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Finnish Centre of Excellence in Physics, Chemistry, Biology and Meteorology of Atmospheric Composition and Climate Change: summary and outlook

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The Finnish Centre of Excellence (FCoE) in "Physics, Chemistry, Biology and Meteorology of Atmospheric Composition and Climate Change" (2008–2013) completed its research activity at the end of 2013. The FCoE research was originally focused on enhanced process-level understanding of various couplings between atmospheric CO_2 concentrations, photosynthesis, biogenic volatile organic compounds (BVOC), aerosol particles and clouds. During the FCoE period, the scientific scope moved gradually towards the so-called all-scale concept. In this paper we summarize its main scientific achievements, and give an outlook for future scientific activities and focus.

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Background

The atmosphere forms a major part of the environment to which life on Earth is sensitively responsive. The atmosphere closely interacts with the biosphere, hydrosphere, cryosphere and lithosphere on time scales from seconds to millennia (Wanner *et al*. 2008). Changes in one of the components are directly or indirectly communicated to the others via intricately linked processes and feedbacks. A good example of this are atmospheric aerosols that have motivated much research in recent years due to their important role in the global radiation budget, cloud formation and human health (e.g. Russell and Brunekreef 2009, IPCC 2013). Concentrations and properties of atmospheric aerosol particles are tightly connected with greenhouse gases and reactive trace gases via physical, chemical and biological processes occurring in the atmosphere, biosphere and at their interface (Arneth *et al*. 2010, Carslaw *et al*. 2010, Mahowald 2011). In addition to various natural feedback mechanisms, complicated couplings between atmospheric aerosols, greenhouse gases, trace gases, air quality and climate are greatly affected by human and societal actions, such as emission policy, forest management and land use change (Arneth *et al*. 2009, Raes *et al*. 2010, Shindell *et al*. 2012).

The backbone of the scientific approach has been the supra-disciplinary research work by different teams from the Departments of Physics, Chemistry and Forest Sciences at the University of Helsinki, the Finnish Meteorological Institute and the Department of Physics at the University of Eastern Finland. Before the beginning of the recent FCoE period in 2008, we had formulated our FCoE objectives in the following way: "Our main objective is to contribute **to the reduction of scientific uncertainties concerning global climate change issues, particularly those related to aerosols and clouds**. We aim at creating a **deep understanding of the dynamics of aerosol particles and ion and neutral clusters in the lower atmosphere, with the emphasis on biogenic formation mechanisms and their linkage to biosphere-atmosphere interaction processes, biogeochemical cycles and trace gases**". The relevance and usage of the results in the context of global-scale modelling, and the development and utilization of the newest measurement techniques are addressed. The cores of the activities are (a) in continuous measurements and database of atmospheric and ecological mass fluxes and aerosol precursors and CO_2/aerosol/trace gas interactions in SMEAR field stations and Pallas-Sodankylä GAW station, and (b) in focused experiments and modelling to understand the observed patterns."

While the FCoE research was originally focused on an enhanced process-level understanding of various couplings between atmospheric CO_2 concentrations, photosynthesis, biogenic volatile organic compounds (BVOC), aerosol particles and clouds (e.g. Kulmala *et al*. 2004), the scientific scope of the FCoE moved gradually towards the so-called all-scale concept (Kulmala *et al*. 2011b). This means in practice that the relevance and usage of the scientific outcome, starting from the molecular scale, can be applied and up-scaled to the context of global-scale phenomena, such as the prediction of climate change. Today, the scientific orientation of the FCoE research community is called "Atmospheric Science — From Molecular and Biological processes to The Global Climate" (ATM), which will also be the name of the new, third FCoE, targeted for the years 2014–2019. Here, we will summarize briefly the scientific progress made during the period 2008–2013 and then outline our new research priorities for the third FCoE period.

Scientific progress

The scientific work of the FCoE was originally divided into five thematic areas closely interacting with each other: (1) formation and growth of atmospheric aerosol particles, (2) ions and clusters, (3) aerosol–cloud interactions, (4) biosphere–atmosphere interactions, and (5) earth system behaviour. For each of the themes, we further defined a few research topics within which we expected scientific breakthroughs during the period 2008–2013. In the following subsections we will summarize our progress toward the anticipated scientific breakthroughs.

Formation and growth of atmospheric aerosol particles

Pre-defined topics for expected scientific breakthroughs:

- Detection of nucleation mode (3–25 nm) particle composition using newly developed novel instrumentation.
- Understanding the formation and growth mechanisms of secondary biogenic aerosol particles.
- Parameterized formation and growth mechanisms for global climate models.

Knowledge about the chemical composition of nucleation mode particles is crucial to the understanding of atmospheric aerosol formation because such information serves as a link between atmospheric chemistry, formation of new particles, their growth and subsequent cloud condensation nuclei (CCN) production (Riipinen *et al*. 2011, Mauldin *et al*. 2012, Kulmala *et al*. 2014a). Globally, the importance of organic compounds has been identified (Jimenez 2009, Crippa *et al*. 2013). However, low mass concentration at sizes of the nucleation mode hinders the specification of the chemical composition of these particles. We developed novel techniques to overcome this boundary (e.g. Parshintsev *et al*. 2011, Ruiz-Jimenez *et al*. 2011). Joint field studies with international collaborators have identified the importance of acid-base chemistry, sulphuric acid, amines as well as organic compounds (Smith *et al*. 2010, Ruiz-Jimenez 2012, Pennington *et al*. 2013, Vogel *et al*. 2013) in the formation and growth of atmospheric aerosol particles.

In order to enhance our understanding of the formation and growth mechanisms of secondary biogenic aerosol particles, we continued long-term measurements at several field sites, initiated new long-term measurements, carried out intensive campaigns at selected sites, and outlined general guidelines for investigating atmospheric nucleation and growth (Kulmala *et al*. 2012). As an overall summary of this work, we introduced a new, observational-based framework on atmospheric aerosol formation (Kulmala *et al*. 2013b). This framework combines in a consistent way (i) molecules, small atmospheric clusters and growing nanoparticles, (ii) sulphuric acid, strong bases and organic vapours, and (iii) various dynamical processes. The framework confirms that the aerosol formation is essentially a two-step process. The first step involves nucleation or, more precisely, the formation of stabilized clusters, and this process seems to take place almost continually. The second step, characterized by an enhanced cluster growth due to the activation of the growing clusters by organic vapours, determines the formation rate of 3 nm particles and is efficient only during periods of active aerosol formation.

The existing nucleation parameterizations based on the classical nucleation theory were shown to have serious problems when compared with field and laboratory experiments (Sipilä *et al*. 2010, Kerminen *et al*. 2010). As a result, we started developing semi-empirical parameterizations where the nucleation rate follows a simple power law dependence on the gaseous sulphuric acid concentration in line with either a kinetic or activation-type nucleation process (Weber *et al*. 1996, Kulmala *et al*. 2006). We derived and tested several forms of such parameterizations based on available field measurement data, and some of the parameterizations included the effect of organic vapours in addition to sulphuric acid (Paasonen *et al*. 2010). We developed further our earlier parameterizations that convert the nucleation rate to the apparent formation rate of particles at some desired diameter (Lehtinen *et al*. 2007, Anttila *et al*. 2010), a feature needed by most large-scale modelling frameworks. Our new parameterizations have been applied widely to investigating the global aerosol budgets and indirect radiative effects caused by nucleation (Wang and Penner 2009, Kazil *et al*. 2010, Makkonen *et al*. 2012a).

Ions and neutral clusters

Pre-defined topics for expected scientific breakthroughs:

- Detection of neutral clusters using novel instrumentation.
- Reliable *ab initio* calculations on formation of neutral clusters.

- Understanding the atmospheric nucleation mechanism.
- Global map of ion cluster properties.

The key to understanding atmospheric nucleation is in direct observations of the concentration and composition of the freshly formed clusters. While the capacity to detect ions has been available for years (Hirsikko *et al*. 2011), neutral clusters have been beyond the measurement techniques until recently (Kulmala *et al*. 2013b). In short, our work within the FCoE approached the critical size range from two different directions.

Firstly, we improved Condensation Particle Counters (McMurry 2000), especially their ability to count particles smaller than 3 nm in diameter. This path includes the improvement of the Pulse-Height Analysis method (Saros *et al*. 1996) to cope with the homogeneous nucleation and still be able to determine concentrations of atmospheric particles at sizes < 3 nm (Sipilä *et al*. 2009, Lehtipalo *et al*. 2009, 2010). The major breakthrough in CPCs was the combination of a mixing type approach (Sgro and Fernández de la Mora 2004) with di-ethylene glycol as the working fluid (Iida *et al*. 2009). This work pinnacled in the Particle Size Magnifier, PSM (Vanhanen *et al*. 2011) which is capable of detecting particles down to about 1 nm in mobility diameter.

The second approach was to develop mass spectrometers capable of sampling from ambient pressure with a high enough throughput while being sensitive enough for atmospheric ion concentrations. This was realized in an Atmospheric Pressure interface Time-Of-Flight mass spectrometer (APiTOF; Junninen *et al*. 2010). Without any pre-treatment of the sample, APiTOF is capable of quantifying the chemical composition of atmospheric cluster ions (Ehn *et al*. 2010). The results indicated that the ion population is dominated by strong acids and bases in negative and positive polarities, respectively. The key development was to accompany APiTOF with chemical ionization (CI) following the design by Eisele and Tanner (1991). This technology is used for sulphuric acid measurements (e.g. Petäjä *et al*. 2009, Sipilä *et al*. 2010). The Chemical Ionization APiTOF (Jokinen *et al*. 2012) has proven to be a robust technique that can detect not only sulphuric acid and its clusters (Petäjä *et al*. 2011, Almeida *et al*. 2013) but also highly-oxidized organic vapours and clusters (Ehn *et al*. 2012, Schobesberger *et al*. 2013).

The benefit of these two approaches is that we are both able to determine the concentration with single-particle resolution and to determine the chemical composition of the particles at the same time. The real benefit of these techniques arises from their utilization together (Kulmala *et al*. 2013b) for probing the initial steps of atmospheric nanoparticle formation. The next steps of the development will include optimization and refinement of the techniques as well as expansion of the ionization to include different reagent ions to widen the spectrum of clusters that we are able to measure. The work is already on-going (Kangasluoma *et al*. 2013, 2014, Lehtipalo *et al*. 2014, Wimmer *et al*. 2013, Taipale *et al*. 2014).

Quantum chemical methods have become a powerful tool for investigating the molecular mechanisms responsible for atmospheric cluster formation, for getting information about the composition and stability of molecular clusters, and for interpreting experimental results. The main results of our quantum chemical studies were predicting, prior to experimental verification, the enhancement of sulphuric acid nucleation by amines at atmospheric trace concentrations (Kurtén *et al*. 2008), and subsequent near-quantitative modelling of the acid-amine cluster formation process (Almeida *et al*. 2013) using our cluster dynamics code (McGrath *et al*. 2012). Another key result was the prediction (Kurtén *et al*. 2011) of a rapid rate constant for the oxidation of SO_2 by Criegee Intermediates, which turned out to have significant implications in ambient conditions (Mauldin *et al*. 2012, Boy *et al*. 2013). We also used quantum chemical data for connecting experimental observations of charged cluster distributions to the corresponding neutral distributions (e.g. Olenius *et al*. 2013a) and assessed the validity of some fundamental assumptions commonly made in both theoretical and experimental nucleation studies (Olenius *et al*. 2013b).

Besides the new observational-based framework on atmospheric aerosol formation discussed in the previous subsection, we received

plenty of new insight into atmospheric nucleation mechanisms. The results indicate that a comprehensive data on precursor concentrations, oxidant levels, cluster concentrations and their composition is needed to understand the atmospheric aerosol formation (Kulmala *et al*. 2013b). Both sulphuric acid and stabilizing bases are needed for the initial formation of clusters with enough stability (Almeida *et al*. 2013). The organic vapours participating already at the clustering stage and enabling the growth to larger sizes add further complexity (Schobesberger *et al*. 2013). Taken together, our findings emphasize the crucial importance of initial clustering and stability of the clusters as well as an important role of organic compounds in atmospheric new particle formation and the usual dominance of neutral nucleation pathways over ion-induced nucleation in the lower troposphere (Kulmala *et al*. 2013b).

By coordinating the application of air ion and cluster mobility spectrometers for more than one year across Europe, we constructed the first large-scale data set on the formation and growth rates of neutral and charged 2-nm diameter aerosol particles (Manninen *et al*. 2010). We also reviewed the published literature on atmospheric ions (Hirsikko *et al*. 2011), with the main focus on measured ion concentrations and their association with atmospheric nucleation.

Aerosol-cloud interactions

Pre-defined topics for expected scientific breakthroughs:

- Quantitative understanding of CCN production from nucleation events in present and pre-industrial conditions.
- The effect of trace gases on CCN activation as a function of pollution.
- Parameterised CCN, cloud droplet and rain droplet formation mechanisms for global models.

We previously demonstrated that atmospheric nucleation is capable of producing new CCN (Lihavainen *et al*. 2003, Laaksonen *et al*. 2005), and that the same particles may ultimately participate in cloud droplet activation (Kerminen *et al*. 2005). During the latest FCoE period, we investigated this topic systematically with the help of field observations (e.g. Sihto *et al*. 2011, Kyrö *et al*. 2013), large-scale model simulations (Spracklen *et al*. 2008, Merikanto *et al*. 2009, Makkonen *et al*. 2012a, 2012b) and combinations of these two approaches (Laakso *et al*. 2013). We found that CCN production associated with atmospheric nucleation is a frequent and widespread phenomenon in the continental boundary layer, likely so in the free troposphere as well, and that the character of this phenomenon has probably changed considerably from pre-industrial times to the present. Our synthesis on this topic revealed that quantifying the role of nucleation on atmospheric nucleation on CCN formation requires (i) better understanding of the factors controlling atmospheric CCN production, (ii) detailed information on the properties of both primary and secondary CCN and their interactions, and (iii) studies where field measurements will be combined with regional or large-scale model simulations (Kerminen *et al*. 2012).

The CCN activity of aerosol can be enhanced by semi-volatile aerosol components that condense onto aerosol particles prior to the cloud droplet formation. The aerosol composition measurements conducted at the Puijo station support the theory by showing that semi-volatile compounds favour the partitioning into cloud droplets inside the cloud (Hao *et al*. 2013). To estimate the climatic significance of this effect, we performed the first global simulations and found that atmospheric nitric acid is causing a radiative forcing of -0.46 ± 0.25 W m^{-2} through aerosol indirect effects (Makkonen *et al*. 2012c). However, the experimental quantification of CCN activity enhancement is still lacking, since in the current state-of-the-art CCN counter design semi-volatile compounds are more likely to evaporate from particles than to condense onto them (Romakkaniemi *et al*. 2013). In several studies, surface-active compounds have been speculated to affect the cloud droplet formation, although the theory of surface partitioning does not support these findings (Sorjamaa and Laaksonen 2006), and even in the case of semi-volatile surfactants the effect on CCN activity is very small (Romakkaniemi *et al*.

2011). We parameterized the surfactant partitioning (Raatikainen and Laaksonen 2011, Prisle *et al*. 2011), and found that in the global scale the effect on global radiation budget was negligible if surfactants are represented with state of the art methods (Prisle *et al*. 2012).

Field measurements conducted at the Hyytiälä, Pallas and Puijo stations showed that in order to derive the CCN number concentration, information about the size-resolved aerosol composition is needed (Anttila *et al*. 2009, 2012, Paramonov *et al*. 2013, Jaatinen *et al*. 2014). Studies on aerosol–cloud interactions in (boreal) boundary-layer clouds showed that at low aerosol concentrations, the cloud droplet number concentration is linearly dependent on the accumulation mode particle number concentration, but due to low updrafts in these clouds the aerosol effect saturates already at concentration exceeding 300 cm^{-3} (Lihavainen *et al*. 2010, Romakkaniemi *et al*. 2012, Ahmad *et al*. 2013). We developed parameterizations with different levels of details to take into account the influence of the aerosol size distribution on cloud droplet formation (Kivekäs *et al*. 2008, Barahona *et al*. 2010).

Biosphere-atmosphere interactions

Pre-defined topics for expected scientific breakthroughs:

- Quantification of oxidized volatile organic compound (OVOC) emissions from boreal ecosystems.
- Detection and understanding of annual patterns of BVOC, other trace gases and aerosol particle fluxes.
- Clarifying and quantifying processes behind BVOC formation and their acclimation to climate and atmospheric variation at different time scales.
- Quantification of the role of root exudates in forest carbon balance.
- Interaction between radiation distribution, soil water and gross primary production (GPP).

Biogenic volatiles contribute to the growth of freshly-formed aerosol particles, and therefore knowledge of variations in BVOC emissions and the drivers controlling their atmospheric concentrations is crucial for understanding the aerosol processes. The most important compounds emitted by the Scots pine are the monoterpenes α-pinene and Δ^3-carene and the sesquiterpenes β-caryophyllene and α-farnesene (Hakola *et al*. 2006, Yassaa *et al*. 2012). Interestingly, each pine tree emits a specific, inherited monoterpene blend, which has important implications for the atmospheric chemistry and emission models, often parameterized using only information from one individual tree (Bäck *et al*. 2012, Yassaa *et al*. 2012, Kajos *et al*. 2013). Large fluxes of OVOCs, including methanol, acetone and acetaldehyde, were measured especially during the period of the fast growth in spring (Aalto *et al*. 2014), as well as from soil fungi and ground vegetation (Bäck *et al*. 2010, Aaltonen *et al*. 2012b). For the first time, freshwater vegetation was also identified as an aerosol-precursor source (Kyrö *et al*. 2013).

The BVOC fluxes are highly variable in time and related to both biological and environmental drivers. In short term, emissions of monoterpenes show a clear temperature-dependency owing to the exponential relationship between their volatility and temperature (Guenther *et al*. 1993). However, about 40% of the recently assimilated carbon in emissions indicates a strong light-dependency and relationship with photosynthetic carbon as well (Ghirardo *et al*. 2010). In addition to short-term drivers, medium-term changes in conditions affect the emission capacity (Lappalainen *et al*. 2013, Peräkylä *et al*. 2014). Emissions and air concentrations of most BVOCs peak in summer, while in winter the concentrations are very low. The diurnal variability in BVOC concentrations is affected by the friction velocity and boundary layer conditions, which cause a build-up in the poorly-mixed layer at night time (Hakola *et al*. 2012).

The forest floor plays a substantial role in the total VOC emissions of the boreal forest ecosystem both in summer and in winter (Aaltonen *et al*. 2011, 2012a, 2012b). The density of the canopy influences the chemical degradation of the VOC flux from the forest floor by hindering transport and thus prolonging the transport

time (Rinne *et al*. 2012), and affects the VOC lifetimes by influencing the concentrations of oxidants. For reactive compounds like caryophyllene, the effect of chemistry is considerable especially at nighttime, and the calculated fluxes are seriously underestimating the actual emissions.

Ecosystems are key components in biogeochemical cycles and contribute to climate change by exchanging carbon, nitrogen and water between atmosphere, vegetation and soil. We quantified the carbon, nitrogen and water balances of the pine forest stand (Ilvesniemi *et al*. 2009, 2010, Kolari *et al*. 2009, Korhonen *et al*. 2013). In a closed-canopy pine stand, trees contribute almost 90% of the ecosystem gross primary production (GPP). 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). A novel approach to measuring photosynthesis and GPP is using optical signals. A theoretical model framework with a number of novel chlorophyll fluorescence parameters allowed us to study in detail the seasonal acclimation of photosynthesis in field conditions (Porcar-Castell *et al*. 2011, 2012).

Trees emit significant amounts of carbon to the soil as root exudates which sustain mycorrhizal fungal symbionts and other groups of specialized micro-organisms, and contribute to the net carbon balance of forest stands (Pumpanen *et al*. 2009, 2012, Heinonsalo *et al*. 2010). Roots and rhizosphere play an important role in the mineralization of soil organic matter that contains large amounts of immobilized nitrogen. The response of the mineralization of old soil organic matter to easily decomposable carbon substrates is much stronger in the presence of living root system than in bare soil without roots (Lindén *et al*. 2013). Thus, the feedbacks between soil, rhizosphere and above ground parts of plants should be taken into consideration in soil organic matter decomposition models which are used as part of larger ecosystem carbon cycle models or coupled climate carbon circulation models. The forest growth analysis underlines the importance of nitrogen (Magnani *et al*. 2008, Hari *et al*. 2013), as the boreal forest ecosystems are far from being nitrogen saturated (Korhonen *et al*. 2013).

With an evolutionary optimization principle in leaf stomatal control, it is possible to predict very accurately the tree gas exchange (Hari *et al*. 2009). The stomata regulate the leaf gas exchange to maximize assimilate transport from leaves to carbohydrate sinks, such as growing tissues or soil (Nikinmaa *et al*. 2013). This links stomata functioning and photosynthetic production mechanistically to plant structure and water transportation, which is vulnerable to freezing and cavitation, i.e. to phase change from liquid to ice or gaseous. We found that xylem anatomy regulates the sizes of nuclei catalysing freezing and drought cavitation within plants thus linking together adaption of plants to cold and drought stress (Lintunen *et al*. 2013).

The annual ecosystem respiration shows substantially greater temperature sensitivities in aquatic ecosystems compared with terrestrial ecosystems (Yvon-Durocher *et al*. 2012). This can be due to differences in the importance of variables other than temperature, such as the primary productivity and terrestrial carbon inputs to the aquatic systems. In boreal regions, lakes are net sources of carbon, and in our case in southern Finland the lake was emitting between 70 and 100 g C m^{-2} each year. When compared with the surrounding forest, which was a net carbon sink, the emissions from the lake were enough to offset about 10% of the forest's annual carbon sequestration (Huotari *et al*. 2011). Part of the carbon entering the soil is transported to the aquatic systems dissolved in the water (Rasilo *et al*. 2012, Dinsmore *et al*. 2013). Thus the lateral transport between terrestrial and aquatic ecosystems should be accounted for when calculating ecosystem carbon balances. The current estimates of the transport of carbon are now probably conservative and underestimated due to the emission factors used in calculating the transport of greenhouse gases from rivers to the atmosphere (Huotari *et al*. 2013).

Earth system behaviour

Pre-defined topics for expected scientific breakthroughs:

- Development of a reliable aerosol module based on our parameterization to be used in climate and air quality models.
- Development of biosphere–atmosphere interaction model to be included in climate models.
- Estimate of climate change impact on boreal surface–atmosphere exchanges.
- Estimate of impact of climate change on aerosol forcing.
- Estimate of climate feedbacks involving natural secondary aerosols.
- Holistic understanding of biosphere-atmosphere interactions over boreal region.
- Reduced uncertainty of radiative forcing of atmospheric aerosols.

As part of our investigations on the Earth system behaviour, we developed and evaluated a new sectional aerosol module for large-scale modelling purposes (Kokkola *et al*. 2008, Bergman *et al*. 2012). We applied several process-level parameterizations in global models to investigate biosphere–atmosphere and air quality–climate interactions under present-day conditions and in changing climate (e.g. Makkonen *et al*. 2012a, 2012b, 2012c, Prisle *et al*. 2012).

The impact of climate change on the sinks and sources of greenhouse gases occurs primarily via changes in the ambient temperature and radiation, both driving the inter-annual variations in these sinks and sources (le Maire *et al*. 2010), and to a lesser extent via the water availability. Changes in the nitrogen deposition and carbon dioxide concentration per se have also impacts. We showed that warm autumns at high latitudes lead to a decreased carbon sink because the soil remains warm and keeps the respiration at a high level, and because the photosynthesis remains low due to the short day (Piao *et al*. 2008, Vesala *et al*. 2010). Warm springs, in turn, enhance the carbon uptake in boreal and temperate forests (Delpierre *et al*. 2009, Thum *et al*. 2009). Warming of the climate has also a significant impact on release of carbon from soils to the atmosphere (Thum *et al*. 2011).

The impact of climate change on the aerosol radiative forcing is coupled tightly with various feedbacks involving natural aerosols (Carslaw *et al*. 2010, IPCC 2013). We focused on two overlapping feedback loops proposed for secondary organic aerosols originating from BVOC emissions, both initiated by increasing CO_2 concentrations (Kulmala *et al*. 2004, 2013a). By combining a large number of field measurement data, we demonstrated that these feedbacks are likely to be active over large spatial scales over the continents, and that their strengths are large enough to warrant detailed future investigations (Paasonen *et al*. 2013, Kulmala *et al*. 2014b).

Our work has contributed to the reduction of the uncertainty in the radiative forcing by atmospheric aerosols (Kulmala *et al*. 2011b, Cappa *et al*. 2012, IPCC 2013). We identified several climate feedback mechanisms associated with terrestrial ecosystems (Arneth *et al*. 2010) and made the first quantitative strength estimates for two such feedbacks (Paasonen 2013, Kulmala *et al*. 2014b). We further showed that the role of natural aerosols in the climate system is affected by anthropogenic activities not only via climate change but also more directly via changes in aerosol and pre-cursor emissions (Arneth *et al*. 2009, Makkonen *et al*. 2012b). We conclude that the atmospheric composition is coupled tightly with both natural phenomena and anthropogenic activities, and that the future climate change and air quality cannot be investigated independently of each other.

International collaboration

One of the backbones of the successful scientific outcome of the FCoE has been our active participation in large-scale, integrated field and laboratory experiments in Europe and worldwide. The European-scale projects, focusing on producing standardized and quality-controlled research and data products, have increased (i) the active use of the FCoE data, (ii) the overall visibility of our research, and (iii) knowledge transfer between the FCoE and research teams elsewhere in the world. Working together with the leading research teams in Europe and United States has enabled us to up-scale local- and regional-scale observations into a global context.

All the thematic areas of the FCoE research have been closely connected to the European Union Framework Programmes (EU-FP). Development and application of new instruments for

detecting small molecular clusters and aerosol particles, which is a central theme in our FCoE, was made in close co-operation with "European Integrated project on aerosol, cloud, climate and air quality interactions" (EUCAARI, Kulmala *et al*. 2011b, *see* also Manninen *et al*. 2010, Mirme *et al*. 2010). The success of the FCoE in developing the nanoparticle measurement technology is largely based on our long-term collaborations with world-leading instrument developers such as Fred Eisele (e.g. Eisele and Tanner 1991), Peter McMurry (e.g. McMurry 2000) and Doug Worsnop (e.g. Jimenez *et al*. 2009). Within the frame of the project "Cosmics leaving Outdoor Droplets" (MarieCurie-ITN-CLOUD), the FCoE teams participated in leading-edge laboratory experiments on the formation of aerosol particles both via neutral and ion induced pathways (Kirkby *et al*. 2011, Almeida *et al*. 2013). Theoretical understanding of the new particle formation process has benefited from the tight collaboration with the European Research Council project "Role of molecular clusters in atmospheric particle formation" (MOCAPAF).

In order to address the large-scale research questions of "biosphere–atmosphere interactions and the "Earth system behavior", the most crucial EU-FP-projects have been "European Supersites for Atmospheric Aerosol Research" (EUSAAR; Philippin *et al*. 2009), "Aerosols, Clouds, and Trace gases Research InfraStructure Network" (ACTRIS-I3), "The Pan-European Gas-AeroSOls Climate Interaction Study" (PEGASOS), "Experimentation in Ecosystem Research" (EXPEER-I3), and the European Research Council project "Atmospheric nucleation: from molecular to global scale" (ATMNUCLE). The EU-FP projects have provided standardized, quality-checked and controlled data products and platform for unique field campaigns such as Zeppelin flights over Finland in 2013. The databases of the EUCAARI, EUSAAR and PEGASOS projects on aerosol properties and clouds, including comprehensive aerosols measurement in developing countries, are being utilized for developing and evaluating regional and global model simulating climate and air quality (e.g. Kulmala *et al*. 2011b). These projects also inspired the joint comprehensive field campaigns with e.g. the Max-Planck Institute (HUMPPA-COPEC 2010) (Williams *et al*. 2011).

The FCoE scientific breakthroughs in understanding the biosphere–atmosphere interactions and the Earth system behavior have been connected to the ATMNUCLE project. ATMNUCLE aims to quantify the mechanisms responsible for atmospheric new particle formation and to discover how important this process is in the context of the global aerosol and climate systems. ATMNUCLE, in co-operation with FCoE, developed continuous data on aerosol properties over the boreal forest region for testing new theories relevant to global models of aerosol particle formation. ATMNUCLE focused on measuring neutral clusters down to sizes of 1 nm as well as gas molecules relevant to new particle formation (Kulmala *et al*. 2013b). The new knowledge on the northern and Arctic aspects of the biosphere–atmosphere interactions, especially in the cryospheric environments, were attained via collaboration between FCoE and the two Nordic centers of excellence: Cryosphere–Atmosphere Interactions in a Changing Arctic Climate (CRAICC) and Impacts of a changing cryosphere — depicting ecosystem-climate feedbacks from permafrost, snow and ice (DEFROST).

The FCoE program on "Physics, Chemistry, Biology and Meteorology of Atmospheric Composition and Climate Change" together with the international collaboration has led to an integrated ensemble of a research approach, research infrastructure development and education activities. This work servers now as a baseline for the planning of the "Pan Eurasian Experiment (PEEX), which is a new type of multidisciplinary research initiative resolving the major uncertainties in the Earth system science and global sustainability questions in the Arctic and boreal pan-Eurasian regions (Kulmala *et al*. 2011a). Also the new initiatives to improve air quality in China are based on FCoE activities (Ding *et al*. 2013a, 2013b).

Future outlook

The work conducted within FCoE over the last six years has demonstrated that atmospheric oxi-

dation, aerosol chemistry and physics, clustering and aerosol formation are central to the cycles of carbon, nitrogen and sulphur in the Earth-atmosphere system.

By quantitative analysis of the initial phase transitions from gas molecules to clusters, we can gauge the atmosphere-ecosystem exchange processes of carbon, nitrogen and sulphur. During the new FCoE period, we aim to describe these complex processes and their feedbacks, starting from mesoscopic clusters and their growth to CCN sizes, their activation into cloud droplets, and ultimately the formation of precipitating clouds. The process-level understanding and mathematically robust descriptions will be applied to reveal the global consequences and feedbacks between the sub-systems in biosphere and atmosphere. To guide this work, we have identified the following topics with potential scientific breakthroughs during the next six years:

1. Atmospheric oxidation, the role of Criegee radicals originating from atmospheric oxidation of biogenic organic compounds (Kiendler-Scharr *et al*. 2009, Mauldin *et al*. 2012).
2. Cluster dynamics and composition particularly in sizes between 0.5–5 nm, including both neutral and ion cluster formation pathways, coagulation, growth and deposition (Winkler *et al*. 2008, Kirkby *et al*. 2011, Kulmala *et al*. 2013b).
3. Aerosol dynamics, including aerosol formation, growth, deposition, and phase transitions in aerosol particles (Virtanen *et al*. 2010, Riipinen *et al*. 2012).
4. Cloud condensation nuclei formation, cloud microphysics and precipitation formation including warm, mixed and ice clouds (Kerminen *et al*. 2012, Rosenfeld *et al*. 2008).
5. Ice nucleation and freezing in the atmosphere.
6. Biospheric phase transitions, including cavitation, freezing and ice nucleation in trees and their feedbacks with tree structure and water transport and VOC production within trees (Lintunen *et al*. 2013).
7. Seasonal variation of snow and ice cover as well as frost/permafrost distributions and their changes at the decadal scale together with secondary satellite data products combining vegetation, aerosols, clouds, radiation and atmospheric chemistry data.
8. Synthesis of BVOCs and organic nitrogen in various environments, including soils, lakes/rivers, peat lands, forests and cryosphere all around the world.
9. Atmospheric turbulence and its effect on atmospheric fluxes of clusters and their precursors including BVOCs, amines, sulphur compounds, atmospheric oxidants, HONO and carbonyl sulphide.
10. Atmospheric boundary layer dynamics and its interactions with greenhouse gas fluxes and biogeochemical cycles of water, carbon, nitrogen and sulphur (Kulmala and Petäjä 2011).
11. Quantification of continental biosphere-atmosphere–cloud–climate feedback loops (Kulmala *et al*. 2013a, Kulmala *et al*. 2014b), including the effects of disturbances (e.g. fire, extreme weather) on biogeochemical cycles.
12. Global and regional importance of the feedback loops, including the effects of changing biosphere, cryosphere and land cover (e.g. Nordbo *et al*. 2012) as well as changes in megacities.

The new FCoE will contribute its scientific findings directly to international assessments, including the assessment reports by the International Panel of Climate Change, as well as to international climate negotiations.

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