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The Center of Excellence in Atmospheric Science (2002–2019) — from molecular and biological processes to the global climate

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The study of atmospheric processes related to climate requires a multidisciplinary approach, encompassing physics, chemistry, meteorology, forest science, and environmental science. The Academy of Finland Centre of Excellence in atmospheric sciences (CoE ATM) responded to that need for 18 years and produced extensive research and eloquent results, which are summarized in this review. The work in the CoE ATM enhanced our understanding in biogeochemical cycles, ecosystem processes, dynamics of aerosols, ions and neutral clusters in the lower atmosphere, and cloud formation and their interactions and feedbacks. The CoE ATM combined continuous and comprehensive long-term in-situ observations in various environments, ecosystems and platforms, ground- and satellite-based remote sensing, targeted laboratory and field experiments, and advanced multi-scale modeling. This has enabled improved conceptual understanding and quantifications across relevant spatial and temporal scales. Overall, the CoE ATM served as a platform for the multidisciplinary research community to explore the interactions between the biosphere and atmosphere under a common and adaptive framework.

Introduction

Life on Earth, the biosphere, interacts closely with the atmosphere, and is sensitive to changes in its composition and functioning. These interactions occur on spatial scales from nanometers to global, and time scales from nanoseconds to millennia (Wanner *et al.* 2008, Kulmala *et al.* 2009a). In addition to the atmosphere, the biosphere interacts with the hydrosphere, cryosphere and lithosphere, and changes in any of these components affect the others through material and energy flows, and linked processes and feedbacks. Accordingly, global climate system is strongly influenced by biosphere through the effect of vegetation cover on surface radiation budget, sensible and latent heat fluxes, and atmospheric composition, and therefore understanding biotic processes is necessary for correctly analyzing the global climate system (Krinner *et al.* 2005, Arneth *et al.* 2010). Although biosphere–atmosphere feedbacks are globally widespread and regionally strong, current Earth system models underestimate these feedbacks (Green *et al.* 2017). One reason for this is that the Earth observations are traditionally rather fragmented. The Earth system can be studied piece by piece, but to be able to study complicated interactions and feedbacks between different Earth components and to foresee changes in the Earth system, a holistic, interdisciplinary approach is needed (Kulmala 2018).

Currently, humanity is facing severe global changes in the Earth system. Each of the last four decades has been successively warmer than

any decade before it since 1850, and the global surface temperature (in 2011–2020) is already 1.09°C above the pre-industrial (1850–1900) level (IPCC 2021). Despite decades of research, there are still many areas of uncertainty in understanding the global climate system and its changes. The critical issues that need continued scientific efforts are called Grand Challenges, and include e.g. the role of clouds and aerosols in the climate system, sea-level rise, water availability, cryospheric processes, and linking extreme events to climate change (Beniston 2013). Fundamental in solving these Grand Challenges is in addition to holistic scientific efforts, an adequate Earth observation network (Kulmala 2018), easy access to climate research data and data compatibility.

Due to the Grand Challenges related to global warming, the remaining allowable net carbon emission to stay below the 1.5˚C Paris temperature target is highly uncertain. The Intergovernmental Panel on Climate Change (IPCC) recognizes atmospheric aerosols as the single largest source of uncertainty in human-driven climate change (IPCC 2013, 2021). The effects of these aerosols include the direct effect of aerosols on radiative forcing, their impact on cloud formation and properties, and their feedbacks in the natural carbon cycle. The Academy of Finland's Center of Excellence in Atmospheric Sciences (CoE ATM) has worked for 18 years to increase the understanding on and to quantify various biogeochemical cycles, ecosystem processes, dynamics of aerosols, ions and neutral clusters in the lower atmosphere, cloud formation and their interactions and feedbacks. The COntinental Biosphere-Aerosol-Cloud-Climate (COBACC) feedback loop demonstrates the complexity of interactions existing between biosphere processes and aerosol formation and growth (Kulmala *et al.* 2013a, 2014, Fig. 1). The COBACC theoretical framework has been developed in the CoE ATM and used to test and develop new theoretical understanding. In COBACC, the higher temperatures and atmospheric CO_2 levels boost ecosystem carbon sink and biomass production, which leads to increased biogenic secondary organic aerosol and cloud condensation nuclei concentrations, causing cooling. Enhanced aerosol and cloud nuclei concentrations further increase the fraction of diffuse solar radiation that feeds back to plant biomass production, because plants can utilize diffuse radiation more efficiently than direct radiation in biomass production. COBACC connects the two main cooling agents, carbon sink and aerosol source, to each other, and combines physical, chemical and biotic phenomena and feedbacks (Arneth *et al.* 2010, Carslaw *et al.* 2010, Mahowald 2011). Human actions, such as emission policies and land use changes, also have a substantial impact on the complex couplings between atmospheric aerosols, trace gases, greenhouse gases (GHG), clouds and climate (Arneth *et al.* 2009, Raes *et al.* 2010, Shindell *et al.* 2012, Boy *et al*. 2019, Artaxo *et al.* 2022). Aerosols link air quality with weather and climate, introducing additional feedbacks, particularly through boundary layer dynamics (Kulmala 2015, Petäjä *et al*. 2016a).

Here we present an overview of the CoE ATM's approach and key scientific advancements from 2002–2019. A unique feature and a key component in the success of the group has been the comprehensive, long-term and co-located measurements of various Earth components and close collaboration with scientists from different disciplines. This interdisciplinary approach is needed to understand the interactions and feedbacks among Earth components and ultimately to solve the Grand Challenges of the Earth.

Approach

Scientific approach

We measure material and energy flows in and between different ecosystems and the atmosphere to understand the biosphere–atmosphere interactions. Mass and energy flows are like an interdisciplinary language which allows an information flow between disciplines. This is especially important between biological/ecological and physical and chemical sciences. Thus, in a fundamental level, the aim of the consecutive CoEs has been to understand the processes and interactions in the biosphere–atmosphere continuum by combining the multidisciplinary

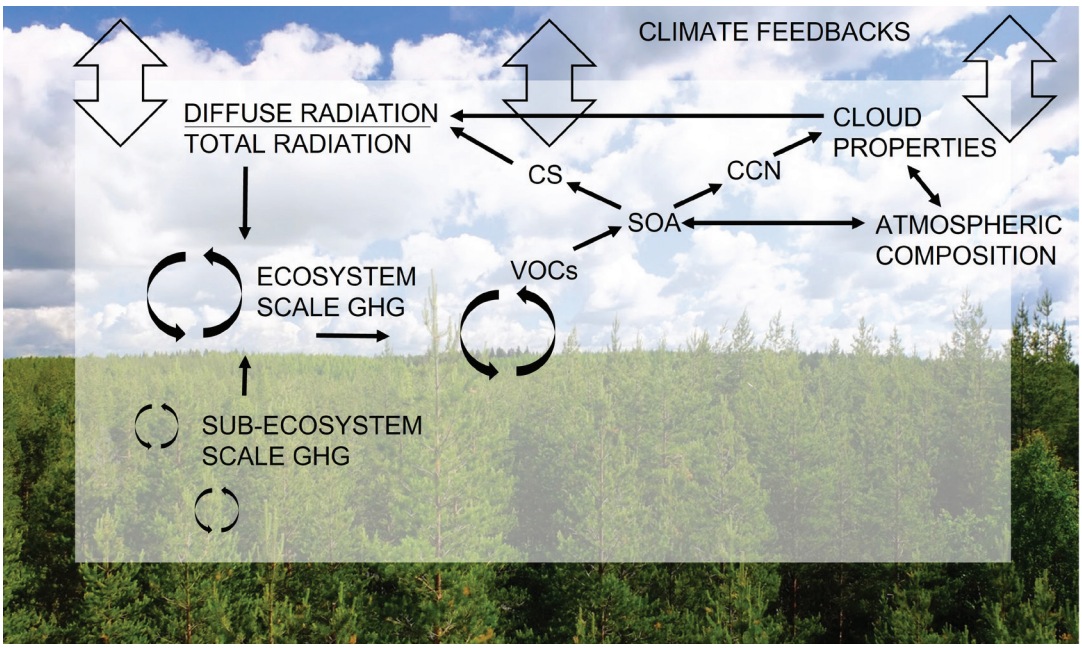

Fig. 1. The CoE ATM has focused on studying the Earth surface–atmosphere interactions, feedbacks and the individual ecosystem and atmospheric processes involved. The emission of volatile organic compounds (VOCs) from an ecosystem (that can be any type of ecosystem, including e.g. urban area or agricultural field) is linked to ecosystem activity and greenhouse gas (GHG) emission. VOCs participate in the formation of secondary organic aerosols (SOA) in the atmosphere. SOA directly affects atmospheric composition, forms cloud condensation nuclei (CCN) affecting cloud formation and properties, and build up condensation sink (CS) that further increases the amount of diffuse radiation the ecosystem is receiving. Diffuse radiation can be used by the ecosystem more efficiently than direct radiation in photosynthesis, thus increasing ecosystem carbon sink. These ecosystem–atmosphere interactions feed back to global climate, which is shown with the big arrows at the top of the picture: Increasing atmospheric CO_2 concentration increases ecosystem carbon assimilation and increasing air temperature affects many of the ecosystem and atmospheric processes involved, whereas increasing cloudiness and CS cools air temperature by reflecting sunlight back to space and increasing ecosystem carbon sink decreases the atmospheric CO_2 concentrations.

expertise of the consortium, i.e. learning from the dialogue between the disciplines and the disciplinary-specific methodologies to provide new insights into the processes (e.g. Kulmala *et al.* 2005, Hari and Kulmala 2008, Kulmala *et al.* 2009b).

The need to understand mass and energy flows led to the development of network of Stations for Measuring Earth Surface–Atmosphere Relations (SMEAR; Hari *et al*. 2016). The first SMEAR station (SMEAR I) was established in Värriö, Northern Finland (Hari *et al*. 1994). At this station, the particular focus was on timely topics at the time, i.e., acidification, acid rain and their effects on forests. Over the following years, more SMEAR stations were constructed with SMEAR II in Hyytiälä being the most comprehensive of them all (Hari and Kulmala 2005). The SMEAR stations have provided the experimental backbone also for studies on urban air quality (SMEAR III, Järvi *et al.* 2009a) and aerosol-cloud interactions (SMEAR IV, Leskinen *et al.* 2009). Pallas-Sodankylä Global Atmospheric Watch station is another station (in addition to SMEAR I) in Northern Finland and provides, e.g., the longest continuous data set of high-precision atmospheric CO_2 concentration in Finland, dating back to 1996 (Hatakka *et al.* 2003, Kilkki *et al.* 2015). The SMEAR concept has expanded also to study atmosphere–forest interactions in a hemi-boreal forest in Estonia (Noe *et al*. 2015), air quality–savannah interactions in South Africa (Venter *et al.* 2018), air quality–boundary layer–precipitation processes

in Eastern China (Ding *et al*. 2013a, 2016a, Petäjä et al. 2016a), and air quality–aerosol–haze processes (Kulmala *et al*. 2021a, Liu *et al*. 2020) in a Chinese gigacity environment (Kulmala *et al.* 2021a). Such comprehensive ground-based observations are needed (Kulmala 2015, Kulmala 2018) together with satellite remote sensing data (e.g. Sun 2017), multiscale modeling and integration of the results (e.g. Kerminen *et al*. 2010, Petäjä *et al.* 2021a, 2021b) to tackle the challenges the societies are facing now and in the future.

Although the fundamental approach has not changed over the years, the thematic focus of the CoE ATM has developed and widened as we have deployed the framework to a variety of research questions in different environments. For example, experimental determination of fluxes between the biosphere and the atmosphere originally enabled us to study photosynthesis and CO_2 exchange (e.g. Hari *et al*. 1999a,b). Over the years, we have extended this approach to include, for example, biogenic volatile organic compounds (BVOCs, Aalto *et al.* 2014, Rantala *et al.* 2015, Héllen *et al.* 2018), methane (Rinne *et al.* 2018, Vainio *et al.* 2021), nitrous oxide (Korkiakoski *et al*. 2020) and carbonyl sulfide (Kohonen *et al.* 2020). Under the SMEAR umbrella, we have studied the material flows in addition to different forests in pristine wetlands (Rinne *et al.* 2018), urban areas (Karttunen *et al.* 2020) and over lakes (Erkkilä *et al.* 2018). Recently, SMEAR-type measurements have started also on agricultural ecosystems (Heimsch *et al.,* 2021) and on a cutover wetland. The analysis has tackled not only vertical exchange but also lateral transport of water and dissolved organics (e.g. Rasilo *et al.* 2015).

With the comprehensive observational data, we have connected the emission of BVOCs from the biosphere, their atmospheric oxidation, the formation of fresh atmospheric aerosol particles and the production of secondary organic aerosol (Kulmala *et al*. 2013b, Ehn *et al.* 2014). In order to observe these processes, we have developed novel aerosol technologies, such as the Particle Size Magnifier (PSM, Vanhanen *et al.* 2011), which is able to detect aerosol particles down to 1 nm in size. At the same time, we have developed atmospheric mass spectrometric techniques to determine the chemical composition of the trace gases and clusters (e.g. Petäjä *et al*. 2009, Sipilä *et al*. 2010, Kulmala *et al*. 2013b, Ehn *et al.* 2014). Furthermore, we have contributed significantly to the development of a Neutral cluster and Air Ion Spectrometer (NAIS) (Manninen *et al.* 2009, 2010, Mirme and Mirme 2013, Kulmala *et al.* 2013b). These techniques are currently deployed worldwide to resolve the chemistry and physics behind secondary aerosol formation in different environments in the atmosphere and in controlled laboratory experiments (e.g. Kerminen *et al*. 2018, Yao *et al.* 2018, Chu *et al.* 2019, Wang *et al.* 2020, Bianchi *et al.* 2021).

The comprehensive observations at SMEAR stations enable us to expand the scale and relevance of individual observations. The interdisciplinary observations from the ecosystem sciences (e.g. photosynthesis, emission of BVOCs), chemistry (e.g. atmospheric oxidation), aerosol science (e.g. formation and growth of aerosol particles), atmospheric and cloud physics (e.g. aerosol activation to cloud droplets) and meteorology (e.g. boundary layer dynamics, formation of precipitation) allow us to explore the influence of biogenic emissions to atmospheric optical properties, precipitation, cloud properties and ultimately to radiative forcing (e.g. Paasonen *et al*. 2013, Kulmala *et al*. 2014, Kulmala *et al.* 2020, Petäjä *et al*. 2021a). Thus, one cornerstone in the CoE ATM approach has been the co-location of different environmental measurements, and we have actively integrated these measurements in Finland and promoted their integration also in Europe and globally.

The CoE ATM research infrastructure (INAR RI) belongs to the Finnish RI roadmap 2021–2024. INAR RI integrates the measurements of four European research infrastructures: Integrated Carbon Observation System (ICOS), Aerosol, Clouds, and Trace Gases Research Infrastructure (ACTRIS), Integrated European Long-Term Ecosystem, Critical Zone & Socio-Ecological Research Infrastructure (eLTER) and Infrastructure for Analysis and Experimentation on Ecosystems (AnaEE). The CoE ATM was providing data and acted for years as the national focal point of Finnish activities in these research infrastructures.

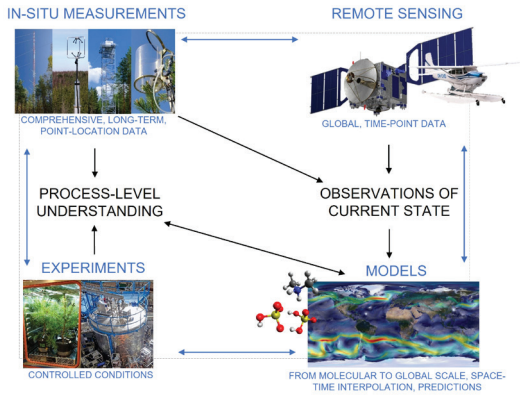

Fig 2. The ATM CoE scientific approach was to combine comprehensive in situ observations in different types of environments, ecosystems and platforms, ground- and satellite-based remote sensing, targeted laboratory and field experiments and multi-scale modeling.

Overall, the CoE ATM has served as a platform for the multidisciplinary research community to explore different aspects in the biosphere–atmosphere interactions under a common and adaptive framework that combines experimental work, theoretical thinking and multiscale modeling following the research philosophy principles of realism. In its traditional form, realism expresses the belief that reality exists independently of us. In the past century, realism has undergone two important developments that also reflect the progress in science (Tuomivaara *et al.* 1994, Bunge 2006, 2018, 2019, Agazzi 2014): 1) the recognition of the importance of theory and its critical use of theory in science; and 2) the systemic way of thinking. Over a long enough period, observations, experimental work, theoretical thinking and multiscale modeling have interacted and evolved in the CoE ATM towards joint and coherent answers to formulated science questions. In the future, this community will have the tools developed during the CoE ATM era to tackle a suite of global grand challenges.

Methods

The key strength of our holistic scientific approach is to combine: 1) continuous and comprehensive long-term in situ observations in different types of environments, ecosystems and platforms; 2) ground- and satellite-based remote sensing; 3) targeted laboratory and field experiments; and 4) advanced multi-scale modeling, in order to provide improved conceptual understanding and quantifications over the relevant spatial and temporal scales (Fig. 2). The holistic approach and innovative method and instrument development allows us to address interdisciplinary research questions related to current and future climate change drivers and their impacts on ecosystem functioning, and to provide harmonized comprehensive data to unravel mechanisms and feedbacks involved in ecosystems' responses and feedback to climate.

The CoE ATM has operated five highly instrumented boreal and sub-arctic research stations in Finland: the four SMEAR stations and the GAW station in Pallas-Sodankylä. The SMEAR concept promotes the integration and co-location of different Earth component measurements from different disciplines. This has enabled us to study the complicated interactions between the different Earth components and led to important new findings, especially regarding aerosol formation and growth. The north-boreal SMEAR I station was set up in 1991 in a Scots pine forest in Eastern Lapland (67.77°N, 29.58°E, Hari *et al.* 1994), and it provides atmospheric, meteorological and ecosystem data. The south-boreal SMEAR II station was set up in 1995 in a Scots pine forest in southern Finland (61.85°N, 24.28°E), and it is the world-leading station on ecosystem-atmosphere data due to its comprehensive research program and its unique time series of aerosol formation and biogeochemical fluxes (Hari and Kulmala 2005, Kulmala *et al.* 2021b). The SMEAR II station has satellite stations on a nearby lake and a wetland site. The urban SMEAR III station was established in 2004 in Helsinki city (60.2°N, 24.97°E, Järvi *et al.* 2006), and it provides in addition to atmospheric and micrometeorological data, also urban vegetation data. A boreal SMEAR IV station (62.9°N, 27.65°E, Leskinen *et al.* 2009) joined the SMEAR network in 2009, and it provides atmospheric data focusing especially on aerosol-cloud interactions. The sub-arctic Pallas-Sodankylä GAW station (67.97°N, 24.12°E) is an atmosphere-ecosys-

tem supersite that comprises satellite sites at a Scots pine forest, wetland, lake and fell-top tundra (Aurela *et al*. 2015a). Atmospheric composition measurements started at the station in 1991. The CoE ATM has been intensively involved in developing a global Earth observatory (Kulmala 2018) to promote multidisciplinary integration, and provide a global reference for synergies, co-location and complementarities already implemented in the CoE ATM.

In addition to continuous field observations, extensive field campaigns and remote measurements have been important to extend the spatial range of the measurements. As an example of field campaigns, our researchers from the Finnish Meteorological Institute and the University of Helsinki have been collaborating in Antarctic research already for two decades (e.g. Koponen *et al.* 2003, Sipilä *et al*. 2016, Jokinen *et al*. 2018). In terms of ground-based, airborne or spaceborne measurements, we have used data from laser scanning (Liu *et al*. 2019), drones (Alekseychik *et al.* 2021a), radars (Li and Moisseev 2020), aircrafts (Petäjä *et al.* 2016b), zeppelin (European project PEGASOS, Lampilahti *et al.* 2021) and satellites (Sun *et al.* 2017). The Earth Observation techniques (space-borne remote sensing) provide spatially continuous, frequent observational data sets that can be applied to the validation of the climate model predictions for the large scale, to the trend analyses of essential climate variables, to the analyses of spatially distributed processes and to develop further the physical models through the improvement of the model parametrization or static background fields (Fig. 2).

CoE ATM research laboratories include Aerosol Laboratories in the Universities of Helsinki and Eastern Finland and in the Finnish Meteorological Institute, and Ecophysiological laboratory and Laboratory for Analytical Chemistry in the University of Helsinki. Research has also been done at The Cosmics Leaving Outdoor Droplets (CLOUD) chamber at CERN particle accelerator facility (Wang *et al.* 2020). In addition to measurements, strong modelling expertise was a central part of the CoE ATM. Modelling was used intensively at different spatial and temporal scales, ranging from modelling molecules and molecular clusters in computational aerosol physics (e.g. Besel *et al.* 2020) to modelling global climate with Earth System Models (Sporre *et al.* 2020) as part of Europe-wide consortium EC-Earth that participates in the Coupled Model Intercomparison Project (CMIP6) featuring climate models for the 2021 IPCC sixth assessment report with different model configurations.

Open data

Open Science is one of the key principles of modern scientific work, and the CoE ATM has played a strong role in the development in this field as well. The CoE has a long experience with open data issues, as the SMEAR station datasets have been from the very beginning openly available to the scientific communities (https://smear.avaa.csc.fi/, Junninen et al. 2009). Practical developments of the original and the later updated SmartSMEAR platforms have created a possibility to openly visualize and access most of the infrastructure-based observations in the stations. In addition, the CoE campaign-type observations are generally available to public in different international repositories, and most of the articles in the CoE have been published in open access journals.

The connection to the European Strategy Forum on Research Infrastructures (ESFRI) developments has been a key part of the open science activities, and the CoE has been leading two large cluster projects (ENVRI PLUS and ENVRI FAIR) on environmental ESFRI open data collaboration, as well been a major participant in similar developments in individual ESFRI RIs, particularly in ACTRIS, ICOS and eLTER. CoE participants are also key personnel in many overall developments in the open science communities. Participation in the Research Data Alliance activities, particularly in the data citation, versioning and identification working groups have been active (e.g. Rauber *et al.* 2015, Klump *et al*. 2020). As a part of ENVRI collaboration, the CoE has also been deeply involved in the development of the European Open Science Cloud (e.g. Petzold and Asmi 2020, Gotz *et al*. 2020).

Collaboration and PEEX initiative

The CoE ATM research community was formed of researchers from the University of Helsinki, the Finnish Meteorological Institute and the University of Eastern Finland, and had direct working connections with ~2000 researchers in ~50 countries over the years. An important part of this collaboration was coordination of environmental research infrastructure networks in Europe and globally. The CoE ATM was also a key partner in the CLOUD consortium at CERN. CoE ATM participated in all CLOUD campaigns, elucidating several new particle formation mechanisms (e.g. Riccobono *et al*. 2014, Dunne *et al*. 2016, Kirkby *et al*. 2016, Tröstl *et al*. 2016, Wang *et al*. 2020, He *et al.* 2021) and was also involved in the cloud and ice formation experiments (e.g. Nichman *et al.* 2017).

To expand the CoE ATM collaboration from Europe and USA to northern Eurasia, a Pan-Eurasian Experiment (PEEX) program (www.atm.helsinki.fi/peex) was initiated in 2012 (Kulmala *et al.* 2015, Lappalainen *et al*. 2014, 2016). Especially regions in Russia, China and the new Silk Road economic regions, have a significant impact on the global climate but a limited environmental data coverage. The PEEX program aims to enhance coordinated, comprehensive and integrated environmental measurements especially in these areas (Alekseychik *et al*. 2016, Hari *et al.* 2016, Kulmala 2018, Lappalainen *et al.* 2018, Vihma *et al.* 2019, Petäjä *et al*. 2021b), and a number of bi-lateral research projects with Russian and Chinese universities and research institutes has been carried out in the PEEX framework (Lappalainen *et al.* 2022a). The collaboration with the Russian organizations has been suspended since March 2022. We have a long on-going collaboration with Estonia particularly organizing joint workshops like Air Ions, Clusters and Atmospheric Aerosols Workshop (Kulmala *et al*. 2016a), and a first SMEAR station outside Finland was established in Järveselja, Estonia in 2014. In 2011, a Station for Observing Regional Processes of the Earth System (SORPES) started operation about 20 km northeast of downtown Nanjing, China (Ding *et al.* 2013b). This SMEAR-type station is a cross-disciplinary research and experiment platform developed by Nanjing University in collaboration with University of Helsinki (Ding *et al.* 2016b). In 2019, another SMEAR-type station ("Beijing Haze", hosted by the Beijing University of Chemical Technology BUCT) started in operation in Beijing (Liu *et al.* 2020). In addition to research activities, PEEX contributes to education, works towards solving the UN sustainable development goals and addresses the role of science diplomacy when collaborating towards these global aims under different geopolitical conditions (Kulmala *et al*. 2021b; Lappalainen *et al*. 2022b). Unique observations already over 10 years on greenhouse gas concentrations (Ivakhov *et al*., 2019) and fluxes (Aurela *et al.* 2015b, Juutinen *et al.* 2017, Tuovinen *et al*. 2019), oceanic and continental aerosol production and black carbon sources (Asmi *et al.* 2016) at Tiksi, Northern Sakha (Yakutia) has extended the observational network to most remote permafrost Siberia on the coast of the Arctic Ocean (Uttal *et al.* 2016).

The scientific core of the PEEX program has been tightly connected to the CoE ATM (Kulmala *et al.* 2015, Lappalainen *et al.* 2016) and expanded the geographical range of the COBACC feedback analysis to the Northern Eurasia, especially to Siberia (Lappalainen *et al*. 2015, 2019). As an example, PEEX collaboration has provided new information e.g. on the interaction between aerosols, solar radiation and the gross primary production (Ezhova *et al.* 2018). As a concrete example, new particle formation (NPF) was studied at Fonovaya in 2016–2018, and the results were compared with other boreal SMEAR-like stations in Europe (Finland and Estonia, Demakova *et al*. 2021). It was confirmed that NPF happens less often in Siberia than in the European boreal sites. The factors driving NPF were similar between the sites (shortwave radiation, condensation sink and SO_2), but NPF at Fonovaya was often associated with the wind direction from the industrial areas of Russia and Kazakhstan (Demakova *et al*. 2021), whereas NPF in the European sites was largely associated with the air masses coming from the clean sector in the North. Other recent research highlights in

PEEX collaboration has focused on permafrost areas of north-west Siberia: it was found that permafrost is thawing at the studied locations due to warm and/or snowy winters (Ezhova *et al*. 2021) and even in locations with the least pronounced snow insulation effect, the temperature on the top of permafrost increased at the rate 0.56°C yr^{-1} (Kukkonen *et al.* 2020). Permafrost thaw was critical for an outbreak of anthrax on Yamal peninsula in 2016 after 70 years of epidemiologically stabile situation (Timofeev *et al*. 2019, Ezhova *et al.* 2021). Finally, increasing probability of tundra fires was demonstrated in nortwest Siberia, where fires has promoted a vegetation shift from tundra to forest (Sizov *et al*. 2021). In addition to the COBACC research framework, the other scientific driver of the PEEX program has been studying air quality in urban environments. While the CoE ATM has studied atmospheric composition in a relatively clean city environment (e.g. Järvi *et al.* 2009a), PEEX collaboration has allowed to study air quality in the most polluted environments in the globe, especially in Chinese megacities and in Beijing (Kulmala 2015, Petäjä *et al.* 2016a).

The link from the CoE ATM scientific breakthroughs towards innovations has been established via close co-operation with enterprises, especially with the three university spin-off companies established during the CoE ATM era: Airmodus Ltd. that provides aerosol particle counters and mass-spectrometer products, Karsa Ltd. that produces detectors of trace amounts of explosives for air cargo and other safety purposes, and SMEAR Ltd. that provides infrastructures and services needed for expanding the SMEAR network with the aim of a global network GlobalSMEAR. The CoE ATM had collaboration also with tens of other enterprises in terms of instrument, method and product testing and development, expertise in environmental services and process understanding, emission measurements, and carbon accounting development. One example of the current SMEAR-based applications is the MegaSense project, where machine learning techniques are used for the calibration of low-cost sensors with a few highly accurate SMEAR-type measurement stations (Motlagh *et al.* 2020).

Scientific progress

Greenhouse gases

Abundance of greenhouse gases (GHG) in the atmosphere makes life on Earth possible, but anthropogenic GHG emissions are the primary cause of current climate change. Carbon dioxide (CO_2) is the most abundant GHG after water vapour (water vapour amplifies the warming produced by the other GHG), and atmospheric CO_2 concentration has increased from the pre-industrial times with ~139 ppm being over 420 ppm in May 2022 (https://smear.avaa.csc.fi/). Another important greenhouse gas is methane (CH_4), which absorbs much more infrared radiation per molecule than CO_2 and thus has a very high global warming potential. However, CH_4 remains in the atmosphere for a much shorter time than CO_2 due to atmospheric oxidation. Increased greenhouse gas concentrations in the atmosphere are changing the climate and thus have a major effect on all living organisms on Earth, including forest ecosystems. Boreal and arctic areas are warming two to four times as fast as the global average (e.g. Mikkonen *et al.* 2015, Ruosteenoja and Räisänen 2021, Rantanen *et al.* 2022). In the short term, the forests in high latitudes can grow even more due to longer growing seasons (Pulliainen *et al.* 2017), whereas soil respiration may increase due to higher temperatures. This seems to lead to increase of the seasonal amplitude of the atmospheric CO_2 concentration due to the different seasonality of photosynthesis and respiration (Hu *et al*., 2021). On a longer and larger scale, it is evident that climate change is a risk for forests due to increased risk for drought (Brodribb *et al*. 2020, Lindroth *et al.* 2020), pest infestations (Kolb *et al*. 2016) and more frequent and intense forest fires (Brando *et al.* 2014). While forest ecosystems are affected by climate change, the climate is also affected by forests.

The most important climate change mitigation potential of forests is linked to their carbon sink. Forests and other terrestrial ecosystems remove about one-third of the CO_2 annually emitted to the atmosphere by human activity (Friedlingstein *et al*. 2020). Boreal regions contain more organic carbon than any

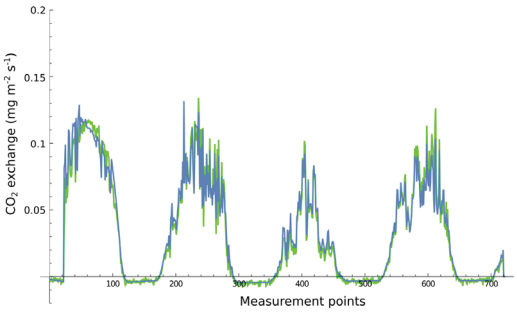

Fig 3. Example of measured (green line) and modelled (blue line) Scots pine branch CO_2 exchange from SMEAR I station. Measured and modelled values are shown for four consecutive days in year 2012, each day having 180 measurement points.

other terrestrial ecosystem due to their large soil carbon stocks (Scharlemann *et al.* 2014). The carbon balance of an ecosystem is the difference between its carbon gains via photosynthesis and losses mainly via respiration from plants and heterotrophs, such as microbes that decompose soil organic matter. The annual carbon uptake in photosynthesis of boreal forests is relatively low, which is a characteristic of these forests — but due to the cool and humid climate, the annual carbon loss in respiration is even lower, which makes the contribution of boreal forests to global terrestrial carbon sink equal to that of tropical forests (Tagesson *et al*. 2020). However, climate change may cause unexpected feedbacks to soil carbon stock due to higher tree growth and increased carbon allocation belowground. There are indications that increased plant growth and nutrient acquisition correlates negatively with soil carbon stocks (Terrer *et al*. 2021), the trade-off being related to soil nutrients mining, which decreases soil organic carbon storage. As climate change induced disturbances (e.g. fires, pathogen or herbivore outbreaks) may become more frequent, more research is needed to better understand how the positive and negative feedbacks such as elevated temperatures and CO_2 levels affect soil carbon stocks in the boreal zone.

Photosynthesis

Photosynthetic CO_2 uptake was one of the first fluxes between the biosphere and the atmosphere that the physicists and forest scientists studied together in the CoE ATM (e.g. Hari *et al*. 1999a,b). Photosynthesis provides material and energy for plant metabolism and growth and is the origin of vast majority of organic material in the living ecosystems. The light reactions of photosynthesis capture energy in solar radiation binding it into short-lived chemical compounds. The carbon reactions assimilate atmospheric CO_2 to convert this energy into stable carbon compounds, sugars. The availability of solar radiation, kinetic energy (related to the temperature of the leaf), carbon dioxide, as well as the physiological state of the plant determine how much sugars can be synthetized by a plant. Accordingly, the dependency of photosynthesis on light, temperature and CO_2 availability can be used to model its dynamic response to the environment (see Hari *et al*. 2014). Models can be further developed by taking into account stomatal functioning as gases enter and exit leaves through small pores, stomata, whose opening and closing are actively regulated by plants. When stomata are open to take CO_2 into leaves, water is simultaneously lost to the atmosphere. In the developed model (Hari *et al*. 1986, 2014, 2018), it was assumed that the transpiration cost for a plant is proportional to the amount of transpiration and that the goal of the stomatal regulation is to maximize the difference between photosynthetic production and transpiration cost during several days. Later on, the model was further developed (Hari *et al*. 2017a,b, 2018) by taking into account seasonal endogenous (i.e. plant internal origin) regulation of photosynthesis that has emerged in evolution and determines the seasonal efficiency of photosynthesis (Fig. 3). To summarize, we constructed our theory and model in three steps: 1) synthesis of sugars; 2) optimal regulation of stomatal action; and 3) seasonal cycles of photosynthesis. Our theory and model is able to predict 90–95% of the variance of the measured photosynthesis.

Simultaneously with the development of the above-described leaf-level photosynthesis model over the years, the theories and measurements have extended to various GHG fluxes and processes, temporal and spatial scales and types of ecosystems.

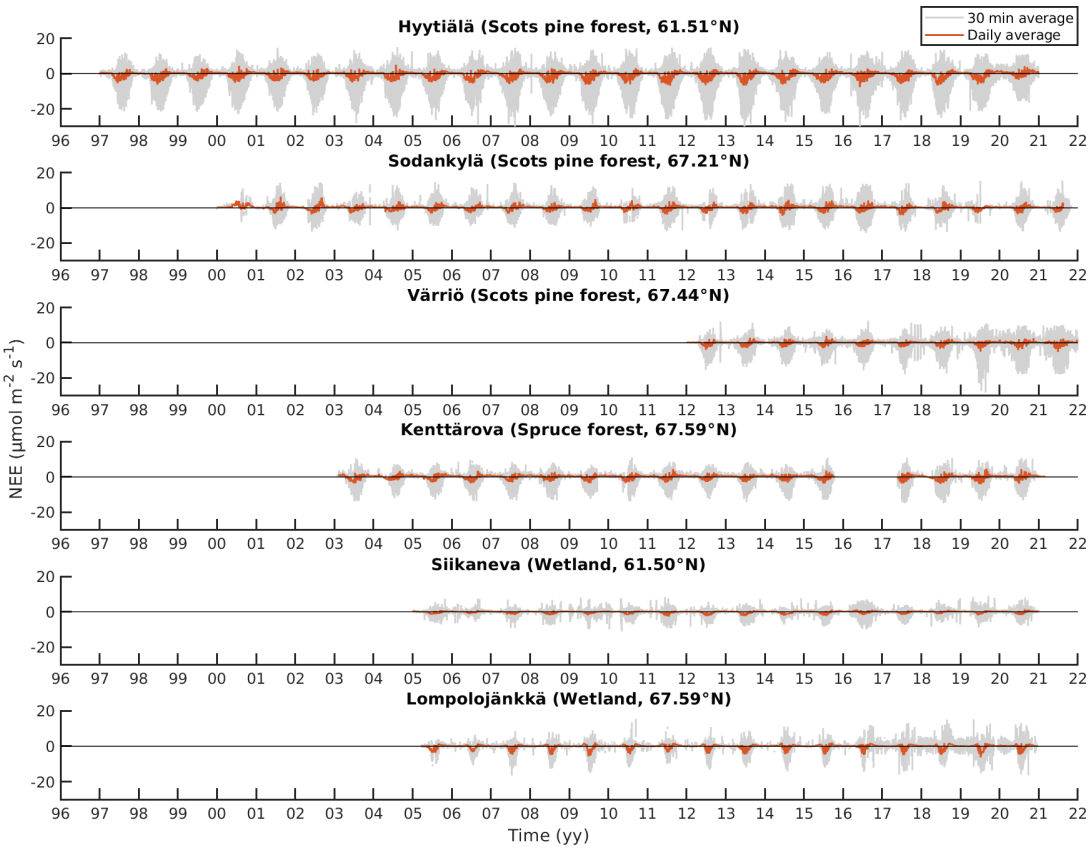

Fig 4. Time series of net ecosystem exchange of CO_2 (NEE) at six different measurement sites in Finland: a south-boreal pine-dominated SMEAR II forest site in Hyytiälä, two subarctic pine-dominated forest sites in Sodankylä and SMEAR I in Värriö, a subarctic spruce-dominated forest site in Kenttärova, a south-boreal open wetland site in Siikaneva and a sub-arctic open wetland site in Lompolonjänkkä. Grey lines show data in original 30 min resolution and red lines show daily averages. NEE is obtained from micrometeorological eddy covariance measurements and is stability-filtered, storage-corrected and gap filled (see Kolari et al. 2009).

Ecosystem-scale greenhouse gas fluxes

We have quantified the carbon and energy balances of the boreal pine forest stand at a south-boreal SMEAR II site, and the seasonal and interannual drivers of the flux components (Kolari *et al.* 2009, Launiainen 2010) using the eddy-covariance (EC) technique (Aubinet *et al.* 2012). When it comes to the seasonal variability of energy partitioning, we found a clear role of the surface conductance (g_s). In spring, because of low values of g_s, the available energy is mainly used as sensible heat flux (the ratio between sensible and latent heat flux = 4–6), while the evapotranspiration takes over in July–August when the ratio has minimum values of 0.7–0.9 (Launiainen 2010). In the SMEAR II forest, the annual net ecosystem exchange (NEE, Fig. 4) of carbon has varied from the years 2001–2017 — from –152 to –309 g C m^{-2} yr^{-1}, the mean annual carbon sink being –252 g C m^{-2} yr^{-1} (Launiainen *et al.* 2022). In forests with closed canopies, trees contribute almost 90% of the ecosystem gross primary production (GPP) and the highest cumulative, annual net carbon uptake coincides with the earliest onset of growing season (Kolari *et al.* 2009), whereas warm autumns result in an increased respiration (Vesala *et al*. 2010). In the sub-arctic SMEAR I and Pallas-Sodankylä sites, the annual CO_2 fluxes are clearly lower leading to less negative NEE, i.e. smaller carbon sink, or even a carbon source over a year (Fig. 4). At Kenttärova spruce forest in Pallas, the annual NEE was close to zero with an interan-

nual variation ranging from a moderate CO_2 uptake (–49 g C m^{-2} yr^{-1}) to a high CO_2 release (+73 g C m^{-2} yr^{-1}) (Aurela *et al.* 2015a). Taking into account the carbon accumulating in the growing trees, the annual NEE estimates suggest that in most years the soil is losing carbon. This conclusion was supported by soil chamber flux measurements conducted with the chamber technique (T. Penttilä, unpubl. data). At Sodankylä, the annual NEE in 2001–2020 varied from +2 to +120 g C m^{-2} d^{-1} with a mean of 69 g C m^{-2} yr^{-1} (M. Aurela unpubl. data), indicating that the soil is losing considerably carbon each year. One, perhaps not full, explanation for this could be the groundwater coming from a nearby wetland percolating the sandy soil and bringing excess carbon to the soil. Part of this carbon is mineralized into CO_2 and exhaled out from the soil at the forest site (Marttila *et al*. 2021).

Besides CO_2, other trace gases have been extensively investigated. The boreal forest is a significant sink of carbonyl sulphide (COS) during the growing season at day-time and the observed fluxes correlate well with GPP estimated from the observed CO_2 fluxes (Kohonen *et al.* 2022,; see also Whelan *et al*. 2018). We observed the five growing season (April–August) average net uptake of COS in the SMEAR II forest totaling 58.0 gS ha^{-1} with 37% inter-annual variability (Vesala *et al.* 2022). COS is a promising proxy for photosynthesis as plants take up COS through their stomata in a similar way as CO_2, but it is not emitted back to the atmosphere unlike CO_2. Regarding ozone (O_3), seasonal and interannual variability of O_3 deposition fluxes over a forest stand were also analysed using a decade of observations. Rannik *et al.* (2012) found the highest O_3 deposition velocity (4 mm s^{-1}) in SMEAR II during the peak growing season, and the lowest during winter (1 mm s^{-1}). The day-time non-stomatal deposition of O_3 varied from 26% to 44% from the start of the growing season until the end. Although the non-stomatal conductance was partly explained by vapour pressure deficit (VPD), temperature and monoterpene concentration, it was concluded that other processes and/or surface chemical reactions with unknown compounds may play a role (Rannik *et al*. 2012).

The dynamics of wetland GHG exchange have been studied in Siikaneva since 2004 (Aurela *et al.* 2007, Rinne *et al*. 2007a, Riutta *et al.* 2007, Fig. 4). Siikaneva is a protected wetland complex of ca. 12 km^2 that mainly consists of oligotrophic fen vegetation (aapa mire) with ombrotrophic raised bog areas. EC measurements were initiated at the fen site (61°500 N 24°120 E) in 2004 for CO_2 and in 2005 for CH_4 and have been carried on ever since. In the bog site (61°5027 N, 24°101 E) that has less water exchange than the fen site, the EC measurements of CO_2 and CH_4 were carried out between 2011 and 2016 (Rinne and Tuittila 2012). At both sites, EC measurements were accompanied with chamber measurements and ecological studies. The annual CO_2 sink was on average higher in the fen than in the bog, but net ecosystem exchange (NEE) also varied more in the fen than in the bog. In the fen site, annual NEE ranged between sink of 96 g C m^{-2} and source of 27 g C m^{-2}, with an 8-year mean of –53 g C m^{-2} (Rinne *et al.* 2018). For the bog site, annual NEE (converted from growing season values to annual estimates based on Wilson *et al.* (2016)) ranged between sink of 50 g C m^{-2} and source of 1 g C m^{-2}, with a 6-year mean of –36 g C m^{-2} (Alekseychik *et al.* 2021b). Methane is important not only to the GHG exchange but for the carbon balance of the wetland systems, being nearly 20% of the annual net CO_2 uptake at these ecosystems (Rinne *et al.* 2018, Korrensalo *et al.* 2018, Alekseychik *et al*. 2021b) produced throughout the whole peat profile (Putkinen *et al.* 2009) and throughout the annual cycle (Juottonen *et al*. 2008). Methane emission was slightly higher in the fen (10-year average 9.7 g C m^{-2}) than in the bog (estimated 6-year average 8.2 g C m^{-2}). Similarly to NEE, methane emission had higher interannual range of 6.0 to 14 g C m^{-2} in the fen than the 7.4 to 9.7 g C m^{-2} in the bog. In the bog, ebullition formed up to 8% of the total methane emission during the peak season (Männistö *et al*. 2019). The temporal variation of ecosystem level methane emission is observed to depend mostly on peat temperature and gross primary production (GPP, Rinne *et al*. 2007a, Rinne *et al.* 2018). Methane emission seems to be quite insensitive to observed short term water table variations, while seasonal droughts have an effect on both

methane and carbon dioxide exchange (Rinne *et al.* 2020). Field observations have been combined with the development of HIMMELI process model for calculating methane emissions from wetlands (Raivonen *et al.* 2017). Model results indicate that the CH_4 emissions largely depend on the prescribed rate of anoxic respiration and the sensitivity of the total CH_4 emission to other input variables is mainly mediated via the concentrations of dissolved gases, in particular, the O_2 concentrations that affect the CH_4 production and oxidation rates.

At Pallas Lompolojänkkä wetland, the annual CO_2 balance averaged –42 g C m^{-2} yr^{-1} in 2006–2010 (Aurela *et al.* 2015a), indicating a slightly smaller CO_2 sink as compared to Siikaneva. The annual CH_4 emission on the other hand, has been significantly higher, up to 20 g CH_4-C m^{-2} yr^{-1}, at Lompolojänkkä as compared to many other Nordic wetlands (Lohila *et al.* 2016, Rinne *et al.* 2020). The spatial variation of CH_4 emission has been shown to be closely linked to the proximity of the stream (Zhang *et al.* 2020).

In contrast to forests and wetlands, lakes in the boreal landscape are carbon sources with distinct flux maxima in the spring and autumn at the time of mixing periods (Huotari *et al*. 2011, Mammarella *et al.* 2015). However, continuous measurements over several years have revealed that contrary to the common paradigm, boreal brown water lakes, surrounded by coniferous forests and wetlands, regularly act as a carbon sink for a short period in mid-summer (Huotari *et al.* 2011, Golub *et al*. 2021). According to the long-term GHG flux measurements, the amount of carbon vented out from a lake, whether surrounded by managed or pristine coniferous forest, corresponds up to 10% of the NEE of the surrounding terrestrial ecosystem (Huotari *et al*. 2011). At Lake Pallasjärvi, which is an oligotrophic subarctic lake, the emission of CO_2 into the atmosphere was about 120 g CO_2 m^{-2} yr^{-1} (Lohila *et al.* 2015), which was considerably less than in the Huotari *et al.* (2011) study at a polyhumic lake in southern Finland.

EC measurements have been extended also to urban areas to continuously monitor the surface fluxes of momentum, energy, water, CO_2 and aerosol particle number (Vesala *et al.* 2008, Järvi *et al.* 2009a, Nordbo *et al.* 2013). Using EC observations from two sites in Helsinki, representing semi-urban and densely built urban neighborhoods, we have shown direct correlation between carbon emissions and urban surface cover (Nordbo *et al.* 2012), derived empirical relationships for urban biogenic emissions and mixed fleet emission factors for CO_2 and particles (Järvi *et al.* 2009b, 2012), and examined uncertainties of urban EC measurements (Järvi *et al*. 2018). Detailed information has been complemented with urban land surface modelling to examine in detail the emission strengths and variability of CO_2 and energy emissions in Helsinki (Järvi *et al*. 2019) and large-eddy simulation modelling to quantify source areas and uncertainties of urban EC observations (Auvinen *et al.* 2017).

Overall, long-term experience has led to substantial advances in GHG and other trace gases measurement techniques, including the development of state of the art tools (Mammarella *et al.* 2016) and methodologies (Mammarella *et al*. 2009, Taipale *et al*. 2010, Nordbo *et al*. 2014, Rantala *et al*. 2014, Rannik *et al*. 2016, Kohonen *et al*., 2019) to estimate ecosystem scale gas fluxes and their uncertainty, also within the context of international infrastructures such as ICOS (e.g. Franz *et al.* 2018, Nemitz *et al.* 2018; Rebmann *et al.* 2018; Sabbatini *et al.* 2018). Beside methodological issues, the underlying physical processes affecting fluxes have been investigated. We have shown that the canopy morphology and atmospheric stability affects the turbulent flow characteristics inside the canopy (Launiainen *et al*. 2007), and how, in particular during stable night-time conditions, the subcanopy flow decouples from the one above canopy (Alekseychik *et al.* 2013), suppressing the vertical turbulent mixing and enhancing the non-turbulent transport of matter (Mammarella *et al.* 2007, Rannik *et al.* 2009a).

Tree greenhouse gas emissions and growth

In addition to ecosystem scale GHG flux measurements, we have developed chamber techniques to measure GHG fluxes from different ecosystem components, mainly tree branches

(Hari *et al.* 1999a, Aalto *et al.* 2014, 2015), stems (Vanhatalo *et al*. 2015, Rissanen *et al*. 2020, Lintunen *et al.* 2021) and forest floor (Pumpanen *et al*. 2004, Pihlatie *et al.* 2013, Koskinen *et al*. 2014, Korkiakoski *et al*. 2017). As described earlier, measuring and modelling photosynthetic process of trees, its environmental drivers and endogeneous regulation have received great attention in the CoE ATM (e.g. Mäkelä *et al.* 2004, Hari *et al.* 2017b). Similarly, its variability across sites and years has been studied e.g. by Kolari *et al.* (2007) and Gea-Izquiedro *et al*. (2010). Long-term studies on trees also allowed development of a new whole-tree level theory on the optimization of stomatal control in leaves based on maximizing the instantaneous phloem transport rate (Nikinmaa *et al.* 2013). Understanding stomatal regulation is crucial in predicting the impact of changing climate on tree gas exchange. Later, we have further developed the process understanding of photosynthesis with an approach that includes the effect of non-stomatal limitations in models of tree gas-exchange (Hölttä *et al.* 2017, Lintunen *et al*. 2020, Salmon *et al*. 2020). Other examples of research related to tree gas exchange processes include studying tree gas exchange during drought (Duursma *et al*. 2008, Salmon *et al*. 2020) or cold stress (Lintunen *et al.* 2014, Lindfros *et al.* 2015), modelling and measuring of CO_2 fluxes from tree stems (Hölttä and Kolari 2009, Lintunen *et al.* 2021), and measuring phloem transport in trees (Paljakka *et al*. 2017) that is linked to tree gas exchange via e.g. the sink control of photosynthesis. In addition to CO_2 and H_2O, we have studied the exchange of CH_4 and N_2O from tree stems and canopies (Machacova *et al*. 2016, 2019) to understand their role in the GHG balance of forests.

Besides improving process understanding, the results of tree ecophysiological studies can be used for constructing ecosystem level GHG flux and budget estimates. Small-scale observations are upscaled to stand level by models that incorporate the responses of photosynthesis and respiration to environmental drivers and the description of the spatial variability of the drivers within forest stand (Mäkelä *et al.* 2006), most notably the variability of light (Vesala *et al*. 2000). The upscaling results also provide independent reference data for evaluating the accuracy and precision of GHG fluxes and budgets obtained by micrometeorological methods. Synthesis of carbon, nitrogen and water balances determined with different methods was presented by Ilvesniemi *et al*. (2009, 2010), Korhonen *et al.* (2013) and Kolari *et al*. (2009). This approach of combining bottom-up upscaling with direct measurement has been also applied for other gases than GHGs, for instance upscaling of O_3 deposition and comparison with EC (Altimir *et al.* 2006). The sub-ecosystem scale studies can also be used for studying the climate sensitivity of different ecosystem components. Kulmala *et al.* (2019) studied year-to-year variation in ecosystem photosynthesis at the subarctic SMEAR I station and found that the year-to-year variation is minor in tree canopy and smaller compared to that of ground vegetation. Similarly, Matkala *et al*. (2021) found using shoot chamber and EC data at SMEAR I that subarctic conifers showed high resilience in the CO_2 and water fluxes to extreme events.

Increasing availability of long-term EC data has made it possible to construct canopy-scale models of photosynthesis that are directly parameterised using EC data. Mäkelä *et al.* (2008) constructed a light use efficiency (LUE) based daily time-step model of canopy GPP that was statistically fitted to data from several flux sites in temperate and boreal regions. This approach was further developed by Peltoniemi *et al.* (2015) to include simple soil water dynamics and evapotranspiration. The resulting model, PRELES, runs with daily weather data with minimum site information including fraction of absorbed photosynthetically active radiation and soil water holding capacity. A Bayesian calibration study including data from several boreal flux sites suggested that the model can be used with generic parameter values across boreal coniferous forests (Minunno *et al.* 2016). A further development was to estimate climate and vegetation type specific parameters for PRELES, extending its use from boreal to global (Tian *et al.* 2020).

In addition to shoot and ecosystem level flux measurements and modelling, we are also investigating and developing the potential of optical methods to track the dynamics of photosynthesis across spatial and temporal scales, from sec-

onds to seasons, from leaves to satellite pixels (Porcar-Castell *et al*. 2021). Being spatially explicit, optical data such as spectral reflectance or chlorophyll fluorescence offer excellent possibilities to integrate biophysical processes across scales, for example, via the assimilation of satellite solar-induced fluorescence into land-surface models to improve surface GPP estimates (Bacour *et al.* 2019, Raczka *et al.* 2019). Importantly, the use of spectral reflectance and chlorophyll fluorescence data requires a sound and quantitative understanding of the mechanistic connection between the signals and the factors that control their variation across scales, which has been one of our core activities (Porcar-Castell *et al.* 2014, 2021). Temporally, we have elucidated the factors that couple/decouple the seasonal connection between the photochemical reflectance index (Porcar-Castell *et al.* 2012) or the emission of chlorophyll fluorescence (Porcar-Castell 2011, Zhang *et al*. 2019) and photosynthesis. Spatially, we use radiative transfer models to quantify how the information content of the signals scales from the leaf to the plant canopy under the action of canopy structure (Liu *et al*. 2019), light gradients (Atherton *et al*. 2017), and their dynamics (Xu *et al.* 2021). Lately, we are also developing new methods for the standardization (Rajewicz *et al.* 2019), measurement (Atherton *et al*. 2019, Van Wittenberghe *et al.* 2021) and interpretation (Atherton *et al.* 2016) of multiscale reflectance and fluorescence data.

The long-term flux measurements together with data on environmental factors have allowed detailed tree growth analysis (Kulmala *et al.* 2016b, 2017a) and combined with modelling, the analyses have revealed that annual changes in photosynthesis are reflected to the secondary and primary growth of pines in different manners (Schiestl-Aalto *et al*. 2015). Growth measurements and modelling have allowed for analyzing how photosynthesized carbon is allocated into the growth of different tree organs, energy production or storages and how these allocation patterns differ seasonally and within longer time scales (Schiestl-Aalto *et al.* 2013, 2015, 2019). We have found that in our conditions, temperature variation alone explains by far the most part of the daily growth variation, whereas other factors such as photosynthesis or drought affect growth processes at longer timescales. The combination of measurements and process-based models has also enabled quantifying the unmeasurable sink of carbon by root symbionts which based on our analysis is about 6% of the annual GPP (Schiestl-Aalto *et al.* 2019).

During the CoE ATM, we developed a better understanding of the dynamics of cambial growth and its short-term responses to environmental conditions and photosynthetic production. Hölttä *et al*. (2010) demonstrated that the dynamics of cambial growth can be modelled using a mechanistic whole tree level framework including photosynthesis and xylem and phloem transport and a detailed description of cell division, enlargement and cell wall thickening. Chan *et al.* (2016) managed to derive the dynamics of short-term cambial growth from simultaneous measurements of xylem and whole tree diameter change, and show how temperature and water availability affects cambial growth. Further, Chan *et al.* (2018) demonstrated how the short-term dynamics of cambial growth and growth respiration were tightly connected to photosynthetic production.

Ding *et al.* (2020) were able to detect the timing and drivers of growth of fine roots with a novel fine root growth measurement technique. Ryhti *et al.* (2021) quantified the partitioning of forest floor CO_2 source flux to components of tree root, ground vegetation, fungal and heterotrophic respiration. Those results increase understanding of belowground processes and also allow for a more comprehensive understanding of whole tree carbon allocation.

Greenhouse gas fluxes from soil and forest floor

To gain a deeper understanding of the net GHG exchange of an ecosystem, we need to study not only tree processes, but also soil and forest floor. Forest floor plays a major role in the carbon balance of forest ecosystems, especially in boreal forests. Due to relatively open tree canopy and high light conditions on the soil surface, the forest floor vegetation may contribute significantly to the CO_2 uptake, especially in young

forests. The GPP of ground vegetation was first quantified by Kolari *et al.* (2006) and later, the momentary rate and drivers for the photosynthesis of the most common ground vegetation species were further studied by Kulmala *et al.* (2008, 2011a). In Värriö at SMEAR I, the share of ground vegetation in the total ecosystem GPP flux is approx. 43–49% (Kulmala *et al*. 2019) whereas in Hyytiälä at SMEAR II, the GPP of ground vegetation is annually approx. 146 g m^{-2} (Kulmala *et al.* 2011b), which is ~13% of the GPP of the whole ecosystem.

Carbon is transferred from soil to the atmosphere in soil respiration, which originates from two major sources; decomposition of soil organic matter (heterotrophic respiration) and respiration of roots and associated mycorrhizal fungi (autotrophic respiration). The majority of soil respiration originates from the soil surface humus layer and the surface of the mineral soil which contributed about 70% of the total soil CO_2 emissions at the SMEAR II station in Hyytiälä (Pumpanen *et al*. 2008). To understand interacting soil processes, the forest floor CO_2 flux was partitioned using the trenching method (root exclusion) combined with vegetation removal treatments. The respiration of tree roots contributed 48%, heterotrophic soil respiration 30%, respiration of ground vegetation 18% and the respiration of hyphae of mycorrhizal fungi 4% of the total soil respiration (Ryhti *et al.* 2021). Heterotrophic respiration increased in the trenched treatments without ground vegetation over time, due to the "Gadgil effect". Our results indicate intense interaction between plant roots and soil microbes that affect forest floor CO_2 efflux (Ryhti *et al.* 2021) and soil carbon stocks. For this reason, the soil microbial communities, microbial interactions between trees and ground vegetation and the impact of decomposition of microbial necromass to soil organic matter stability were investigated at SMEAR II site or in the greenhouse and laboratory experiments (Adamczyk *et al.* 2016, 2019a,b, Kieloaho *et al.* 2016, Santalahti *et al.* 2016, Sietiö *et al.* 2018, 2019). Depending on the development stage of the forest, the proportion of autotrophic and heterotrophic respiration may be remarkably different and has a large seasonal variation as a major part of the autotrophic respiration is originating from the recently assimilated carbon which is transported to the root system and emitted to soil in the respiration of fine roots and the associated fungi (Palonen *et al.* 2018, Pumpanen *et al*. 2015, Ryhti *et al*. 2022).

Disturbances affect significantly the carbon fluxes in forest ecosystems. For example, wildfire or windthrow may kill a large proportion of trees and decrease the photosynthetic capacity and may increase the decomposable material in soil surface. Clear-cutting can cause huge emissions of CO_2 and N_2O and minor emissions of CH_4 in drained wetland forest, while the partial harvest does not significantly affect the soil GHG fluxes (Korkiakoski *et al*. 2019, 2020). Similar to clear-cutting and burning of slash (Virkkula *et al*. 2014), wildfires also result in drastic changes in the soil microbiome (Sun *et al.* 2016, Zhou *et al.* 2020) and change the greenhouse gas fluxes between soil and the atmosphere (Köster *et al.* 2014, 2015a, 2016). Especially, the CO_2 effluxes decrease as a result of fire as a large proportion is originating from roots system which is adversely affected by fire (Köster *et al.* 2014) but the fire also increases the soil CH_4 sink (Köster *et al.* 2015a).

At SMEAR II station in Hyytiälä, long-term measurements of forest floor exchange show that the site has been a strong CH_4 sink during the almost 20 years of measurements (Skiba *et al.* 2009, Vainio *et al.* 2021), and a small but significant source of N_2O (Pihlatie *et al*. 2007). Extensive forest floor flux measurements revealed that small moist patches of the forest floor emit significant amounts of CH_4 during wet periods, however, these wet spots do not markedly decrease the sink strength of the soil in the forest. Similar to mature trees in the forest (Machacova *et al.* 2016), forest floor shrubs *Vaccinium myrtillus*, *Vaccinium uliginosum* and small Scots pine seedlings emit small amounts of CH_4 while the belowground parts consume CH_4 (Halmeenmäki *et al.* 2017) but the net fluxes of the soil surface still showed net CH_4 uptake into the forest floor.

At Kenttärova spruce forest growing on podzolic moraine in Pallas, the whole forest floor turned into a CH_4 source after a very wet growing season for more than half a year time (Lohila *et al.* 2016). As a result, the landscape scale

methane emission — 10% of the landscape being open wetlands — more than doubled during a wet year as compared to a more normal year, and that was due to the upland forest soil, not the wetlands. Recently it was confirmed that in a meteorologically normal year, at the peak growing season, the whole catchment acts as a source of CH_4 (Räsänen *et al.* 2021). For CH_4 production to take place in upland soils, inundated conditions and input of fresh carbon is needed, which was proven in lab experiments done with Kenttärova soils (Korkiakoski *et al.* 2021).

Herbivory also affects the greenhouse gas fluxes between the soil and the atmosphere. Our studies in reindeer grazed areas close to the SMEAR I station in Värriö in Eastern Lapland have shown that grazing significantly reduced lichen coverage and biomass and increased the biomass and coverage of mosses (Köster *et al.* 2015b, 2017, 2018). These grazing-induced changes in vegetation composition affect both positively and negatively GHG fluxes from the forest field layer in a northern boreal forest (Köster *et al.* 2018). Higher CO_2 emissions and CH_4 uptake were detected from grazed areas, and the gas fluxes were affected by soil temperature, moss and lichen coverage (Köster *et al*. 2015b, 2018). Non-grazed areas were a small N_2O sink.

Volatile organic compounds (VOC)

Volatile organic compounds emitted by natural ecosystems have major effects on atmospheric chemistry and new particle formation. Therefore, their sources and emissions have been widely studied in the CoE ATM. We have developed new methods for dynamic chamber (Kolari *et al.* 2012, Aaltonen *et al.* 2013, Vanhatalo *et al*. 2015, Barreira *et al*. 2017) and ecosystem scale flux measurements (Rantala *et al.* 2014, 2015) as well as for reactivity measurements (Mogensen *et al.* 2015, Praplan *et al*. 2017) and measurements of new compound groups (Hellén *et al*. 2012, Kieloaho *et al*. 2013, 2016, Helin *et al*. 2017, 2020). The observations cover long-term ecosystem scale fluxes (e.g. Rantala *et al*. 2015, Schallhart *et al*. 2018) and leaf level measurements from main tree species (e.g. Hakola *et al*. 2006, 2017, Hellén *et al*. 2021) as well as other

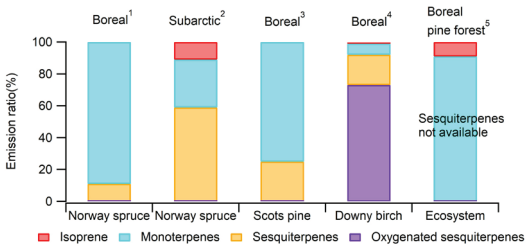

Fig 5. Relative contributions of different compound groups in the foliar mass emissions of different tree species and in a Scots pine forest stand during the peak emission period (July). References: [1]measured at SMEAR II site in 2019 (246 samples) using the methods described in Hellén et al. (2021), [2]measured at Pallas in Northern Finland in 2020 (383 samples) using the methods described in Hellén et al. (2021), [3]Hakola et al. (2006), [4]Hellén et al. (2021), [5]Schallhart et al. (2018).

forest compartments, such as tree trunks, soil and understory vegetation (e.g. Kieloaho *et al.* 2013, 2017, Rissanen *et al.* 2016, 2020, Mäki *et al.* 2017, 2019a, b, Vanhatalo *et al*. 2015). We have also conducted field and laboratory experiments to reveal processes behind emissions and their linkages with biotic and abiotic drivers (e.g. Ehn *et al.* 2014, Bianchi *et al*. 2019). Once emitted to the atmosphere, VOCs can react with oxidants to form less volatile oxidation products. These reactions take place at very different rates depending on the precise VOC and oxidant, and the products can have highly variable tendencies to form aerosols (e.g. Ehn *et al*. 2014, Bianchi *et al.* 2019). In addition to monoterpenes (MTs), we have succeeded in measuring also very reactive compounds, such as sesquiterpenes (SQT), in the emissions and ambient air and in describing the temporal variability in concentrations (Hellén *et al.* 2018, 2020).

VOC emissions from trees and forest floor

MTs are clearly the most abundant terpenoid compound group (in mass) emitted from boreal forest vegetation (Rinne *et al.* 2009, Rantala *et al.* 2015, Schallhart *et al*. 2018). The summary in Fig. 5 shows relative contributions of different terpenoid groups emitted from different tree species and whole ecosystem scale. The ecosystem

scale measurements do not contain SQTs and oxygenated sesquiterpenes were only measured in downy birch and boreal Norway spruce emissions. The emission rates are following the diurnal and seasonal temperatures, and in a few cases also variations in irradiation levels, as is the case with isoprene. Therefore, highest emission rates are usually measured in mid-summer and during the daytime.

Content and emission of different MTs has been observed also to vary a lot within each coniferous tree species, reflecting the inherited "chemotype" of the trees (Bäck *et al*. 2012, Hakola *et al*. 2017). Two main compounds, α-pinene and Δ^3-carene formed together 40–97% of the monoterpene proportions in both the branch emissions of Scots pines and in the air concentrations in the surrounding forest. The emissions were dominated either by α-pinene (ca. 10% of the trees), or Δ^3-carene (ca. 20%). This has large implications to the models describing the boundary layer atmospheric chemistry, since these MT isomers have very different chemical reactivities with the main oxidants and considerable differences in secondary organic aerosol (SOA) formation (Kurtén *et al*. 2017, Thomsen *et al.* 2021).

Continuous, long term observations on Scots pine needle and shoot VOC emissions has revealed that the conventional view on temperature and light as emission controlling factors is likely too simplifying. For example, new foliar biomass growth has been found to be a significant source of many VOCs (Aalto *et al.* 2014). Also the photosynthetic recovery in spring, especially during the period when light use efficiency is still low after the cold winter season, can cause a transient increase in both MT synthesis and emissions (Aalto *et al*. 2015).

In addition to forest canopy, Scots pine stems have been identified as a small source of MT (Vanhatalo *et al.* 2015). The stem MT emissions are generally highest at mid-summer and in daytime, but large emission peaks may occur in spring (Vanhatalo *et al*. 2015). Drivers for stem MT emissions include temperature, soil humidity and potentially pressure of resin (high in MT content) within the stem (Rissanen *et al.* 2016, 2020, Vanhatalo *et al*. 2015).

The forest floor accounted 2–93% of the forest stand MT emissions (Mäki *et al*. 2019a). The highest MT emissions were measured from the decomposing litter in autumn, whereas the understorey vegetation was a MT sink due to adsorption to the leaf surfaces (Mäki *et al.* 2017). MT and SQT concentrations were high in the organic soil and the top mineral soil due to high abundance of decomposing litter, tree roots and root-associated microbes (Mäki *et al*. 2019b). Stand type regulates MT and SQT emissions from the forest floor in hemiboreal and boreal climates (Mäki *et al.* 2019c). The decaying wood is an MT and SQT source as fungal species synthesize these compounds to interact with other fungal species based on the laboratory experiment (Mali *et al*. 2019). Noteworthy emissions of MT and SQT were also observed in situ measurements from the decaying wood disks and logs.

Barreira *et al.* (2017) observed that α-pinene and Δ^3-carene constituted 80-90% of the measured monoterpenes in the soil chambers. Solid phase microextraction technique was successfully utilized for the sampling of BVOCs together with portable gas-chromatograph-mass spectrometer (GC-MS) analysis on the site. A good correlation was found with the Proton-transfer-reaction mass spectrometer (PTR-MS) results. Nearby sawmill was found to be one emission source affecting the results when the wind was from southeast (Barreira *et al*. 2017, Liao *et al.* 2011). We have also found that soil at Hyytiälä is a source of dimethylacetamide and a sink of diethanolamine (DEA), while the direction of estimated flux was dependent on soil temperature, soil water content and soil pH, but also on soil solution concentration of alkyl amines (Kieloaho *et al*. 2013, 2017).

Wetlands have been found to be strong emitters of isoprene (Haapanala *et al*. 2006, Hellén *et al.* 2006, 2020) while MT emissions of only 10% of isoprene emissions (Hellén *et al.* 2020) have been observed at a subarctic wetland. However, SQT emissions at the subarctic wetland site were several times higher than MT emissions, especially in early summer. Main emitted SQTs were tentatively identified as α-farnesene, cadinene and β-cadinene. Due to their high reactivity, they have strong impact on local atmospheric chemistry.

Ecosystem scale emission measurements of VOCs

Since 2000, the potential of the EC method to measure VOCs exchange also at the ecosystem scale has been evident (e.g. Rinne *et al.* 2001, Karl *et al.* 2002). Initially, only two days of methanol flux data was published. However, the development of improved methods and more powerful instrumentation led to longer datasets, which covered a greater range of VOCs. Rinne *et al*. (2007b) presented four days of ecosystem exchange of four VOCs, as well as their calculated chemical degradation. These first measurements of ecosystem exchange in Hyytiälä were obtained using Disjunct Eddy Covariance (DEC; Rinne and Ammann 2012, Rinne *et al.* 2021) and they confirmed the enclosure measurement results, that the major emitted VOCs are the monoterpenes, methanol and acetone. From 2006 onwards, the DEC measurements were made semi-continuously in Hyytiälä. In 2010, the VOC flux measurement method was changed to the gradient technique, as Rantala *et al.* (2014) showed that with the used setup of the measurements and the low concentration environment, the gradient technique was more reliable than DEC. Schallhart *et al.* (2018) reported the first 10Hz EC fluxes using a Proton-Transfer-Reaction — Time Of Flight (PTR-TOF) in Hyytiälä. Due to the capability to the PTR-TOF to measure whole VOC spectra with high frequency, this setup was already able to measure the exchange of 25 different VOCs.

The micrometeorological flux measurements have shown a wide variety of VOCs emitted from pine forest ecosystems (Schallhart *et al*. 2018). A combination of laboratory experiments and analysis of micrometeorological MT flux data have indicated that around half of the monoterpenes emitted from pine forest originate as de novo emission directly from synthesis, the rest being evaporative emission from resin (Ghirardo *et al*. 2010, Taipale *et al.* 2011, Rantala *et al.* 2015). Micrometeorological flux measurements have also revealed a seasonal trend in the normalized emission potential on monoterpenes (Taipale *et al.* 2011, Rantala *et al.* 2015). The earlier micrometeorological flux measurents using surface layer gradient technique and gas chromatography — mass spectrometry analysis has revealed the MT emissions from Scots pine forests to be dominated by α-pinene (60-70%) and Δ^3-carene (12-26%) (Rinne *et al*. 1999, 2000). The chemical reactions strongly modify the above-canopy fluxes of highly reactive compounds, such as some sesquiterpenes, but are less important for compounds with chemical lifetimes about half-an hour or more, such as isoprene and monoterpenes (Rinne *et al*. 2007b, 2012).

Development of air sampling and analyses techniques of VOCs

In addition to emission measurements of VOCs, we measure VOC concentrations in the atmosphere. We have developed techniques to sample atmospheric VOCs, and in this section, we describe in more detail some of this sampling development. Due to their important role in the new particle formation, aliphatic amines are an especially interesting group of VOCs. To measure the low gas phase concentrations of e.g. ammonia, primary, secondary and tertiary amines, solid phase microextraction (SPME) with on-fiber derivatization was developed for air samples (Parshintsev *et al.* 2015). Further developments include SPME Arrow sampling for the collection of amines (Helin *et al.* 2015) and functionalized mesoporous silica coated SPME Arrow sampling for aliphatic amines (Lan *et al*. 2019a). The developed technique enabled collection of samples even at quite low temperatures and to determine diethylamine and triethylamine with very low concentrations. We have also developed a fully automated online dynamic in-tube extraction (ITEX) for continuous sampling and analysis of VOCs in air. With this sampling approach combined with gas chromatography mass spectrometer, we were able to observe day/night variations for very low concentrations of anthropogenic VOCs such as toluene, p-xylene, and 1,2,3-trimethylbenzene (Lan *et al*. 2019b).

The applicability of a drone as a carrier for miniaturized air sampling systems (SPME Arrow and ITEX) has been proved in the recent years in the CoE ATM (Ruiz-Jimenez *et al*. 2019, Lan *et al.* 2021). The versatility, flexibility and easy

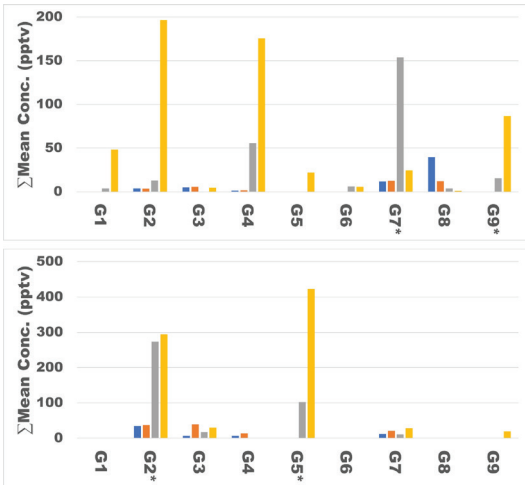

Fig 6. Variation of the gas phase (upper figure) and aerosol particle composition (lower figure) with the sampling height for samples collected at the SMEAR II station (blue: 5 m and orange: 50 m sampling heights) and Qvidja farm (grey 50 m and yellow 150 m sampling heights). G1, Organic oxygen compounds; G2, lipid-like molecules; G3, carbonyl compounds; G4, alcohols; G5, organonitrogen compounds; G6, benzene and its substituted derivates; G7, ketones; G8, aldehydes; and G9, prenol lipids. *Mean concentration found in the samples divided by 10 (Lan et al. 2021).

operation of the sampling systems with a drone allow sampling at different heights and positions even in places that are difficult to access, being a good tool also for low cost vertical profiling studies and offering an interesting alternative to other traditional sampling platforms such as building/tower, balloons, and aircrafts. In case of ITEX, high selectivity can be obtained with careful selection of the trapping material and by small accessories prior the sorbent trapping of VOCs (particle removal, removal of ozone, water and other non-targeted compounds). Utilizing drone sampling platform, air samples were collected during summer 2019 at the SMEAR II station and agricultural Qvidja farm (Finland) using SPME Arrow coated with two different coatings, and ITEX packed with 10% polyacrylonitrile. The total number of VOCs identified was 347. Quantitative/semi-quantitative results were achieved by the use of individual Partial Least Squares Regression models developed using the gas phase standard compounds provided by a permeation system (Ruiz-Jimenez et al., 2020). The simultaneous sampling by active (ITEX) and passive (SPME) systems allowed the clarification of the VOCs distribution between gas phase and particles. Average gas phase VOC concentrations were from 0.1 (toluene, the SMEAR II station) to 680 ng L^{-1} (acetone, Qvidja farm). In aerosols, average VOC concentrations ranged from 0.1 (1,4-cyclohexadiene, the SMEAR II station) to 2287 ng L^{-1} (megastigma-4,6,8-triene, Qvidja farm). Clear differences were observed for gas phase and aerosol particles based on the sampling site and height (see Fig. 6, Lan *et al*. 2021).

Ambient VOC concentrations and their atmospheric chemistry

The long-term observations allow us to draw conclusions of the interannual variability and the seasonal cycle of ambient concentrations of VOCs. The concentration measurements reflect both the emission rates from all sources at the concentration footprint area, which is larger than flux footprint, but also the impact of chemical reactions and turbulent meteorological factors on the chemistry and mixing of the atmospheric boundary layer. The concentrations in the boreal forest at SMEAR II site are dominated by monoterpenes, methanol, acetone and acetaldehyde, and frequently peak in July, but considerable year-to-year variation exists (Fig. 7).

Species specific ambient air concentrations have been measured in the boreal forest at SMEAR II during two years using in situ GC-MS (Hellén *et al.* 2018). These molecular level results enabled detailed characterization of importance of detected VOCs on local atmospheric chemistry (Fig. 8). Both mono- and sesquiterpene oxidation was dominated by ozone. Major terpenes producing oxidation products at the site were β-caryophyllene and α-pinene, which both are known to have high SOA yields. Oxidation of other VOCs than terpenes showed very minor contributions. Species specific in situ GC-MS measurements were conducted also at a sub-arctic wetland surrounded by the coniferous forests in Northern Finland in 2018 (Hellén *et al.* 2020). Daily mean ambient air concentrations of MTs and

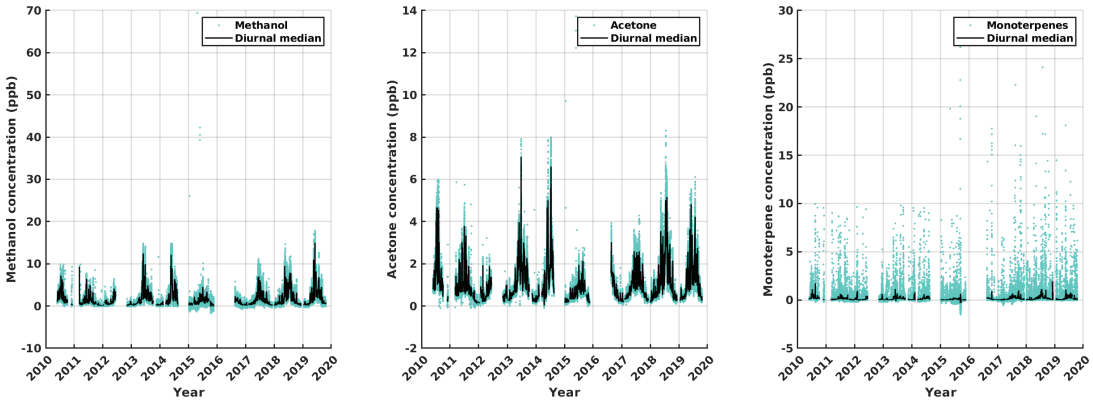

Fig 7. All data (points) and diurnal median concentrations (solid line; measured with the PTR-MS) of methanol, acetone and monoterpenes at 33 m height (> 10 m above the canopy top) over the 10-year measurement period, 2010-2019.

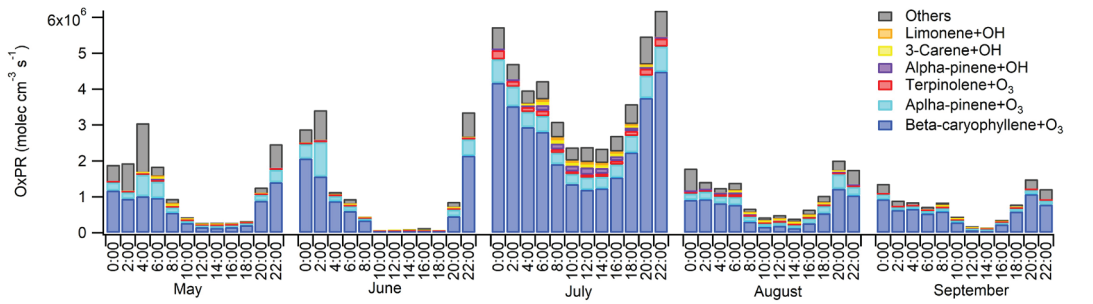

Fig 8. Diurnal variations in production rates for secondary organic compounds (OxPRs) from the oxidation of terpenes with different oxidants (hydroxyl radical, OH; ozone, O_3; and nitrate radical, NO_3) in the boreal forest at SMEAR II in 2016. OxPRs were calculated from the concentrations of VOCs and O_3 measured at the site and from the modelled concentrations of OH and NO_3 as described in Hellén et al. (2018).

SQTs at this site had strong correlation with ozone concentrations indicating that BVOC emissions from these ecosystems have a strong impact also on the local ozone levels.

Although the oxidation of other VOCs than terpenes have smaller contributions to SOA formation, clear enhancement of NPF and SOA formation has been seen in the presence of dimethylamine (DMA). Laboratory and field studies on reaction between pinonaldehyde and DMA, and on the ozonolysis of α-pinene in the presence of DMA have detected several mid to low vapor pressure nitrogen-containing compounds, and some of the oxidation products (e.g. $C_{11}H_{20}O_2N$ and $C_{12}H_{22}O_2N$) obtained in the flow reactor experiments were also detected in the aerosol particles collected in the SMEAR II station utilizing high resolution mass spectrometer techniques (Fig. 9, Duporté *et al.* 2016, 2017). In addition, oxidation products of sesquiterpenes have been synthesized in the laboratory and one of them, β-nocaryophyllone aldehyde, could also be detected from the aerosol particles (Parshintsev *et al.* 2008).

Total hydroxyl radical (OH) reactivity can be used to investigate how well reactive compounds (in ambient air or from emissions) are chemically characterized. Comparing total OH reactivity measured experimentally with the OH reactivity expected from the known chemical compounds present in the air (or in emissions) provide information about the fraction of reactive compounds that are not detected. Total OH reactivity studies have found high fraction of missing (i.e., unexplained) OH reactivity in the boreal forest air (Nölscher *et al.* 2012, Yang *et al.* 2016, Praplan *et al.*

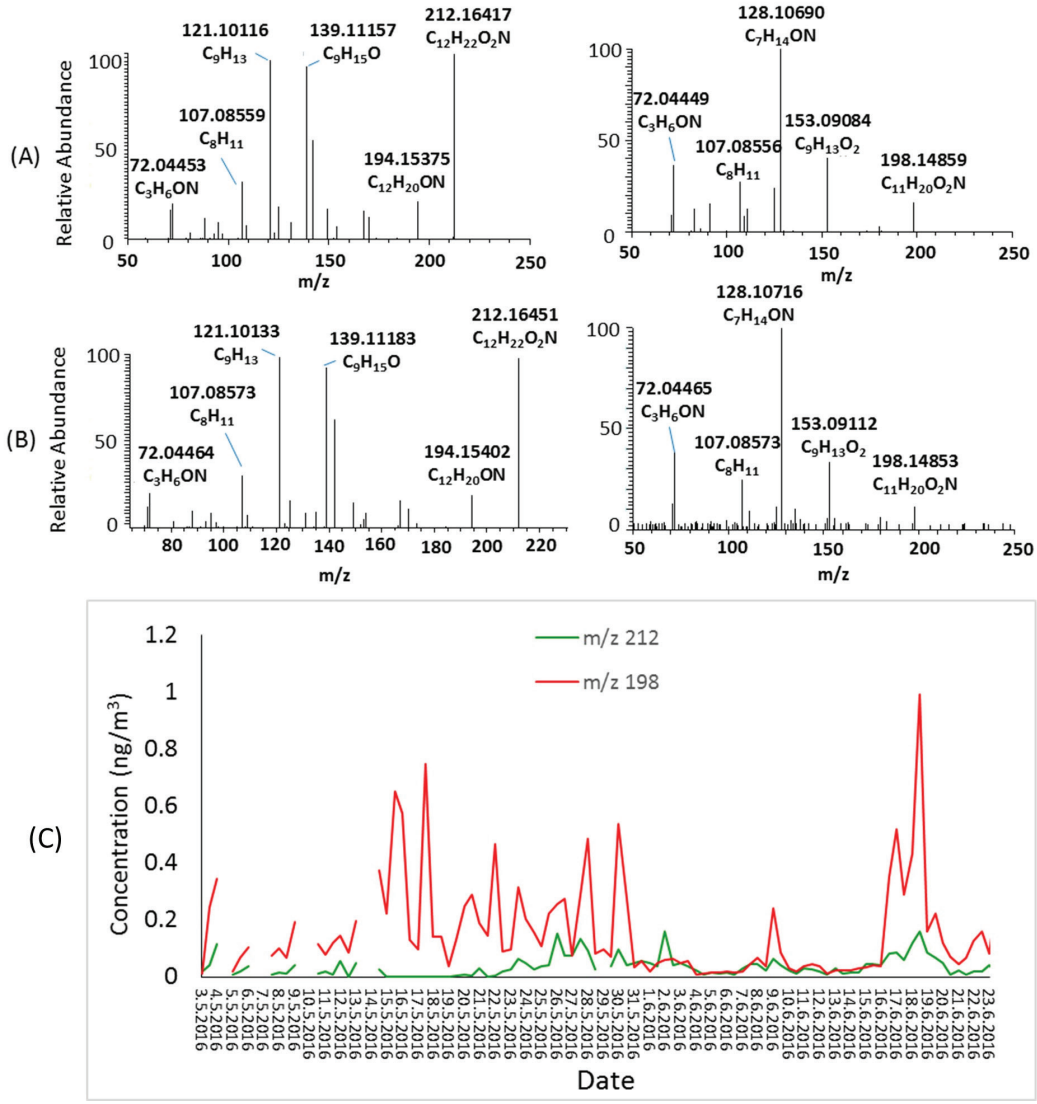

Fig 9. Liquid chromatography (LC)-positive electrospray ionization (ESI+)-Orbitrap tandem mass spectrometry (MS-MS) fragmentation pattern of product ions at m/z 198.148 and 212.164 from (**A**) α-pinene + O_3 + DMA flow reactor experiment and (**B**) PM_1 (i.e. diameter less than 1 micron) sample collected in SMEAR II station in Hyytiälä, Finland (23 May 2016 day), (bottom figure) long time (3 May–23 June 2016) monitoring of the two product ions in the PM_1 samples collected in Hyytiälä (Duporté *et al.* 2017).

2019). This so-called missing reactivity has been suggested to stem from unknown primary emissions or oxidation products of VOCs (often, only the precursors are routinely measured). In our recent studies we showed that, based on model estimates, currently known oxidation products are only able to explain a minor fraction (< 4.5%) of the missing reactivity in the boreal forest air (Praplan *et al.* 2019). Moreover, using young trees, we found large fractions of missing reactivity directly in the direct emissions of main boreal tree species, in particular under stress conditions (Praplan *et al.* 2020).

Aerosol formation and growth

In the previous section, we focused on VOC emissions and their atmospheric concentrations that have a major role in the formation of new atmospheric particles. In this section, we proceed from the VOCs to the formation and growth of atmospheric aerosol particles, because this phenomenon has a dominant role in the global particle number concentration budget (Dunne *et al.* 2016) and it is a major contributor to atmospheric cloud condensation nuclei (CCN) concentrations (Spracklen *et al*. 2008, Merikanto *et al*. 2009, Gordon *et al*. 2017). As a result, atmospheric new particle formation (NPF) has a potential to influence clouds and climate (Makkonen *et al.* 2009, Kazil *et al*. 2010), and to participate in several climatically-relevant feedback mechanisms. In our CoE, we have investigated aerosol formation and growth under laboratory conditions and using various computational methods, developed new instruments for this purpose, and applied all this knowledge to field measurements and their interpretation. The combined effect of all these activities has been a greatly enhanced understanding of atmospheric NPF and its potential influences, along with a number of new research tools to be used in future from process-level atmospheric studies to global climate simulations.

Aerosol precursors

The formation and growth of new aerosol particles is driven by nucleation and condensation of low-volatile vapors (Kulmala *et al.* 2013b). The most important of these aerosol precursors are typically low-volatile organics for the condensational growth (Ehn *et al.* 2014), and sulfuric acid for the initial formation of small clusters (Sipilä *et al.* 2010), often in conjunction with some stabilizing bases such as ammonia or amines (Almeida *et al.* 2013). Under certain atmospheric conditions, also iodine-containing acids can play a key role (Sipilä *et al.* 2016; He *et al*. 2021). The upcoming sections detail the findings of CoE ATM in studying aerosol formation from these, and also other, precursors. However, much of the understanding has developed from the detailed direct observations of these aerosol precursors in the gas phase, which in turn has been largely driven by the developments within the field of mass spectrometry.

The mass spectrometer development within CoE ATM got started with the first atmospheric pressure interface — time of flight (APi-TOF) mass spectrometer that w.e characterized for ambient ion chemistry measurements in 2010 (Junninen *et al*. 2010). This new technique provided a near universal interface for atmospheric pressure sampling. The APi-TOF allows both anion and cation measurements and these key features have now been utilized in laboratory and field studies. The APi-TOF is a superior tool for the detection of molecular ion clusters and has provided detailed information on acid-base (mainly sulfuric acid-ammonia or sulfuric acid-amines) nucleation mechanisms (e.g. Kirkby *et al.* 2011).

The next step in instrumental development features the integration of a chemical ionization inlet on the APi-TOF (CI-APi-TOF). Jokinen *et al.* (2012) demonstrate how this set up is capable of detecting atmospheric sulfuric acid in very low quantities (~4×10^4 molecules cm^{-3}). This technique has since been utilized in dozens of field and laboratory studies, yielding scientific breakthroughs such as the first ambient observations of iodic acid driven particle formation in coastal areas (Sipilä *et al*. 2016) and Highly Oxygenated organic Molecules (HOM) that are key players in growing atmospheric nanoparticles (Ehn *et al.* 2014). The meachanism forming HOM, namely autoxidation (Crounse *et al.* 2013), had been completely underestimated in the atmospheric oxidation of VOCs. Later studies have found that autoxidation is crucial for forming condensable and low-volatile vapors from a variety of VOC types, including monoterpenes (Jokinen *et al.* 2014), sesquiterpenes (Jokinen *et al.* 2016), aromatics (Wang *et al.* 2017), and even alkanes (Wang *et al.* 2021).

The first ambient APi-TOF deployment took place at SMEAR II from April–May 2009 (Ehn *et al.* 2010), while the first CI-APi-TOF measurements were conducted at the same site from March–April 2011 (Jokinen *et al.* 2012). Continuous measurements of aerosol precursors using a CI-APi-TOF began at SMEAR II in 2014 (Sulo

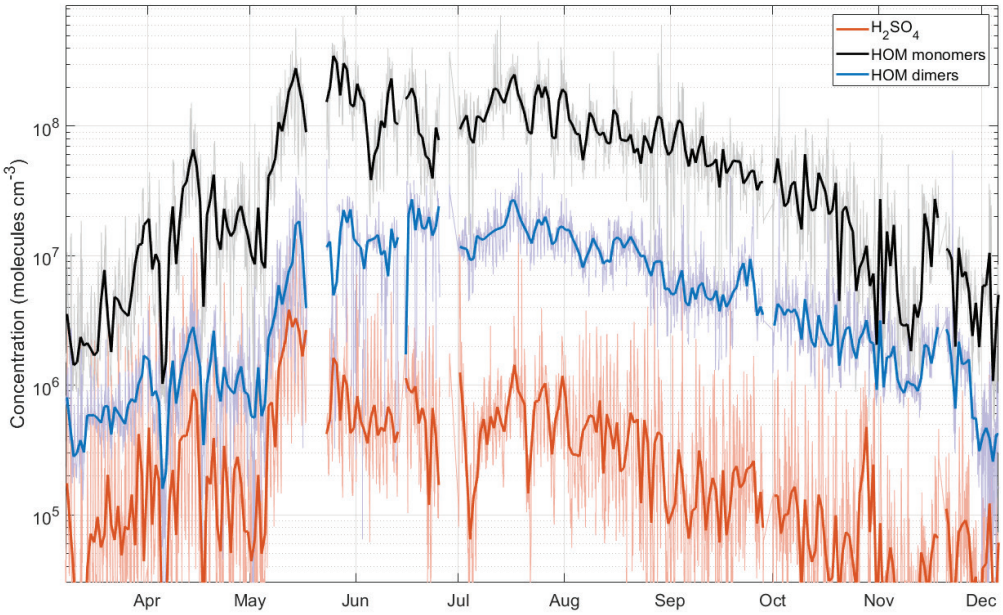

Fig 10. The daily median concentrations of precursor gases measured with CI-APi-TOF at SMEAR II station at 35 m altitude in 2018. The 30 min average of sulfuric acid (H_2SO_4), HOM monomers (m/z 290–430 Th) and dimers (m/z 430–620 Th) is shown in shaded colors displaying the diurnal variation. The HOMs are divided to monomers and dimers based on assumption that most of them are oxidation products of monoterpenes. The used data is analysed by unit mass resolution and the calibration constant is received from sulfuric acid calibration.

et al. 2021), and at SMEAR I in 2019 (Jokinen *et al.* 2022). These data provide crucial support for a variety of aerosol formation studies, as it provides key information on the concentrations of different aerosol precursors like sulfuric acid and HOM (Fig. 10).

Using the knowledge we gained from the early APi-TOF development and observations of sulfuric acid clustering with ambient bases, we developed a high sensitivity detector for ammonia and amines using sulfuric acid and its clusters as the CI-APi-TOF reagent ion (Sipilä *et al*. 2015). We deployed this instrument at the SMEAR II station gaining information of dimethyl amine with concentrations even down to 70 ppqv (parts per quadrillion, 0.07 pptv). Understanding the role of ambient bases and developing detection technologies is an ongoing project. Since contaminants and memory effects often complicate the detection of bases, we took a step to develop chemical ionization inlets to a sheath flow-free design. The result was a multi-scheme chemical ionization inlet (MION, Rissanen *et al.* 2019) that we coupled with the APi-TOF. This inlet also allows fast switching between ion mode and CI-mode that was not possible before, and required separate instruments. We have also been involved in development of online analysis of gas and particle composition (Lopez-Hilfiger *et al.* 2014) using a filter inlet for gases and aeorosols (FIGAERO) inlet that allows the detection of both gas phase aerosol precursors and particle molecular composition simultaneously.

Laboratory studies on new particle formation and growth

The teams participating in our CoE have more than 30-year experience in conducting nucleation experiments and related studies under laboratory conditions. The first results from experimental homogeneous nucleation studies using a laminar flow diffusion chamber from Finnish Meteorological Institute were published in 2000 (Lihavainen *et al.* 2000), as a part of a worldwide joint experiment on homogeneous nucleation.

The objective of this experiment was to measure homogenous nucleation rates of n-pentanol with helium as a carrier gas with all available tested instruments at the time. Measurements with the developed laminar flow diffusion chamber were expanded to a series of alcohols from n-propanol to n-octanol (Hyvärinen *et al.* 2004, Görke *et al.* 2014). Several studies on carrier gas pressure and kind were also carried out (e.g. Brus *et al*. 2008). Homogenous nucleation of water in laminar flow diffusion chamber was measured to extend the earlier experimental data sets, and the measurements were found to agree surprisingly well (Manka *et al.* 2010). The experience from unary homogenous nucleation experiments was utilized in building a flow chamber (Brus *et al.* 2010), which was used for investigating water-sulfuric acid nucleation (Sipilä *et al.* 2010). The roles of different bases on water-sulfuric acid nucleation was also investigated, however no effect was found at the relatively high concentrations used in these experiments (Neitola *et al*. 2014).

Surface tensions and densities of H_2SO_4 + NH_3 + water solutions were measured for the first time as a part of atmospheric nucleation and growth studies (Hyvärinen *et al.* 2005). Through surface tension measurements, we investigated the role of other substances typically found in atmospheric aerosols during new particle formation and growth events. These included sulphuric acid- dimethylamine- water solutions (Hyvärinen *et al.* 2004) as well as aqueous solutions of oxalic, malonic, succinic, maleic, malic and cis-pinonic acids (Hyvärinen *et al*. 2006).

In 2009, the Cosmics Leaving Outdoors Droplets (CLOUD) project started its first experiments at the CLOUD chamber facility at CERN (European Center for Nuclear Research) in Switzerland. The project aims to investigate whether and how ionization from galactic cosmic rays can influence particle and cloud formation, with potential implications for climate. Due to the high cleanliness and controllability of the CLOUD chamber and its suitability for conducting nucleation experiments at atmospherically relevant levels of precursor vapors, the experiments in the CLOUD chamber have greatly advanced our basic understanding of the chemical and physical mechanisms of new particle formation.

In the first major publication from CLOUD, Kirkby *et al.* (2011) showed that binary sulfuric acid-water nucleation, which was previously thought to be one of the main nucleation mechanisms, is inefficient at sulfuric acid levels relevant for atmospheric boundary layer. However, small amounts of ammonia or amines (Kirkby *et al.* 2011, Almeida *et al.* 2013) was found to greatly enhance particle formation rates, making ternary nucleation one of the important pathways. Other major breakthroughs from the CLOUD experiment have been e.g. showing that highly oxygenated organic molecules (HOMs) from alpha-pinene ozonolysis can form (Kirkby *et al.* 2016) and grow (Tröstl *et al.* 2016) particles without the involvement of sulfuric acid, and that nitric acid could participate in rapidly growing particles in cold polluted climates (Wang *et al.* 2020). Dunne *et al.* (2016) presents the most comprehensive evaluation so far on the importance of the CLOUD results on the global atmosphere.

The CoE has had an important contribution to the CLOUD experiment since the beginning, especially by providing the direct measurements of the size distribution and composition of the forming clusters and ions using recently developed instrumentation (see the section about instrument development related to aerosol formation and growth) and lately also by leading the experiments focusing on simulating boreal-forest-type (Lehtipalo *et al*. 2018, Yan *et al.* 2020) and marine-type (He *et al.* 2021) particle formation. Figure 11 shows an example from the experiments aiming to replicate NPF in boreal forest. The Finnish CLOUD team have especially concentrated on probing the chemical composition of the forming clusters and particles (e.g. Keskinen *et al.* 2013, Schobesberger *et al.* 2013), explaining the particle growth mechanisms (Lehtipalo *et al.* 2016, Stolzenburg *et al.* 2020), and studying the effect of ions (Franchin *et al*. 2015, Wagner *et al.* 2017).

Computational aerosol physics

Prior to 2002, particle formation in the atmosphere was modelled almost exclusively based on a classical thermodynamic liquid drop description of even the smallest clusters. Some early quantum

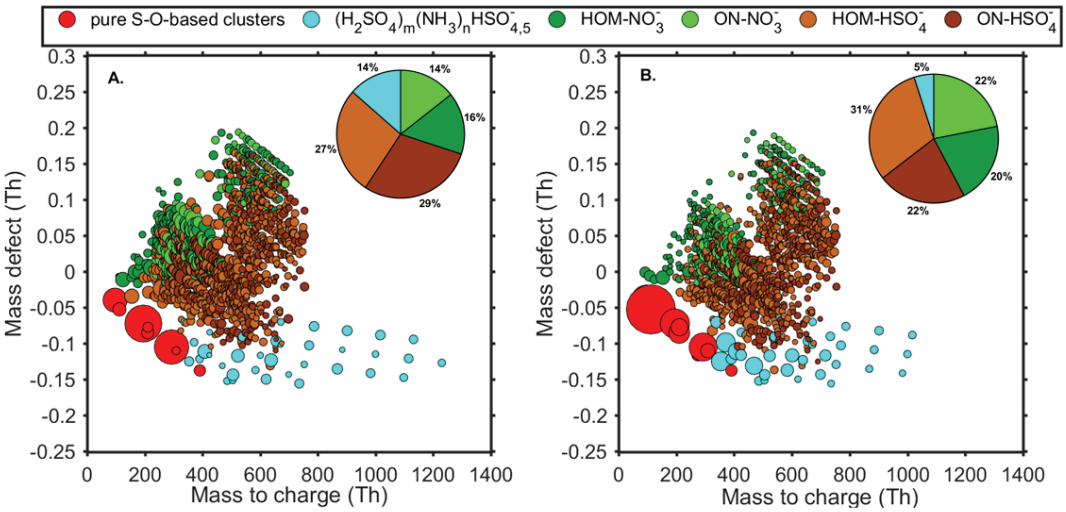

Fig 11. The composition of negative ions and ion clusters recorded with the APi-TOF during CLOUD experiments simulating boreal-forest NPF (left) and during a springtime NPF event in Hyytiälä (right), showing a remarkable similarity. Different groups of compounds are identified with different colors. Figure from Lehtipalo et al. 2018.

chemical calculation had already been performed for individual cases, such as the smallest clusters of sulfuric acid and water or ammonia (Kurdi and Kochanski 1989, Bandy and Ianni 1998, Beichert and Schrems 1998, Ianni and Bandy 1999, 2000, Re *et al.* 1999). Though it was not widely realized at the time, the structural and energetic data of these early studies already demonstrated that the liquid drop model fails catastrophically for the smallest sulfuric acid–base clusters due to an incorrect description of proton transfer (Elm *et al.* 2020). While these and other early insights from first principles calculations (e.g. Kurtén *et al*. 2008) were useful, the development of actual quantum chemistry-based particle formation models required the systematic calculation of formation thermodynamics for a sufficiently large set of clusters (e.g. Herb *et al.* 2011, Ortega *et al.* 2012, Henschel *et al.* 2014, DePalma *et al.* 2014), using progressively more accurate methods (as described below), and the combination of thermodynamic data with cluster population dynamics models solving the birth–death equations.

The flagship cluster dynamic model of the CoE ATM is the Atmospheric Cluster Dynamics Code (ACDC, McGrath *et al.* 2012). Until now, the collision rates used by ACDC have been taken from kinetic gas theory and existing ion-molecule parametrizations, while the evaporation rates are derived via detailed balance from formation free energies calculated using quantum chemistry (Ortega *et al*. 2012). Work is underway to compute more accurate collision rates taking long-range interactions such as dipole-dipole attraction into account using molecular dynamics (Halonen *et al.* 2019). Our computational scheme was validated by comparison to experimental results from the CLOUD chamber (Almeida *et al.* 2013, Olenius *et al.* 2013). A key finding of these joint experimental-modelling studies has been that the rate of sulfuric acid-driven new particle formation can be immensely increased by extremely low (part-per-trillion or even lower) concentrations of for example basic vapors such as amines — as we predicted already in 2008 (Kurtén *et al*. 2008). It is noteworthy that the threshold concentrations at which these vapors affect new-particle formation may be far below their detection limits using many common measurement techniques. Also, while chemical ionization-based mass spectrometry is an immensely powerful tool for studying atmospheric new particle formation, both the ionization process, and the subsequent high-energy collisions in the ion optics, may bias the

results. The charged cluster distributions detected by chemical ionization mass spectrometers may thus not match the original, usually predominantly neutral, distribution. Fortunately, quantum chemical calculations combined with ion cluster fragmentation models can be used to address both issues, and ultimately reconstruct an unbiased cluster distribution (Passananti *et al.* 2019, Zapadinsky *et al.* 2019).

Our breakthroughs in understanding of atmospheric new-particle formation at a molecular level have been facilitated by the combination of the conceptual and methodological frameworks of computational and physical chemistry with those of atmospheric aerosol physics. On the computational side, key developments allowing the construction of quantum chemistry-based particle formation models include the continuous increase of computing power, the emergence of density functional methods capable of describing intermolecular interactions sufficiently accurately to compute reliable structures and vibrational frequencies of molecular clusters with at least several tens of atoms, and the development of cost-effective variants of coupled-cluster theory (e.g. DLPNO-CCSD(T)) allowing — albeit often only after extensive benchmarking and tweaking of settings — the calculation of near-quantitative binding energies for these clusters. See Elm *et al.* (2020) for a review of the relevant computational methods. As the system size for which accurate quantum chemical calculations are possible has grown, the bottleneck for ACDC-type modelling has become the conformational sampling of molecular cluster structures (Kubečka *et al.* 2019). This problem has been accentuated as focus has expanded from structurally relatively simple sulfuric acid–base systems to clusters containing also large and flexible organic oxidation products (Elm 2019, Shi *et al.* 2019, Zanca *et al*. 2020). We have accordingly developed a systematic and automated approach for the configurational sampling of molecular clusters.

Instrument development related to aerosol formation and growth

Condensation particle counter (CPC) development in the University of Helsinki began around year 2000 when the pulse height counting technique (Saros *et al*. 1996), developed at the University of Minnesota, was modified to the commercial CPCs maybe by TSI (USA), and this pulse height CPC was used in field measurements to detect newly formed particles in the atmosphere. The pulse height CPC was used in Mace Head, Ireland, and in Hyytiälä SMEAR II station, Finland, to monitor the nanoparticle size distributions in the size range of 3–10 nm (Aalto *et al.* 2001, O'Dowd *et al.* 2002). To detect nanoparticles even smaller than 3 nm, the flow profile in the commercial TSI CPC growth stage was made turbulent on purpose (Mordas *et al*. 2008a). This allowed detecting nanoparticles close to 1 nm without major interference from homogeneous droplet formation, and was utilized as a battery of four CPCs at SMEAR II to detect atmospheric clusters (Kulmala *et al.* 2007a). Development of the pulse height CPC technology was continued by decreasing the size of the smallest detectable particle size down to 1 nm by increasing the supersaturation ratio over the limit of homogeneous butanol droplet formation. The pulse height analysis was used to separate homogenously nucleated droplets from ambient particles. This version of the pulse height CPC was used in SMEAR II station to detect atmospheric cluster size distributions even down to the molecular size around 1 nm (Sipilä *et al*. 2008, 2009, Lehtipalo *et al*. 2009).

The next major step in the development of CPCs was the adaption of the flow mixing technology (Okuyama *et al.* 1984, Sgro and Fernandéz de la Mora 2004) to the monitoring of atmospheric clusters. Vanhanen *et al*. (2011) constructed a particle size magnifier (PSM) that is capable of activating and growing even 1 nm particles for the detection in an external CPC. When used in so-called supersaturation scanning mode, it enables the continuous measurement of particle size distributions in the size range of 1–3 nm in the atmosphere (Lehtipalo *et al.* 2022). The PSM has played a major role in elucidating the particle formation mechanisms together with the mass spectrometers in several different atmospheric locations, and chamber and laboratory studies.

Parallel to the CPC development, the CoE was active in advancing the mobility particle

sizing techonology. Especially, the collaboration with University of Tartu has led to developments in the ion mobility spectrometers for atmospheric size distribution measurements (Kulmala *et al*. 2016a). The first ion spectrometers used in the CoE to measure the atmospheric ion siz distribution were the Balanced Scanning Mobility Analyzer (BSMA) (Tammet 2006) and Air Ion Spectrometer (AIS) (Mirme et al. 2007), both manufactured by AIREL Ltd., Estonia. The next big step forward was the development of the Neutral cluster and Air Ion Spectrometer (NAIS), which utilizes unipolar charging prior to detection with electrical mobility analyzers (Mirme and Mirme 2013). This enables the measurement of the total aerosol size distribution in the size range from ~2–42 nm in particle diameter. Due to its robustness and easy operation, the NAIS has been widely used around the world both for long-term measurements and during specific measurement campaigns (e.g. Manninen *et al*. 2009, 2010).

Since the rapid development of several applications to measure the particle concentration and size distribution of newly formed particles, efforts were aimed at narrowing down the uncertainties in these measurements, which is still a scientific challenge especially in the sub-5 nm size range (Kangasluoma *et al*. 2020). This has required development also in the methods to test and validate the instrumentation. Several new methods to produce and size select particles were adopted, including mobility standards produced by electrospraying, wire-generators and high-resolution differential mobility analyzers (e.g. Sipilä *et al.* 2009, Kangasluoma *et al.* 2013, 2016). Several international instrument intercomparison and calibration workshops have been organized in the aerosol laboratory in Helsinki (Mordas *et al.* 2008b, Asmi *et al.* 2009, Gagne *et al.* 2011, Kangasluoma *et al*. 2017). This work will continue in future under the ACTRIS Cluster Calibration Center at University of Helsinki, which is part of the ACTRIS Center for Aerosol In-Situ Measurements.

In situ atmospheric observations and their analysis

Observations indicating atmospheric new particle formation (NPF) date back more than a century (see Malila 2018, and references therein). However, it was not until the late 1990s when this phenomenon was recognized to extend beyond local scales (Mäkelä *et al.* 1997), often taking simultaneously place over tens to hundreds of kilometers in horizontal direction. At the beginning of our CoE ATM, we had already developed the basic tools to characterize regional NPF, including the particle formation and growth rates, condensation and coagulation sinks, and categorizing how frequently and intensively this phenomenon occurs in a certain environment (Kulmala *et al*. 2001a, 2001b). It soon became apparent that atmospheric NPF is predominantly a daytime phenomenon, typically starting soon after the sunrise and continuing for a few hours, whereas the growth of newly-formed particles to larger sizes remains visible up to couple of days in atmospheric observations (Kulmala *et al*. 2004a).

Observing and characterizing atmospheric NPF has been one of the central research topics in our CoE, so that over the years we have measured NPF in tens of different locations covering all the continents (Kerminen *et al*. 2018, Nieminen *et al*. 2018, Chu *et al*. 2019). Perhaps the most important finding of these studies has been the relatively frequent and wide-spread occurrence of regional NPF in almost all continental environments ranging from polar areas to highly-polluted megacities and mountain-top locations. Currently, we have long-term, continuous observations of atmospheric NPF from three boreal forest locations (Asmi *et al.* 2011, Kyrö *et al*. 2014, Nieminen *et al.* 2014), Crete in the Mediterranean (Kalivitis *et al.* 2019), a polluted Po Valley area in Italy (Hamed *et al.* 2007), and from an urban Budapest, Hungary (Mikkonen *et al.* 2020, Salma *et al.* 2021). In addition to making it possible to conduct detailed analyses on the factors influencing atmospheric NPF over very different time scales, these observations have revealed influences of both anthropogenic (mainly sulfur) and natural (mainly biogenic aerosol precursor compounds) emission changes on atmospheric NPF.

Measurements of atmospheric NPF have advanced considerably over the years as a result of instrumental developments (see the previous section). During the first years of CoE, investi-

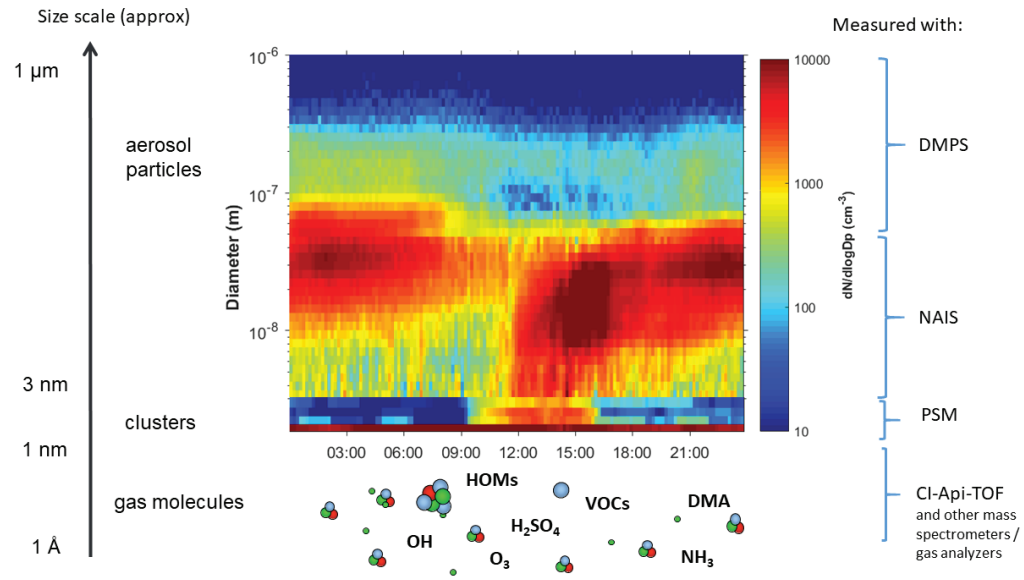

Fig 12. The development in measurement technology during CoE has enabled continuous measurements of the most important aerosol precursors vapors, clusters and the particle size distribution starting from ~1 nm, which are crucial for studying atmospheric new particle formation.

gating atmospheric NPF was based entirely on particle number size distribution measurements starting from particle diameters of 3–10 nm (Dal Maso *et al*. 2005). Later, measurements were extended to the sub-3 nm size range, first to ions (Laakso *et al.* 2004), then to neutral clusters and aerosol particles (e.g. Kulmala *et al.* 2007b, 2013b, Manninen *et al*. 2009), and eventually to molecules and small clusters participating in the very early steps of atmospheric NPF (Ehn *et al*. 2014). These advances are illustrated in Fig. 12. Our CoE was the first to measure the initial steps of atmospheric NPF molecule by molecule (Sipilä *et al*. 2016). More recently, we obtained similar information from Antarctica (Jokinen *et al.* 2018), high-latitude arctic (Beck *et al*. 2021), mountain-top region in Himalayas (Bianchi *et al*. 2021), and heavily polluted megacities in China (Yao *et al*. 2018, Cai *et al.* 2021, Yan *et al*. 2021). One important conclusion based on these studies is the apparent diversity of dominating cluster formation mechanisms in different atmospheric environments.

Concurrent with the development of atmospheric observations, more details were brought into identifying, characterizing and classifying atmospheric NPF (Leino *et al.* 2016, Dada *et al*. 2018, Joutsensaari *et al.* 2018). New theoretical tools and model frameworks were developed to understand the dynamics of molecular clusters (see the section on computational aerosol physics), and the subsequent aerosol formation and growth in the sub 3 to 5-nm size range, including semi-empirical formulas which connect new particle formation rates with concentrations of nucleating vapours (Sihto *et al*. 2006, Paasonen *et al.* 2010) and nano-Köhler theory which describes the activation of molecular clusters for further growth (Kulmala *et al.* 2004b, Kontkanen *et al*. 2018). A crucial factor in determining whether NPF may occur or not in any atmospheric environment is the competition between the cluster growth and their removal by coagulation onto larger pre-existing particles (McMurry and Friedlander 1979, McMurry 1983). During our CoE ATM, we have developed several expressions to estimate how this phenomenon influences newly formed aerosol particle populations (Kerminen and Kulmala 2002, Lehtinen *et al.* 2007, Korhonen *et al*. 2014). In spite of this progress, we still do not have a full scientific understanding on why atmospheric NPF is able

to take place under heavily polluted conditions encountered in some of the megacities (Kulmala *et al.* 2017b).

The growth of newly formed particles to cloud condensation nuclei (CCN), corresponding to a particle diameter of at least a few tens of nm, takes typically from a few hours to a couple of days in the lower atmosphere (Kerminen *et al.* 2018). Our CoE reported the first, direct observational evidence linking atmospheric NPF to CCN production (Kurtén *et al*. 2003, Lihavainen *et al.* 2003), and later more quantitative data on atmospheric CCN production resulting from NPF were obtained from several sites around the world (Sihto *et al*. 2011, Kerminen *et al.* 2012, and references therein). In polluted environments, atmospheric NPF followed by particle growth can contribute to a particle population that forms a haze (Guo *et al.* 2014, Kulmala *et al.* 2021a). We found indications that by reducing NPF by emission control, we might delay haze formation and thereby substantially reduce the annual number of haze days in polluted environments (Kulmala *et al*. 2021a).

While there are evidently multiple compounds and processes influencing the particle growth to larger sizes (Mikkonen *et al.* 2011, Riipinen *et al*. 2012, Vakkari *et al*. 2015), the important role of organic vapours of largely biogenic origin in this growth was recognized already at the early stages of our CoE (Kulmala *et al*. 2001a, Tunved *et al*. 2006). Our most recent observations, combined with laboratory experiments and model simulations, have brought much new insight into the growth of newly formed particles to larger sizes (Ehn *et al*. 2014, Jokinen *et al*. 2015, Tröstl *et al.* 2016, Lehtipalo *et al.* 2018, Paasonen *et al.* 2018). In particular, HOM have been shown to be important contributors to NPF (Ehn *et al.* 2014, Tröstl *et al*. 2016), yet the ability of these to grow new particles of different sizes is highly sensitive to the atmospheric conditions under which they were formed (Yan *et al.* 2020). The oxidation processes converting any VOC into condensable HOM is strongly influenced by factors such as temperature (Quéléver *et al.* 2019), atmospheric mixing (Zha *et al.* 2018), nitrogen oxides (Yan *et al.* 2020), and photochemistry (Yan *et al.* 2016). Further studies are needed to quantitatively link both aerosol formation and growth to various atmospheric precursor compounds and conditions.

Aerosol composition and bioaerosols

Measuring the bulk composition of aerosol particles in the accumulation mode provides a way to estimate the main sources for the aerosols at a given location. The SMEAR II site has therefore had continuous online measurements of aerosol chemical composition since 2012 (Heikkinen *et al.* 2020). Thanks to the long data set, we could distinguish how the aerosol has a bimodal pattern across a year. One mass concentration peak occurs in the winter, mainly from inorganic species such as sulfate and nitrate, and is due to an increased need to domestic heating and different combustion processes. However, the largest mass concentrations are measured in summer, and those are ~80% organic, being SOA formed from the oxidation of the abundant BVOCs that are emitted from the forest. While some of the organics can result from athropogenic activities, we were able to show that this is a very small (< 10%) contribution at this site (Heikkinen *et al.* 2021).These links were only made possible due to the wealth of supporting data available at the station, such as direct VOC measurements to identify biogenic sources, and black carbon and CO for identifying combustion sources.

Here, we focus in more detail into two specific aerosol components, primary biological aerosol particles and soot. Plant fragments, pollen, spores, algae, bacteria and viruses are typical constituents of primary biological aerosol particles (PBAPs, Després *et al.* 2012, Šantl-Temkiv *et al*. 2020). The amount and distribution of PBAPs in the atmosphere have gained more interest, mainly because of the possible underestimation of their levels and their atmospheric impact (Morris *et al.* 2011, Estillore *et al*. 2016). Globally, PBAPs constitute ca. 30% of the mass of particles larger than 1 μm in urban and rural air (Fröhlich-Nowoisky *et al*. 2016), up to 65% in boreal forest air (Manninen *et al.* 2014), and up to 80% in tropical forest air (Elbert *et al.* 2007).

We have successfully determined chemical tracers (amino acids, saccharides, see Fig. 13) and gene copy numbers (16S and 18S for bacteria and fungi, respectively) to access bioaerosols in size-segregated aerosol samples collected at the SMEAR II station (Helin *et al*. 2017, Ruiz-Jimenez *et al.* 2021). According to our results (Helin *et al*. 2017), the spring pollen season influenced the levels of free amino acids as well as the bacterial abundances. Elevated PBAP abundances occured during the pollen season (Manninen *et al*. 2014), and based on our estimation even up to 77% of the total particle mass may be of biological origin in the studied site. We also observed clear variations in the composition of bioaerosols as a function of the particle size (Ruiz-Jimenez *et al*. 2021). In most cases, the highest concentration values and gene copy numbers (in the case of microbes) were observed for 2.5–10 µm particles, followed by > 10, 1–2.5, and < 1.0 µm particles. Variables related to the air and soil temperature, UV radiation, and the amount of water in the soil all affected the composition of bioaerosols. Positive correlations were also observed between gas-phase VOCs (acetone, toluene, methanol, and 2-methyl-3-buten-2-ol) and the gene copy numbers of microbes in biogenic aerosols.

Notable ingredient in the urban atmospheric aerosols is soot, which originates from combustion processes and motor vehicle emissions (Harley *et al.* 2005). It may be referred to black carbon or elemental carbon (Bond and Bergstrom 2006). Although soot and aerosol particles in general have a short atmospheric lifetime, they contribute to both regional and global climate change (Jacobson 2001, Ramanathan and Carmichael 2008). More recently, various studies have been made to resolve the uncertainties related to the effect of soot/black carbon on cloud formation and global warming (e.g. Lohmann *et al*. 2020, Fierce *et al.* 2020). Using specifically developed laser desorption-ionization aerosol mass spectrometer we were able to quantify carbon clusters from C_{14} to C_{19} in 50 nm urban air particles. The average fraction of carbon clusters per sample was 7.2%, ranging from 0.01% to 30.8% (19 pg to 6465 pg). Fullerenes from C_{50} to C_{100} in the 50 nm urban air particles were also detected with this technique (Laitinen *et al.* 2014).

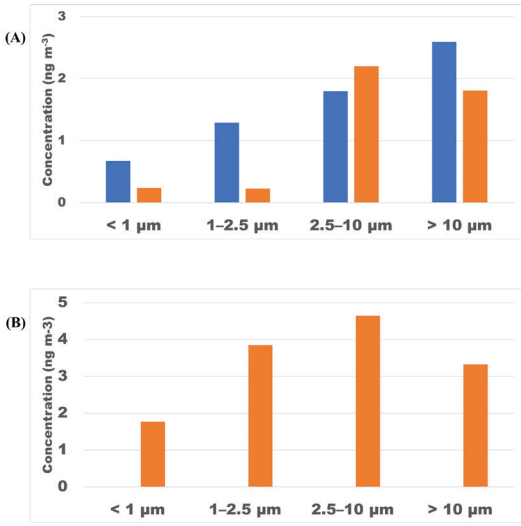

Fig 13. Average free amino acid (upper figure) and saccharides (lower figure) concentrations in different size fractions determined at the SMEAR II station during 2014 (blue) (1 Feb. 2014–1 Nov. 2014) and 2017 (orange) (4 Sep. 2017–22 Nov. 2017) campaigns. Note: no data available for average saccharides concentration from 2014 campaign (Helin *et al.* 2017 and Ruiz-Jimenez *et al.* 2021).

Cloud formation

Aerosols acting as cloud condensation nuclei are capable of modulating cloud droplet number concentration, and therefore its radiative properties. Additionally, there is evidence that perturbation in aerosol concentrations cause changes in cloud cover or liquid water path. Given that clouds are a major part of the daily weather and the global climate system and that boreal forest acts as a source of biogenic aerosol particles that grow to CCN sizes, impact of these particles on cloud formation and evolution is a topic of current scientific interest. Representation of cloud feedbacks is a major source of uncertainty in climate models and accounts for a large inter-model variability in predicted climate scenarios (Coupled Model Intercomparison Project (CMIP) Phase 3 (Bony *et al.* 2006, Dufresne and Bony 2008), Phase 5 (Andrews *et al.* 2012) and Phase 6 (Zelinka *et al*. 2020)). Aerosol–cloud–precipitation interactions is one of the major reasons for this uncertainty.

In situ measurements of cloud formation

CoE partner institutes maintain two stations with continuous in situ observations for aerosol–cloud interactions, SMEAR IV station in Kuopio is located at the top of the Puijo tower 306 m above sea level and 230 m above the surrounding lake level in a semi-urban environment and Pallas-Sodankylä Global Atmospheric Watch (GAW) station in northern Finland is located at the Sammaltunturi measurement site on top of a fell 300 m above the surrounding area. These two stations measure aerosol size distribution and number concentration, light extinction (scattering and absorption) by aerosol particles, cloud droplet size distribution, trace gas concentration, and several meteorological parameters. The aerosol size distribution is measured through two inlets allowing the sampling of the interstitial aerosol and total aerosol including both the interstitial particles and cloud droplet residuals. The difference between the total aerosol and interstitial aerosol gives then the fraction of activated particles as a function of particle size. These measurements enable detailed in situ studies of cloud droplet activation and the effect of aerosol properties on it (Komppula *et al.* 2005, Anttila *et al*. 2009, 2012, Portin *et al*. 2014).

Aerosol chemical composition plays a central role in cloud droplet activation. Väisänen *et al*. (2016) illustrated the sensitivity of cloud droplet formation to aerosol hygroscopicity by investigating activation efficiencies of externally mixed aerosol having high and less hygroscopic modes at size range relevant for activation. They showed that the less hygroscopic mode remained unactivated reducing the potential of particle population to activate cloud droplets by 70% compared to case where all particle would belong to high hygroscopic mode. Also Ruuskanen *et al.* (2021) investigated the on-cloud scavenging of scattering and absorbing aerosol particles. In their study, the absorbing aerosol consisted mostly of black carbon. Their analysis showed that the scavenging coefficients for scattering and absorbing aerosol particles were 0.85 and 0.55 respectively and higher fraction of black carbon is in smaller particle sizes so that at least 20–30% of interstitial particles within clouds consisted of absorbing material. Chemistry plays a role not only in activation, but also in other cloud processes as the aerosol particles may transform chemically in clouds due to aqueous phase chemistry (e.g. Ervens 2015). Hao *et al*. (2013) studied in more detail the effects of chemical composition on activation and further the effects of cloud processing on aerosol composition in low level stratus clouds. They observed higher fraction of low volatile oxygenated organic aerosol in cloud droplet residuals than in interstitial aerosol. Also clear differences in particle acidity were observed: cloud residues were clearly less acidic than interstitial aerosol. According to their analysis, these observations indicates possible joint effect of activation process and cloud processing.

Radar measurements of cloud formation

Combining in-situ and ground-based remote-sensing measurements collected during Biogenic Aerosols–Effects on Clouds and Climate (BAECC) campaign at SMEAR II in 2014 (Petäjä *et al.* 2016b), allowed investigating the influence of biogenic aerosols on clouds (Petäjä *et al*. 2021a). Using a combination of comprehensive ground-based aerosol measurements it was found that the marine air transported over forested areas increase aerosol load by factors of about 2 to 8 for the air mass transport times of 20 hours and 75 hours, respectively. Corresponding cloud remote sensing observations detect an increase in the droplet number concentration, liquid water path and optical depth of warm, low-level clouds, which are staying connected to the ground, by factors of up to two (Petäjä *et al.* 2021a). This study shows that the boreal forest emissions may play an important role by modulating warm, low-level cloud properties and therefore affect cloud radiative effects.

In mid- to high-latitudes, majority of precipitation originates from ice clouds (Field and Heymsfield 2015). Given that a large fraction of these clouds contains supercooled liquid water, riming may have a significant impact on surface precipitation. Using surface-based observations collected in SMEAR II in Hyytiälä, Moisseev *et al*. (2017) have shown that 5–40% of snowfall mass is gained by riming. This is in line with

other geographical locations, i.e. 30%–40% in the Sierra Nevada (Mitchell *et al.* 1990), and more than 50% in Japan (Harimaya and Sato 1989). It was also shown that riming modifies snowflake shape, when the mass of accreted water exceeds 50% of the total particle mass ice particles become more spherical (Li *et al.* 2018). To further advance our knowledge of riming occurrence at different geographic location, a method to estimate riming fraction from Doppler cloud radar observations was derived (Kneifel and Moisseev 2020). This method was applied to cloud radar observations collected at several ACTRIS stations. It was found that riming occurs in 1–8% of the non-convective ice containing clouds. The dependence of riming on aerosol loading however was not as clear as was expected from previous studies.

We have also conducted long-term radar measurements in the city of Helsinki. Using 11 years of University of Helsinki, Kumpula campus radar observations, it was found that airplanes arriving to or departing from the Helsinki-Vantaa airport could in certain condition increase surface precipitation intensity (Moisseev *et al.* 2019). Adiabatic cooling of air behind airplane wings leads to the production of ice particles (Woodley *et al.* 2003), which seed lower mixed-phase cloud layer. This process leads to the formation of larger snowflakes, by aggregation, and result in heavier precipitation intensity. It was found that precipitation in the affected areas could be 6–14 times higher than in the background large-scale precipitation.

Modelling of cloud formation

Cloud modelling covers a vast range of scales starting from the microphysical properties of a single cloud hydrometeor ending up to atmospheric cloud systems with varying cloud types. In the CoE ATM, cloud modelling concentrated mainly on the aerosol effect on the liquid phase clouds, where we first studied how different chemical compounds affect the droplet formation.

Semivolatile aerosol components produce an increase in cloud droplet number concentration in a similar manner as the classical Twomey effect forecasts related to the changes in aerosol number concentration. In the CoE ATM, we have explained the enhanced activation through Köhler theory (Kokkola *et al*. 2003), estimated the effect in atmospheric clouds with parcel modeling framework, and finally, related to nitric acid, also paramerized the effect and produced the first estimate of –0.46 W m^{-2} for global indirect radiative forcing (Romakkaniemi *et al*. 2005, Makkonen *et al*. 2012a). Köhler theory also forecasts that certain surface active compounds lower the surface tension of liquid aerosol solute prior and during the cloud droplet formation process, and thus these compounds have potential also to enhance droplet concentration. However, accounting for different competing effects during the activation process, the effect is estimated to be small especially at a global scale (Sorjamaa *et al.* 2004, Prisle *et al.* 2012), although different results have been presented for some conditions (e.g. Ovadnaite *et al*. 2017).

During the CoE, we have taken steps towards simulating aerosol indirect effects at cloud resolving scale with a coupled aerosol-large-eddy model UCLALES-SALSA (Tonttila *et al.* 2017). The core of the model is based on the well-established large-eddy simulation platform (UCLALES) intended for idealized cloud studies. To account for aerosol it is coupled with SALSA aerosol microphysics (Kokkola *et al*. 2008), which is further updated to cover both liquid and solid hydrometeors of different sizes (Tonttila *et al*. 2017, Ahola *et al*. 2020). To this end UCLALES-SALSA has been employed in different applications including smog events, fog formation, warm and mixed phase boundary layer clouds, and also moderate convection. Common to all of these is the need for explicit representation of aerosol properties in different atmospheric processes, and interaction with cloud and atmospheric dynamics. We have shown that the employment of cloud droplet parameterization in fogs leads to an overestimate in fog droplet number concentration affecting the fog lifetime in numerical weather prediction and also estimates of aerosol radiative forcing caused through changes in fog properties (Boutle *et al*. 2018). Related to drizzle formation in marine stratocumulus, the role of aerosol size distribu-

tion and especially the giant CCN particles is shown to be evident (Tonttila *et al.* 2021).

Dynamic meteorology

Meteorology is a branch of atmospheric sciences that operates on especially large scales. At the start of the CoE ATM, the link between in situ studies and meteorology was initially weak. At the planetary scale, it is obvious that the Earth system components, such as atmosphere and biosphere, are in perpetual interaction. However, when considering local scale processes, it can be quite hard to perceive and appreciate these large-scale couplings. Process studies, for instance, typically concentrate on some specific phenomena that are studied experimentally and theoretically (e.g. Viskari *et al.* 2012a, 2012b). At this scale, the process seems to be embedded in a large-scale environment that is given, thus easily masking the feedback to the processes controlling the environment. More concretely, dynamics and physics of global weather systems control the time evolution of atmospheric state. These provide the environment for aerosol microphysical processes and air chemistry where the key ambient variables are temperature, humidity, and intensity of solar and terrestrial radiation. In such process studies, the role of meteorology is weak since it only provides expertise regarding the environment — the curiosity is directed to the microphysical processes and their theoretical formulation, and less to the feedbacks affecting the large-scale environment and the large-scale system itself.

Widening the study scope can have an immediate impact on the curiosity towards the larger-scale environment. Roll vortices is a universal fluid dynamical phenomenon in convectively active boundary layers (Lampilahti *et al*. 2020). In the atmospheric boundary layer, the vortices ventilate near-surface air and enhance long-range transport, thus controlling trace gas and aerosol concentrations. The CoE organized a major measurement campaign which involved air-borne and surface in situ observations and produced complicated measurement data in four dimensions. In this case, data analysis and interpretation were equally rewarding both for scientists involved in local processes and larger-scale flow dynamics — expertise in all disciplines were necessary to interpret the data, even though the impact of increased aerosol concentration in the ascending branches on the cloud formation and roll vortex intensity could not eventually be determined.

Core research objectives in microphysical process studies are the new formulations of theories that explain the observed phenomena (e.g. Kokkola *et al.* 2008). Long-term support of the CoE enabled development of large-scale models of the atmosphere and Earth system where these new microphysical process formulations can be implemented (Makkonen *et al.* 2009). On one hand, this enables us to test how general the theories are in explaining events outside the immediate surroundings of the in situ measurement sites. Even more importantly, these large-scale models open a direct route to investigate the feedbacks between, e.g., the aerosol population and the large-scale environment. These models can be used in very wide variety of research questions, and this is exactly what has been accomplished in the CoE ATM.

The CoE has enhanced inter-disciplinarity, and incorporation of weather system understanding to microphysical research. A shining example is related to deep convection which transports moisture to the upper troposphere where it has a major impact on the radiative transfer of the Earth system (Riuttanen *et al.* 2016). Aerosols have microphysical and thermo-dynamical impacts on deep convective clouds, and any changes in atmospheric aerosols can have direct climate feedback. Therefore, there is a need to detect the existence and understand the character of this feedback mechanism. The interdisciplinary approach fostered by the CoE has been instrumental in enabling feedback studies that were previously not possible.

In summary, a major benefit of this CoE is that participating scientists have learned to appreciate the importance of the mutual couplings between the Earth system components. Despite some interactions and feedbacks appearing to be beyond control in certain research contexts, the interactions and feedbacks do existence and their importance have been recognized.

Remote sensing for investigating Earth System processes

Earth Observation data available from space-borne remote sensing instruments can be used to investigate climate processes on a global scale. Daily data records from microwave radiometers start as early as 1978 providing brightness temperatures at a coarse spatial resolution of tens of kilometers (Njoku *et al.* 1980, Hollinger *et al.* 1990). We used these data sets to quantify hemispheric processes of the terrestrial cryosphere and to study their relation to carbon cycle. This was accomplished by fusing satellite data sets, in situ data and model predictions. The main investigated variables included snow cover and soil freeze-thaw status that both affect the seasonal cycle of CO_2 and CH_4 in the boreal and arctic region. Detailed comparison of remote sensing signatures and land-atmosphere coupling were carried out in the Sodankylä site that incorporates in situ sensor networks and a suite of satellite reference instruments monitoring both ground/vegetation and the properties of atmosphere, e.g. columnar concentrations of greenhouse gases (Rautiainen *et al.* 2014, Colliander *et al.* 2017, Hannula *et al*. 2020, Lorente *et al.* 2021). Based on these investigations, a novel method to monitor circum-polar soil freezing and thawing processes from L-band microwave radiometer satellite data was established and implemented in co-operation with the European Space Agency (Rautiainen *et al.* 2016).

The employment of microwave radiometer data together with global in situ observations of snow depth and snow mass (given as snow water equivalent in individual samples) led to a major advance in assessing Earth's snow mass and its trend for regional to hemispheric scales for the period 1980–2018 (Pulliainen *et al*. 2020). The obtained results indicate that in Eurasia the multi-decadal trend in the seasonal peak snow mass has been negligible, whereas a decreasing trend is evident in North America. However, regional differences are strong. For example, results for East Siberia indicate exceptionally high levels of snow mass for the years 2017 and 2018, possibly due to absence of sea ice on the East Siberian Sea during these years. Further, the earlier uncertainty in hemispheric snow mass estimates was reduced from 33% to 7.4% (concerning hemispheric non-alpine areas). Another key finding was the estimation of advancement boreal forest spring recovery due to earlier snow melt by linking microwave radiometer data-derived snow melt information to carbon uptake (Pulliainen *et al.* 2017). This was carried out by the combined use of satellite data, EC measurements on CO_2 fluxes across Eurasia and North America, and simulations with a land ecosystem model coupled to a global circulation model (ECHAM/JSBACH). The results show that the spring recovery of photosynthesis advanced about two days per decade for the circum-polar boreal forest zone from 1979–2014. We estimated that this advance of spring has increased the carbon uptake of conifer-dominated boreal forests by 4–6% per decade (for the first half of the year) providing a negative feedback mechanism to ongoing global warming.

Climate feedbacks

Climate feedbacks are defined as interactions in which a perturbation in one climate variable causes a change in the second, and the change in the second quantity ultimately leads to an additional change in the first (IPCC 2013, 2021). The Earth's climate system is affected by numerous feedback mechanisms that can either amplify (positive feedback) or suppress (negative feedback) the ongoing global warming (Bony *et al*. 2006, Heineman and Reichstein 2008, Ceppi *et al.* 2017, Williams *et al.* 2019). During our CoE, we have concentrated on climate feedbacks involving the continental biosphere, and introduced new potentially important feedback mechanisms, and we have investigated the strength of these feedbacks using both long-term observations and large-scale model simulations.

Background to our feedback studies

The interest in climate feedbacks associated with biosphere-atmosphere interactions originates largely from the work by Charlson *et al.* (1987), who suggested that increased sulfur emissions from the oceans in a warming climate

would lead to increased aerosol concentrations and cloud albedos, causing a negative feedback that would slow down the warming of the climate. Kulmala *et al.* (2004c) hypothesized that a negative feedback mechanism involving natural aerosol particles would also operate in a continental biosphere. Central components of this feedback are increased emissions of biogenic volatile organic compounds (BVOCs) from the continental biosphere in a warming climate and enhanced formation of secondary organic aerosol (SOA) resulting from the atmospheric oxidations of these BVOCs, causing an increased atmospheric aerosol loading and larger negative radiative effect that would slow down the global warming (Fig. 1).

The potentially important role of continental ecosystems in the cycling of water, carbon and nitrogen in the Earth system, and thereby in climate feedbacks, were gradually recognized (e.g. Barth *et al.* 2005, Heineman and Reichstein 2008). Our CoE participated actively in constructing a review paper that summarized our understanding on feedback loops connecting terrestrial biogeochemical cycles, biosphere-atmosphere exchange, atmospheric composition and climate (Arneth *et al.* 2010). One of the main conclusions of that paper was that the strength of biogeochemical feedbacks, while highly uncertain, could be comparable to those of more widely-studied feedbacks in the physical climate system.

Higher atmospheric aerosol loadings increase the ratio of diffuse to total radiation reaching the Earth's surface, which in turn tends to enhance the carbon uptake by photosynthesis in many continental ecosystems (Gu *et al.* 2002, Mercado *et al*. 2009, Cirino *et al*. 2014, Ezhova *et al*. 2018). Kulmala *et al.* (2013a) combined this idea with the work by Kulmala *et al*. (2004c), and introduced the so-called continental biosphere-aerosol-cloud-climate (COBACC) feedback mechanism. This mechanism has two partly overlapping feedback loops, both initiated by higher atmospheric CO_2 concentrations, and both of them having a suppressing effect on global warming (Fig. 1). The overlapping part of the two loops is related to higher atmospheric aerosol loadings due to higher BVOC emissions in a world with higher CO_2 concentrations and ambient temperatures.

The latest development in our feedback work is the introduction of the concept CarbonSink+ (Kulmala *et al*. 2020). It builds on the idea of using forest management in climate change mitigation (e.g. Grassi *et al*. 2017), with a couple of new extensions. More specifically, CarbonSink+ aims to take into account: 1) the influences of changing atmospheric composition on the forest carbon uptake over time scales comparable to typical afforestation or reforestation activities; and 2) the combined climatic effects by which forests act as a carbon sink and change either the surface albedo or atmospheric aerosol loading. Using the boreal forest environment as an illustrative example, Kulmala *et al*. (2020) and Kalliokoski *et al*. (2020) demonstrated the importance of considering all these factors when mitigating the climate change through forest management.

Lessons from long-term observations

Our CoE has vast experience in using observational data for investigating the different parts of the COBACC feedback mechanism discussed in the previous section. Using several years of particle number size distribution measurements from three sites, Tunved *et al*. (2006) were able to establish a firm link between summer-time aerosol concentrations inside the boreal forest zone and SOA formation associated with BVOC emissions from the forest. Later we showed that, due to the strong response of BVOC emissions to the ambient temperature, higher temperatures lead to larger aerosol loadings in a boreal forest environment (Paasonen *et al.* 2013, Liao *et al*. 2014, Lihavainen *et al.* 2015).

Closing the original branch of the COBACC feedback mechanism requires estimating the radiative effect associated with biogenic SOA formation and its response to increasing ambient temperatures. We have estimated this radiative effect due to both aerosol-cloud and aerosol-radiation interactions using different measurement data sets (Kurtén *et al.* 2003, Lihavainen *et al.* 2009, Paasonen *et al.* 2013). For the cloud albedo feedback due to biogenic secondary organic aerosol, Paasonen *et al.* (2013) derived local estimates in the range from –0.4 W m^{-2} K^{-1}

to –0.7 W m^{-2} K^{-1} at four boreal forest sites, and the global value of about –0.01 W m^{-2} K^{-1}. Lihavainen *et al.* (2015) investigated the feedback due aerosol-radiation interactions in a boreal forest site, and derived a feedback strength that was clearly smaller than the corresponding cloud albedo feedback.

The strength of the feedback loop related to carbon uptake in the COBACC feedback mechanism was estimated first by Kulmala *et al*. (2014). They found a potentially large gain for this feedback in the boreal forest environment, however with a large uncertainty range. In SMEAR II, a 10% increase in ecosystem GPP driven by increased atmospheric CO_2 concentration led to a significant positive feedback to GPP by increasing atmospheric particle growth rate due to organic vapours (+7.3%), the amount of biogenic secondary organic aerosols measured with condensation sink (+6.5%), the ratio between the diffuse and global radiation (+8.5%) and finally the GPP (+3%, Kulmala *et al.* 2014). However, in a more recent analysis Launiainen et al. (2022) found that only 30–40% of the observed ecosystem GPP increase over 2001-2017 could be attributed to CO_2 fertilization at SMEAR II. Ezhova *et al.* (2018) used measurement data from five boreal and hemi-boreal sites, and found that the presence of atmospheric aerosols can increase the carbon uptake by photosynthesis by up to 10-15% under clear sky conditions. Recently, Yli-Juuti *et al*. (2021) presented direct evidence on the COBACC feedback mechanism utilizing long term in situ aerosol composition measurements and remote sensing on aerosol and cloud properties. Their results show a significant negative feedback of −0.63 Wm^{-2} $°C^{-1}$ from temperature-dependent BSOA formation via strengthening of both direct and cloud albedo effects of aerosols. The cooling effect of the feedback equals to ~18% of the current effective radiative forcing of anthropogenic aerosols over boreal forest area in summer (Yli-Juuti *et al*. 2021).

Quantification of the different parts of this feedback loop using measurement data alone, or the overall strength of this feedback, has turned out to be difficult. The main reason for this are challenges in controlling the influences of factors other than those originally ascribed to this feedback, including the presence of clouds or anthropogenic pollutants, and possible synergistic effects caused by the ambient temperature or some other meteorological variables.

Model investigations

Model simulations have been used widely to study climate feedbacks (e.g. Carslaw *et al*. 2010, Webb *et al*. 2013, Hu *et al.* 2020). The advantage of this approach over observations is a better control of the factors and processes influencing the studied feedback, and a possibility to make future predictions. The drawback is that models include only a subset of all the processes operating in the Earth system, so it is possible that a model simulation could miss some factor having an important influence on a feedback.

The first global modeling effort by our CoE ATM was related to the COBACC feedback mechanism and was conducted by Makkonen *et al.* (2012b), who demonstrated that the radiative effect caused by biogenic SOA is sensitive to BVOC emissions and atmospheric new particle formation under both present-day conditions and in the future. Scott *et al*. (2014) diagnosed climate feedbacks caused by terrestrial natural aerosols from global model simulations. Their global estimate on the climate feedback caused by biogenic SOA was –0.03 W m^{-2} K^{-1}, dominated by aerosol-radiation interactions, while the part of this feedback caused by the cloud albedo effect was similar to the observation-based estimate by Paasonen *et al.* (2013) (see Sect. 3.7.2). Using a global aerosol-climate model, Arneth *et al.* (2016) found a global cloud radiative forcing of –0.03 W m^{-2} due to about 40% increase of BVOC, although the forcing was much stronger (–0.50 W m^{-2}) over the Eastern Siberian study region. In Boy *et al*. (2019), a coupled atmosphere-ocean-land model was used to quantify BVOC-aerosol feedback in an idealized doubled CO_2 concentration. Over boreal forest, monoterpene emissions increased from 18 to 28 Tg yr^{-1} leading to increased aerosol and cloud droplet number concentrations. Sporre *et al.* (2019) used an Earth system model to investigate the whole COBACC feedback mechanism and found it to have a significant impact on climate. They did

not find any boost in the global carbon uptake by photosynthesis (GPP) associated with the lower branch of the COBACC feedback, probably because BSOA effects on clouds seemed to dominate the simulated changes in GPP. Finally, using three Earth system models, Sporre *et al*. (2020) showed that the radiative effects by biogenic SOA are sensitive to how BVOC and resulting SOA treated in the models.

In addition to biogenic SOA, concentrations and thereby radiative effects of many other natural aerosol types change with changing climate (Carslaw *et al.* 2010). Model simulations by Korhonen *et al*. (2010) indicate that the increased wind speeds associated with the observed acceleration of the westerly jet in the Southern Hemisphere have caused higher sea spray aerosol flux from the Sourthern Ocean to the atmosphere since 1980's, resulting in a substantial negative climate feedback. Scott *et al*. (2014) estimated that in a warming climate, the climate feedback due to increased forest fire aerosol emissions could be even higher than the feedback due to increased biogenic SOA formation.

Anthropogenic contribution to the future atmospheric aerosol load is very likely to decrease as a result of air pollution control, which is likely to have strong climatic implications (Brasseur and Roeckner 2005, Partanen *et al*. 2018, Samset 2018). Our CoE was among the first ones to show that air pollution control can cause accelerated warming in the future, and that this effect is coupled with the future behavior of the natural aerosol system (Makkonen *et al.* 2012c). We also investigated how different air pollution control strategies would influence near-future changes in aerosol radiative effects (Pietikäinen *et al*. 2015). The results showed that from a climate warming point of view, targeted emission reduction measures would be a much better choice than overall high reductions globally.

Global model studies provide insight that there is potential for strong Earth system feedbacks via biogenic aerosols influencing climate projections of the 21st century. Strong BVOC emission increases are predicted in at least some global emission models of gases and aerosols from nature (MEGAN-based models, e.g. Sporre *et al.* 2019), although the response in some process-based dynamic vegetation-terrestrial ecosystem models (e.g. LPJ-GUESS) or other model approaches might yield lower emission response under future pathways (e.g. Makkonen *et al.* 2012b). While models agree fairly well in aerosol concentration and optical depth responses to simple BVOC perturbations (Sporre *et al.* 2020), results remain mixed when analyzing the changes in aerosol size distributions, CCN concentrations or cloud properties. Hence, the radiative flux perturbation related to BVOC increase varies significantly between models and can even be of opposite sign.

Conclusions and future outlook

Feedback mechanisms are essential components of our climate system, as they either accelerate or slow down changes in climate-related quantities in the presence of external forcings. During the nearly two decades of CoE ATM work, combining the gained process level understanding with comprehensive, long-term field measurement enabled us to provide the first quantitative estimates regarding the COBACC feedback in boreal and hemi-boreal environments (Kulmala *et al*. 2014, Ezhova *et al.* 2018, Yli-Juuti *et al.* 2021). This feedback loop demonstrates how important biospheric processes are not only for carbon and aerosol budgets, but also for the whole climate system, as the COBACC feedback mitigates global warming, thereby giving the mankind a window of opportunity to reduce global carbon emissions. It is important to find out how important this feedback loop is globally and particularly in the boreal and arctic environments. The strength of the COBACC feedback is connected tightly with the functioning of the biosphere, including the observed yet poorly understood greening and other vegetation changes currently taking place in boreal and arctic environments (Buermann *et al.* 2014, Pearson *et al.* 2013, Reich *et al.* 2015). Therefore, more long-term measurements are needed globally to understand and quantify the COBACC feedback loop in different climate regions and ecosystem types, and to predict how the ecosystem and atmosphere interaction

will change in the future. Our analysis demonstrated the power of using comprehensive field measurements in investigating the complicated couplings between the biosphere and atmosphere on one hand, and on the other hand, the need for complementary approaches relying on the combination of field data, satellite observations and model simulations.

Results from atmospheric research can also be directly applied to human health (Mielonen *et al*. 2015, Yang *et al*. 2018); in Finland the small particles in the atmosphere shorten the average lifetime expectancy by few months, but e.g. in China the effect is several years (Lelieveld *et al*. 2015). Climate change and poor air quality form a pair of problems that must be mitigated simultaneously in the coming decades. Changes in climate and air quality depend on both natural and man-made emissions, as well as various physical, chemical and biological processes occurring in the atmosphere, biosphere and at their interface. Air quality, trace gases, aerosols, and climate are tightly linked via atmospheric processes — e.g. reduction of aerosol emissions causes additional warming due to a reduction in scattering of radiation and changes in cloud properties (Kulmala 2015). On the other hand, absorbing and toxic carbonaceous aerosols increase warming and simultaneously cause health effects. Poor understanding of the atmospheric interactions may thus lead to unwanted climate impacts and various negative health effects (Fiore *et al.* 2012, Fuzzi *et al.* 2015, Kulmala 2015). The fact that new particle formation does occur in polluted Chinese megacities like Beijing and Shanghai (Chu *et al.* 2019, Kulmala *et al.* 2021a) suggests that there are several major physical and chemical mechanisms in a heavily-polluted atmosphere that have not been recognized before and may not even operate in clean or moderately-polluted environments. This requires contributions from a new type of science that links disciplines, knowledge systems and societal partners to support a more agile innovation system. Thus, we are directing increasing efforts to study air quality and its feedbacks with atmospheric processes in the era after the CoE ATM. The work of the CoE ATM community continues in a new Atmosphere and Climate Competence Center (ACCC) flagship.

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