhang et al., 2017; Liu et al., 2019). Photosynthesis is also enhanced due to lengthening of the growing season and migration of woody plant species both northward and upslope, allowing prominent changes to the local vegetation diversity especially at high latitudes where the warming is faster than the global average (Chmielewski and Rötzer, 2001; Peñuelas and Boada, 2003; Tape et al., 2006; Jump et al., 2009; IPCC, 2021; Sizov et al., 2021).

Plants allocate some of the carbon, fixed in photosynthesis, to the production of BVOCs, which are emitted both by aerial and below ground plant parts. Plant species have unique emission profiles, which are further modified by environmental conditions, stresses, and even variations in genotypes, thus leading to thousands of different BVOCs (Dudareva et al., 2006; Laothawornkitkul et al., 2009; Bäck et al., 2012; Guenther et al., 2012; Fineschi et al., 2013; Monson, 2013; Aalto et al., 2014, 2015; Maja et al., 2014; Faiola et al., 2015). The emissions of many BVOCs depend exponentially on temperature due to increased enzyme activity, saturation vapor pressure, and diffusion of compounds from leaf tissue (Tingey et al., 1991; Guenther et al., 1995; Fuentes et al.,

2000; Li and Sharkey, 2013; Harley, 2013). However, the temperature dependency of the emissions can naturally be exponential only if the plant is not damaged by the heat (Kleist et al., 2012).

Elevated CO_2 concentration may suppress the emissions of some BVOCs (Peñuelas and Staudt, 2010; Holopainen et al., 2018). Particularly isoprene emissions have been shown to decrease in exposure to elevated CO_2 concentration (366–2500 ppm) (Peñuelas and Staudt, 2010, Suppl., and references therein), because isoprene formation is then uncoupled from photosynthesis (Wilkinson et al., 2009; Peñuelas and Staudt, 2010; Calfapietra et al., 2013; Potosnak et al., 2014). In case of other BVOCs, only few studies with divergent results exist (Peñuelas and Staudt, 2010; Calfapietra et al., 2013; Holopainen et al., 2018). For example, in one study monoterpene emissions of Scots pines (*Pinus sylvestris*) were not affected by the elevated CO_2 concentration (700 ppm), but increased significantly when air temperature was simultaneously risen (2–6 °C) (Räisänen et al., 2008). In their study, temperature increase alone resulted in decrease in monoterpene emission rate. In addition, many monoterpenes emitting trees have special storage pools for monoterpenes, e.g. in resin ducts, and thus the compounds are not necessarily emitted into the atmosphere instantly after the production, which can partly explain why the effect of elevated CO_2 concentration is not detected (Ghirardo et al., 2010; Taipale et al., 2011; Calfapietra et al., 2013; Vanhatalo et al., 2018).

2.1.1 Plant stresses

Plants experience stress when they are exposed to unfavorable growing conditions such as drought, heat wave and air pollution (abiotic stressors), or pathogens and herbivores (biotic stressors) (Dudareva et al., 2006; Vickers et al., 2009). Stresses can induce production of BVOCs, increase or decrease constitutive plant emissions, or turn off emissions of some BVOCs (Dicke et al., 2009; Peñuelas and Staudt, 2010; Faiola and Taipale, 2020). The modulations in BVOC emission profiles are part of a defense response of plants, and thus help them to mitigate stresses (Holopainen, 2004, 2011; Dudareva et al., 2006; Yuan et al., 2009; Fineschi and Loreto, 2012). The stress-induced BVOCs can, for example, reduce oxidative stress of the leaf tissue by reacting with oxidants, repel herbivores and pathogens, attract herbivore parasites and predators, or even warn the neighboring trees of the stress (Dudareva et al., 2006; Mäntylä et al., 2008; Yuan et al., 2009; Dicke and Baldwin, 2010; Holopainen and Gershenzon, 2010;

Loreto and Schnitzler, 2010; Fineschi and Loreto, 2012; Holopainen and Blande, 2013; Possell and Loreto, 2013; Trowbridge and Stoy, 2013; Koski et al., 2015). Due to their multiple functions, stress-induced BVOCs are highly plant species and stress dependent (Kesselmeier and Staudt, 1999; Vuorinen et al., 2007; Dicke et al., 2009; Niinemets, 2010b; Kreuzwieser and Rennenberg, 2013; Faiola and Taipale, 2020). Moreover, the timing and duration, as well as stress severity impact the BVOC induction (Niinemets, 2010a,b; Niinemets et al., 2013; Miresmailli et al., 2012; Maja et al., 2014; Rieksta et al., 2020). For example, boreal forest trees can suffer from a cold spell during growing season but they are accustomed to cold temperatures during wintertime. Co-occurring or successive stresses can modify the emissions of BVOCs in a different way than the stresses individually, since plants may acclimate to stresses, or one stress may lower plants' tolerance for other stresses (Dicke et al., 2009; Niinemets, 2010b).

As a consequence of climate change, many alterations in the growing conditions are occurring: temperature and CO_2 concentration are rising, but also for example ozone concentration, precipitation patterns, and nutrient, mainly nitrogen, availability may change (Law and Stohl, 2007; Peñuelas and Staudt, 2010; Holopainen et al., 2018; IPCC, 2021). Furthermore, warming climate enables migration of plant, herbivore and pathogen species to areas where they did not thrive earlier (Ammunét et al., 2012; Hunter et al., 2014; Pureswaran et al., 2015; Lehmann et al., 2020). These changes may substantially alter the total amount and fractions of different BVOCs in the atmosphere. For example, emissions of BVOCs from biotically stressed plants are often significantly increased compared to the emissions from stress-free plants (Joutsensaari et al., 2005, 2015; Yli-Pirilä et al., 2016; Zhao et al., 2017; Kari et al., 2019; Li et al., 2019; Rieksta et al., 2020), and simultaneous warming experiment have resulted in strong increase in the emissions of BVOCs (Kivimäenpää et al., 2016; Zhao et al., 2017; Li et al., 2019).

Mountain birch–autumnal moth relationship

Laboratory and modeling studies have found increase in aerosol load from the emissions of biotically stressed trees due to higher concentrations and different blend of BVOCs (Joutsensaari et al., 2005, 2015; Hamilton et al., 2009; Berg et al., 2013; Mentel et al., 2013; Bergström et al., 2014; Yli-Pirilä et al., 2016; Zhao et al., 2017; Ylisirniö et al., 2020; Taipale et al., 2021). However, in some studies the aerosol mass yield has de-

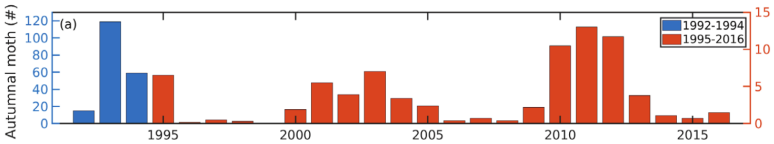

Figure 2: Autumnal moth population density estimation near a measurement station in Finnish Lapland in 1992–2016. The population density is estimated by counting the found larvae in 10 minutes of search (see Sect. 3.2.4). Note that the y-axis is different for the first three years due to the much higher population densities. The figure is found in **Paper II** as Fig. 3a.

creased compared to the mass yield from BVOC emissions from healthy trees (Mentel et al., 2013; Faiola et al., 2018, 2019). This has been related to the unfavorable blend of BVOCs, since their ability to form suitable low volatility vapors is diverse. So far, field studies on the effects of biotic stresses on aerosol processes do not exist, though they are vital for validating the results of laboratory and modeling studies. Thus, this thesis opens the field observation analyses on the topic, by studying formation and growth of aerosol particles at a measurement site in Finnish Lapland where common biotic stress is defoliation of mountain birches (*Betula pubescens* var. *pumila*, formerly spp. *czerepanovii*) due to feeding by autumnal moth larvae (*Epirrita autumnata*, Lepidoptera: Geometridae) (**Paper II**). In the following paragraph, the selected herbivore and tree species are introduced.

Autumnal moths are univoltine, meaning that they can have one generation per year, moths that feed on deciduous trees and shrubs, but in the northern Fennoscandia their main host plant is mountain birch (Haukioja et al., 1988). Autumnal moths overwinter as eggs, hatch concurrently with budbreak, taking place in early June in the study area, and start feeding on leaves (Kaitaniemi et al., 1997; Kaitaniemi and Ruohomäki, 1999). Larvae go through five development stages, instars, within 4–7 weeks, after which they pupate, and merge flying in August to breed (Kaitaniemi and Ruohomäki, 1999). In the northern Fennoscandia, autumnal moths have about ten years long periodic cycle in population density (Haukioja et al., 1988), which enables the investigation of periods with different stress severity. Autumnal moth population density from the study site, estimated with larval index (see Sect. 3.2.4), is presented in Fig. 2.

Yli-Pirilä et al. (2016) have studied emissions of BVOCs from mountain birches, infested by autumnal moths, and ability of the emitted gases to produce secondary or-

ganic aerosol (SOA) in laboratory conditions. They found that the SOA mass formed from stress-induced emissions was up to 150-fold compared to emissions from control trees. The emission rates of BVOCs from mountain birches depend on the level of defoliation, and thus the number of larvae and their rate of feeding (Haukioja, 2003; Yli-Pirilä et al., 2016; Taipale et al., 2021). However, in case of severe defoliation, the total emissions of BVOCs can decrease due to extensive suppression of leaf area (Taipale et al., 2021). The total level of defoliation in the northern Fennoscandia is usually less than 15 %, though during severe mass outbreaks total defoliation and even mortality of trees has occurred (Bylund, 1995; Tenow and Bylund, 2000). Additionally, mountain birches have been shown to emit defense compounds for several years after infestation, indicating that they have an immunological memory for stresses (Kaitaniemi et al., 1998; Ruuhola et al., 2007; Haapanala et al., 2009).

2.2 New particle formation

Atmospheric new particle formation is a process where new nanometer sized clusters and particles are formed from gas phase precursors (Vehkamäki and Riipinen, 2012; Kerminen et al., 2018). The precursor compounds produce first small molecular clusters, which gradually grow into larger sizes. Though the mechanisms behind NPF vary at different locations, commonly the initiation of NPF requires sulfuric acid (H_2SO_4), stabilizing bases, such as ammonia (NH_3) and amines, and organic compounds (Kulmala et al., 2014b; Lehtipalo et al., 2018; Olenius et al., 2018; Yan et al., 2021). Particles reaching sizes of around 100 nm have a high climatic relevance since they may scatter radiation or act as CCN (Kerminen et al., 2012; Riipinen et al., 2015; Bellouin et al., 2020). Hence, NPF is the key linking precursor compounds to the climate feedback mechanisms due to CCN production, cloud properties, and radiation interactions.

NPF occurs almost everywhere in the world, and it is a prominent source of small particles and CCN into the atmosphere (Kerminen et al., 2018). Small particles dominate aerosol number concentration but due to their small size, aerosol mass concentration is dominated by large particles (Kerminen et al., 2018). Depending on the field of study, different variables are relevant in research. For instance, number concentration of particles is an essential variable when studying cloud formation whereas mass concentration is relevant in air pollution and health related research (e.g. Kerminen et al., 2012; Guo et al., 2014; Brauer et al., 2016).

2.2.1 Organic compounds

Organic compounds from biogenic and athropogenic sources are oxidized in the atmosphere most importantly by OH and NO_3 radicals, and ozone (Atkinson and Arey, 2003; Mogensen et al., 2011, 2015; Hellén et al., 2018). The oxidation products have lower vapor pressures and are, therefore, less volatile, enabling some of them to contribute to formation and growth of aerosol particles (Riipinen et al., 2011, 2012; Donahue et al., 2013; Ehn et al., 2014).

The groups of BVOCs commonly emitted by boreal forest trees, monoterpenes and sesquiterpenes, have oxidation products that are efficient in growing particles (Tunved et al., 2006; Hao et al., 2009; Jimenez et al., 2009; Peräkylä et al., 2014, 2020; Heikkinen et al., 2020; Simon et al., 2020; Qiao et al., 2021), whereas the most common BVOC, isoprene, have been shown to suppress NPF when found at high concentrations (Kiendler-Scharr et al., 2009, 2012; McFiggans et al., 2019; Heinritzi et al., 2020). Hence, boreal forests are an important source of SOA, and have considerable effect on climate via terrestrial feedback mechanisms (Scott et al., 2018; Yli-Juuti et al., 2021). Moreover, as described in Sect. 2.1, at high CO_2 concentrations the emission levels of isoprene may be lower than the present day levels, which thereby can enhance NPF and growth of particles in the future.

2.2.2 Sulfuric acid

Sulfuric acid (H_2SO_4) is a key precursor for NPF, and hence knowing its atmospheric concentration precisely is essential when studying atmospheric chemical and physical processes. Sulfuric acid is formed from the oxidation of sulfur dioxide (SO_2) by OH radicals and stabilized Criegee intermediates, produced in the ozonolysis of alkenes from biogenic or anthropogenic sources (Mauldin III et al., 2012; Boy et al., 2013; Percival et al., 2013; Sipilä et al., 2014). The oxidation by OH radicals has been known already for a long time, whereas the importance of oxidation by stabilized Criegree intermediates was discovered more recently. Sulfuric acid is lost in formation and growth of atmospheric aerosol particles by condensing due to the low vapor pressure of sulfuric acid (Vakkari et al., 2015; Stolzenburg et al., 2020). Figure 3 illustrates sources and sinks of sulfuric acid. Hence, sulfuric acid is linked to terrestrial climate feedback mechanisms in two ways: due to production of Criegee intermediates from BVOCs,

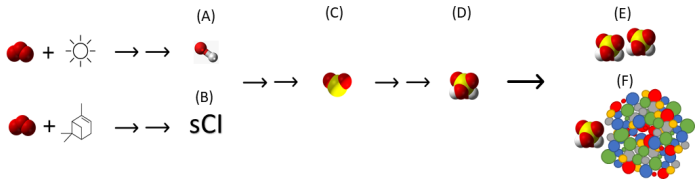

Figure 3: Schematic representation of sources and sinks of sulfuric acid. Various steps of chemical reactions have been excluded in order to keep the illustration simple (double arrows). OH radicals (A) are formed when ozone is dissociated by light, whereas stabilized Criegee intermediates (B) are produced in the ozonolysis of alkenes. OH and stabilized Criegee intermediates can oxidize sulfur dioxide (C), forming eventually sulfuric acid (D). Dimer formation (E) and condensation on particles (F) are sinks for sulfuric acid.

and contribution to aerosol formation and growth processes to form particles relevant for cloud formation and scattering of solar radiation.

The ambient sulfuric acid concentration is low ($10^4 - 10^7$ molecules cm^{-3}), which makes sulfuric acid a difficult and expensive compound to measure, despite the developments in mass spectrometric instruments (Junninen et al., 2010; Mikkonen et al., 2011). Therefore, especially long-term observations of sulfuric acid concentration are sparse, and there is a demand for proxies estimating sulfuric acid concentration accurately in different locations (Petäjä et al., 2009; Mikkonen et al., 2011; Lu et al., 2019). As an example, in **Paper II** we used proxy by Petäjä et al. (2009) to estimate the sulfuric acid concentration in Finnish Lapland as it was not measured at the station during the time of the study (Jokinen et al., 2022).

Earlier proxies accounted for formation of sulfuric acid from oxidation of sulfur dioxide by OH radicals, and loss due to condensing on preexisting particles (condensation sink, CS) (Petäjä et al., 2009; Mikkonen et al., 2011; Lu et al., 2019). However, the recent findings showing the relevance of Criegee intermediates for sulfuric acid formation, and suggesting the dimer formation to be the first step of NPF (Almeida et al., 2013; Yao et al., 2018; Yan et al., 2021), call for an addition of a new source and sink terms for sulfuric acid in the proxies. Therefore, in **Paper III** we propose a revised sulfuric acid proxy, and study the importance of different sources and sinks of sulfuric acid in various environments.

2.3 Aerosol–cloud–radiation interactions

The increasing atmospheric CO_2 concentration has initiated a terrestrial feedback loop where photosynthesis, first enhanced by increasing CO_2 concentration and temperature, is further enhanced due to aerosol–cloud–radiation interactions (**Paper I**; Kulmala et al., 2004, 2013, 2014a; Arneth et al., 2010; Makkonen et al., 2012a; Paasonen et al., 2013; Rap et al., 2018; Scott et al., 2018; Sporre et al., 2019; Yli-Juuti et al., 2021; Artaxo et al., 2022). As described above, BVOCs emitted by forests can help in forming and growing new aerosol particles but also contribute in the production of another important precursor, sulfuric acid, via formation of Criegee intermediates. When formed particles grow into sizes around 100 nm in diameter, they can alter the radiative budget of the Earth directly by scattering solar radiation and indirectly by acting as CCN, and thereby changing cloud formation and properties of clouds, and linking eventually back to the ecosystem processes due to feedback mechanisms, described in detail below (Merikanto et al., 2009; Kerminen et al., 2012; IPCC, 2021).

Cloud droplets form around CCN, which are seeds on top of which water condenses. In high CCN concentration water is distributed over a large number of nuclei, leading to small cloud droplets. These small cloud droplets have a high surface area, which makes them scatter more light compared to larger and fewer in number cloud droplets formed in areas with lower CCN concentration (Twomey effect) (Twomey, 1977). Thus, small cloud droplets increase the albedo of clouds, which has a cooling effect on climate (Paasonen et al., 2013; IPCC, 2021; Yli-Juuti et al., 2021). In addition, clouds with small cloud droplets have longer lifetime because it takes then longer for small cloud droplets to grow into rain drops, and eventually fall out from the sky (Albrecht effect) (Albrecht, 1989; Gryspeerdt et al., 2014; Rosenfeld et al., 2014; Haywood et al., 2021; IPCC, 2021). Hence, the clouds have a longer time span to scatter radiation, and thereby they can further enhance the cooling effect by clouds.

Clouds interact also with long-wave thermal radiation emitted by the Earth. The altitude and optical properties of clouds determine whether they are effectively cooling the climate, i.e. scattering solar radiation and emitting long-wave radiation to space, or whether they have a warming effect by passing most of the solar radiation and emitting only little thermal energy to space (Schneider and Dennett, 1975; IPCC, 2021). Low level, optically thick clouds are efficient in cooling the climate, whereas optically thin clouds at high altitudes have a net warming effect on climate. Cloud

distribution over the Earth additionally affects their ability to cool the climate. Clouds forming above surfaces with low albedo, such as oceans, can increase the albedo of the area, and thereby increase the backscattering of solar radiation and help in mitigating global warming. Thus, when considering also the Twomey and Albrect effects, CCN concentration has an important role in cloud processes and can significantly contribute to the radiative budget of the Earth.

Besides the impact on air temperature, scattering of solar radiation modifies the light conditions on the Earth, which can affect the photosynthetic activity of vegetation for several reasons. Firstly, under diffuse radiation conditions light saturation of the upper leaves, meaning that they are exposed to excess energy which they cannot utilize for photosynthesis, occurs seldom as radiation is distributed more evenly inside the canopy (Gu et al., 2002). Secondly, diffuse radiation has been suggested to increase light and water use efficiencies, which describe the efficiency to use incident light for photosynthesis and water for metabolism, respectively (Gu et al., 2002, 2003; Cheng et al., 2015, 2016; Kivalov and Fitzjarrald, 2019). Thirdly, partitioning of energy to diffuse radiation can alter other environmental parameters, for example decrease air temperature, leading to reduction in leaf and soil respiration (Gu et al., 1999, 2002, 2003). Hence, net carbon uptake in the ecosystem scale can be enhanced under diffuse light conditions, i.e. in the presence of aerosol particles and clouds (**Paper I**; Gu et al., 1999, 2002, 2003; Niyogi et al., 2004; Mercado et al., 2009; Cheng et al., 2015, 2016; Kivalov and Fitzjarrald, 2019; Artaxo et al., 2022).

The radiative conditions on the Earth can additionally affect atmospheric photochemistry, and subsequently NPF (Dada et al., 2017). For example, formation of OH radicals, needed for oxidation of e.g. sulfur dioxide and BVOCs into low volatility vapors that can participate in NPF, requires UVB radiation, which is limited in the presence of clouds (Petäjä et al., 2009; Peräkylä et al., 2014). Therefore, the emissions of BVOCs, NPF and CCN production are crucially linked to clouds as well as ecosystem processes (Fig. 1b). To further increase understanding on aerosol–cloud–radiation interactions, and to improve analyses on the effects of clouds on boundary layer processes, an algorithm for cloud type classification is developed in **Paper IV**.

2.3.1 Cloud type classification

Because different cloud types have variant impacts on many processes occurring in the boundary layer, these processes need to be studied considering also the presence of clouds. In comprehensive data analysis, based on long-term data sets, automated cloud classification is necessity since the algorithms produce data inexpensively, and with high temporal and spatial resolution, compared to human observations. Several types of instruments can provide data for classification algorithms, including both ground-based and satellite instrumentation (Tapakis and Charalambides, 2013). The feasibility of the instrumentation for the algorithm depends on the application of the cloud classification. For example, satellite instrumentation capture large scale phenomena while algorithms based on ground-level observations can describe local variation in cloudiness (Tapakis and Charalambides, 2013).

A simple cloud algorithm based on the existing instrumentation of a measurement station in southern Finland is developed in **Paper IV** for ecosystem–atmosphere interaction studies. The algorithm is an adaptation of the work by Duchon and O'Malley (1999), where different types of clouds were distinguished by their ability to attenuate global radiation and cause temporal fluctuations in global radiation. In **Paper IV** cloud base height data from a ceilometer is added to the algorithm, enabling to increase the number of different cloud classes, as clouds with similar features on radiation can be distinguished. Radiation and cloud base height measurements are rather common at different measurement sites, and therefore the cloud algorithm may also be used in other environments (Illingworth et al., 2007; Tapakis and Charalambides, 2013).

3 Materials and methods

This thesis is based on data collected at several field stations, located both inside and outside of the boreal region. In the following sections, the stations (Sect. 3.1), the used instrumentation (Sect. 3.2), and the calculation of derived variables (Sect. 3.3) are introduced.

3.1 Measurement stations

The key measurement stations of this thesis are SMEAR I (**Papers I**–**II**) in eastern Finnish Lapland and SMEAR II (**Papers I**; **III**–**IV**) in southern Finland. They are described here in detail, whereas other stations located in various environments ranging from Siberian taiga to the megacity of Beijing (**Papers I**; **III**) are briefly introduced.

3.1.1 SMEAR I

SMEAR (Station for Measuring Ecosystem–Atmosphere Relations) sites have been established for comprehensive monitoring of atmospherical and ecological variables, aiming for better understanding of versatile interactions occurring in nature (Hari et al., 2013). SMEAR I is located in Värriö strict nature reserve in eastern Finnish Lapland (67°44'N, 29°37'E). There are only minor local pollution sources, though occasionally pollution from industrial areas in Kola peninsula in Russia are transported to the station (Kyrö et al., 2014; Sipilä et al., 2021). The atmospheric measurements are conducted on top of Kotovaara hill (390 m a.s.l.), which is surrounded by about 70-years-old Scots pine (*Pinus sylvestris*) dominated forest with average canopy height of 10 m. Nearby there are also several fells (the closest is Värriö I, about 1.5 km south-west from Kotovaara) with slopes forested by mountain birches. The coldest month is January (average temperature −11.4 °C) and the warmest is July (13.1 °C). The sun does not set approximately between 27 May and 15 July.

3.1.2 SMEAR II

SMEAR II site is located in Hyytiälä in southern Finland (61°51'N, 24°17' E, 181 m a.s.l.). SMEAR II is a rural background measurement site. The closest city (Tampere,

240 000 inhabitants) is located about 50 km south-west from the station. Other local pollution sources are sawmills, the closest locating about 6 km south-east from the station. The forest around the station is dominated by about 60-years-old Scots pines with canopy height of about 20 m. The coldest month is January (−7.2 °C) and the warmest is July (16.0 °C).

3.1.3 Other stations

In **Paper I** data from SMEAR Estonia (Järvselja, Estonia) as well as two stations in Siberia: Zotino (Krasnoyarsk region, Russia) and Fonovaya (Tomsk region, Russia) were included in the analysis. SMEAR Estonia is located in the hemiboreal region. The forest around the station is mixed, containing Scots pine, Norway spruce (*Picea abies*), silver birch (*Betula pendula*), and downy birch (*Betula pubescens*). The station in Zotino is surrounded by old-grown Scots pine forest, whereas the forest in Fonovaya is mixed containing Scots pines, birch (*Betula verrucósa*), and aspen (*Populus tremula*).

In the development of sulfuric acid proxies (**Paper III**), we used also data from Agia Marina (Cyprus), Kilpilahti (Finland), and three different sized cities: SMEAR III in Helsinki (Finland), Budapest (Hungary), and Beijing (China). The site in Agia Marina is a rural background station, while measurements in Kilpilahti were collected in an industrial area near an oil refinery. SMEAR III is an urban background station, whereas the measurement platform in Budapest is in the city center, and in Beijing near a busy street.

3.2 Key measurements

The results of this thesis are based on versatile measurements, including in-situ and remote-sensed aerosol data, radiation and cloud parameters, as well as phenological data on herbivore population densities. In this section, the key instrumentation is outlined topic-wise.

3.2.1 Aerosol particles

At SMEAR sites, aerosol number size distribution is measured with differential mobility particle sizer (DMPS) (Aalto et al., 2001). It consists of two differential mobility analyzers (DMA), which employ an electric field to select certain sized particles. The size selection is based on the electrical mobility of particles, charged with a radioactive source. The separated particles are then transported to condensation particle counters (CPC), where the particles are grown by condensation of butanol. After that the particles can be counted optically by directing them through a laser beam and measuring the scattered radiation. At SMEAR I, the lower cut-off size of the DMPS system was decreased from 8 nm to 3 nm in 2003, and the higher cut-off size increased from 500 nm to 850 nm in 2005. At SMEAR II, the measured size range is from 3 nm to 1 000 nm. Aerosol size distribution data from SMEAR I was used in **Paper II**. In **Papers I–III**, growth and formation rates as well as condensation sink, derived from the number size distribution data (see Sect. 3.3), were utilized. Aerosol total concentration data in **Paper II** was obtained as an integral over the DMPS size distribution data, and additionally from a separate CPC set to measure outdoor air without size selection.

Aerosol optical depth (AOD) and precipitable water (PW) are retrieved from spectral solar and sky radiation measurements by ground-based sun photometer (Cimel) conducted by AERONET (Holben et al., 1998). Version 2, level 2 (quality assured and cloud screened) data was used. AOD and PW were employed in the Solis clear-sky model (see Sect. 3.3.2), which was used in the clear-sky analysis (**Paper I**) and in derivation of parameters for the cloud classification algorithm (**Paper IV**).

3.2.2 Trace gases

The concentration of sulfuric acid in **Paper III** is measured with chemical ionization atmospheric-pressure-interface time-of-flight mass spectrometer (CI-APi-TOF). The instrument applies chemical ionization by nitrate ions NO_3^- to charge the molecular clusters in the sample (Jokinen et al., 2012). The sample ions are guided through a quarupole chamber, which focuses the beam for the time-of-flight mass analyzer. The mass spectrum of the sample is hence calculated based on the time the ions spend in the flight tube, the elapsed time being longer for molecules with a larger mass-to-charge ratio. The time-of-flight mass spectrometers have a high resolving power, which allows

them to identify isobaric compounds, i.e. compounds with a similar nominal mass but with a different molecular formula. The data of CI-APi-TOF is analyzed using tofTools package on MATLAB software (Junninen et al., 2010).

VOC concentrations in **Paper III** are measured with quardrupole proton-transfer reaction mass spectrometer (PTR-MS, Ionicon Analytik GmbH) at SMEAR II (Lindinger and Jordan, 1998). The instrument ionizes water vapor to produce hydronium ions (H_3O^+), which are used for chemical ionization of VOCs via proton transfer from the hydronium ions. The charged VOCs are guided through a quadrupole chamber, and are detected based on their mass-to-charge ratio. Instead of solving the full mass spectrum of the sample, the quadrupole mass spectrometers can only scan over predetermined mass-to-charge ratios. The resolving power of quadrupole mass spectrometers is lower than that of time-of-flight mass spectrometers, and therefore they cannot separate neither isobaric or isomeric (e.g. different monoterpenes) compounds. The PTR-MS at SMEAR II is calibrated regularly as described in Taipale et al. (2008). The instrument measures continuously concentrations of various VOCs from several heights between 4.2 m and 125 m, but in **Paper III** data measured at 16.8 m was used. Because the instrument is additionally used for measuring VOC concentrations from soil and shoot chambers, the atmospheric concentrations are measured only every third hour.

SO_2 concentration, used in sulfuric acid proxy (**Paper III**) and in **Paper II**, is measured with pulsed fluorescence analyzer (Thermo Fisher Scientific, USA) at SMEAR sites and in Beijing. The detection limit of the instrument is 0.1 ppb. Ozone concentration, used for Criegee radical estimation, is measured with UV absorption gas analyzers (Thermo Fisher Scientific, USA) at SMEAR sites and in Beijing (**Paper III**). The detection limit of the instrument is 1 ppb.

3.2.3 Radiation

Global and UVB radiation are measured with pyranometers in energy units ($\mathrm{W\,m^{-2}}$) (**Papers I–IV**). Photosynthetically active radiation (PAR), the part of global radiation relevant for photosynthesis (400–700 nm), is measured with quantum sensors (therefore called also photosynthetic photon flux density, PPFD), and is hence reported in quantum units ($\mu\mathrm{mol\,s^{-1}\,m^{-2}}$). Except for SMEAR II and SMEAR Estonia, at the measurement stations in **Paper I** only either PAR or global radiation was measured. Therefore, to be able to compare global radiation measured by pyranometers and PAR

measured by quantum sensors, we applied quantum efficiency χ for the conversion. It was obtained from the measurements from SMEAR II by dividing observed PAR values with observed global radiation measurements (Ross and Sulev, 2000). The mean value over growing season was $\chi_{\text{glob}} = 2.06\ \mu\text{mol}\,\text{s}^{-1}\,\text{W}^{-1}$, which was then used in the conversion between PAR and global radiation.

3.2.4 Autumnal moths

Population density of autumnal moths in **Paper II** was estimated in three ways: 1) time-relative collection method, 2) collection with a net, and 3) catching with light traps. In time-relative collection a "larval index" was obtained by counting autumnal moth larvae found from mountain birch branches in 10 minutes of search (Klemola et al., 2016). The search was conducted once per year. With the second method, autumnal moth larvae were collected approximately every fifth day by sweeping with a net 100 times from mountain birch branches and another 100 times from bilberry branches on the northern slope of Värriö I fell (Fig. 4a). All the found caterpillars were counted, but autumnal moths is by far the most common species in the area (Hunter et al., 2014). These two methods count the autumnal moth larvae, whereas the third method is based on attracting adult moths into a light trap (Fig. 4b). Light traps consist of bright light (500 W, switched on between 20:00 and 08:00) on top of a funnel which directs nocturnal moths into a catch (Leinonen et al., 1998; Hunter et al., 2014). The caught moths are stored, and later separated to species and counted.

3.2.5 Other measurements

Cloud base height used in **Paper IV** was retrieved from ceilometer data (Vaisala CL31). The ceilometer can detect up to three cloud layers, with a maximum detection height of 7 500 m, from the back-scattering profile of a laser pulse. Fully or partially obscured data points have been removed from the analysis to prevent bias in cloud base height measurements.

Ecosystem scale photosynthesis is quantified by gross primary production (GPP), which describes the amount of carbon fixed by the ecosystem from the atmosphere. GPP in **Paper I** is calculated from total ecosystem respiration (TER) and net ecosystem exchange (NEE), obtained from eddy covariance (SMEAR and Zotino stations) or

Figure 4: Collection of autumnal moths for population density estimation by (a) sweeping with a net, and (b) with Jalas model light trap. Figures courtesy of Veli Pohjonen (a) and Esko Karvinen (b). The figures are found in **Paper II** as Fig. 1c and 1d.

gradient method (Fonovaya station) measurements as

$$\mathrm{GPP} = \mathrm{TER} - \mathrm{NEE} \tag{1}$$

(Kolari et al., 2009). TER is determined from nighttime CO_2 flux partitioning using soil (SMEAR I and II) or air temperature (SMEAR Estonia, Zotino and Fonovaya) (Reichstein et al., 2005; Kolari et al., 2009).

3.3 Derived variables

3.3.1 Growth rate, formation rate, and condensation sink

Growth rate (GR) and formation rate (J) are measures of NPF events. Growth rate describes how fast newly formed particles grow into larger sizes, and formation rate quantifies how fast particles are formed, i.e. the strength of the NPF event, which is why they are used in **Paper II** when assessing the effects of biotic stresses on aerosol processes. Condensation sink in turn characterizes aerosol population and its ability to act as condensing surface for low-volatility vapors that could potentially contribute to NPF. All of these variables are derived from aerosol number size distribution data (Kulmala et al., 2012). NPF event classification in **Paper II** was determined from DMPS data using the method suggested by Dal Maso et al. (2005).

Growth rate is obtained by fitting log-normal distributions to the particle distribution modes, and calculating the geometric mean diameters of the fits. Growth rate is then the slope of line fitted to the mean diameters (Kulmala et al., 2012). Formation rate is the rate at which particles grow through a certain size range:

$$J_{D_\mathrm{P}} = \frac{\mathrm{d}N_{D_\mathrm{P}}}{\mathrm{d}t} + \mathrm{CoagS}_{D_\mathrm{P}} \cdot N_{D_\mathrm{P}} + \frac{\mathrm{GR}}{\Delta D_\mathrm{P}} + S_\mathrm{losses}, \tag{2}$$

where $\mathrm{d}N_{D_\mathrm{P}} / \mathrm{d}t$ describes the time evolution of the particle number concentration, $\mathrm{CoagS}_{D_\mathrm{P}} \cdot N_{D_\mathrm{P}}$ determines the loss of particles due to coagulation with other particles, $\mathrm{GR}/\Delta D_\mathrm{P}$ accounts for growth of particles to larger sizes, and S_losses describes other possible loss terms (Kulmala et al., 2012). D_P refers to the diameter of a particle. In **Paper II** growth and formation rates were calculated for 8–25 nm sized particles.

The condensation sink is calculated as:

$$\mathrm{CS} = 2\pi D_\mathrm{v} \int_0^{D_{\mathrm{P,max}}} D_\mathrm{P} \beta N(D_\mathrm{P}) \mathrm{d}D_\mathrm{v}, \tag{3}$$

where D_v is the diffusion coefficient of the condensing vapor (sulfuric acid), β is Fuchs–Sutugin coefficient, and $N(D_\mathrm{P})$ is the number size distribution of particles (Kulmala et al., 2012). In **Paper III** condensation sink was calculated without correction for hygroscopic growth (Laakso et al., 2004), though for SMEAR II the proxy was additionally derived using corrected condensation sink.

3.3.2 Solis clear-sky model and cloudiness

Solis clear-sky model determines the theoretical intensity of global radiation on a horizontal surface at ground-level in cloud-free conditions:

$$I_\mathrm{gh} = R_\mathrm{g,mod} = I_0' \cdot \exp\left(\frac{-\tau_\mathrm{g}}{\cos^g(\mathrm{SZA})}\right) \cos(\mathrm{SZA}). \tag{4}$$

For direct radiation, the equation becomes:

$$I_\mathrm{dir} = I_0' \cdot \exp\left(\frac{-\tau_\mathrm{b}}{\cos^b(\mathrm{SZA})}\right). \tag{5}$$

I_0' is the AOD and PW modified solar flux density at top of the atmosphere, τ_g and τ_b are the global total and direct optical depths, g and b are AOD and PW related fitting

parameters, and SZA is solar zenith angle (Ineichen, 2008). Diffuse radiation can be modeled as a difference between horizontal and direct irradiances:

$$I_{\mathrm{dh}} = R_{\mathrm{d,mod}} = I_{\mathrm{gh}} - I_{\mathrm{dir}} \cos(\mathrm{SZA}). \tag{6}$$

In **Paper I** clear-sky cases were determined using two conditions: the ratio of measured global radiation to the modeled clear-sky radiation at the top of the atmosphere ($R_{\mathrm{g,meas}}/R_{\mathrm{g,mod}} \geq 0.9$) and the ratio of measured diffuse radiation to the modeled diffuse radiation ($0.8 \leq R_{\mathrm{d,meas}}/R_{\mathrm{d,mod}} \leq 1.2$). Using only global radiation criterion, cloudy points could still interfere the results due to global radiation enhancement occurring in the cloud droplets (Pecenak et al., 2016). In this case, global radiation is close to the theoretical maximum, but diffuse irradiacnce is still increased due to the presence of clouds.

Transmittance and patchiness

The development of the cloud algorithm in **Paper IV** was based on three parameters: measured cloud base height, transmittance and patchiness. Transmittance and patchiness are derived from the Solis clear-sky model and measured global radiation as follows. Transmittance quantifies how much radiation is penetrated through clouds:

$$\mathrm{TR} = \left\langle \frac{R_{\mathrm{g,meas}}}{R_{\mathrm{g,mod}}} \right\rangle_{21\mathrm{min}}. \tag{7}$$

It is determined as a 21 min running average. The length of the time interval comes from empirical experience: a few tens of minutes is a relevant time scale for clouds because then one time interval is not described by only one passing cloud nor is the time so long that the cloud type is changed within one interval (Duchon and O'Malley, 1999).

Patchiness describes the variability in the cloud layer. As an example, it is high for cumulus clouds, which create sharp cloudy–cloud-free patterns. Patchiness is the running standard deviation of scaled measured global radiation:

$$\mathrm{PA} = \sigma \left(\frac{1\,400\ \mathrm{W\,m^{-2}} \cdot R_{\mathrm{g,meas}}}{R_{\mathrm{g,mod}}} \right)_{21\mathrm{min}}. \tag{8}$$

The radiation was scaled because the oscillations in radiation are different in different times of day due to changing amount of incoming radiation. $1\,400\ \mathrm{W\,m^{-2}}$ was selected

as it is slightly higher than the theoretical maximum radiation (Duchon and O'Malley, 1999).

The data by the cloud algorithm is valid for moments when SZA is less than 70°. With larger SZA, the global radiation measurements can be biased, and therefore cannot reliably be used for cloud detection. In case of SMEAR II, the solar zenith angle limit leads to exclusion of wintertime data from the middle of October to the end of February.

3.3.3 Sulfuric acid proxies

Petäjä et al. (2009) formulated the first sulfuric acid proxies based on measurement data from SMEAR II. Their proxies accounted for sulfuric acid formation from the oxidation of sulfur dioxide by OH radicals and loss of sulfuric acid due to condensing on preexisting particles (CS):

$$\frac{\mathrm{d}[\mathrm{H_2SO_4}]}{\mathrm{d}t} = k[\mathrm{SO_2}][\mathrm{OH}] - \mathrm{CS}[\mathrm{H_2SO_4}], \tag{9}$$

where k is an empirically derived scaling factor. The authors confirmed the results by Berresheim et al. (2002) and Rohrer and Berresheim (2006) showing that OH concentration correlates well with UVB and global radiation, and hence in the absence of measurements, OH concentration can be estimated using UVB or global radiation. In **Paper II** we used the UVB radiation based proxy for estimating sulfuric acid concentration in Värriö.

In **Paper III**, we performed a revision of the sulfuric acid proxies, and added terms describing sulfuric acid formation due to oxidation of sulfur dioxide by stabilized Criegee intermediates and loss due to dimer formation. The proxy was derived using global radiation (Fit 1):

$$\frac{\mathrm{d}[\mathrm{H_2SO_4}]}{\mathrm{d}t} = k_1[\mathrm{SO_2}]R_{\mathrm{g,meas}} + k_2[\mathrm{SO_2}][\mathrm{O_3}][Alkenes] - \mathrm{CS}[\mathrm{H_2SO_4}] - k_3[\mathrm{H_2SO_4}]^2. \tag{10}$$

We assumed steady state conditions, meaning that the sources and sinks of sulfuric acid are in balance at short time scales, in which case the left side of Eq. (10) could be assigned to zero. The equation was then solved for sulfuric acid concentration, and thereby an equation for modeled sulfuric acid concentration was reached:

$$[\mathrm{H_2SO_4}]_{\mathrm{proxy}} = -\frac{\mathrm{CS}}{2k_3} + \sqrt{\left(\frac{\mathrm{CS}}{2k_3}\right)^2 + \frac{[\mathrm{SO_2}]}{k_3}(k_1 R_{\mathrm{g,meas}} + k_2[\mathrm{O_3}][Alkene])}. \tag{11}$$

Measured sulfur dioxide, ozone and alkene concentrations, condensation sink and global radiation were given as input for the proxy, which was compared with the measured sulfuric acid concentration to find the fitting coefficients k_1, k_2 and k_3. The search for the optimum values for the fitting coefficients was done by minimizing the sum of squared logarithm of the ratio between the modeled and measured sulfuric acid concentration using MATLAB's built-in function *fminsearch* (Lagarias et al., 1998).

At SMEAR II the most common alkenes are monoterpenes (Rinne et al., 2005; Hellén et al., 2018) and in Beijing alkenes from anthropogenic emissions (Li et al., 2015). Here we used butylene, butadiene, isoprene, pentene, and hexene. In Agia Marina and Budabest VOCs were not measured, and hence for those sites the Criegee term was excluded from the analyses (see Fit 2 below, Eq. 12).

To estimate the importance of Criegee and dimer formation terms in sulfuric acid proxies, three additional proxies without these terms were fitted: 1) a proxy without the Criegee term (Fit 2):

$$\frac{\mathrm{d[H_2SO_4]}}{\mathrm{d}t} = k_1[\mathrm{SO_2}]R_{\mathrm{g,meas}} - \mathrm{CS[H_2SO_4]} - k_3[\mathrm{H_2SO_4}]^2, \tag{12}$$

2) a proxy without the dimer formation term (Fit 3):

$$\frac{\mathrm{d[H_2SO_4]}}{\mathrm{d}t} = k_1[\mathrm{SO_2}]R_{\mathrm{g,meas}} + k_2[\mathrm{SO_2}][\mathrm{O_3}][Alkenes] - \mathrm{CS[H_2SO_4]}, \tag{13}$$

and 3) a proxy without either of those terms (Fit 4):

$$\frac{\mathrm{d[H_2SO_4]}}{\mathrm{d}t} = k_1[\mathrm{SO_2}]R_{\mathrm{g,meas}} - \mathrm{CS[H_2SO_4]}. \tag{14}$$

Additionally, the sulfuric acid proxies by Petäjä et al. (2009) (Eq. 4 therein) and Mikkonen et al. (2011) (Eq. 11 therein) were compared with the formulated proxies.

4 Overview of key results

4.1 Can stress-induced emissions of BVOCs increase the ambient aerosol load?

Organic compounds emitted by forests compose a large fraction of aerosol mass in the boreal region (Tunved et al., 2006; Jimenez et al., 2009). Laboratory and modeling studies investigating biotic stresses have suggested enhanced SOA mass yield from stress-induced emissions of BVOCs, though the magnitude of the increase varies, and some studies have even shown decrease due to alterations in BVOC emission blend (Joutsensaari et al., 2005, 2015; Hamilton et al., 2009; Berg et al., 2013; Mentel et al., 2013; Bergström et al., 2014; Yli-Pirilä et al., 2016; Zhao et al., 2017; Faiola et al., 2018, 2019; Ylisirniö et al., 2020; Taipale et al., 2021). Therefore, in **Paper II**, 25 years of field data comprising several aerosol related variables, temperature, relative humidity, radiation and trace gases was analyzed to determine whether enhanced aerosol processes could be observed during or after intensified biotic stress events. Because VOCs were not measured at the station, the possible elevation in VOC concentration due to stress-induced emissions of BVOCs could not be distinguished directly, but rather we searched for a relation between autumnal moth population density and aerosol variables.

We could not find a clear correlation between the number of NPF event days, aerosol number concentration, or formation and growth rates and autumnal moth population density. However, we found some evidence showing that the aerosol load could be higher a few years after the peak population densities, indicating of a possible delayed induced stress effect of mountain birches (Kaitaniemi et al., 1998; Ruuhola et al., 2007) (Fig. 5). The low correlation between aerosol-derived variables and autumnal moth population density could result from the small biomass of mountain birches at the site combined with relatively low autumnal moth population densities during the study period. Taipale et al. (2021) used a model to study how various biotic stresses affect aerosol particle concentration in different environmental conditions. They found that a mountain birch stand infested by autumnal moths could lead to higher aerosol particle concentration, though the effect is considerably higher in for example oak forest infested by European gypsy moth (*Lymantria dispar*) in central European conditions. The leaf area index (LAI) used in Taipale et al. (2021) for a mountain birch stand

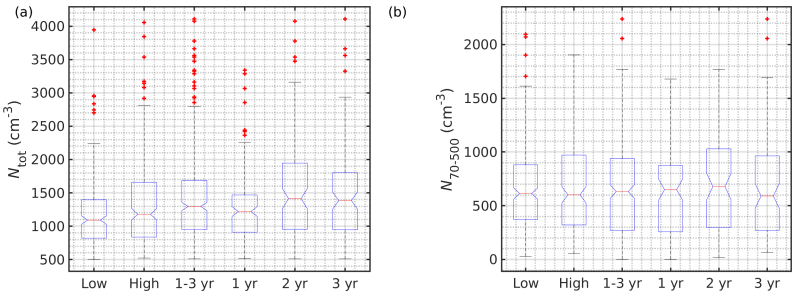

Figure 5: June–July daytime (9:00–17:00 UTC+3) (a) total particle concentration and (b) concentration of CCN sized particles (70–500 nm) from low (Low) and high (High) autumnal moth population density years, from the first three years the peak population densities (1-3 yr), and from those three years separately (1 yr, 2 yr, and 3 yr). The figure is found in **Paper II** as Fig. 8.

was 2 m^2m^{-2}, which is in the higher end of estimated LAI values by Dahlberg et al. (2004, and references therein) (1.44–2.06 m^2m^{-2}) and Heiskanen (2006) (0–2.7 m^2m^{-2}, mean 0.76 m^2m^{-2}), which could at least partly explain the observed increase in aerosol particle concentration in the modeling results.

Organic compounds from vegetation and sulfuric acid are both prominent factors affecting aerosol formation and growth processes. In **Paper II**, the relation between sulfuric acid and total particle concentration with respect to autumnal moth population density was determined to analyze their relation at SMEAR I. We also wanted to ensure there were not such changes in the relation that would affect the conclusions of the data analysis. Sulfuric acid concentration was estimated using the proxy by Petäjä et al. (2009). **Paper III** shows that the proxy underestimates the daytime values at SMEAR II, and the same could be true for the SMEAR I site. However, in **Paper II** the study period comprised only June and July, and therefore almost during the whole measurement time there was constant sunlight which, in theory, could increase the performance of the proxy. On the other hand, the proxy was not originally derived for SMEAR I, and thereby the fitting parameters could be different for the SMEAR I site. In **Paper III**, the parameters for different environments were mostly within a factor of 10 from each other. Furthermore, also the condensation sink is low at the study station which can bias the proxy values (Petäjä et al., 2009). Hence, the low correlation found between sulfuric acid concentration and aerosol total particle concentration could at least partially be explained by the discrepancies in the sulfuric acid

concentration estimates. Clear relation to autumnal moth population density was not detected.

4.2 How do different source and sink terms contribute to the estimated sulfuric acid concentration?

As sulfuric acid is one of the key components in NPF, information of its atmospheric concentration is valuable in research related to atmospheric chemistry and physics. However, the measurements of sulfuric acid concentration are rather sparse, and hence the concentration is often estimated with a proxy, which is an equation derived from the main sources and sinks of sulfuric acid. In **Paper III**, a sulfuric acid proxy accounting for formation of sulfuric acid from oxidation of sulfur dioxide by OH radicals and stabilized Criegee intermediates, and loss by condensing on preexisting particles and dimer formation was formulated. The proxy was tested in different environments, showing good correlation with measured sulfuric acid concentration, and the importance of different source and sink terms were determined.

We found that oxidation by stabilized Criegee intermediates was an important source of sulfuric acid during nighttime (Fig. 6a), and in fact crucial for estimating sulfuric acid concentration during dark hours at least in the environments where we had VOC measurements and formation of Criegee intermediates could be estimated. OH is a short-lived oxidant that is formed in the presence of sunlight. Therefore, OH radicals do not exist during dark hours, and proxies based only on the formation of sulfuric acid from OH oxidation of sulfur dioxide cannot estimate the concentrations of sulfuric acid during dark hours.

Dimer formation was found to be increasingly important when shifting from rural to urban environments (Fig. 6b), resulting probably from high concentrations of sulfuric acid monomers and stabilizing bases (Yan et al., 2018; Yao et al., 2018). At SMEAR II, the relative contribution of the sink terms stayed rather constant throughout the year while in Beijing the strength of the condensation sink term increased during winters, which likely corresponds to the higher particle pollution occurring during winters. On a daily scale, the condensation sink term was more important during nights at both sites highlighting the importance of daytime cluster formation and higher nighttime condensation sink due to boundary layer dynamics (Kulmala et al., 2012). The diur-

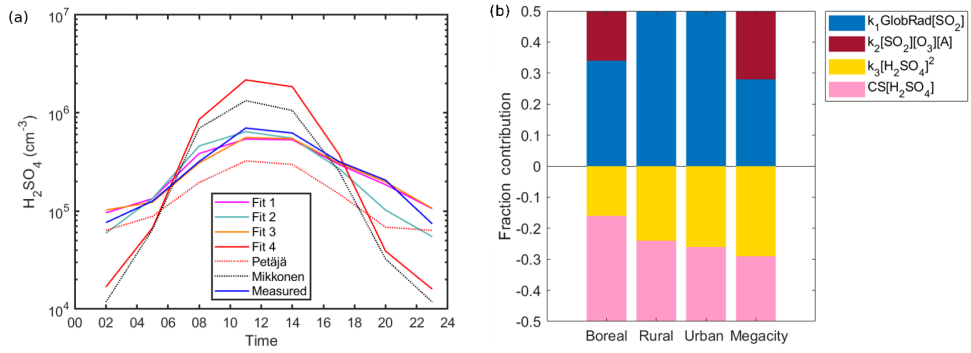

Figure 6: (a) Diurnal variations of measured and predicted sulfuric acid concentrations using different proxies fitted with data from SMEAR II. Calculated condensation sink has been corrected for hygroscopic growth. Fit 1 includes sulfuric acid formation by OH and stabilized Criegee intermediates (terms $k_1 GlobRad[SO_2]$ and $k_2[SO_2][O_3][A]$ in subplot b), and loss by condensation and dimer formation (terms $CS[H_2SO_4]$ and $k_3[H_2SO_4]^2$ in subplot b). Fit 2 does not have Criegee term, Fit 3 dimer formation term, and Fit 4 neither of them. Petäjä refers to proxy by Petäjä et al. (2009), and Mikkonen to proxy by Mikkonen et al. (2011). (b) The relative fractions of different source and sink terms displayed for all measurement stations. Notice that at Agia Marina and Budabest sites VOCs are not measured, and therefore the Criegee intermediate term is not shown. Boreal = SMEAR II, Rural = Agia Marina, Urban = Budabest, and Megacity = Beijing. Figure (a) is found in **Paper III** as Fig. S12, and (b) as Fig. 11a.

nal variation of the source terms displays the importance of daytime radiation driven formation of sulfuric acid due to oxidation by OH radicals, and nighttime formation due to Criegee intermediates, as expected.

Stabilized Criegee intermediates are formed in the ozonolysis of alkenes, which are dominated by BVOCs in the boreal region and anthropogenic VOCs in urban areas. Due to the climate feedback mechanisms, the emissions of BVOCs can be enhanced, whereas due to air pollution control the emissions of anthropogenic VOCs and sulfuric acid diminished, which can then potentially lead to changes in ambient sulfuric acid concentration and subsequently in NPF. Studies have already shown that reduction in sulfuric acid concentration can decrease NPF (e.g. Kyrö et al., 2014), and thereby lead to heating of the planet (Arneth et al., 2009; Makkonen et al., 2012b; Acosta Navarro et al., 2016). Overall, in order to clean the atmosphere safely, it is important to

understand the interactions behind different atmospheric components (Kulmala, 2015; Torkmahalleh et al., 2021; Wei et al., 2021). With proxies separating the sources and sinks of sulfuric acid, also changes in the term contributions can be analyzed and utilized in atmospheric chemistry, air pollution, and NPF related research.

4.3 How is photosynthesis affected by aerosol load and clouds?

Clouds have an important role in Earth's radiative budget since they can interact with both short and long-wave radiation, which impacts directly the temperature and radiative conditions on the Earth (e.g. Kulmala et al., 2013; IPCC, 2021). There is evidence that photosynthesis is enhanced under diffuse radiation conditions, though it is not yet clear what is the contribution of aerosol particles, clouds, or different cloud types on the increment in photosynthesis (Gu et al., 1999, 2002, 2003; Niyogi et al., 2004; Mercado et al., 2009; Cheng et al., 2015, 2016). Moreover, clouds have been associated with NPF and transport of particles from the upper troposphere to the ground level (Perry and Hobbs, 1994; Waddicor et al., 2012; Murphy et al., 2015; Wehner et al., 2015; Lampilahti et al., 2020; Bardakov et al., 2021), which further highlights the importance of the connection between ecosystem and atmosphere processes. In **Paper IV**, we developed an algorithm for cloud type classification. The information of different cloud types can be used in quantifying the effects of different clouds on ecosystem and atmospheric chemical and physical processes (Fig. 1b). In **Paper I**, we studied the quantified the impact of aerosol particles and clouds on radiation and photosynthesis separately.

SMEAR II site is particularly designed for ecosystem–atmosphere relation studies, and hence it is convenient to study also cloud processes there. As there is not continuous total sky imagining at SMEAR II, we developed an inexpensive but effective cloud algorithm based on the existing instruments. In the developing and validation process the cloud type data given by the algorithm was compared with total sky images from Hyytiälä taken during Biogenic Aerosols — Effects on Cloud and Climate (BAECC) campaign in spring and summer 2014 (Petäjä et al., 2016). We found good agreement with the algorithm and observations, overall about 70 % and for one class (nimbostratus) up to 100 %. The least performing cloud types were those having only weak influence on radiative conditions, such as cirrus clouds. In general, the algorithm shows that low level overcasting or patchy clouds are common in southern Finland. The high-

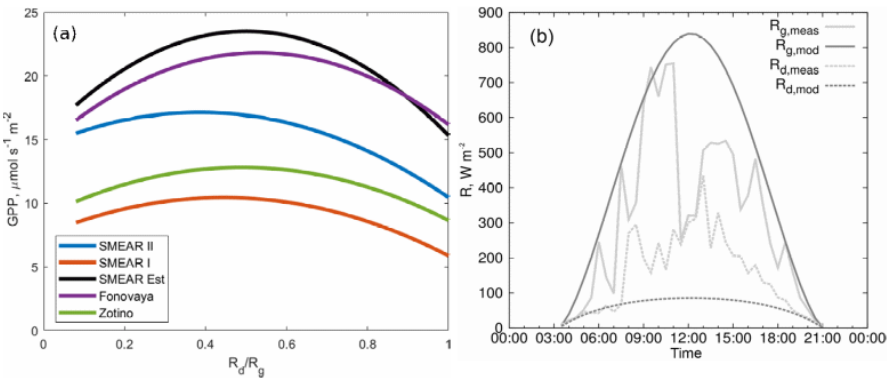

Figure 7: (a) Relation between GPP and the fraction of diffuse radiation from global radiation (R_d/R_g) at different measurement sites. (b) Evolution of measured global ($R_{g,meas}$) and diffuse radiation ($R_{d,meas}$) compared to the modeled values ($R_{g,mod}$ and $R_{d,mod}$, respectively) on a cloudy day at SMEAR Estonia. Figure (a) is found in **Paper I** as Fig. 8, and (b) as Fig. 2b.

est cloud fraction occurs during wintertime with an average of 79 %, and the lowest during summertime with an average of 56 %.

In **Paper I**, we separated clear-sky and cloudy moments based on measured and modeled irradiance at five measurement stations in the boreal and hemiboreal region. When studying the clear-sky conditions, we could describe how aerosol particles affect the radiative conditions and how the radiative conditions affect photosynthesis of the forest ecosystems. Correspondingly, when including also cloudy moments, we could quantify the combined effect of clouds and aerosol particles on radiation and photosynthesis. Both aerosol particles and clouds were found to scatter solar radiation effectively: the diffuse fraction due to aerosol particles varied between 0.1 (low aerosol load) to 0.27 (high aerosol load), and the diffuse fraction in the presence of clouds reached up to 0.35.

Light use efficiency (LUE) can be defined as GPP per unit absorbed PAR (APAR), and thus GPP = LUE $\cdot$ APAR (Cheng et al., 2016). We found that light use efficiency increases with increasing diffuse fraction, and PAR intensity decreases with increasing diffuse fraction, hence leading to a parabolic relation between GPP and the fraction of diffuse radiation, which is presented in Fig. 7 for the five measurement sites. We

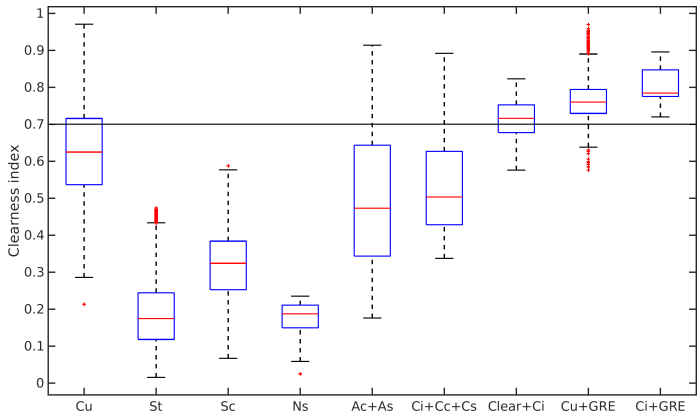

Figure 8: Clearness index values associated with different cloud types. The data is from 2014 and 2016–2017 June–August at 9:00–15:00 (UTC+3). Low clouds: Cu = cumulus, St = stratus, Sc = stratocumulus, and Ns = nimbostratus. Middle clouds: Ac+As = altocumulus and altostratus. High clouds: Ci+Cc+Cs = cirrus, cirrocumulus and cirrostratus. Clear+Ci = clear-sky and cirrus clouds, Cu+GRE = cumulus with GRE, and Ci+GRE = cirrus with GRE. The figure is found in **Paper IV** as Fig. 6.

can see that the optimal diffuse fraction for photosynthesis is about 0.4–0.5 in the boreal and hemiboreal region. The study showed that GPP increased by 6–14 % due to increasing aerosol load, and by 21 % (coniferous forest) to 33 % (mixed forest) in the presence of clouds, showing clearly that forests can benefit from the increasing fraction of diffuse radiation compared to clear-sky and clean atmosphere conditions.

In **Paper I**, we separated clear-sky and cloudy moments by comparing both measured global and diffuse radiation to modeled global and diffuse radiation. In many studies clear-sky moments have been separated using only global radiation criterion (clearness index), i.e. ratio of measured global radiation to the modeled radiation. When the clearness index has been above a certain limit, usually 0.6 or 0.7, the sky has been assumed to be clear from clouds (Kulmala et al., 2014a; Dada et al., 2017). However, as clouds scatter radiation effectively, they can cause global radiation enhancement (GRE), which means that clouds "focus" radiation, leading to cases when the measured global radiation is similar to, or even higher than, the modeled radiation (Pecenak et al., 2016). These moments can falsely be classified as clear-sky conditions when using only the clearness index. Figure 7b shows an example of a cloudy day at SMEAR Estonia where there are moments when the global radiation is close to the modeled radiation

but the diffuse radiation deviating from the modeled diffuse radiation level reveals the presence of clouds. Hence, using both global and diffuse radiation as criteria, clear-sky and cloudy moments can be separated more accurately.

In **Paper IV**, we indeed showed that the conditions, when the clearness index exceeded 0.6 or 0.7, were associated with cumulus, middle level altostratus or altocumulus, or cirrus clouds (Fig. 8) — fair weather clouds which have previously been associated with VOC oxidation, and aerosol formation and transport processes (Perry and Hobbs, 1994; Waddicor et al., 2012; Wehner et al., 2015; Lampilahti et al., 2020). Those cloud types can also be related to more active photosynthesis due to scattering of solar radiation (Cheng et al., 2016; Kivalov and Fitzjarrald, 2019), which is why they can lead to increases in aerosol formation and growth processes via increased emissions of BVOCs.

Figure 9 demonstrates GPP distribution on transmittance–patchiness plane in the presence low clouds. Low patchiness and transmittance values are caused by optically thick and overcasting clouds, such as stratus, and are hence associated with low GPP values. The highest GPP values in Fig. 9 are linked from moderate to high transmittance and patchiness values caused by cumulus clouds. Patchy cumulus clouds are effectively scattering radiation and creating light–shadow patterns that can increase light and water use efficiencies, and thus benefit photosynthesis (Gu et al., 2002; Cheng et al., 2015, 2016; Kivalov and Fitzjarrald, 2019; Artaxo et al., 2022).

Dada et al. (2017) analyzed the environmental circumstances on NPF event and non-event days, and concluded that NPF is more likely to occur under clear-sky. However, they used the clearness index to separate clear-sky and cloudy moments, and therefore these moments could additionally be biased by clouds. On the other hand, the cloud algorithm shows that in spring the fraction of clear-sky and cirrus clouds is high, which could thereby in part explain the higher fraction of event days observed during spring (Dal Maso et al., 2005; Dada et al., 2018). Yet, the possibility of cloud interactions on aerosol processes requires further research.

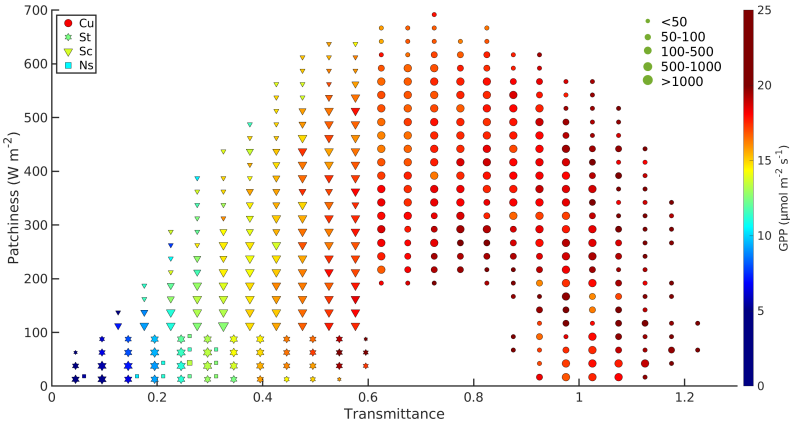

Figure 9: Patchiness as a function of transmittance colored by GPP shown for low clouds (Cu = cumulus, Sc = stratocumulus, St = stratus, and Ns = nimbostratus). Data is from 9:00–15:00 (UTC+3) June–August from years 2014, 2016–2017. The original GPP data in 30 min resolution was linearly interpolated into 1 min resolution to match the time resolution of the cloudiness data. The number of data points was then reduced by dividing data into cells and showing the median transmittance, patchiness and GPP value for each cell. Marker size shows the number of data points taken into account within each cell.

5 Review of papers and the author's contribution

Paper I aims to separate the effects of clouds and aerosol on solar irradiance, and quantify how changes in irrandiance influence GPP in boreal environments. We found that GPP can increase 6–14 % due to direct aerosol effect and by 21–33 % when including also cloud effects. We showed that GPP has a parabolic dependence on the diffuse fraction of solar irradiance, which cannot exceed 0.7 for maximum GPP occurrence. I calculated CS and performed CS–radiation analysis for SMEAR I, and also commented the manuscript.

Paper II studies whether emissions of BVOCs from mountain birches infested by autumnal moth larvae can have an effect on aerosol load at SMEAR I. We could not find a clear correlation between aerosol processes and autumnal moth population density. However, we found some evidence of possible delayed induced defense mechanism where the emissions of induced defense compounds stay on a higher level even for several years after the peak moth densities. For this paper, I performed the data analysis and wrote the paper with input from all co-authors. Additionally, I was part of a team collecting autumnal moths with a net and light traps in summer 2017.

Paper III describes derivation of a new sulfuric acid proxy, which includes two source and sink terms: formation of sulfuric acid due to oxidation of sulfur dioxide by OH and stabilized Criegee intermediates, and loss due to condensing on preexisting particles and formation of dimer clusters. The proxy was tested in several environments with good agreement with measurements. For this paper, I initiated the analysis by developing the first codes for the proxy and analyzing data from SMEAR II. I wrote about the instrumentation at SMEAR II and proxy development, and commented the manuscript.

Paper IV quantifies cloud types and properties over SMEAR II. We developed an algorithm for cloud type estimation based on solar irradiance and cloud base height measurements. Most common cloud types were low level patchy and overcasting clouds such as cumulus, stratocumulus and stratus. The overall performance of the algorithm was good with nearly 70 % agreement with visual inspection. The best performance was reached with cloud types having well-distinguishable effects on solar radiation, such as thick nimbostratus clouds. In this paper, I participated and finalized the algorithm development, analyzed the data, and wrote the paper with contributions from all co-authors.

6 Conclusions and future directions

Previous studies have found evidence of climate feedback mechanisms between ecosystem and atmosphere. The connection implies that the emissions of BVOCs, promoted by activated photosynthesis and increase in temperature due to rising CO_2 concentration, lead to enhanced formation of nanoparticles. The change in emissions of BVOCs may have a contribution also to the production of sulfuric acid, a key component in aerosol formation, because one pathway of sulfuric acid production requires formation of Criegee intermediates from atmospheric alkenes. Some of the nanoparticles formed in the atmosphere may act as seed particles in cloud formation, thereby altering cloud properties, or scatter solar radiation reducing the incoming radiation, and increasing the fraction of diffuse radiation on the Earth. These changes link back to temperature and photosynthetic activity, and therefore to the emissions of BVOCs.

In this thesis, we seek for further insights into the feedback mechanisms between ecosystem and atmosphere. We study the underlying processes and preconditions for aerosol formation by determining the impact of stress-induced emissions of BVOCs on aerosol load, and by quantifying the sources and sinks of sulfuric acid and providing a revised proxy to estimate sulfuric acid concentration. We formulate a cloud type classification algorithm, which can be used in the future to analyze the effects of different clouds on the terrestrial feedback mechanisms by, for example, retrieving the interlinkages between cloud type, caused diffuse radiation, alteration in photosynthesis and BVOC emissions, and aerosol formation. Finally, we quantified the effect of aerosols and clouds on diffuse radiation in the boreal region, and how ecosystem scale photosynthesis was affected by the change in diffuse radiation. The main findings are summarized below.

1) We assessed the effect of biotic stress by autumnal moth larvae feeding on mountain birches on aerosol processes by analyzing 25 years of field data from SMEAR I. We hypothesize that direct enhancement of aerosol processes due to increasing autumnal moth population cannot be observed due to small biomass of the mountain birches and relatively low autumnal moth population densities during the study period. However, we found indications that delayed induced stress effect may cause elevated aerosol load for a few years after peak infestation years. It is important to keep in mind that climate change is altering the environment fast at high latitudes, resulting in higher basal emissions of BVOCs due to rise in air temperature, increasing biomass due to lengthening of growing season, migration of woody plant species to higher latitudes and

altitudes and changes in tree species, and intensifying and more frequent plant stresses. Due to theses changes, it is possible that in the future the stress caused by larval feeding can be detected as increased aerosol load in the northern Fennoscandia. However, more field studies in different environments are required to verify the atmospheric relevance of enhanced SOA formation due to stress-induced plant emissions.

2) We added new source and sink terms to sulfuric acid proxy, and evaluated the importance of different terms on the estimated sulfuric acid concentration in several different environments. We conclude that including formation of sulfuric acid due to oxidation of sulfur dioxide by stabilized Criegee intermediates is crucial, especially during dark hours when sulfuric acid formation is dominated by Criegee intermediate oxidation. Loss of sulfuric acid due to dimer formation is an important sink for sulfuric acid in urban environments where the high concentration of stabilizing bases enhances cluster formation. The revised sulfuric acid proxy improved our previous estimates of the sulfuric acid concentration in the atmosphere, particularly during night and wintertime, and increased our understanding on sulfuric acid formation and loss in different environments.

3) We developed an algorithm to classify different cloud types at SMEAR II based on global radiation and cloud base height measurements. We conclude that the algorithm performs well with overall agreement of nearly 70 % with observations. We show that the fraction of fair weather clouds increases during summer, and could thereby be associated with enhanced photosynthesis and aerosol formation and growth processes. In the future, the role of clouds and different cloud types on various boundary layer processes should be analyzed carefully, since the uncertainty of aerosol–cloud–radiation interactions is high. The cloud algorithm is designed to be a new tool in these types of analyses.

4) We studied the direct aerosol and cloud effect on radiation separately, and quantified the effect of radiation on ecosystem scale photosynthesis at several locations in the boreal and hemiboreal region. We conclude that the fraction of diffuse radiation increases both in the presence of aerosol particles (up to 0.27) and clouds (up to 0.35), and that ecosystem scale photosynthesis is enhanced under diffuse radiation by up to 33 %. The analysis shows that the maximal enhancement in photosynthesis occurs around a diffuse fraction of 0.4–0.5. The study highlights the importance of the relations between atmosphere and ecosystem. In the future, it is relevant to separate the effect of aerosol and clouds, and even cloud types on radiation and their meaning in

different boundary layer processes to deepen our understanding on the various linkages between ecosystem and atmosphere.

This thesis introduces holistically carbon-based terrestrial climate feedback mechanisms. It covers interactions between photosynthesis, emissions of BVOCs, aerosol formation and growth processes, and clouds. The presented results pinpoint the relevance to validate the impacts of stress-induced BVOC emissions on aerosol processes in different environments, particularly accounting for the changes caused by global warming, and the role of different cloud types on aerosol and ecosystem processes. The development of cloud classification algorithm and sulfuric acid proxy aim to promote further research on aerosol–cloud–radiation interactions.

References

Aalto, J., Kolari, P., Hari, P., Kerminen, V.-M., Schiestl-Aalto, P., Aaltonen, H., Levula, J., Siivola, E., Kulmala, M., and Bäck, J. (2014). New foliage growth is a significant, unaccounted source for volatiles in boreal evergreen forests. *Biogeosciences*, 11(5):1331–1344.

Aalto, J., Porcar-Castell, A., Atherton, J., Kolari, P., Pohja, T., Hari, P., Nikinmaa, E., Petäjä, T., and Bäck, J. (2015). Onset of photosynthesis in spring speeds up monoterpene synthesis and leads to emission bursts. *Plant, Cell & Environment*, 38(11):2299–2312.

Aalto, P., Hämeri, K., Becker, E., Weber, R., Salm, J., Mäkelä, J. M., Hoell, C., O'dowd, C. D., Hansson, H.-C., Väkevä, M., Koponen, I., Buzorius, G., and Kulmala, M. (2001). Physical characterization of aerosol particles during nucleation events. *Tellus B: Chemical and Physical Meteorology*, 53(4):344–358.

Acosta Navarro, J. C., Varma, V., Riipinen, I., Seland, Ø., Kirkevåg, A., Struthers, H., Iversen, T., Hansson, H.-C., and Ekman, A. M. L. (2016). Amplification of Arctic warming by past air pollution reductions in Europe. *Nature Geoscience*, 9(4):277–281.

Albrecht, B. A. (1989). Aerosols, Cloud Microphysics, and Fractional Cloudiness. *Science*, 245(4923):1227–1230.

Almeida, J., Schobesberger, S., Kürten, A., Ortega, I. K., Kupiainen-Määttä, O., Praplan, A. P., Adamov, A., Amorim, A., Bianchi, F., Breitenlechner, M., David, A., Dommen, J., Donahue, N. M., Downard, A., Dunne, E., Duplissy, J., Ehrhart, S., Flagan, R. C., Franchin, A., Guida, R., Hakala, J., Hansel, A., Heinritzi, M., Henschel, H., Jokinen, T., Junninen, H., Kajos, M., Kangasluoma, J., Keskinen, H., Kupc, A., Kurtén, T., Kvashin, A. N., Laaksonen, A., Lehtipalo, K., Leiminger, M., Leppä, J., Loukonen, V., Makhmutov, V., Mathot, S., McGrath, M. J., Nieminen, T., Olenius, T., Onnela, A., Petäjä, T., Riccobono, F., Riipinen, I., Rissanen, M., Rondo, L., Ruuskanen, T., Santos, F. D., Sarnela, N., Schallhart, S., Schnitzhofer, R., Seinfeld, J. H., Simon, M., Sipilä, M., Stozhkov, Y., Stratmann, F., Tomé, A., Tröstl, J., Tsagkogeorgas, G., Vaattovaara, P., Viisanen, Y., Virtanen, A., Vrtala, A., Wagner, P. E., Weingartner, E., Wex, H., Williamson, C., Wimmer, D., Ye, P., Yli-Juuti, T., Carslaw, K. S., Kulmala, M., Curtius, J., Baltensperger, U., Worsnop,

D. R., Vehkamäki, H., and Kirkby, J. (2013). Molecular understanding of sulphuric acid-amine particle nucleation in the atmosphere. *Nature*, 502(7471):359–363.

Ammunét, T., Kaukoranta, T., Saikkonen, K., Repo, T., and Klemola, T. (2012). Invading and resident defoliators in a changing climate: cold tolerance and predictions concerning extreme winter cold as a range-limiting factor. *Ecological Entomology*, 37(3):212–220.

Arneth, A., Harrison, S. P., Zaehle, S., Tsigaridis, K., Menon, S., Bartlein, P. J., Feichter, J., Korhola, A., Kulmala, M., O'Donnell, D., Schurgers, G., Sorvari, S., and Vesala, T. (2010). Terrestrial biogeochemical feedbacks in the climate system. *Nature Geoscience*, 3(8):525–532.

Arneth, A., Unger, N., Kulmala, M., and Andreae, M. O. (2009). Clean the Air, Heat the Planet? *Science*, 326(5953):672–673.

Artaxo, P., Hansson, H.-C., Andreae, M., Bäck, J., Alves, E., Barbosa, H., Bender, F., Bourtsoukidis, E., Carbone, S., Chi, J., Decesari, S., Despres, V., Ditas, F., Ezhova, E., Fuzzi, S., Hasselquist, N., Heintzenberg, J., Holanda, B., Guenther, A., Hakolal, H., Heikkinen, L., Kerminen, V.-M., Kontkananen, J., Krejci, R., Kulmala, M., Lavric, J., de Leeuw, G., Lehtipalo, K., Machado, L., McFiggans, G., Franco, M., Meller, B., Morais, F., Mohr, C., Morgan, W., Nilsson, M., Peichl, M., Petäjä, T., Praß, M., Pöhlker, C., Pöhlker, M., Pöschl, U., von Randow, C., Riipinen, I., Rinner, J., Rizzo, L., Rosenfeld, D., Silva Dias, M., Sogecheva, L., Stier, P., Swietlicki, E., Sörgel, M., Tunved, P., Virkkula, A., Wang, J., Weber, B., Yáñez-Serrano, A. M., Zieger, P., Mikhailov, E., Backes Meller, B., Gonçalves Morais, F., Smith, J. N., and Kesselmeier, J. (2022). Tropical and boreal forest - atmosphere interactions: A review. *Tellus B: Chemical and Physical Meteorology*, 74:24–163.

Atkinson, R. and Arey, J. (2003). Gas-phase tropospheric chemistry of biogenic volatile organic compounds: a review. *Atmospheric Environment*, 37:197–219.

Bäck, J., Aalto, J., Henriksson, M., Hakola, H., He, Q., and Boy, M. (2012). Chemodiversity of a Scots pine stand and implications for terpene air concentrations. *Biogeosciences*, 9(2):689–702.

Bardakov, R., Thornton, J. A., Riipinen, I., Krejci, R., and Ekman, A. M. L. (2021). Transport and chemistry of isoprene and its oxidation products in deep convective clouds. *Tellus B: Chemical and Physical Meteorology*, 73(1):1–21.

Bellouin, N., Quaas, J., Gryspeerdt, E., Kinne, S., Stier, P., Watson-Parris, D., Boucher, O., Carslaw, K. S., Christensen, M., Daniau, A.-L., Dufresne, J.-L., Feingold, G., Fiedler, S., Forster, P., Gettelman, A., Haywood, J. M., Lohmann, U., Malavelle, F., Mauritsen, T., McCoy, D. T., Myhre, G., Mülmenstädt, J., Neubauer, D., Possner, A., Rugenstein, M., Sato, Y., Schulz, M., Schwartz, S. E., Sourdeval, O., Storelvmo, T., Toll, V., Winker, D., and Stevens, B. (2020). Bounding Global Aerosol Radiative Forcing of Climate Change. *Reviews of Geophysics*, 58(1):e2019RG000660.

Berg, A. R., Heald, C. L., Huff Hartz, K. E., Hallar, A. G., Meddens, A. J. H., Hicke, J. A., Lamarque, J.-F., and Tilmes, S. (2013). The impact of bark beetle infestations on monoterpene emissions and secondary organic aerosol formation in western North America. *Atmospheric Chemistry and Physics*, 13(6):3149–3161.

Bergström, R., Hallquist, M., Simpson, D., Wildt, J., and Mentel, T. F. (2014). Biotic stress: a significant contributor to organic aerosol in Europe? *Atmospheric Chemistry and Physics*, 14(24):13643–13660.

Berresheim, H., Elste, T., Tremmel, H. G., Allen, A. G., Hansson, H.-C., Rosman, K., Dal Maso, M., Mäkelä, J. M., Kulmala, M., and O'Dowd, C. D. (2002). Gas-aerosol relationships of H2SO4, MSA, and OH: Observations in the coastal marine boundary layer at Mace Head, Ireland. *Journal of Geophysical Research: Atmospheres*, 107(D19):PAR 5–1–PAR 5–12.

Boy, M., Mogensen, D., Smolander, S., Zhou, L., Nieminen, T., Paasonen, P., Plass-Dülmer, C., Sipilä, M., Petäjä, T., Mauldin, L., Berresheim, H., and Kulmala, M. (2013). Oxidation of SO_2 by stabilized Criegee intermediate (sCI) radicals as a crucial source for atmospheric sulfuric acid concentrations. *Atmospheric Chemistry and Physics*, 13(7):3865–3879.

Brauer, M., Freedman, G., Frostad, J., van Donkelaar, A., Martin, R. V., Dentener, F., Dingenen, R. v., Estep, K., Amini, H., Apte, J. S., Balakrishnan, K., Barregard, L., Broday, D., Feigin, V., Ghosh, S., Hopke, P. K., Knibbs, L. D., Kokubo, Y., Liu, Y., Ma, S., Morawska, L., Sangrador, J. L. T., Shaddick, G., Anderson, H. R., Vos, T., Forouzanfar, M. H., Burnett, R. T., and Cohen, A. (2016). Ambient Air Pollution Exposure Estimation for the Global Burden of Disease 2013. *Environmental Science & Technology*, 50(1):79–88.

Bylund, H. (1995). *Long-term interactions between the autumnal moth and mountain*

birch: The roles of resources, competitors, natural enemies, and weather. PhD Thesis, Swedish University of Agricultural Sciences.

Calfapietra, C., Pallozzi, E., Lusini, I., and Velikova, V. (2013). Modification of BVOC Emissions by Changes in Atmospheric [CO2] and Air Pollution. In Niinemets, Ü. and Monson, R. K., editors, *Biology, Controls and Models of Tree Volatile Organic Compound Emissions*, pages 253–284. Springer Netherlands, Dordrecht.

Cheng, S. J., Bohrer, G., Steiner, A. L., Hollinger, D. Y., Suyker, A., Phillips, R. P., and Nadelhoffer, K. J. (2015). Variations in the influence of diffuse light on gross primary productivity in temperate ecosystems. *Agricultural and Forest Meteorology*, 201:98–110.

Cheng, S. J., Steiner, A. L., Hollinger, D. Y., Bohrer, G., and Nadelhoffer, K. J. (2016). Using satellite-derived optical thickness to assess the influence of clouds on terrestrial carbon uptake. *Journal of Geophysical Research: Biogeosciences*, 121(7):1747–1761.

Chmielewski, F.-M. and Rötzer, T. (2001). Response of tree phenology to climate change across Europe. *Agricultural and Forest Meteorology*, 108(2):101–112.

Dada, L., Chellapermal, R., Buenrostro Mazon, S., Paasonen, P., Lampilahti, J., Manninen, H. E., Junninen, H., Petäjä, T., Kerminen, V.-M., and Kulmala, M. (2018). Refined classification and characterization of atmospheric new-particle formation events using air ions. *Atmospheric Chemistry and Physics*, 18(24):17883–17893.

Dada, L., Paasonen, P., Nieminen, T., Buenrostro Mazon, S., Kontkanen, J., Peräkylä, O., Lehtipalo, K., Hussein, T., Petäjä, T., Kerminen, V.-M., Bäck, J., and Kulmala, M. (2017). Long-term analysis of clear-sky new particle formation events and nonevents in Hyytiälä. *Atmospheric Chemistry and Physics*, 17(10):6227–6241.

Dahlberg, U., Berge, T. W., Petersson, H., and Vencatasawmy, C. P. (2004). Modelling biomass and leaf area index in a sub-arctic Scandinavian mountain area. *Scandinavian Journal of Forest Research*, 19(1):60–71.

Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P. P., and Lehtinen, K. E. (2005). Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland. *Boreal Environment Research*, 10(5):323.

Dicke, M. and Baldwin, I. T. (2010). The evolutionary context for herbivore-induced plant volatiles: beyond the 'cry for help'. *Trends in Plant Science*, 15(3):167–175.

Dicke, M., Van Loon, J. J., and Soler, R. (2009). Chemical complexity of volatiles from plants induced by multiple attack. *Nature Chemical Biology*, 5(5):317–324.

Donahue, N. M., Ortega, I. K., Chuang, W., Riipinen, I., Riccobono, F., Schobesberger, S., Dommen, J., Baltensperger, U., Kulmala, M., Worsnop, D. R., and Vehkamäki, H. (2013). How do organic vapors contribute to new-particle formation? *Faraday Discussions*, 165(0):91–104.

Duchon, C. E. and O'Malley, M. S. (1999). Estimating Cloud Type from Pyranometer Observations. *Journal of Applied Meteorology*, 38(1):132 – 141.

Dudareva, N., Negre, F., Nagegowda, D. A., and Orlova, I. (2006). Plant volatiles: recent advances and future perspectives. *Critical Reviews in Plant Sciences*, 25(5):417–440.

Ehn, M., Thornton, J. A., Kleist, E., Sipilä, M., Junninen, H., Pullinen, I., Springer, M., Rubach, F., Tillmann, R., Lee, B., Lopez-Hilfiker, F., Andres, S., Acir, I.-H., Rissanen, M., Jokinen, T., Schobesberger, S., Kangasluoma, J., Kontkanen, J., Nieminen, T., Kurtén, T., Nielsen, L. B., Jørgensen, S., Kjaergaard, H. G., Canagaratna, M., Dal Maso, M., Berndt, T., Petäjä, T., Wahner, A., Kerminen, V.-M., Kulmala, M., Worsnop, D. R., Wildt, J., and Mentel, T. F. (2014). A large source of low-volatility secondary organic aerosol. *Nature*, 506(7489):476–479.

Faiola, C. and Taipale, D. (2020). Impact of insect herbivory on plant stress volatile emissions from trees: a synthesis of quantitative measurements and recommendations for future research. *Atmospheric Environment: X*, 5:100060.

Faiola, C. L., Buchholz, A., Kari, E., Yli-Pirilä, P., Holopainen, J. K., Kivimäenpää, M., Miettinen, P., Worsnop, D. R., Lehtinen, K. E. J., Guenther, A. B., and Virtanen, A. (2018). Terpene Composition Complexity Controls Secondary Organic Aerosol Yields from Scots Pine Volatile Emissions. *Scientific Reports*, 8(1):3053–13.

Faiola, C. L., Jobson, B. T., and VanReken, T. M. (2015). Impacts of simulated herbivory on volatile organic compound emission profiles from coniferous plants. *Biogeosciences*, 12(2):527–547.

Faiola, C. L., Pullinen, I., Buchholz, A., Khalaj, F., Ylisirniö, A., Kari, E., Miettinen, P., Holopainen, J. K., Kivimäenpää, M., Schobesberger, S., Yli-Juuti, T., and Virtanen, A. (2019). Secondary Organic Aerosol Formation from Healthy and Aphid-Stressed Scots Pine Emissions. *ACS Earth and Space Chemistry*, 3(9):1756–1772.

FAO and UNEP (2020). *The State of the World's Forests 2020. Forests, biodiversity and people.* Rome. https://doi.org/10.4060/ca8642en.

Fernández-Martínez, M., Vicca, S., Janssens, I. A., Ciais, P., Obersteiner, M., Bartrons, M., Sardans, J., Verger, A., Canadell, J., Chevallier, F., Wang, X., Bernhofer, C., Curtis, P. S., Gianelle, D., Grünwald, T., Heinesch, B., Ibrom, A., Knohl, A., Laurila, T., Law, B. E., Limousin, J., Longdoz, B., Loustau, D., Mammarella, I., Matteucci, G., Monson, R., Montagnani, L., Moors, E., Munger, J., Papale, D., Piao, S., and Peñuelas, J. (2017). Atmospheric deposition, CO2, and change in the land carbon sink. *Scientific Reports*, 7(9632):1–13.

Fineschi, S. and Loreto, F. (2012). Leaf volatile isoprenoids: an important defensive armament in forest tree species. *Iforest-Biogeosciences and Forestry*, 5(1):13.

Fineschi, S., Loreto, F., Staudt, M., and Peñuelas, J. (2013). Diversification of Volatile Isoprenoid Emissions from Trees: Evolutionary and Ecological Perspectives. In Ninemets, Ü. and Monson, R. K., editors, *Biology, Controls and Models of Tree Volatile Organic Compound Emissions*, pages 1–20. Springer Netherlands, Dordrecht.

Fuentes, J. D., Lerdau, M., Atkinson, R., Baldocchi, D., Bottenheim, J. W., Ciccioli, P., Lamb, B., Geron, C., Gu, L., Guenther, A., Sharkey, T. D., and Stockwell, W. (2000). Biogenic Hydrocarbons in the Atmospheric Boundary Layer: A Review. *Bulletin of the American Meteorological Society*, 81(7):1537 – 1576.

Ghirardo, A., Koch, K., Taipale, R., Zimmer, I., Schnitzler, J.-P., and Rinne, J. (2010). Determination of de novo and pool emissions of terpenes from four common boreal/alpine trees by 13CO2 labelling and PTR-MS analysis. *Plant, Cell & Environment*, 33(5):781–792.

Gryspeerdt, E., Stier, P., and Partridge, D. G. (2014). Satellite observations of cloud regime development: the role of aerosol processes. *Atmospheric Chemistry and Physics*, 14(3):1141–1158.

Gu, L., Baldocchi, D., Verma, S. B., Black, T., Vesala, T., Falge, E. M., and Dowty, P. R. (2002). Advantages of diffuse radiation for terrestrial ecosystem productivity. *Journal of Geophysical Research: Atmospheres*, 107(D6):ACL 2–1 – ACL 2–23.

Gu, L., Baldocchi, D. D., Wofsy, S. C., Munger, J. W., Michalsky, J. J., Urbanski, S. P., and Boden, T. A. (2003). Response of a deciduous forest to the Mount Pinatubo eruption: Enhanced photosynthesis. *Science*, 299(5615):2035–2038.

Gu, L., Fuentes, J. D., Shugart, H. H., Staebler, R. M., and Black, T. A. (1999). Responses of net ecosystem exchanges of carbon dioxide to changes in cloudiness: Results from two North American deciduous forests. *Journal of Geophysical Research: Atmospheres*, 104(D24):31421–31434.

Guenther, A., Hewitt, C. N., Erickson, D., Fall, R., Geron, C., Graedel, T., Harley, P., Klinger, L., Lerdau, M., McKay, W., Pierce, T., Scholes, B., Steinbrecher, R., Tallamraju, R., Taylor, J., and Zimmerman, P. (1995). A global model of natural volatile organic compound emissions. *Journal of Geophysical Research: Atmospheres*, 100(D5):8873–8892.

Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., and Wang, X. (2012). The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions. *Geoscientific Model Development*, 5(6):1471–1492.

Guo, S., Hu, M., Zamora, M. L., Peng, J., Shang, D., Zheng, J., Du, Z., Wu, Z., Shao, M., Zeng, L., Molina, M. J., and Zhang, R. (2014). Elucidating severe urban haze formation in China. *Proceedings of the National Academy of Sciences*, 111(49):17373–17378.

Haapanala, S., Ekberg, A., Hakola, H., Tarvainen, V., Rinne, J., Hellén, H., and Arneth, A. (2009). Mountain birch – potentially large source of sesquiterpenes into high latitude atmosphere. *Biogeosciences*, 6(11):2709–2718.

Hamilton, J. F., Lewis, A. C., Carey, T. J., Wenger, J. C., Borrás i Garcia, E., and Muñoz, A. (2009). Reactive oxidation products promote secondary organic aerosol formation from green leaf volatiles. *Atmospheric Chemistry and Physics*, 9(11):3815–3823.

Hao, L. Q., Yli-Pirilä, P., Tiitta, P., Romakkaniemi, S., Vaattovaara, P., Kajos, M. K., Rinne, J., Heijari, J., Kortelainen, A., Miettinen, P., Kroll, J. H., Holopainen, J. K., Smith, J. N., Joutsensaari, J., Kulmala, M., Worsnop, D. R., and Laaksonen, A. (2009). New particle formation from the oxidation of direct emissions of pine seedlings. *Atmospheric Chemistry and Physics*, 9(20):8121–8137.

Hari, P., Nikinmaa, E., Pohja, T., Siivola, E., Bäck, J., Vesala, T., and Kulmala, M. (2013). Station for Measuring Ecosystem-Atmosphere Relations: SMEAR. In Hari, P., Heliövaara, K., and Kulmala, L., editors, *Physical and Physiological Forest Ecology*, pages 471–487. Springer Netherlands, Dordrecht.

Harley, P. C. (2013). The Roles of Stomatal Conductance and Compound Volatility in Controlling the Emission of Volatile Organic Compounds from Leaves. In Niinemets, Ü. and Monson, R. K., editors, *Biology, Controls and Models of Tree Volatile Organic Compound Emissions*, pages 181–208. Springer Netherlands, Dordrecht.

Haukioja, E. (2003). Putting the insect into the birch–insect interaction. *Oecologia*, 136(2):161–168.

Haukioja, E., Neuvonen, S., Hanhimäki, S., and Niemelä, P. (1988). The Autumnal Moth in Fennoscandia. In Berryman, A. A., editor, *Dynamics of Forest Insect Populations: Patterns, Causes, Implications*, pages 163–178. Springer US, Boston, MA.

Haywood, J. M., Abel, S. J., Barrett, P. A., Bellouin, N., Blyth, A., Bower, K. N., Brooks, M., Carslaw, K., Che, H., Coe, H., Cotterell, M. I., Crawford, I., Cui, Z., Davies, N., Dingley, B., Field, P., Formenti, P., Gordon, H., de Graaf, M., Herbert, R., Johnson, B., Jones, A. C., Langridge, J. M., Malavelle, F., Partridge, D. G., Peers, F., Redemann, J., Stier, P., Szpek, K., Taylor, J. W., Watson-Parris, D., Wood, R., Wu, H., and Zuidema, P. (2021). The CLoud–Aerosol–Radiation Interaction and Forcing: Year 2017 (CLARIFY-2017) measurement campaign. *Atmospheric Chemistry and Physics*, 21(2):1049–1084.

Heikkinen, L., Äijälä, M., Riva, M., Luoma, K., Dällenbach, K., Aalto, J., Aalto, P., Aliaga, D., Aurela, M., Keskinen, H., Makkonen, U., Rantala, P., Kulmala, M., Petäjä, T., Worsnop, D., and Ehn, M. (2020). Long-term sub-micrometer aerosol chemical composition in the boreal forest: inter- and intra-annual variability. *Atmospheric Chemistry and Physics*, 20(5):3151–3180.

Heinritzi, M., Dada, L., Simon, M., Stolzenburg, D., Wagner, A. C., Fischer, L., Ahonen, L. R., Amanatidis, S., Baalbaki, R., Baccarini, A., Bauer, P. S., Baumgartner, B., Bianchi, F., Brilke, S., Chen, D., Chiu, R., Dias, A., Dommen, J., Duplissy, J., Finkenzeller, H., Frege, C., Fuchs, C., Garmash, O., Gordon, H., Granzin, M., El Haddad, I., He, X., Helm, J., Hofbauer, V., Hoyle, C. R., Kangasluoma, J., Keber, T., Kim, C., Kürten, A., Lamkaddam, H., Laurila, T. M., Lampilahti, J., Lee, C. P., Lehtipalo, K., Leiminger, M., Mai, H., Makhmutov, V., Manninen, H. E., Marten, R., Mathot, S., Mauldin, R. L., Mentler, B., Molteni, U., Müller, T., Nie, W., Nieminen, T., Onnela, A., Partoll, E., Passananti, M., Petäjä, T., Pfeifer, J., Pospisilova, V., Quéléver, L. L. J., Rissanen, M. P., Rose, C., Schobesberger, S., Scholz, W., Scholze, K., Sipilä, M., Steiner, G., Stozhkov, Y., Tauber, C., Tham, Y. J., Vazquez-Pufleau, M., Virtanen, A., Vogel, A. L., Volkamer, R., Wagner, R., Wang, M., Weitz, L., Wimmer, D., Xiao, M., Yan, C., Ye, P., Zha, Q., Zhou, X., Amorim, A., Baltensperger, U., Hansel, A., Kulmala, M., Tomé, A., Winkler, P. M., Worsnop, D. R., Donahue, N. M., Kirkby, J., and Curtius, J. (2020). Molecular understanding of the suppression of new-particle formation by isoprene. *Atmospheric Chemistry and Physics*, 20(20):11809–11821.

Heiskanen, J. (2006). Estimating aboveground tree biomass and leaf area index in a mountain birch forest using ASTER satellite data. *International Journal of Remote Sensing*, 27(6):1135–1158.

Hellén, H., Praplan, A. P., Tykkä, T., Ylivinkka, I., Vakkari, V., Bäck, J., Petäjä, T., Kulmala, M., and Hakola, H. (2018). Long-term measurements of volatile organic compounds highlight the importance of sesquiterpenes for the atmospheric chemistry of a boreal forest. *Atmospheric Chemistry and Physics*, 18(19):13839–13863.

Holben, B. N., Eck, T. F., Slutsker, I., Tanré, D., Buis, J. P., Setzer, A., Vermote, E., Reagan, J. A., Kaufman, Y. J., Nakajima, T., Lavenu, F., Jankowiak, I., and Smirnov, A. (1998). AERONET—A Federated Instrument Network and Data Archive for Aerosol Characterization. *Remote Sensing of Environment*, 66(1):1–16.

Holopainen, J. (2011). Can forest trees compensate for stress-generated growth losses by induced production of volatile compounds? *Tree Physiology*, 31:1356–1377.

Holopainen, J. K. (2004). Multiple functions of inducible plant volatiles. *Trends in Plant Science*, 9(11):529–533.

Holopainen, J. K. and Blande, J. D. (2013). Where do herbivore-induced plant volatiles go? *Frontiers in Plant Science*, 4:185.

Holopainen, J. K. and Gershenzon, J. (2010). Multiple stress factors and the emission of plant VOCs. *Trends in Plant Science*, 15(3):176–184.

Holopainen, J. K., Virjamo, V., Ghimire, R. P., Blande, J. D., Julkunen-Tiitto, R., and Kivimäenpää, M. (2018). Climate Change Effects on Secondary Compounds of Forest Trees in the Northern Hemisphere. *Frontiers in Plant Science*, 9:1445.

Hunter, M. D., Kozlov, M. V., Itämies, J., Pulliainen, E., Bäck, J., Kyrö, E.-M., and Niemelä, P. (2014). Current temporal trends in moth abundance are counter to predicted effects of climate change in an assemblage of subarctic forest moths. *Global Change Biology*, 20(6):1723–1737.

Illingworth, A. J., Hogan, R. J., O'Connor, E. J., Bouniol, D., Brooks, M. E., Delanoé, J., Donovan, D. P., Eastment, J. D., Gaussiat, N., Goddard, J. W. F., Haeffelin, M., Baltink, H. K., Krasnov, O. A., Pelon, J., Piriou, J.-M., Protat, A., Russchenberg, H. W. J., Seifert, A., Tompkins, A. M., Zadelhoff, G.-J. v., Vinit, F., Willén, U., Wilson, D. R., and Wrench, C. L. (2007). Cloudnet: Continuous Evaluation of Cloud Profiles in Seven Operational Models Using Ground-Based Observations. *Bulletin of the American Meteorological Society*, 88(6):883 – 898.

Ineichen, P. (2008). A broadband simplified version of the Solis clear sky model. *Solar Energy*, 82(8):758–762.

IPCC (2021). *Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prevot, A. S. H., Zhang, Q., Kroll, J. H., DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., Aiken, A. C., Docherty, K. S., Ulbrich, I. M., Grieshop, A. P., Robinson, A. L., Duplissy, J., Smith, J. D., Wilson, K. R., Lanz, V. A., Hueglin, C., Sun, Y. L., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J. M., Collins, D. R., Cubison, M. J., E., Dunlea, J., Huffman, J. A., Onasch, T. B., Alfarra, M. R., Williams, P. I., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A.,

Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J. Y., Zhang, Y. M., Dzepina, K., Kimmel, J. R., Sueper, D., Jayne, J. T., Herndon, S. C., Trimborn, A. M., Williams, L. R., Wood, E. C., Middlebrook, A. M., Kolb, C. E., Baltensperger, U., and Worsnop, D. R. (2009). Evolution of Organic Aerosols in the Atmosphere. *Science*, 326(5959):1525–1529.

Jokinen, T., Lehtipalo, K., Thakur, R. C., Ylivinkka, I., Neitola, K., Sarnela, N., Laitinen, T., Kulmala, M., Petäjä, T., and Sipilä, M. (2022). Measurement report: Long-term measurements of aerosol precursor concentrations in the Finnish subarctic boreal forest. *Atmospheric Chemistry and Physics*, 22(4):2237–2254.

Jokinen, T., Sipilä, M., Junninen, H., Ehn, M., Lönn, G., Hakala, J., Petäjä, T., Mauldin III, R. L., Kulmala, M., and Worsnop, D. R. (2012). Atmospheric sulphuric acid and neutral cluster measurements using CI-APi-TOF. *Atmospheric Chemistry and Physics*, 12(9):4117–4125.

Joutsensaari, J., Loivamäki, M., Vuorinen, T., Miettinen, P., Nerg, A.-M., Holopainen, J. K., and Laaksonen, A. (2005). Nanoparticle formation by ozonolysis of inducible plant volatiles. *Atmospheric Chemistry and Physics*, 5(6):1489–1495.

Joutsensaari, J., Yli-Pirilä, P., Korhonen, H., Arola, A., Blande, J. D., Heijari, J., Kivimäenpää, M., Mikkonen, S., Hao, L., Miettinen, P., Lyytikäinen-Saarenmaa, P., Faiola, C. L., Laaksonen, A., and Holopainen, J. K. (2015). Biotic stress accelerates formation of climate-relevant aerosols in boreal forests. *Atmospheric Chemistry and Physics*, 15(21):12139–12157.

Jump, A. S., Mátyás, C., and Peñuelas, J. (2009). The altitude-for-latitude disparity in the range retractions of woody species. *Trends in Ecology & Evolution*, 24(12):694–701.

Junninen, H., Ehn, M., Petäjä, T., Luosujärvi, L., Kotiaho, T., Kostiainen, R., Rohner, U., Gonin, M., Fuhrer, K., Kulmala, M., and Worsnop, D. R. (2010). A high-resolution mass spectrometer to measure atmospheric ion composition. *Atmospheric Measurement Techniques*, 3(4):1039–1053.

Kaitaniemi, P. and Ruohomäki, K. (1999). Effects of autumn temperature and oviposition date on timing of larval development and risk of parasitism in a spring folivore. *Oikos*, 84(3):435–442.

Kaitaniemi, P., Ruohomäki, K., and Haukioja, E. (1997). Consequences of defoliation on phenological interaction between Epirrita autumnata and its host plant, mountain birch. *Functional Ecology*, 11(2):199–208.

Kaitaniemi, P., Ruohomäki, K., Ossipov, V., Haukioja, E., and Pihlaja, K. (1998). Delayed induced changes in the biochemical composition of host plant leaves during an insect outbreak. *Oecologia*, 116(1):182–190.

Kari, E., Faiola, C. L., Isokääntä, S., Miettinen, P., Yli-Pirilä, P., Buchholz, A., Kivimäenpää, M., Mikkonen, S., Holopainen, J. K., and Virtanen, A. (2019). Time-resolved characterization of biotic stress emissions from Scots pines being fed upon by pine weevil by means of PTR-ToF-MS. *Boreal Environment Research*, 24:25–49.

Kerminen, V.-M., Chen, X., Vakkari, V., Petäjä, T., Kulmala, M., and Bianchi, F. (2018). Atmospheric new particle formation and growth: review of field observations. *Environmental Research Letters*, 13(10):103003.

Kerminen, V.-M., Paramonov, M., Anttila, T., Riipinen, I., Fountoukis, C., Korhonen, H., Asmi, E., Laakso, L., Lihavainen, H., Swietlicki, E., Svenningsson, B., Asmi, A., Pandis, S. N., Kulmala, M., and Petäjä, T. (2012). Cloud condensation nuclei production associated with atmospheric nucleation: a synthesis based on existing literature and new results. *Atmospheric Chemistry and Physics*, 12(24):12037–12059.

Kesselmeier, J. and Staudt, M. (1999). Biogenic volatile organic compounds (VOC): an overview on emission, physiology and ecology. *Journal of Atmospheric Chemistry*, 33(1):23–88.

Kiendler-Scharr, A., Andres, S., Bachner, M., Behnke, K., Broch, S., Hofzumahaus, A., Holland, F., Kleist, E., Mentel, T. F., Rubach, F., Springer, M., Steitz, B., Tillmann, R., Wahner, A., Schnitzler, J.-P., and Wildt, J. (2012). Isoprene in poplar emissions: effects on new particle formation and OH concentrations. *Atmospheric Chemistry and Physics*, 12(2):1021–1030.

Kiendler-Scharr, A., Wildt, J., Dal Maso, M., Hohaus, T., Kleist, E., Mentel, T. F., Tillmann, R., Uerlings, R., Schurr, U., and Wahner, A. (2009). New particle formation in forests inhibited by isoprene emissions. *Nature*, 461(7262):381–384.

Kivalov, S. N. and Fitzjarrald, D. R. (2019). Observing the whole-canopy short-term dynamic response to natural step changes in incident light: Characteristics of tropical and temperate forests. *Boundary-Layer Meteorology*, 173(1):1–52.

Kivimäenpää, M., Ghimire, R. P., Sutinen, S., Häikiö, E., Kasurinen, A., Holopainen, T., and Holopainen, J. K. (2016). Increases in volatile organic compound emissions of Scots pine in response to elevated ozone and warming are modified by herbivory and soil nitrogen availability. *European Journal of Forest Research*, 135(2):343–360.

Kleist, E., Mentel, T. F., Andres, S., Bohne, A., Folkers, A., Kiendler-Scharr, A., Rudich, Y., Springer, M., Tillmann, R., and Wildt, J. (2012). Irreversible impacts of heat on the emissions of monoterpenes, sesquiterpenes, phenolic BVOC and green leaf volatiles from several tree species. *Biogeosciences*, 9(12):5111–5123.

Klemola, T., Andersson, T., and Ruohomäki, K. (2016). No regulatory role for adult predation in cyclic population dynamics of the autumnal moth, Epirrita autumnata. *Ecological Entomology*, 41(5):582–589.

Kolari, P., Kulmala, L., Pumpanen, J., Launiainen, S., Ilvesniemi, H., Hari, P., and Nikinmaa, E. (2009). CO2 exchange and component CO2 fluxes of a boreal Scots pine forest. *Boreal Environment Research*, 14:761–783.

Koski, T.-M., Laaksonen, T., Mäntylä, E., Ruuskanen, S., Li, T., Girón-Calva, P. S., Huttunen, L., Blande, J. D., Holopainen, J. K., and Klemola, T. (2015). Do Insectivorous Birds use Volatile Organic Compounds from Plants as Olfactory Foraging Cues? Three Experimental Tests. *Ethology*, 121(12):1131–1144.

Kreuzwieser, J. and Rennenberg, H. (2013). Flooding-Driven Emissions from Trees. In Niinemets, Ü. and Monson, R. K., editors, *Biology, Controls and Models of Tree Volatile Organic Compound Emissions*, pages 237–252. Springer Netherlands, Dordrecht.

Kulmala, M. (2015). Atmospheric chemistry: China's choking cocktail. *Nature*, 526(7574):497–499.

Kulmala, M., Ezhova, E., Kalliokoski, T., Noe, S., Vesala, T., Lohila, A., Liski, J., Makkonen, R., Bäck, J., Petäjä, T., and Kerminen, V.-M. (2020). CarbonSink+—Accounting for multiple climate feedbacks from forests. *Boreal Environment Research*, 25:145–159.

Kulmala, M., Nieminen, T., Chellapermal, R., Makkonen, R., Bäck, J., and Kerminen, V.-M. (2013). Climate feedbacks linking the increasing atmospheric CO2 concentration, BVOC emissions, aerosols and clouds in forest ecosystems. In Niinemets,

Ü. and Monson, R. K., editors, *Biology, controls and models of tree volatile organic compound emissions*, pages 489–508. Springer Netherlands, Dordrecht.

Kulmala, M., Nieminen, T., Nikandrova, A., Lehtipalo, K., Manninen, H. E., Kajos, M. K., Kolari, P., Lauri, A., Petäjä, T., Krejci, R., Hansson, H.-C., Swietlicki, E., Lindroth, A., Christensen, T. R., Arneth, A., Hari, P., Bäck, J., Vesala, T., and Kerminen, V.-M. (2014a). CO2-induced terrestrial climate feedback mechanism: From carbon sink to aerosol source and back. *Boreal Environment Research*, 19(suppl. B):122–131.

Kulmala, M., Petäjä, T., Ehn, M., Thornton, J., Sipilä, M., Worsnop, D., and Kerminen, V.-M. (2014b). Chemistry of atmospheric nucleation: on the recent advances on precursor characterization and atmospheric cluster composition in connection with atmospheric new particle formation. *Annual Review of Physical Chemistry*, 65:21–37.

Kulmala, M., Petäjä, T., Nieminen, T., Sipilä, M., Manninen, H. E., Lehtipalo, K., Dal Maso, M., Aalto, P. P., Junninen, H., Paasonen, P., Riipinen, I., Lehtinen, K. E. J., Laaksonen, A., and Kerminen, V.-M. (2012). Measurement of the nucleation of atmospheric aerosol particles. *Nature Protocols*, 7(9):1651–1667.

Kulmala, M., Suni, T., Lehtinen, K. E. J., Dal Maso, M., Boy, M., Reissell, A., Rannik, Ü., Aalto, P., Keronen, P., Hakola, H., Bäck, J., Hoffmann, T., Vesala, T., and Hari, P. (2004). A new feedback mechanism linking forests, aerosols, and climate. *Atmospheric Chemistry and Physics*, 4(2):557–562.

Kyrö, E.-M., Väänänen, R., Kerminen, V.-M., Virkkula, A., Petäjä, T., Asmi, A., Dal Maso, M., Nieminen, T., Juhola, S., Shcherbinin, A., Riipinen, I., Lehtipalo, K., Keronen, P., Aalto, P. P., Hari, P., and Kulmala, M. (2014). Trends in new particle formation in eastern Lapland, Finland: effect of decreasing sulfur emissions from Kola Peninsula. *Atmospheric Chemistry and Physics*, 14(9):4383–4396.

Laakso, L., Petäjä, T., Lehtinen, K. E. J., Kulmala, M., Paatero, J., Hõrrak, U., Tammet, H., and Joutsensaari, J. (2004). Ion production rate in a boreal forest based on ion, particle and radiation measurements. *Atmospheric Chemistry and Physics*, 4(7):1933–1943.

Lagarias, J. C., Reeds, J. A., Wright, M. H., and Wright, P. E. (1998). Convergence Properties of the Nelder–Mead Simplex Method in Low Dimensions. *SIAM Journal on Optimization*, 9(1):112–147.

Lampilahti, J., Manninen, H. E., Leino, K., Väänänen, R., Manninen, A., Buenrostro Mazon, S., Nieminen, T., Leskinen, M., Enroth, J., Bister, M., Zilitinkevich, S., Kangasluoma, J., Järvinen, H., Kerminen, V.-M., Petäjä, T., and Kulmala, M. (2020). Roll vortices induce new particle formation bursts in the planetary boundary layer. *Atmospheric Chemistry and Physics*, 20(20):11841–11854.

Laothawornkitkul, J., Taylor, J. E., Paul, N. D., and Hewitt, C. N. (2009). Biogenic volatile organic compounds in the Earth system. *New Phytologist*, 183(1):27–51.

Law, K. S. and Stohl, A. (2007). Arctic air pollution: Origins and impacts. *Science*, 315(5818):1537–1540.

Lehmann, P., Ammunét, T., Barton, M., Battisti, A., Eigenbrode, S. D., Jepsen, J. U., Kalinkat, G., Neuvonen, S., Niemelä, P., Terblanche, J. S., Økland, B., and Björkman, C. (2020). Complex responses of global insect pests to climate warming. *Frontiers in Ecology and the Environment*, 18(3):141–150.

Lehtipalo, K., Yan, C., Dada, L., Bianchi, F., Xiao, M., Wagner, R., Stolzenburg, D., Ahonen, L. R., Amorim, A., Baccarini, A., Bauer, P. S., Baumgartner, B., Bergen, A., Bernhammer, A.-K., Breitenlechner, M., Brilke, S., Buchholz, A., Buenrostro Mazon, S., Chen, D., Chen, X., Dias, A., Dommen, J., Draper, D. C., Duplissy, J., Ehn, M., Finkenzeller, H., Fischer, L., Frege, C., Fuchs, C., Garmash, O., Gordon, H., Hakala, J., He, X., Heikkinen, L., Heinritzi, M., Helm, J. C., Hofbauer, V., Hoyle, C. R., Jokinen, T., Kangasluoma, J., Kerminen, V.-M., Kim, C., Kirkby, J., Kontkanen, J., Kürten, A., Lawler, M. J., Mai, H., Mathot, S., Mauldin, 3rd, R. L., Molteni, U., Nichman, L., Nie, W., Nieminen, T., Ojdanic, A., Onnela, A., Passananti, M., Petäjä, T., Piel, F., Pospisilova, V., Quéléver, L. L. J., Rissanen, M. P., Rose, C., Sarnela, N., Schallhart, S., Schuchmann, S., Sengupta, K., Simon, M., Sipilä, M., Tauber, C., Tomé, A., Tröstl, J., Väisänen, O., Vogel, A. L., Volkamer, R., Wagner, A. C., Wang, M., Weitz, L., Wimmer, D., Ye, P., Ylisirniö, A., Zha, Q., Carslaw, K. S., Curtius, J., Donahue, N. M., Flagan, R. C., Hansel, A., Riipinen, I., Virtanen, A., Winkler, P. M., Baltensperger, U., Kulmala, M., and Worsnop, D. R. (2018). Multicomponent new particle formation from sulfuric acid, ammonia, and biogenic vapors. *Science Aadvances*, 4(12):eaau5363.

Leinonen, R., Söderman, G., Itämies, J., Rytkönen, S., and Rutanen, I. (1998). Intercalibration of different light-traps and bulbs used in moth monitoring in northern Europe. *Entomologica Fennica*, 9(1):37–51.

Li, L., Xie, S., Zeng, L., Wu, R., and Li, J. (2015). Characteristics of volatile organic compounds and their role in ground-level ozone formation in the Beijing-Tianjin-Hebei region, China. *Atmospheric Environment*, 113:247–254.

Li, T., Holst, T., Michelsen, A., and Rinnan, R. (2019). Amplification of plant volatile defence against insect herbivory in a warming Arctic tundra. *Nature Plants*, 5(6):568–574.

Li, Z. and Sharkey, T. D. (2013). Molecular and Pathway Controls on Biogenic Volatile Organic Compound Emissions. In Niinemets, Ü. and Monson, R. K., editors, *Biology, Controls and Models of Tree Volatile Organic Compound Emissions*, pages 119–151. Springer Netherlands, Dordrecht.

Lindinger, W. and Jordan, A. (1998). Proton-transfer-reaction mass spectrometry (PTR–MS): on-line monitoring of volatile organic compounds at pptv levels. *Chemical Society Reviews*, 27(5):347–375.

Liu, Y., Piao, S., Gasser, T., Ciais, P., Yang, H., Wang, H., Keenan, T. F., Huang, M., Wan, S., Song, J., Wang, K., Janssens, I. A., Peñuelas, J., Huntingford, C., Wang, X., Altaf Arain, M., Fang, Y., Fisher, J. B., Huang, M., Huntzinger, D. N., Ito, A., Jain, A. K., Mao, J., Michalak, A. M., Peng, C., Poulter, B., Schwalm, C., Shi, X., Tian, H., Wei, Y., Zeng, N., Zhu, Q., and Wang, T. (2019). Field-experiment constraints on the enhancement of the terrestrial carbon sink by CO2 fertilization. *Nature Geoscience*, 12(10):809–814.

Loreto, F. and Schnitzler, J.-P. (2010). Abiotic stresses and induced BVOCs. *Trends in Plant Science*, 15(3):154–166.

Lu, Y., Yan, C., Fu, Y., Chen, Y., Liu, Y., Yang, G., Wang, Y., Bianchi, F., Chu, B., Zhou, Y., Yin, R., Baalbaki, R., Garmash, O., Deng, C., Wang, W., Liu, Y., Petäjä, T., Kerminen, V.-M., Jiang, J., Kulmala, M., and Wang, L. (2019). A proxy for atmospheric daytime gaseous sulfuric acid concentration in urban Beijing. *Atmospheric Chemistry and Physics*, 19(3):1971–1983.

Maja, M. M., Kasurinen, A., Yli-Pirilä, P., Joutsensaari, J., Klemola, T., Holopainen, T., and Holopainen, J. K. (2014). Contrasting responses of silver birch VOC emissions to short- and long-term herbivory. *Tree Physiology*, 34(3):241–252.

Makkonen, R., Asmi, A., Kerminen, V.-M., Boy, M., Arneth, A., Guenther, A., and Kulmala, M. (2012a). BVOC-aerosol-climate interactions in the global aerosol-climate model ECHAM5.5-HAM2. *Atmospheric Chemistry and Physics*, 12(21):10077–10096.

Makkonen, R., Asmi, A., Kerminen, V.-M., Boy, M., Arneth, A., Hari, P., and Kulmala, M. (2012b). Air pollution control and decreasing new particle formation lead to strong climate warming. *Atmospheric Chemistry and Physics*, 12(3):1515–1524.

Mäntylä, E., Alessio, G. A., Blande, J. D., Heijari, J., Holopainen, J. K., Laaksonen, T., Piirtola, P., and Klemola, T. (2008). From Plants to Birds: Higher Avian Predation Rates in Trees Responding to Insect Herbivory. *PLOS ONE*, 3(7):1–8.

Mauldin III, R., Berndt, T., Sipilä, M., Paasonen, P., Petäjä, T., Kim, S., Kurtén, T., Stratmann, F., Kerminen, V.-M., and Kulmala, M. (2012). A new atmospherically relevant oxidant of sulphur dioxide. *Nature*, 488(7410):193–196.

McFiggans, G., Mentel, T. F., Wildt, J., Pullinen, I., Kang, S., Kleist, E., Schmitt, S., Springer, M., Tillmann, R., Wu, C., Zhao, D., Hallquist, M., Faxon, C., Le Breton, M., Hallquist, Å. M., Simpson, D., Bergström, R., Jenkin, M. E., Ehn, M., Thornton, J. A., Alfarra, M. R., Bannan, T. J., Percival, C. J., Priestley, M., Topping, D., and Kiendler-Scharr, A. (2019). Secondary organic aerosol reduced by mixture of atmospheric vapours. *Nature*, 565(7741):587–593.

Mentel, T. F., Kleist, E., Andres, S., Dal Maso, M., Hohaus, T., Kiendler-Scharr, A., Rudich, Y., Springer, M., Tillmann, R., Uerlings, R., Wahner, A., and Wildt, J. (2013). Secondary aerosol formation from stress-induced biogenic emissions and possible climate feedbacks. *Atmospheric Chemistry and Physics*, 13(17):8755–8770.

Mercado, L. M., Bellouin, N., Sitch, S., Boucher, O., Huntingford, C., Wild, M., and Cox, P. M. (2009). Impact of changes in diffuse radiation on the global land carbon sink. *Nature*, 458(7241):1014–1017.

Merikanto, J., Spracklen, D. V., Mann, G. W., Pickering, S. J., and Carslaw, K. S. (2009). Impact of nucleation on global CCN. *Atmospheric Chemistry and Physics*, 9(21):8601–8616.

Mikkonen, S., Romakkaniemi, S., Smith, J. N., Korhonen, H., Petäjä, T., Plass-Duelmer, C., Boy, M., McMurry, P. H., Lehtinen, K. E. J., Joutsensaari, J., Hamed,

A., Mauldin III, R. L., Birmili, W., Spindler, G., Arnold, F., Kulmala, M., and Laaksonen, A. (2011). A statistical proxy for sulphuric acid concentration. *Atmospheric Chemistry and Physics*, 11(21):11319–11334.

Miresmailli, S., Gries, R., Gries, G., Zamar, R. H., and Isman, M. B. (2012). Population density and feeding duration of cabbage looper larvae on tomato plants alter the levels of plant volatile emissions. *Pest Management Science*, 68(1):101–107.

Mogensen, D., Gierens, R., Crowley, J. N., Keronen, P., Smolander, S., Sogachev, A., Nölscher, A. C., Zhou, L., Kulmala, M., Tang, M. J., Williams, J., and Boy, M. (2015). Simulations of atmospheric OH, O_3 and NO_3 reactivities within and above the boreal forest. *Atmospheric Chemistry and Physics*, 15(7):3909–3932.

Mogensen, D., Smolander, S., Sogachev, A., Zhou, L., Sinha, V., Guenther, A., Williams, J., Nieminen, T., Kajos, M. K., Rinne, J., Kulmala, M., and Boy, M. (2011). Modelling atmospheric OH-reactivity in a boreal forest ecosystem. *Atmospheric Chemistry and Physics*, 11(18):9709–9719.

Monson, R. K. (2013). Metabolic and Gene Expression Controls on the Production of Biogenic Volatile Organic Compounds. In Niinemets, Ü. and Monson, R. K., editors, *Biology, Controls and Models of Tree Volatile Organic Compound Emissions*, pages 153–179. Springer Netherlands, Dordrecht.

Morison, J. I. L. and Lawlor, D. W. (1999). Interactions between increasing CO2 concentration and temperature on plant growth. *Plant, Cell & Environment*, 22(6):659–682.

Murphy, B. N., Julin, J., Riipinen, I., and Ekman, A. M. L. (2015). Organic aerosol processing in tropical deep convective clouds: Development of a new model (CRM-ORG) and implications for sources of particle number. *Journal of Geophysical Research (Atmospheres)*, 120(19):10,441–10,464.

Nieminen, T., Asmi, A., Dal Maso, M., Aalto, P., Keronen, P., Petäjä, T., Kulmala, M., and Kerminen, V. (2014). Trends in atmospheric new-particle formation: 16 years of observations in a boreal-forest environment. *Boreal Environment Research*, 19(Suppl. B):191–214.

Niinemets, Ü. (2010a). Mild versus severe stress and BVOCs: thresholds, priming and consequences. *Trends in Plant Science*, 15(3):145–153.

Niinemets, Ü. (2010b). Responses of forest trees to single and multiple environmental stresses from seedlings to mature plants: past stress history, stress interactions, tolerance and acclimation. *Forest Ecology and Management*, 260(10):1623–1639.

Niinemets, Ü., Kännaste, A., and Copolovici, L. (2013). Quantitative patterns between plant volatile emissions induced by biotic stresses and the degree of damage. *Frontiers in Plant Science*, 4:262.

Niyogi, D., Chang, H.-I., Saxena, V. K., Holt, T., Alapaty, K., Booker, F., Chen, F., Davis, K. J., Holben, B., Matsui, T., Meyers, T., Oechel, W. C., Pielke Sr., R. A., Wells, R., Wilson, K., and Xue, Y. (2004). Direct observations of the effects of aerosol loading on net ecosystem CO2 exchanges over different landscapes. *Geophysical Research Letters*, 31(20).

Olenius, T., Yli-Juuti, T., Elm, J., Kontkanen, J., and Riipinen, I. (2018). New Particle Formation and Growth: Creating a New Atmospheric Phase Interface. In Faust, J. A. and House, J. E., editors, *Physical Chemistry of Gas-Liquid Interfaces*, Developments in Physical & Theoretical Chemistry, pages 315–352. Elsevier.

Paasonen, P., Asmi, A., Petäjä, T., Kajos, M. K., Äijälä, M., Junninen, H., Holst, T., Abbatt, J. P., Arneth, A., Birmili, W., van der Gon, H. D., Hamed, A., Hoffer, A., Laakso, L., Laaksonen, A., Leaitch, W. R., Plass-Dülmer, C., Pryor, S. C., Räisänen, P., Swietlicki, E., Wiedensohler, A., Worsnop, D. R., Kerminen, V.-M., and Kulmala, M. (2013). Warming-induced increase in aerosol number concentration likely to moderate climate change. *Nature Geoscience*, 6(6):438–442.

Patoulias, D., Fountoukis, C., Riipinen, I., and Pandis, S. N. (2015). The role of organic condensation on ultrafine particle growth during nucleation events. *Atmospheric Chemistry and Physics*, 15(11):6337–6350.

Pecenak, Z. K., Mejia, F. A., Kurtz, B., Evan, A., and Kleissl, J. (2016). Simulating irradiance enhancement dependence on cloud optical depth and solar zenith angle. *Solar Energy*, 136:675–681.

Peñuelas, J. and Boada, M. (2003). A global change-induced biome shift in the Montseny mountains (NE Spain). *Global Change Biology*, 9(2):131–140.

Peñuelas, J. and Staudt, M. (2010). BVOCs and global change. *Trends in Plant Science*, 15(3):133–144.

Peräkylä, O., Riva, M., Heikkinen, L., Quéléver, L., Roldin, P., and Ehn, M. (2020). Experimental investigation into the volatilities of highly oxygenated organic molecules (HOMs). *Atmospheric Chemistry and Physics*, 20(2):649–669.

Peräkylä, O., Vogt, M., Tikkanen, O., Laurila, T., Kajos, M., Rantala, P., Patokoski, J., Aalto, J., Yli-Juuti, T., Ehn, M., Sipilä, M., Paasonen, P., Rissanen, M., Nieminen, T., Taipale, R., Keronen, P., Lappalainen, H. K., Ruuskanen, T. M., Rinne, J., Kerminen, V.-M., Kulmala, M., Bäck, J., and Petäjä, T. (2014). Monoterpenes' oxidation capacity and rate over a boreal forest: temporal variation and connection to growth of newly formed particles. *Boreal Environment Research*, 19(Suppl. B):293–310.

Percival, C. J., Welz, O., Eskola, A. J., Savee, J. D., Osborn, D. L., Topping, D. O., Lowe, D., Utembe, S. R., Bacak, A., M c Figgans, G., Cooke, M. C., Xiao, P., Archibald †, A. T., Jenkin, M. E., Derwent, R. G., Riipinen, I., Mok, D. W. K., Lee, E. P. F., Dyke, J. M., Taatjes, C. A., and Shallcross, D. E. (2013). Regional and global impacts of Criegee intermediates on atmospheric sulphuric acid concentrations and first steps of aerosol formation. *Faraday Discuss.*, 165(0):45–73.

Perry, K. D. and Hobbs, P. V. (1994). Further evidence for particle nucleation in clear air adjacent to marine cumulus clouds. *Journal of Geophysical Research: Atmospheres*, 99(D11):22803–22818.

Petäjä, T., Mauldin III, R., Kosciuch, E., McGrath, J., Nieminen, T., Paasonen, P., Boy, M., Adamov, A., Kotiaho, T., and Kulmala, M. (2009). Sulfuric acid and OH concentrations in a boreal forest site. *Atmospheric Chemistry and Physics*, 9(19):7435–7448.

Petäjä, T., O'Connor, E. J., Moisseev, D., Sinclair, V. A., Manninen, A. J., Väänänen, R., Lerber, A. v., Thornton, J. A., Nicoll, K., Petersen, W., Chandrasekar, V., Smith, J. N., Winkler, P. M., Krüger, O., Hakola, H., Timonen, H., Brus, D., Laurila, T., Asmi, E., Riekkola, M.-L., Mona, L., Massoli, P., Engelmann, R., Komppula, M., Wang, J., Kuang, C., Bäck, J., Virtanen, A., Levula, J., Ritsche, M., and Hickmon, N. (2016). BAECC: A Field Campaign to Elucidate the Impact of Biogenic Aerosols on Clouds and Climate. *Bulletin of the American Meteorological Society*, 97(10):1909 – 1928.

Possell, M. and Loreto, F. (2013). The Role of Volatile Organic Compounds in Plant Resistance to Abiotic Stresses: Responses and Mechanisms. In Niinemets, Ü. and

Monson, R. K., editors, *Biology, Controls and Models of Tree Volatile Organic Compound Emissions*, pages 209–235. Springer Netherlands, Dordrecht.

Potosnak, M. J., LeStourgeon, L., and Nunez, O. (2014). Increasing leaf temperature reduces the suppression of isoprene emission by elevated CO2 concentration. *Science of The Total Environment*, 481:352–359.

Pureswaran, D. S., De Grandpré, L., Paré, D., Taylor, A., Barrette, M., Morin, H., Régnière, J., and Kneeshaw, D. D. (2015). Climate-induced changes in host tree–insect phenology may drive ecological state-shift in boreal forests. *Ecology*, 96(6):1480–1491.

Qiao, X., Yan, C., Li, X., Guo, Y., Yin, R., Deng, C., Li, C., Nie, W., Wang, M., Cai, R., Huang, D., Wang, Z., Yao, L., Worsnop, D. R., Bianchi, F., Liu, Y., Donahue, N. M., Kulmala, M., and Jiang, J. (2021). Contribution of Atmospheric Oxygenated Organic Compounds to Particle Growth in an Urban Environment. *Environmental Science & Technology*, 55(20):13646–13656.

Räisänen, T., Ryyppö, A., and Kellomäki, S. (2008). Effects of elevated CO2 and temperature on monoterpene emission of Scots pine (Pinus sylvestris L.). *Atmospheric Environment*, 42(18):4160–4171.

Rantala, P., Aalto, J., Taipale, R., Ruuskanen, T. M., and Rinne, J. (2015). Annual cycle of volatile organic compound exchange between a boreal pine forest and the atmosphere. *Biogeosciences*, 12(19):5753–5770.

Rap, A., Scott, C., Reddington, C., Mercado, L., Ellis, R., Garraway, S., Evans, M., Beerling, D., MacKenzie, A., Hewitt, C., and Spracklen, D. (2018). Enhanced global primary production by biogenic aerosol via diffuse radiation fertilization. *Nature Geoscience*, 11(9):640–644.

Reichstein, M., Falge, E., Baldocchi, D., Papale, D., Aubinet, M., Berbigier, P., Bernhofer, C., Buchmann, N., Gilmanov, T., Granier, A., Grünwald, T., Havránková, K., Ilvesniemi, H., Janous, D., Knohl, A., Laurila, T., Lohila, A., Loustau, D., Matteucci, G., Meyers, T., Miglietta, F., Ourcival, J.-M., Pumpanen, J., Rambal, S., Rotenberg, E., Sanz, M., Tenhunen, J., Seufert, G., Vaccari, F., Vesala, T., Yakir, D., and Valentini, R. (2005). On the separation of net ecosystem exchange into assimilation and ecosystem respiration: review and improved algorithm. *Global Change Biology*, 11(9):1424–1439.

Riccobono, F., Schobesberger, S., Scott, C. E., Dommen, J., Ortega, I. K., Rondo, L., Almeida, J., Amorim, A., Bianchi, F., Breitenlechner, M., David, A., Downard, A., Dunne, E. M., Duplissy, J., Ehrhart, S., Flagan, R. C., Franchin, A., Hansel, A., Junninen, H., Kajos, M., Keskinen, H., Kupc, A., Kürten, A., Kvashin, A. N., Laaksonen, A., Lehtipalo, K., Makhmutov, V., Mathot, S., Nieminen, T., Onnela, A., Petäjä, T., Praplan, A. P., Santos, F. D., Schallhart, S., Seinfeld, J. H., Sipilä, M., Spracklen, D. V., Stozhkov, Y., Stratmann, F., Tomé, A., Tsagkogeorgas, G., Vaattovaara, P., Viisanen, Y., Vrtala, A., Wagner, P. E., Weingartner, E., Wex, H., Wimmer, D., Carslaw, K. S., Curtius, J., Donahue, N. M., Kirkby, J., Kulmala, M., Worsnop, D. R., and Baltensperger, U. (2014). Oxidation products of biogenic emissions contribute to nucleation of atmospheric particles. *Science*, 344(6185):717–721.

Rieksta, J., Li, T., Junker, R. R., Jepsen, J. U., Ryde, I., and Rinnan, R. (2020). Insect Herbivory Strongly Modifies Mountain Birch Volatile Emissions. *Frontiers in Plant Science*, 11:1671.

Riipinen, I., Pierce, J. R., Yli-Juuti, T., Nieminen, T., Häkkinen, S., Ehn, M., Junninen, H., Lehtipalo, K., Petäjä, T., Slowik, J., Chang, R., Shantz, N. C., Abbatt, J., Leaitch, W. R., Kerminen, V.-M., Worsnop, D. R., Pandis, S. N., Donahue, N. M., and Kulmala, M. (2011). Organic condensation: a vital link connecting aerosol formation to cloud condensation nuclei (CCN) concentrations. *Atmospheric Chemistry and Physics*, 11(8):3865–3878.

Riipinen, I., Rastak, N., and Pandis, S. N. (2015). Connecting the solubility and CCN activation of complex organic aerosols: a theoretical study using solubility distributions. *Atmospheric Chemistry and Physics*, 15(11):6305–6322.

Riipinen, I., Yli-Juuti, T., Pierce, J. R., Petäjä, T., Worsnop, D. R., Kulmala, M., and Donahue, N. M. (2012). The contribution of organics to atmospheric nanoparticle growth. *Nature Geoscience*, 5(7):453–458.

Rinne, J., Bäck, J., and Hakola, H. (2009). Biogenic volatile organic compound emissions from the Eurasian taiga: current knowledge and future directions. *Boreal Environment Research*, 14:807–826.

Rinne, J., Ruuskanen, T. M., Reissell, A., Taipale, R., Hakola, H., and Kulmala, M. (2005). On-line PTR-MS measurements of atmospheric concentrations of volatile

organic compounds in a European boreal forest ecosystem. *Boreal Environment Research*, 10:425–436.

Rohrer, F. and Berresheim, H. (2006). Strong correlation between levels of tropospheric hydroxyl radicals and solar ultraviolet radiation. *Nature*, 442(7099):184–187.

Rose, C., Rissanen, M. P., Iyer, S., Duplissy, J., Yan, C., Nowak, J. B., Colomb, A., Dupuy, R., He, X.-C., Lampilahti, J., Tham, Y. J., Wimmer, D., Metzger, J.-M., Tulet, P., Brioude, J., Planche, C., Kulmala, M., and Sellegri, K. (2021). Investigation of several proxies to estimate sulfuric acid concentration under volcanic plume conditions. *Atmospheric Chemistry and Physics*, 21(6):4541–4560.

Rosenfeld, D., Andreae, M. O., Asmi, A., Chin, M., de Leeuw, G., Donovan, D. P., Kahn, R., Kinne, S., Kivekäs, N., Kulmala, M., Lau, W., Schmidt, K. S., Suni, T., Wagner, T., Wild, M., and Quaas, J. (2014). Global observations of aerosol-cloud-precipitation-climate interactions. *Reviews of Geophysics*, 52(4):750–808.

Ross, J. and Sulev, M. (2000). Sources of errors in measurements of PAR. *Agricultural and Forest Meteorology*, 100(2):103–125.

Ruuhola, T., Salminen, J.-P., Haviola, S., Yang, S., and Rantala, M. J. (2007). Immunological memory of mountain birches: effects of phenolics on performance of the autumnal moth depend on herbivory history of trees. *Journal of Chemical Ecology*, 33(6):1160–1176.

Schneider, S. H. and Dennett, R. D. (1975). Climatic barriers to long-term energy growth. *Ambio*, pages 65–74.

Scott, C., Arnold, S., Monks, S., Asmi, A., Paasonen, P., and Spracklen, D. (2018). Substantial large-scale feedbacks between natural aerosols and climate. *Nature Geoscience*, 11(1):44–48.

Scott, C. E., Rap, A., Spracklen, D. V., Forster, P. M., Carslaw, K. S., Mann, G. W., Pringle, K. J., Kivekäs, N., Kulmala, M., Lihavainen, H., and Tunved, P. (2014). The direct and indirect radiative effects of biogenic secondary organic aerosol. *Atmospheric Chemistry and Physics*, 14(1):447–470.

Simon, M., Dada, L., Heinritzi, M., Scholz, W., Stolzenburg, D., Fischer, L., Wagner, A. C., Kürten, A., Rörup, B., He, X.-C., Almeida, J., Baalbaki, R., Baccarini, A., Bauer, P. S., Beck, L., Bergen, A., Bianchi, F., Bräkling, S., Brilke, S., Caudillo, L.,

Chen, D., Chu, B., Dias, A., Draper, D. C., Duplissy, J., El-Haddad, I., Finkenzeller, H., Frege, C., Gonzalez-Carracedo, L., Gordon, H., Granzin, M., Hakala, J., Hofbauer, V., Hoyle, C. R., Kim, C., Kong, W., Lamkaddam, H., Lee, C. P., Lehtipalo, K., Leiminger, M., Mai, H., Manninen, H. E., Marie, G., Marten, R., Mentler, B., Molteni, U., Nichman, L., Nie, W., Ojdanic, A., Onnela, A., Partoll, E., Petäjä, T., Pfeifer, J., Philippov, M., Quéléver, L. L. J., Ranjithkumar, A., Rissanen, M. P., Schallhart, S., Schobesberger, S., Schuchmann, S., Shen, J., Sipilä, M., Steiner, G., Stozhkov, Y., Tauber, C., Tham, Y. J., Tomé, A. R., Vazquez-Pufleau, M., Vogel, A. L., Wagner, R., Wang, M., Wang, D. S., Wang, Y., Weber, S. K., Wu, Y., Xiao, M., Yan, C., Ye, P., Ye, Q., Zauner-Wieczorek, M., Zhou, X., Baltensperger, U., Dommen, J., Flagan, R. C., Hansel, A., Kulmala, M., Volkamer, R., Winkler, P. M., Worsnop, D. R., Donahue, N. M., Kirkby, J., and Curtius, J. (2020). Molecular understanding of new-particle formation from α-pinene between -50 and +25 C. *Atmospheric Chemistry and Physics*, 20(15):9183–9207.

Sipilä, M., Jokinen, T., Berndt, T., Richters, S., Makkonen, R., Donahue, N. M., Mauldin III, R. L., Kurtén, T., Paasonen, P., Sarnela, N., Ehn, M., Junninen, H., Rissanen, M. P., Thornton, J., Stratmann, F., Herrmann, H., Worsnop, D. R., Kulmala, M., Kerminen, V.-M., and Petäjä, T. (2014). Reactivity of stabilized Criegee intermediates (sCIs) from isoprene and monoterpene ozonolysis toward SO_2 and organic acids. *Atmospheric Chemistry and Physics*, 14(22):12143–12153.

Sipilä, M., Sarnela, N., Neitola, K., Laitinen, T., Kemppainen, D., Beck, L., Duplissy, E.-M., Kuittinen, S., Lehmusjärvi, T., Lampilahti, J., Kerminen, V.-M., Lehtipalo, K., Aalto, P. P., Keronen, P., Siivola, E., Rantala, P. A., Worsnop, D. R., Kulmala, M., Jokinen, T., and Petäjä, T. (2021). Wintertime subarctic new particle formation from Kola Peninsula sulfur emissions. *Atmospheric Chemistry and Physics*, 21(23):17559–17576.

Sizov, O., Ezhova, E., Tsymbarovich, P., Soromotin, A., Prihod'ko, N., Petäjä, T., Zilitinkevich, S., Kulmala, M., Bäck, J., and Köster, K. (2021). Fire and vegetation dynamics in northwest Siberia during the last 60 years based on high-resolution remote sensing. *Biogeosciences*, 18(1):207–228.

Sporre, M. K., Blichner, S. M., Karset, I. H. H., Makkonen, R., and Berntsen, T. K. (2019). BVOC–aerosol–climate feedbacks investigated using NorESM. *Atmospheric Chemistry and Physics*, 19(7):4763–4782.

Spracklen, D. V., Bonn, B., and Carslaw, K. S. (2008). Boreal forests, aerosols and the impacts on clouds and climate. *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*, 366(1885):4613–4626.

Stolzenburg, D., Simon, M., Ranjithkumar, A., Kürten, A., Lehtipalo, K., Gordon, H., Ehrhart, S., Finkenzeller, H., Pichelstorfer, L., Nieminen, T., He, X.-C., Brilke, S., Xiao, M., Amorim, A., Baalbaki, R., Baccarini, A., Beck, L., Bräkling, S., Caudillo Murillo, L., Chen, D., Chu, B., Dada, L., Dias, A., Dommen, J., Duplissy, J., El Haddad, I., Fischer, L., Gonzalez Carracedo, L., Heinritzi, M., Kim, C., Koenig, T. K., Kong, W., Lamkaddam, H., Lee, C. P., Leiminger, M., Li, Z., Makhmutov, V., Manninen, H. E., Marie, G., Marten, R., Müller, T., Nie, W., Partoll, E., Petäjä, T., Pfeifer, J., Philippov, M., Rissanen, M. P., Rörup, B., Schobesberger, S., Schuchmann, S., Shen, J., Sipilä, M., Steiner, G., Stozhkov, Y., Tauber, C., Tham, Y. J., Tomé, A., Vazquez-Pufleau, M., Wagner, A. C., Wang, M., Wang, Y., Weber, S. K., Wimmer, D., Wlasits, P. J., Wu, Y., Ye, Q., Zauner-Wieczorek, M., Baltensperger, U., Carslaw, K. S., Curtius, J., Donahue, N. M., Flagan, R. C., Hansel, A., Kulmala, M., Lelieveld, J., Volkamer, R., Kirkby, J., and Winkler, P. M. (2020). Enhanced growth rate of atmospheric particles from sulfuric acid. *Atmospheric Chemistry and Physics*, 20(12):7359–7372.

Taipale, D., Kerminen, V.-M., Ehn, M., Kulmala, M., and Niinemets, Ü. (2021). Modelling the influence of biotic plant stress on atmospheric aerosol particle processes throughout a growing season. *Atmospheric Chemistry and Physics*, 21(23):17389–17431.

Taipale, R., Kajos, M. K., Patokoski, J., Rantala, P., Ruuskanen, T. M., and Rinne, J. (2011). Role of de novo biosynthesis in ecosystem scale monoterpene emissions from a boreal Scots pine forest. *Biogeosciences*, 8(8):2247–2255.

Taipale, R., Ruuskanen, T. M., Rinne, J., Kajos, M. K., Hakola, H., Pohja, T., and Kulmala, M. (2008). Technical Note: Quantitative long-term measurements of VOC concentrations by PTR-MS - measurement, calibration, and volume mixing ratio calculation methods. *Atmospheric Chemistry and Physics*, 8(22):6681–6698.

Tapakis, R. and Charalambides, A. (2013). Equipment and methodologies for cloud detection and classification: A review. *Solar Energy*, 95:392–430.

Tape, K., Sturm, M., and Racine, C. (2006). The evidence for shrub expansion in Northern Alaska and the Pan-Arctic. *Global Change Biology*, 12(4):686–702.

Tenow, O. and Bylund, H. (2000). Recovery of a Betula pubescens forest in northern Sweden after severe defoliation by Epirrita autumnata. *Journal of Vegetation Science*, 11(6):855–862.

Tharammal, T., Bala, G., Devaraju, N., and Nemani, R. (2019). A review of the major drivers of the terrestrial carbon uptake: model-based assessments, consensus, and uncertainties. *Environmental Research Letters*, 14(9):093005.

Tingey, D. T., Turner, D. P., and Weber, J. A. (1991). Factors Controlling the Emissions of Monoterpenes and Other Volatile Organic Compounds. In Sharkey, T. D., Holland, E. A., and Mooney, H. A., editors, *Trace Gas Emissions by Plants*, Physiological Ecology, pages 93–119. Academic Press, San Diego.

Torkmahalleh, M. A., Akhmetvaliyeva, Z., Omran, A. D., Omran, F. F. D., Kazemitabar, M., Naseri, M., Naseri, M., Sharifi, H., Malekipirbazari, M., Adotey, E. K., Gorjinezhad, S., Eghtesadi, N., Sabanov, S., Alastuey, A., Andrade, M. d. F., Buonanno, G., Carbone, S., Cárdenas-Fuentes, D. E., Cassee, F. R., Dai, Q., Henríquez, A., Hopke, P. K., Keronen, P., Khwaja, H. A., Kim, J., Kulmala, M., Kumar, P., Kushta, J., Kuula, J., Massagué, J., Mitchell, T., Mooibroek, D., Morawska, L., Niemi, J. V., Ngagine, S. H., Norman, M., Oyama, B., Oyola, P., Öztürk, F., Petäjä, T., Querol, X., Rashidi, Y., Reyes, F., Ross-Jones, M., Salthammer, T., Savvides, C., Stabile, L., Sjöberg, K., Söderlund, K., Raman, R. S., Timonen, H., Umezawa, M., Viana, M., and Xie, S. (2021). Global Air Quality and COVID-19 Pandemic: Do We Breathe Cleaner Air? *Aerosol and Air Quality Research*, 21(4):200567.

Trowbridge, A. M. and Stoy, P. C. (2013). BVOC-Mediated Plant-Herbivore Interactions. In Niinemets, Ü. and Monson, R. K., editors, *Biology, Controls and Models of Tree Volatile Organic Compound Emissions*, pages 21–46. Springer Netherlands, Dordrecht.

Tunved, P., Hansson, H.-C., Kerminen, V.-M., Ström, J., Dal Maso, M., Lihavainen, H., Viisanen, Y., Aalto, P. P., Komppula, M., and Kulmala, M. (2006). High Natural Aerosol Loading over Boreal Forests. *Science*, 312(5771):261–263.

Twomey, S. (1977). The Influence of Pollution on the Shortwave Albedo of Clouds. *Journal of Atmospheric Sciences*, 34(7):1149 – 1152.

Vakkari, V., Tiitta, P., Jaars, K., Croteau, P., Beukes, J. P., Josipovic, M., Kerminen, V.-M., Kulmala, M., Venter, A. D., van Zyl, P. G., Worsnop, D. R., and

Laakso, L. (2015). Reevaluating the contribution of sulfuric acid and the origin of organic compounds in atmospheric nanoparticle growth. *Geophysical Research Letters*, 42(23):10,486–10,493.

Vanhatalo, A., Ghirardo, A., Juurola, E., Schnitzler, J.-P., Zimmer, I., Hellén, H., Hakola, H., and Bäck, J. (2018). Long-term dynamics of monoterpene synthase activities, monoterpene storage pools and emissions in boreal Scots pine. *Biogeosciences*, 15(16):5047–5060.

Vehkamäki, H. and Riipinen, I. (2012). Thermodynamics and kinetics of atmospheric aerosol particle formation and growth. *Chem. Soc. Rev.*, 41(15):5160–5173.

Vickers, C. E., Gershenzon, J., Lerdau, M. T., and Loreto, F. (2009). A unified mechanism of action for volatile isoprenoids in plant abiotic stress. *Nature Chemical Biology*, 5(5):283–291.

Vuorinen, T., Nerg, A.-M., Syrjälä, L., Peltonen, P., and Holopainen, J. K. (2007). Epirrita autumnata induced VOC emission of silver birch differ from emission induced by leaf fungal pathogen. *Arthropod-Plant Interactions*, 1(3):159.

Waddicor, D. A., Vaughan, G., Choularton, T. W., Bower, K. N., Coe, H., Gallagher, M., Williams, P. I., Flynn, M., Volz-Thomas, A., Pätz, H.-W., Isaac, P., Hacker, J., Arnold, F., Schlager, H., and Whiteway, J. A. (2012). Aerosol observations and growth rates downwind of the anvil of a deep tropical thunderstorm. *Atmospheric Chemistry and Physics*, 12(14):6157–6172.

Wehner, B., Werner, F., Ditas, F., Shaw, R. A., Kulmala, M., and Siebert, H. (2015). Observations of new particle formation in enhanced UV irradiance zones near cumulus clouds. *Atmospheric Chemistry and Physics*, 15(20):11701–11711.

Wei, D., Dada, L., Zhao, J., Chen, X., Daellenbach, K. R., Xie, C., Wang, W., He, Y., Cai, J., Yao, L., Zhang, Y., Wang, Q., Xu, W., Wang, Y., Tang, G., Cheng, X., Kokkonen, T. V., Zhou, W., Yan, C., Chu, B., Zha, Q., Hakala, S., Kurppa, M., Järvi, L., Liu, Y., Li, Z., Ge, M., Fu, P., Nie, W., Bianchi, F., Petäjä, T., Paasonen, P., Wang, Z., Worsnop, D. R., Kerminen, V.-M., Kulmala, M., and Sun, Y. (2021). A 3D study on the amplification of regional haze and particle growth by local emissions. *npj Climate and Atmospheric Science*, 4(1):4.

Wilkinson, M. J., Monson, R. K., Trahan, N., Lee, S., Brown, E., Jackson, R. B., Polley, H. W., Fay, P. A., and Fall, R. (2009). Leaf isoprene emission rate as a function of atmospheric CO2 concentration. *Global Change Biology*, 15(5):1189–1200.

Yan, C., Dada, L., Rose, C., Jokinen, T., Nie, W., Schobesberger, S., Junninen, H., Lehtipalo, K., Sarnela, N., Makkonen, U., Garmash, O., Wang, Y., Zha, Q., Paasonen, P., Bianchi, F., Sipilä, M., Ehn, M., Petäjä, T., Kerminen, V.-M., Worsnop, D. R., and Kulmala, M. (2018). The role of H2SO4-NH3 anion clusters in ion-induced aerosol nucleation mechanisms in the boreal forest. *Atmospheric Chemistry and Physics*, 18(17):13231–13243.

Yan, C., Yin, R., Lu, Y., Dada, L., Yang, D., Fu, Y., Kontkanen, J., Deng, C., Garmash, O., Ruan, J., Baalbaki, R., Schervish, M., Cai, R., Bloss, M., Chan, T., Chen, T., Chen, Q., Chen, X., Chen, Y., Chu, B., Dällenbach, K., Foreback, B., He, X., Heikkinen, L., Jokinen, T., Junninen, H., Kangasluoma, J., Kokkonen, T., Kurppa, M., Lehtipalo, K., Li, H., Li, H., Li, X., Liu, Y., Ma, Q., Paasonen, P., Rantala, P., Pileci, R. E., Rusanen, A., Sarnela, N., Simonen, P., Wang, S., Wang, W., Wang, Y., Xue, M., Yang, G., Yao, L., Zhou, Y., Kujansuu, J., Petäjä, T., Nie, W., Ma, Y., Ge, M., He, H., Donahue, N. M., Worsnop, D. R., Kerminen, V.-M., Wang, L., Liu, Y., Zheng, J., Kulmala, M., Jiang, J., and Bianchi, F. (2021). The Synergistic Role of Sulfuric Acid, Bases, and Oxidized Organics Governing New-Particle Formation in Beijing. *Geophysical Research Letters*, 48(7):e2020GL091944.

Yang, L., Nie, W., Liu, Y., Xu, Z., Xiao, M., Qi, X., Li, Y., Wang, R., Zou, J., Paasonen, P., Yan, C., Xu, Z., Wang, J., Zhou, C., Yuan, J., Sun, J., Chi, X., Kerminen, V.-M., Kulmala, M., and Ding, A. (2021). Toward Building a Physical Proxy for Gas-Phase Sulfuric Acid Concentration Based on Its Budget Analysis in Polluted Yangtze River Delta, East China. *Environmental Science & Technology*, 55(10):6665–6676.

Yao, L., Garmash, O., Bianchi, F., Zheng, J., Yan, C., Kontkanen, J., Junninen, H., Buenrostro Mazon, S., Ehn, M., Paasonen, P., Sipilä, M., Wang, M., Wang, X., Xiao, S., Chen, H., Lu, Y., Zhang, B., Wang, D., Fu, Q., Geng, F., Li, L., Wang, H., Qiao, L., Yang, X., Chen, J., Kerminen, V.-M., Petäjä, T., Worsnop, D. R., Kulmala, M., and Wang, L. (2018). Atmospheric new particle formation from sulfuric acid and amines in a Chinese megacity. *Science*, 361(6399):278–281.

Yli-Juuti, T., Mielonen, T., Heikkinen, L., Arola, A., Ehn, M., Isokääntä, S., Keskinen,

H.-M., Kulmala, M., Laakso, A., Lipponen, A., Luoma, K., Mikkonen, S., Nieminen, T., Paasonen, P., Petäjä, T., Romakkaniemi, S., Tonttila, J., Kokkola, H., and Virtanen, A. (2021). Significance of the organic aerosol driven climate feedback in the boreal area. *Nature Communications*, 12(1):1–9.

Yli-Pirilä, P., Copolovici, L., Kannaste, A., Noe, S., Blande, J. D., Mikkonen, S., Klemola, T., Pulkkinen, J., Virtanen, A., Laaksonen, A., Joutsensaari, J., Niinemets, Ü., and Holopainen, J. K. (2016). Herbivory by an outbreaking moth increases emissions of biogenic volatiles and leads to enhanced secondary organic aerosol formation capacity. *Environmental Science & Technology*, 50(21):11501–11510.

Ylisirniö, A., Buchholz, A., Mohr, C., Li, Z., Barreira, L., Lambe, A., Faiola, C., Kari, E., Yli-Juuti, T., Nizkorodov, S. A., Worsnop, D. R., Virtanen, A., and Schobesberger, S. (2020). Composition and volatility of secondary organic aerosol (SOA) formed from oxidation of real tree emissions compared to simplified volatile organic compound (VOC) systems. *Atmospheric Chemistry and Physics*, 20(9):5629–5644.

Yuan, J. S., Himanen, S. J., Holopainen, J. K., Chen, F., and Stewart Jr, C. N. (2009). Smelling global climate change: mitigation of function for plant volatile organic compounds. *Trends in Ecology & Evolution*, 24(6):323–331.

Zhang, Z., Zhang, R., Cescatti, A., Wohlfahrt, G., Buchmann, N., Zhu, J., Chen, G., Moyano, F., Pumpanen, J., Hirano, T., Takagi, K., and Merbold, L. (2017). Effect of climate warming on the annual terrestrial net ecosystem CO2 exchange globally in the boreal and temperate regions. *Scientific Reports*, 7(1):2045–2322.

Zhao, D. F., Buchholz, A., Tillmann, R., Kleist, E., Wu, C., Rubach, F., Kiendler-Scharr, A., Rudich, Y., Wildt, J., and Mentel, T. F. (2017). Environmental conditions regulate the impact of plants on cloud formation. *Nature Communications*, 8(1):14067–14067.