

# Overview of the biosphere–aerosol–cloud–climate interactions (BACCI) studies

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## ABSTRACT

Here we present research methods and results obtained by the Nordic Centre of Excellence Biosphere–Aerosol–Cloud–Climate Interactions (BACCI) between 1 January 2003 and 31 December 2007. The centre formed an integrated attempt to understand multiple, but interlinked, biosphere–atmosphere interactions applying inter and multidisciplinary approaches in a coherent manner. The main objective was to study the life cycle of aerosol particles and their importance on climate change. The foundation in BACCI was a thorough understanding of physical, meteorological, chemical and ecophysiological processes, providing a unique possibility to study biosphere–aerosol–cloud–climate interactions.

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Continuous measurements of atmospheric concentrations and fluxes of aerosol particles and precursors and CO<sub>2</sub>/aerosol trace gas interactions in different field stations (e.g. SMEAR) were supported by models of particle thermodynamics, transport and dynamics, atmospheric chemistry, boundary layer meteorology and forest growth. The main progress was related to atmospheric new particle formation, existence of clusters, composition of nucleation mode aerosol particles, chemical precursors of fresh aerosol particles, the contribution of biogenic aerosol particles on the global aerosol load, transport, transformation and deposition of aerosol particles, thermodynamics related to aerosol particles and cloud droplets, and the microphysics and chemistry of cloud droplet formation.

## 1. Background and objectives

Atmospheric aerosol particles and trace gases affect the quality of our life in many different ways. In polluted urban environments, they influence human health and deteriorate visibility (e.g. Pope and Dockery, 2006; Hand and Malm, 2007). On regional and global scales, aerosol particles and trace gases have a potential to change climate patterns and the hydrological cycle (Chung et al., 2005; Lohmann and Feichter, 2005; IPCC, 2007). Aerosol particles also influence the radiation intensity distribution reaching the surface of the Earth, and thus have a direct influence on the terrestrial carbon sink (Gu et al., 2002). Better understanding of the various effects in the atmosphere requires detailed information on how different sources (including those of the biosphere) and transformation processes modify properties of aerosol particles and trace gases. Trace gases and atmospheric aerosols are tightly connected with each other via physical, chemical, meteorological and biological processes occurring in the atmosphere and at the atmosphere-biosphere interface. An important phenomenon, for example, is atmospheric aerosol formation, which involves the production of nanometre-size particles by nucleation, and their growth to detectable sizes (Kulmala, 2003). Human actions, such as emission policy, forest management and land use change, and various natural feedback mechanisms involving the biosphere and atmosphere have an impact on the coupling between the aerosols and trace gases.

In 2007, the intergovernmental panel on climate change (IPCC) estimated the global annual radiative forcing due to greenhouse gases and aerosols, along with natural changes associated with solar radiation. Emphasis was placed on the complexity of the combined direct and indirect forcing from both aerosols and gases, as well as on the importance of improving our understanding of the role that each of these three individual components plays in radiative forcing in an integrated system. Such knowledge would reduce the uncertainty in current estimates of radiative forcing and enable a better prediction of the effects of anthropogenic activity on global change. The most important issue to resolve is how the different components affecting radiative forcing interact with each other. It is promising that since the previous report of IPCC (2001), overlapping with the duration and achievements of the BACCI program, the role of aerosols in radiative forcing has been better elucidated, although the uncertainties related to the radiative forcing related to aerosols are still large.

One of the key issues related to aerosol dynamics is the formation and growth of atmospheric aerosol particles. Although aerosol formation has been observed all over the world (Kulmala et al., 2004a) and although it seems to be frequent (see e.g. Dal Maso et al., 2005; 2007), there are still several gaps in our knowledge on this process. The status of current knowledge is that atmospheric aerosol formation is a result of photochemical reactions in the gas phase, particularly the formation of sulphuric acid (Kulmala et al., 2006; Sihto et al., 2006; Riipinen et al., 2007a) and other vapours of very low volatility, such as multifunctional organic compounds and iodine oxides (e.g. O'Dowd et al., 2002a,b, 2005; Allan et al., 2006). Pre-existing aerosol particles, on the other hand, act as a sink for these vapours and nucleated clusters, thus inhibiting atmospheric aerosol formation. Aerosol formation also seems to be affected by several meteorological parameters and phenomena, including the intensity of solar radiation and atmospheric mixing processes such as the evolution of a continental boundary layer or the mixing of stratospheric and tropospheric air near the tropopause.

We have recently proposed a mechanism coupling the climatic effects of CO<sub>2</sub> and aerosol particles (Kulmala et al., 2004b). This suggestion is based on connections between photosynthesis, emissions of non-methane biogenic volatile organic compounds (BVOCs), and their ability to form aerosol particles. Photosynthesis drives plant gross primary production (GPP), the difference between net ecosystem exchange of CO<sub>2</sub> (NEE) and total ecosystem respiration (TER) (see e.g. Valentini et al., 2000). In the boreal zone, photosynthesis occurs predominantly in sunlight during the growing season (Hari and Mäkelä, 2003) and is inhibited in winter. Forest ecosystems are usually sinks of CO<sub>2</sub>, and a direct negative feedback (the higher the CO<sub>2</sub> concentration, the higher the rate of photosynthesis) exists between increasing atmospheric CO<sub>2</sub> concentrations and photosynthesis, whereas a positive feedback lies between ecosystem respiration and temperature. With higher temperatures, water becomes a more important factor influencing both GPP and TER. On the other hand, forest ecosystems also act as significant sources of atmospheric aerosols. Terrestrial vegetation contributes substantially to emissions of a variety of BVOCs (Fuentes et al., 2000) via processes closely linked with photosynthesis (e.g. Bäck et al., 2005), and newly formed particles in forested areas have been found to contain large amounts of organic material (O'Dowd et al., 2002a; Tunved et al., 2006a,b). The

carbon lost as BVOC emissions related to assimilation is generally only a few percent (Grace and Rayment, 2000), but with the significant process couplings due to climate change, the formation and emissions of several BVOCs may increase and possibly modify the aerosol particle formation routes. Besides being an important aerosol source, vegetation also acts as a substantial sink for particles, due to the dependence of dry deposition on particle size (Pryor et al., 2008) reshaping the particle size distribution.

The mechanism described above, linking terrestrial photosynthesis and aerosol forcing in a novel way, is only one among many connections and feedbacks in the Earth system. The climatic and other effects of aerosols and trace gases are coupled together both via human actions, such as emission policy and land use changes, and via various natural feedback mechanisms involving the biosphere and atmosphere (see e.g. Hari et al., 2003; Magnani et al., 2007). As an example, reduced aerosol loads due to actions aiming to improve air quality already seem to have affected climate in certain regions of the world (Brasseur and Roeckner, 2005). Aforestation enhances carbon sinks but at the same it decreases the surface albedo, which tends to compensate for the effect of carbon sink feedback (Gibbard et al., 2005). It is also very probable that some new feedbacks or new pathways for components of biogeochemical cycling (such as that proposed by Keppler et al., 2006) may appear in the future.

The new aerosol particles formed by atmospheric nucleation events (see e.g. Kulmala et al., 2004a) become climatically important only if they grow to sizes larger than about 50–100 nm in diameter. Particles of this size and larger can scatter sunlight back to the space, and they also can act as nuclei for cloud drop formation, having thereby an indirect effect on the albedo of the earth. Furthermore, the health effects of particles are related not only to the toxicity of the particle material but also to the particle size, since the size determines whether the particles are able to penetrate to the lungs and further to the blood circulation. According to our studies on the growth of freshly formed particles both in pristine (Pallas, Finland; Lihavainen et al., 2003; Komppula et al., 2005; Kerminen et al., 2005) and highly polluted (Po Valley, Italy; Laaksonen et al., 2005) environments, nucleation can produce a significant fraction of cloud condensation nuclei regardless of the regional emission strength of primary particulate matter.

The influence of aerosol particles on clouds and Earth's radiation budget is strongly tied with the number concentrations of aerosol particles activating into cloud droplets during the cloud formation. Several chemical and microphysical effects on cloud droplet formation have been identified (e.g. Kulmala et al., 1997; Charlson et al., 2001; Nenes et al., 2002). These effects include surface tension lowering of water droplets due to various organic aerosol species and uptake of trace gases such as nitric acid during cloud formation. Our studies (Sorjamaa et al., 2004; Kokkola et al., 2006) have revealed that, due to intricate effects of surfactant molecule partitioning between the interior and the surface of

minuscule cloud droplets, the surface tension influence on cloud formation is much smaller than what was previously thought (e.g. Facchini et al., 1999). We have also advanced considerably the understanding of the effect of nitric acid on cloud formation (Romakkaniemi et al., 2005b) and parameterized the effect in such a way that it can now be explored using climate models (Romakkaniemi et al., 2005a).

The main scientific objective for BACCI has been to study the importance of aerosol particles on climate change and also on human health. Particularly, we have focused on: (i) formation and growth of atmospheric aerosols, aerosol and ion dynamics; (ii) the effect of secondary biogenic aerosols on regional and global aerosol loads; (iii) aerosol–cloud–climate interactions and (iv) the relations between atmospheric and different ecosystem (particularly boreal forest and also oceans). In this paper we will summarise our recent scientific findings, discuss our original scientific aims and questions and provide answers to these questions.

## 2. Scientific approach and research methods

The system involving atmospheric circulation, composition and impacts is tremendously complex. It requires a diverse range of scientific and technological expertise in the areas of chemistry, physics, biology and meteorology, and involves laboratory studies, ground, ship and airborne field studies, satellite remote-sensing and numerical modelling studies ranging from the molecular *ab initio* level to the global scale Earth system models. Individual efforts to address climate change and impacts of pollutants are clearly advancing, but in a disjointed manner, which has inhibited an efficient exchange of knowledge between different communities. In addition, databases from disconnected field experiments are difficult to merge successfully to tackle issues related to both atmospheric composition and climatic impacts. To be able to meet our objectives and to answer our research questions, we have therefore aimed at inter, multi and cross-disciplinary research with a high level of technological and scientific innovation.

Our scientific approach spans the range from basic nucleation theories followed by models of detailed aerosol dynamic/atmospheric chemistry and vegetation–atmosphere exchange to well-defined laboratory experiments. Next in chain are versatile continuous and field measurements in our research stations, including intensive measurement periods as well as intensive field experiments such as airborne campaigns. The results are extended to models from process oriented box models to global-scale modelling. Our approach thus covers both experimental (laboratory and field experiments) and theoretical (basic theories, simulations, model development, parameterizations) aspects, and it can be described using a research chain (see Kulmala et al., 2005b) that aims to advance our understanding of climate and air quality through a series of connected activities beginning at the molecular scale and extending to the regional

and global scale. We focus on those aspects of the research chain where uncertainties are largest.

In practice, we have started from molecular simulations that are needed to understand the basic nucleation, kinetic and thermodynamic processes (Quantum Chemical, Monte Carlo and Molecular Dynamics; e.g. Merikanto et al., 2004; Kurten et al., 2007a,b,c). To quantify the aerosol dynamical processes related to new particle formation, we have developed a detailed model University of Helsinki multicomponent aerosol model (UHMA; Korhonen et al., 2004). The UHMA model has been coupled further to various other types of atmospheric models, such as a chemical transport model (Grini et al., 2005), outdoor/indoor model (Hussein et al., 2005), Lagrangian air parcel model (Tunved et al., 2006a,b) and column model (Boy et al., 2006). New approaches for interpretation of laboratory data have been developed (e.g. Winkler et al., 2004; Berndt et al., 2005; Riipinen et al., 2006). We have also developed models simulating photosynthesis, autotrophic respiration,  $\text{NO}_x$  exchange and BVOC synthesis in cellular, stomatal and leaf (or shoot for conifers) scales, and tested these models with chamber measurements under laboratory and field conditions (e.g. Hari et al., 2003).

The fundamental aerosol and carbon cycle processes need to be understood to quantify aerosol radiative properties and the influence of aerosols on cloud microphysics and dynamics at the scale of individual clouds and to understand changes in carbon uptake dynamics (e.g. Kulmala et al., 2004b). At larger scales, advances in our understanding of boundary layer meteorology are needed to clarify atmospheric aerosol transport, trace gas (e.g.  $\text{CO}_2$ , methane,  $\text{N}_2\text{O}$ ,  $\text{O}_3$ ,  $\text{SO}_2$ ,  $\text{NO}_x$ , VOCs) and water vapor exchange and deposition processes (e.g. Pryor et al., 2007b). Boundary layer studies form a link to regional-scale processes and further to global-scale phenomena. To be able to simulate global climate and air quality, the most recent progress on this chain of processes must be compiled, integrated and implemented in Climate Change (CC) and Air Quality (AQ) numerical models via novel parameterizations (e.g. Spracklen et al., 2006; Hoyle et al., 2007).

The work in BACCI has been divided into 10 work packages (Fig. 1). The core activity in BACCI is long-term field measurements, which include continuous measurements, intensive measurement periods and separate measurement campaigns. Continuous or campaign-wise measurements have been conducted in all our stations (SMEAR I–III, Puijo tower in Kuopio, Stockholm, Copenhagen, Oslo, Pallas, Aspvreten, Birkenes, Vindeby, Lille Valby, Sorø, Vavihill and Ny Ålesund; see Fig. 2). These stations differ in a variety of environmental factors, and therefore, the data obtained can be used to form a comprehensive picture of the processes involved. The measured parameters include particle number size distribution, trace gas concentrations and basic meteorological variables. During intensive periods, very detailed measurements of trace gases, aerosol chemistry and various boundary layer parameters have been conducted. Sep-

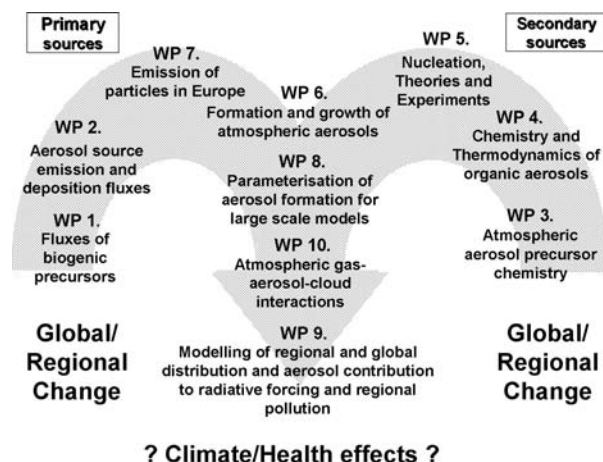


Fig. 1. BACCI work packages and their connections.



Fig. 2. The research stations operated by BACCI.

arate continuous measurements and field campaigns have been made in different locations all over the world (e.g. the Galathea3 cruise, airborne aerosol measurement campaigns in the Arctic and Antarctica, TransSiberian train expedition, field measurements in the Amazon rain forest), either as BACCI co-operative basis or as part of other international projects.

The continuous data, together with coupled Lagrangian particle dynamic and chemical box-model studies, have been used to evaluate the role of nucleation, condensation, coagulation and dry deposition (e.g. Tunved et al., 2004a, b, Tunved et al., 2006b). These studies have demonstrated that box model designs can be successfully used to assess the importance of dynamic processes affecting the aerosol particle population over large land

areas in the boreal region. The successful deployment of the used Lagrangian process models indicate the necessity and value of combining regional measurement strategies with detailed aerosol models both for validating process descriptions as well as for development of parameterizations for future implementation in regional models.

The processes and properties in terrestrial ecosystems and atmosphere are strongly connected. For example, the atmospheric CO<sub>2</sub> concentration influences photosynthesis and VOC emissions from trees and therefore has profound effects on aerosol formation (e.g. Kulmala et al., 2004b). Simultaneous measurements of several phenomena in a forest ecosystem enables combination of different processes and analysis of the connections between the components of the system such as: (i) carbon fluxes connecting the atmosphere, trees and forest soil with each other; (ii) volatile carbon compounds emitted by trees or forest soil are evidently important for the formation of aerosol particles and (iii) rainfall, transpiration, evaporation and run-off connect the atmosphere, trees and soil (see also Hari and Kulmala, 2005).

The rapid development of measurement techniques has enabled versatile field measurements (Hari and Kulmala 2005). New trace compounds can be measured under field conditions, the measuring accuracy and precision are increasing and the required response time is decreasing. For example, plant-atmosphere exchange of more than 10 gases can be monitored with the present instrumentation using dynamic shoot chamber techniques, which requires a response time of only a few minutes. In addition, large measuring systems can be automated with digital techniques.

Laboratory studies on aerosol properties are required for two reasons; first, realistic input parameters are needed for models, and second, in certain cases experimental work is necessary to verify theoretical and model approaches (Kulmala, 2003). The aerosols can be generated either from well-defined pure or mixed compounds or by processes imitating those in the atmosphere (e.g. ozonolysis of plant-released VOCs in controlled laboratory environment, dispersion of collected rain, sea, lake water samples, etc.). Properties that have been studied include: (i) water vapor uptake at varying relative humidities, especially for nm-sized particles (Winkler et al., 2004); (ii) cloud condensation efficiency at varying supersaturations (e.g. Svenningsson et al., 2006) and (iii) the chemical composition of particles (e.g. aerosol mass spectra of known particle types for reference to atmospheric studies, composition of plant derived particles, ethanol uptake and volatility of different particle types; e.g. Joutsensaari et al., 2005).

### 3. Scientific results

At the beginning of BACCI, we identified several key questions to be addressed (presented in Kulmala and Tammet, 2007). In the following we provide a brief summary of our main scientific findings.

#### 3.1. Observations of aerosol formation events and atmospheric clusters

Probably the most significant result is the observation that the formation of new atmospheric aerosol particles and their subsequent growth is ubiquitous (Kulmala et al., 2004a, and references therein). In fact, aerosol formation has been observed to take place in very different lower-tropospheric environments, including Antarctica and the surrounding oceans, forested areas, other remote and rural continental areas, coastal environments, various urban centres and even heavily-polluted regions and megacities. However, new particle formation is rare in the marine boundary layer. Formation of new aerosol particles has also been observed in the free troposphere, being a frequent phenomenon in the upper troposphere and in cloud-outflow regions. The first and longest continuous data set on atmospheric aerosol formation events is available from the SMEAR II station (Kulmala et al., 2001, Dal Maso et al., 2005, 2007).

An example of a typical aerosol formation event, as seen from traditional particle number size distribution measurements, is shown in Fig. 3. From such measurements one may calculate the apparent source rate of 3-nm particles and particle growth rate (Kulmala et al., 2005c). According to our literature review (Kulmala et al., 2004a), the formation rate of 3-nm particles is often in the range of 0.01–10 cm<sup>-3</sup> s<sup>-1</sup> in the boundary layer. However, in urban areas formation rates are often higher (up to 100 cm<sup>-3</sup> s<sup>-1</sup>), and rates as high as 10<sup>4</sup>–10<sup>5</sup> cm<sup>-3</sup> s<sup>-1</sup> can be observed in coastal areas and in industrial plumes. Typical particle growth rates are in the range 1–20 nm h<sup>-1</sup> at mid-latitudes depending on the temperature and the availability of condensable vapours. Over polar areas the growth rate can be as low as 0.1 nm h<sup>-1</sup>. Nucleation can lead to a significant increase in the number concentration of cloud condensation nuclei, even at highly polluted regions such as Po Valley in Italy (Laaksonen et al., 2005) and New Delhi, India (Mönkkönen et al., 2005). As a summary, atmospheric nucleation is a global phenomenon taking place over large areas, and it appears to be frequent as well. However, campaign-based observations in the Amazon suggest that in the pristine tropical rainforest environment, nucleation events do not occur as regularly as in other parts of the world, and therefore, the aerosol formation in this part of the world is a challenging issue for future research (Krejci et al., 2005; Rissler et al., 2006).

Since atmospheric nucleation starts from particle size below 3 nm in diameter, it is important to know what happens in this size range. Ion spectrometers are able to reveal part of the evolution and dynamics of small ion clusters, and therefore, we have developed several ion spectrometers during the recent years (e.g. Mirme et al., 2007). Ion clusters seem to be present everywhere and in practise all the time (e.g. Kulmala and Tammet, 2007). Using ion spectrometers (see Fig. 4), the initial steps of particle formation and growth can be detected (e.g. Kulmala et al., 2004c; Hirsikko et al., 2005).

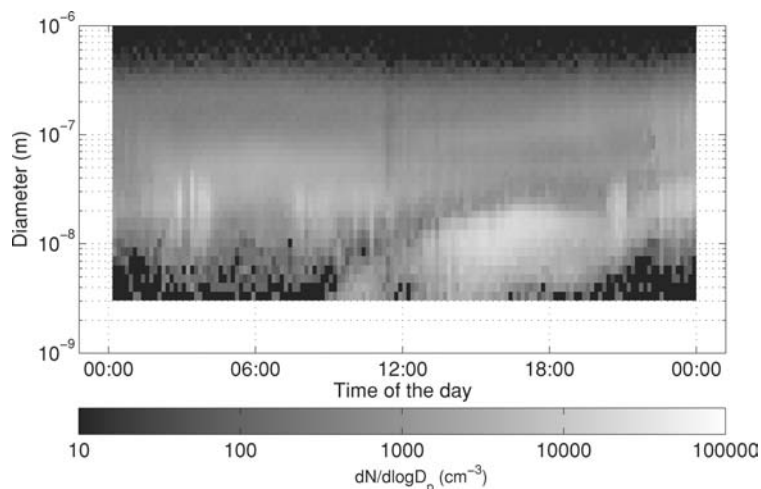


Fig. 3. New particle formation event observed at SMEAR II station on 9 April 2007, by using differential mobility particle sizer.

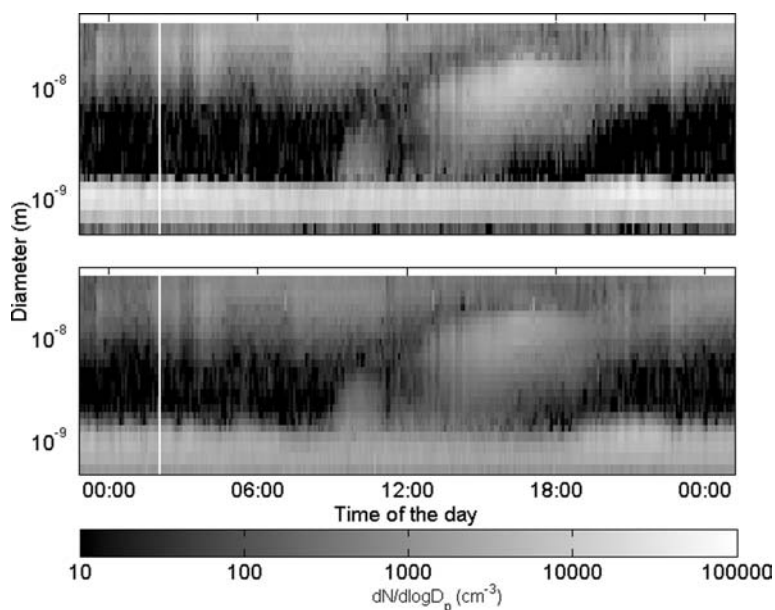


Fig. 4. New particle formation event observed at the SMEAR II station on 9 April 2007, by using an air ion spectrometer (AIS; Mirme et al., 2007). The dynamics of ion clusters is readily seen.

The existence of ion-induced nucleation related to atmospheric particle formation was found by estimation of charge balance on nanometre aerosol particles or the ratio of negative to positive intermediate ions (Hörrak et al., 2004, Vana et al., 2006). The charge preference was found—the nucleation occurred mainly on negative cluster ions. Estimating ion production rate by small ion balance equation and by the measurements of external radiation and radon also showed existence of ion induced nucleation in boreal forest (Laakso et al., 2004a,b).

Unfortunately, traditional ion spectrometers cannot reveal whether there are neutral clusters in atmosphere or explain their role in atmospheric nucleation. Already several years ago, model calculations predicted the existence of such neutral clusters

(Kulmala et al., 2000). However, during the BACCI period, several efforts towards detecting atmospheric neutral clusters have been undertaken. Already some years ago, Kulmala et al. (2005a) found evidence on the existence of neutral clusters *in vivo*. After that we have developed new instruments capable of both detecting neutral clusters and estimating their number concentrations in different atmospheric environments (Kulmala et al., 2007a). As a result, atmospheric nucleation can be seen to start from around 1.5 to 2.0 nm (Fig. 5). Very recently, neutral clusters have also been detected using condensation particle counter techniques (Fig. 6).

In association with BACCI, atmospheric nucleation studies have been performed in a great variety of different environments around the world. A map showing the location of these

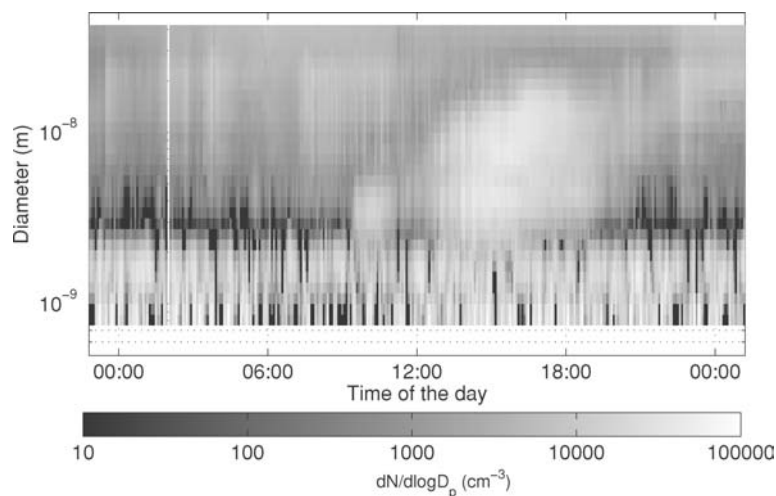


Fig. 5. New particle formation event observed at SMEAR II station on 9 April, 2007, by using a neutral cluster and air ion spectrometer (NAIS). The atmospheric nucleation can be observed to start from around 2 nm size (see also Kulmala et al., 2007a).

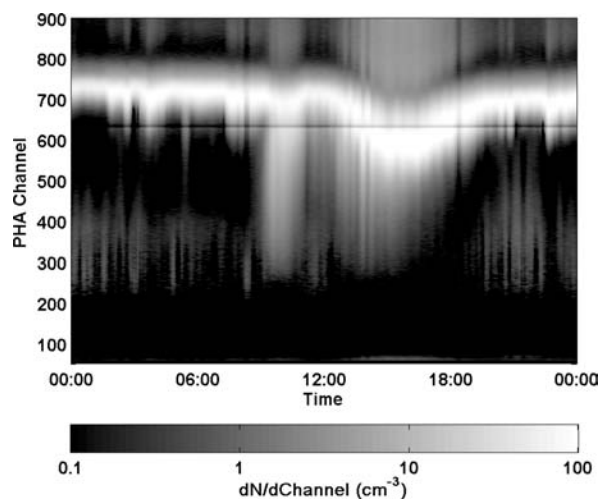


Fig. 6. Aerosol particles and clusters observed by using a pulse-height condensation particle counter (PH-CPC) (observed at SMEAR II station on 9 April, 2007). The clusters are shown at around the channel 300. Particles larger than about 5 nm in diameter are accumulating to the channels of around 600. Additional diffusion battery measurements have shown that the average size of detected clusters is approximately 1.8 nm. The detection efficiency of clusters is not taken into account.

measurement sites, along with the corresponding sites summarized in our earlier review (Kulmala et al., 2004a), is shown in Fig. 7.

### 3.2. Aerosol precursors and related thermodynamics

Although there is strong evidence that sulphuric acid participates in aerosol formation processes (Kulmala et al., 2006, Sihto et al., 2006, Riipinen et al., 2007a), sulphuric acid alone cannot explain the condensation growth of freshly-nucleated particles under typical atmospheric conditions (Kulmala et al. 2001, Kulmala et al. 2004a). Our field observations provide clear evidence that organic vapours participate in aerosol formation or at least in the growth of particles to larger sizes (Allan et al.,

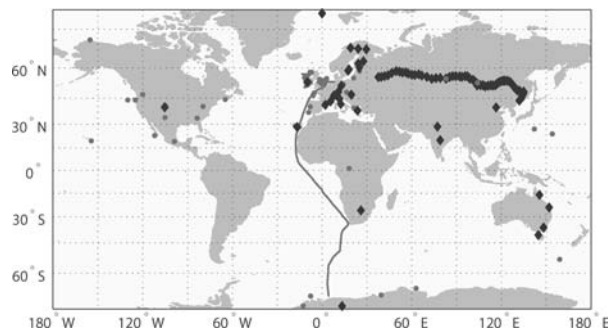


Fig. 7. Location of the sites in which atmospheric aerosol formation has been measured and observed. Quadrangles denote the sites operated by BACCI partners using either differential mobility size spectrometers or ion spectrometers whereas circles denote other sites included in our literature review (Kulmala et al., 2004a).

2006; Tunved et al., 2006a; Laaksonen et al., 2007). Also our (first ever) laboratory study of the ozonolysis of plant emitted VOC's indicates that condensable organics can cause particle growth but are unlikely to nucleate at atmospheric conditions (Joutsensaari et al., 2005).

Unfortunately, it is not possible to get direct information about the chemical composition of freshly-nucleated aerosol particles or clusters. As a result, we have developed new in situ methods, such as the CPC battery (Kulmala et al., 2007b), for studying the water solubility of nucleated particles. This way the composition, or composition changes, of particles in the nucleation mode can be found out indirectly. Instruments like HTMA, VTDMA and TDCIMS provide further information about the chemical properties of nucleated particles (Swietlicki et al., 2008; Smith et al., 2005). In addition to measurements, we have made quantum chemical calculations to predict the probable composition of the atmospheric clusters (e.g. Kurtén et al., 2007a,b).

One way to investigate which compounds might participate in aerosol formation is to study the thermodynamical

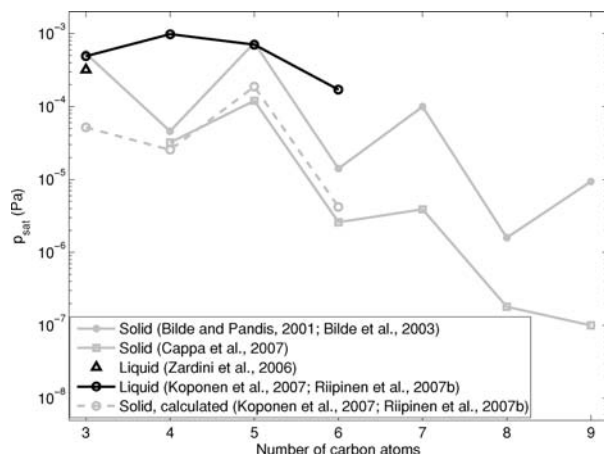


Fig. 8. The saturation vapour pressures of subcooled (red) and solid (black) C3–C9 dicarboxylic acids. The black markers with red connecting lines correspond to solid state values calculated from corresponding subcooled values. It can be seen that the subcooled state values obtained by Riipinen et al. (2006), Koponen et al. (2007) and Riipinen et al. (2007b) are consistent with the subcooled malonic acid vapour pressure reported by Zardini et al. (2006), the solid state values reported by Cappa et al. (2007) and, in case of even acids, also with the results of Bilde et al. (2003).

properties of these compounds. Although we know that the saturation concentration of condensing vapours seems to be around  $1\text{--}3 \times 10^6 \text{ cm}^{-3}$  (Kulmala et al., 1998), deep insight into the thermodynamic properties of atmospheric clusters and condensing vapours is still missing. However, more laboratory experiments close to real atmospheric conditions are needed. As an example, Fig. 8 shows that the saturation vapour pressures of dicarboxylic acids are clearly too high to explain atmospheric nucleation and growth events. At the beginning of BACCI, this question was identified as being very uncertain, and currently, the thermodynamics involved in new-particle formation remains ambiguous. Anyhow, thermodynamic studies are the best way to increase our fundamental understanding of the nature of the compounds participating in atmospheric nucleation and growth of particles.

In addition to aerosol formation, thermodynamics is important to cloud microphysics and chemistry. Facchini et al. (1999) suggested that the surface tension depression by organic substances would influence cloud droplet formation. However, detailed considerations of surface thermodynamics have shown that finite-size effects in cloud drops reduce surface tension effects to almost negligible magnitudes (Sorjamaa et al., 2004; Kokkola et al., 2006; Sorjamaa and Laaksonen 2006). This may reduce the need for elaborate surface tension measurements of numerous multicomponent dilute mixtures.

### 3.3. Regional atmosphere–biosphere interactions

The BACCI community has made an integrated attempt to understand various, but interlinked, biosphere–atmosphere inter-

actions by applying inter and multidisciplinary approaches. We have collected exceptionally long data series on biosphere–atmosphere interactions, especially for atmospheric aerosol formation (e.g. Dal Maso et al., 2005; 2007), and also for carbon dioxide, methane and water vapour over different ecosystems.

The evolution of the aerosol population, as transported over the forested areas over Scandinavia, has been investigated using Lagrangian box models with current knowledge of processes. The used models simulate aerosol dynamics during both the inflow of clean marine air and the outflow of more polluted air over Scandinavia. In general, the model results are in a very good agreement with measurements. Emission of organics seems to be determining the evolution of both number and mass of particles across the selected transects (Tunved et al., 2004a, b; Tunved et al., 2006b). Back trajectory studies indicate that aerosol formation is better correlated with solar radiation in clean, arctic air masses, as compared with continental air where particulate pollution suppresses nucleation by acting as a sink for condensable vapours (Fig. 9, Sogacheva et al., 2005). In summary, the boreal forest acts as a source of the particle number and mass during the southerly transport of clean marine air. On the other hand, northerly transport is often accompanied by a reduction in the particle number but an increase in the particle mass.

Over the Boreal region of Nordic countries, it has been shown that the forest can rapidly establish and support an aerosol population of 1000–2000 particles, including both Aitken and accumulation mode particles via gas-to-particle transformation (Tunved et al., 2006a). This significant natural particle source is most likely capable of competing also with anthropogenic cloud condensation nuclei loadings transported over these areas. These findings support further the idea that the forest is a significant source of both aerosols mass and number. The studies place the boreal region in a central position in evaluation of climate effects originating from natural aerosol emissions. Regarding aerosol–cloud interactions, we have found direct evidence that aerosol particles formed in the atmosphere are, after their subsequent growth, able to take part in cloud droplet formation and thereby alter the climatically important cloud properties (Kerminen et al., 2005).

We have also improved our understanding on the functional dependencies of the dry deposition of atmospheric particles (Pryor 2006, Pryor et al. 2007a). This was partly motivated by the findings showing that whereas the observed size dependence of particle dry deposition velocities over low roughness vegetation is in good accord with models, this is not the case for high roughness vegetated surfaces—that is, forests (Pryor et al. 2008). We used conditional sampling and also comparative analyses of forests with differing architecture to explore these functional dependencies. Among other things, our results indicate that the size variation of dry deposition velocities in the diameter range 40–1000 nm over forests is markedly over stated in models such as that by Slinn (1982).



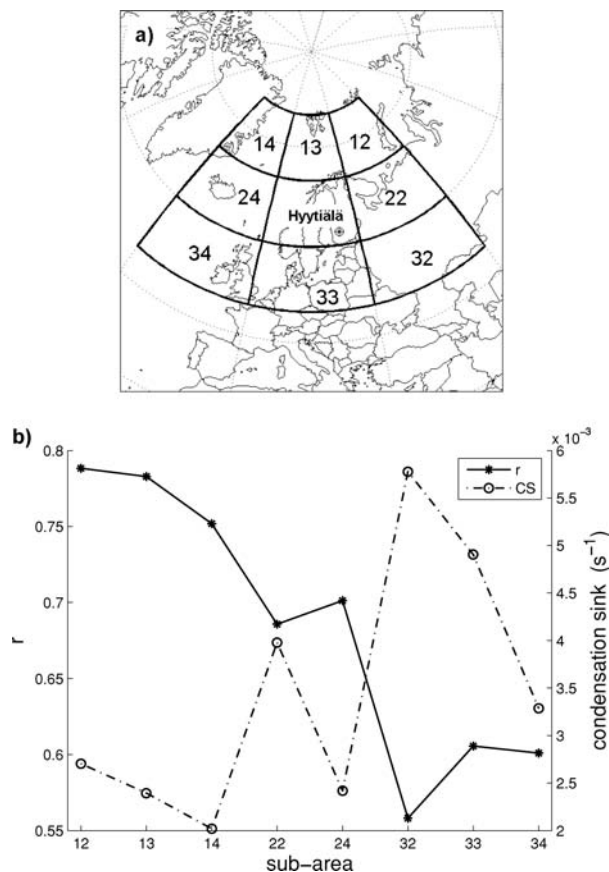


Fig. 9. (a) Division of 4 d back trajectory origins into subareas. (b) Correlation coefficients ( $r$ ) between the monthly number of back trajectories corresponding to nucleation event days in each subarea and the number of all back trajectories from the same subareas multiplied by normalized mean monthly PAR (photosynthetically active radiation) value, and the average condensation sink (CS) associated with back trajectories passed through each subarea. The points are connected for clarity. For the clean Arctic air masses, the correlation between the number of event trajectories and the radiation parameter is quite high (0.7–0.8), whereas for air masses which contain products of the anthropogenic activity, the correlation is lower (0.5–0.6). Thus, for the northern air masses we can predict the monthly number of nucleation days reasonably well by considering radiation level. The increase of the anthropogenic activity makes the prediction worse—the aerosol condensation sink behaves almost exactly opposite to the correlation coefficient, increasing toward more polluted subareas. The only exception is subarea 34, which includes both open ocean and the British Isles, and therefore, the trajectories passed through the subarea represent a mixture of clean and polluted air masses, which explains a simultaneous low correlation coefficient (effect of polluted air masses) and a relatively low average condensation sink (effect of clean air masses).

### 3.4. Implications for large-scale modelling and related applications

Global climate models have gradually reached a state at which aerosol microphysical processes can be explicitly treated in sim-

ulating the future composition of the atmosphere (e.g. Stier et al., 2006; Ghan and Schwartz, 2007; Shindell et al., 2007). This puts the work performed within the BACCI community into a slightly new perspective. First of all, we ultimately have an opportunity to test how important our regional findings are to the global climate system now and in the future. Second, there is an urgent need for accurate yet computationally efficient new parameterizations describing the key aerosol processes in these models.

During the recent years, we have developed a number of new parameterizations for detailed (e.g. Merikanto et al., 2007) and more simplified (e.g. Kulmala et al., 2006; Sihto et al., 2006; Riipinen et al., 2007a) atmospheric nucleation mechanism. We have also developed an effective way of treating atmospheric new-particle formation in large-scale modelling frameworks (Kerminen et al., 2004; Lehtinen et al., 2007). Concerning aerosol–cloud interactions, we have developed a parameterization on the effect of nitric acid on cloud droplet activation (Romakkaniemi et al., 2005a) and studied how the activation process should be dealt with in models having a crude representation of the aerosol size distribution (Korhonen et al., 2005). Relationships between aerosol content and cloud cover for some regions have indicated strong aerosol–cloud interactions with a prolonged cloud lifetime (Kaufman et al., 2005; Koren et al., 2005). During BACCI, relationships between MODIS satellite data of aerosols and clouds have been established on the global scale, showing that the increase in cloud cover with aerosol content is a global phenomenon, with the strongest increase in cloud cover occurring for small aerosol contents (Myhre et al., 2007b). By combining MODIS satellite data and global aerosol models, the study shows that hygroscopic growth of aerosols in the vicinity of clouds influences the relationship between aerosol content and cloud cover but probably to an insufficient extent to describe the relationship seen by the satellite data.

The amount and representativeness of field data collected within BACCI have allowed us to test the overall performance of new-particles formation parameterizations in a global chemical transport model (Spracklen et al., 2006). The same study has demonstrated that atmospheric new-particles formation is likely to contribute significantly (about 30%) to the total particle number concentration in the global troposphere. Our most recent studies indicate that also the global budget of cloud condensation nuclei, a quantity relevant to the aerosol climatic effects, may be significantly affected by atmospheric aerosol formation (Spracklen et al., unpublished).

Significant development has been made during the BACCI period in implementing schemes for calculations of secondary particle production (nitrate and secondary organics) and distribution in global scale models (Oslo CTM2) (Myhre et al., 2006; Hoyle et al., 2007). Modelling of nitrate is more complex than for sulphate since it depends more on other aerosols and aerosol precursors. Ammonium nitrate can be formed when there is excess of ammonia after neutralization of sulphate, but this reaction is

2004 Surface SOA

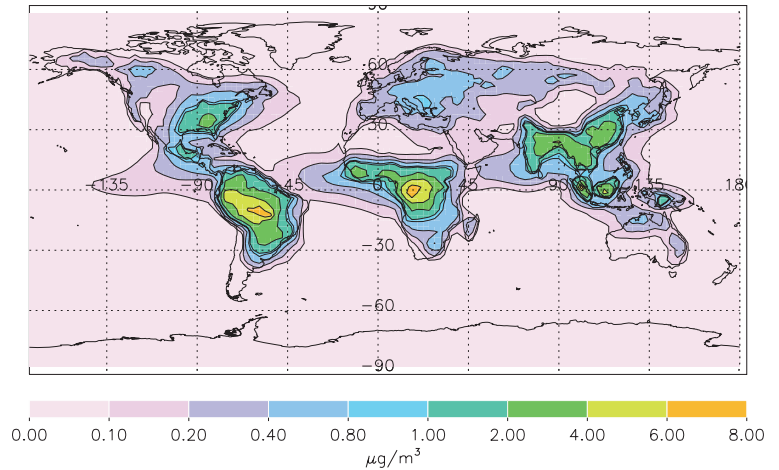


Fig. 10. The surface concentration of SOA for 2004, as modelled by the Oslo CTM2.

temperature dependent (Metzger et al., 2002). Nitrate may also be produced on larger aerosols such as sea salt and dust particles (Metzger et al., 2006). The surface concentration of nitrate in the Oslo CTM2 compares well with observations (Myhre et al., 2006), and importantly, the fine mode nitrate over ocean is very small, which is also in accordance with measurements (Quinn and Bates, 2005). Observations show that coarse mode nitrate over oceans is substantial (Quinn and Bates, 2005) and this is also the case in the Oslo CTM2. This feature is important since nitrate on coarse sea salt particles has a negligible anthropogenic radiative forcing (Myhre et al., 2006). With the Oslo CTM2 it is shown that the secondary organic aerosols (SOA) constitute a significant fraction of the total organic matter in the atmosphere (Hoyle et al., 2007). Measurements in the industrialized regions show a dominance of the SOA compared with primary OC dur-

ing summer (Crosier et al., 2007; Gelencser et al., 2007), and the model reproduces this characteristic. The SOA will have a substantial radiative forcing that is not accounted for in studies so far or in the IPCC reports.

The present day (2004) concentration of SOA at the surface, from the Oslo CTM2, is shown in Fig. 10, and the fraction change in SOA concentrations, since 1750 is shown in Fig. 11. The present day concentration of SOA is highest in regions with both large emissions of the natural precursors, and large emissions of primary organic aerosols (POA) into which the SOA will partition. Therefore, the largest concentrations are found in South America, southern parts of Africa and Indonesia. Anthropogenic emissions also contribute significantly to SOA, as shown by the elevated SOA concentrations in South-East Asia, and the western USA. A significant increase in surface SOA concentrations since

2004/1750 Surface SOA

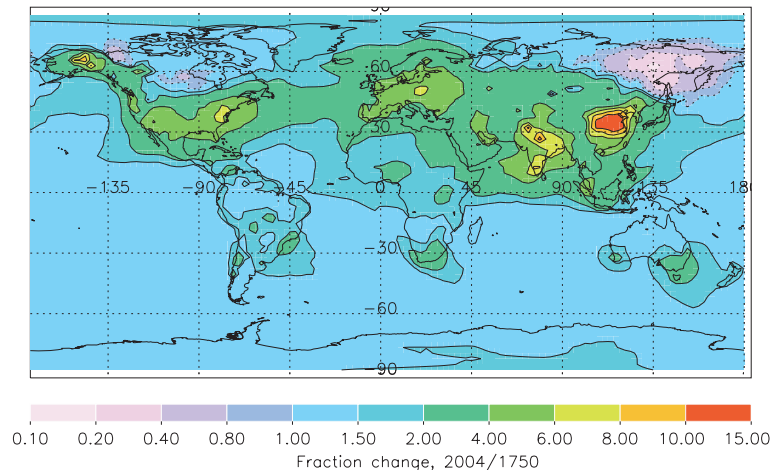


Fig. 11. The fraction increase in surface SOA concentrations between 1750 and 2004, as modelled by the Oslo CTM2.

1750 can be seen in Fig. 11, especially around the industrialized areas such as the west coast of the USA and Asia. This is partly a result of increased POA mass in these areas, from fossil fuel and bio fuel burning, but also due to emissions of aromatic species, which are oxidized and form SOA. Additionally, an increase in ozone and  $\text{NO}_3$  in industrialized regions leads to more oxidation of natural precursors in these areas, further increasing the regional SOA concentrations. Thus, the anthropogenic contribution to increased SOA concentrations, especially near industrialized areas, is related to both direct emissions and a modification of the oxidizing capacity of the atmosphere. The large areas of reduced SOA concentrations north of Japan are related to the reduction in POA emissions in the biomass burning inventories between 1750 and 2004.

Preliminary results based on Hoyle et al. (2007) indicate that the radiative forcing may be twice as strong as that of the primary OC from fossil and bio fuel emissions; a forcing which is included in the IPCC estimate. A complex mixture of primary and secondary aerosol components have thus been implemented in the Oslo CTM2 during BACCI, and over the main industrialized regions the secondary components (sulphate, nitrate and SOA) dominate over the primary components (BC, primary OC, sea salt and mineral dust). This is in accordance with measurements in some of the most polluted European regions such as the Po valley (Crosier et al., 2007).

Input to the IPCC process has been provided through the Aerocom initiative: Oslo CTM2 participated as one of nine global aerosol models that contributed with calculations of composition changes of sulphate and black and organic carbon from fossil fuel and biomass burning and radiative forcing and is one of 16 models simulating the aerosol distribution for present con-

ditions (Kinne et al., 2006; Schulz et al., 2006; Textor et al., 2006; Textor et al., 2007). Comparisons with satellite derived aerosol optical depth show reasonable agreement in the global mean values between models and satellite data, but the global aerosol models have substantial differences with regard to aerosol types (Kinne et al., 2006). Differences between the global aerosol models are often more related to intermodel diversity (such as transport of aerosol and their precursors and their removal) than to emissions of aerosols and their precursors (Textor et al., 2006; Textor et al., 2007). Fig. 12 shows the annual mean geographical distribution of the radiative forcing due to the direct aerosol effect of anthropogenic aerosols (sulphate, BC and primary OC, and carbonaceous aerosols from biomass burning) for the Oslo CTM2, taken from (Schulz et al., 2006). The strongest radiative forcing is found in industrialized and biomass burning regions, with small regions containing a positive radiative forcing, the latter often associated with high surface reflectivity. The global mean radiative forcing from Oslo CTM2 was  $-0.34 \text{ W m}^{-2}$  and was stronger than the mean of the nine models in Aerocom ( $-0.22 \text{ W m}^{-2}$ ) with identical emissions of aerosols and their precursors (Schulz et al., 2006). In Oslo CTM2 the estimate of radiative forcing for sulphate, BC and primary OC was  $-0.35$ ,  $0.22$  and  $-0.16 \text{ W m}^{-2}$ , respectively. Comparisons with observations have given the possibility to evaluate model performances. An important outcome of the Aerocom intercomparison studies is that uncertainties in concentration and radiative forcing estimates from aerosols have been reduced.

The Oslo CTM2 has also been validated against remote sensed data of aerosol optical depth and radiative effect (Myhre et al., 2007a). Despite significant differences between various satellite

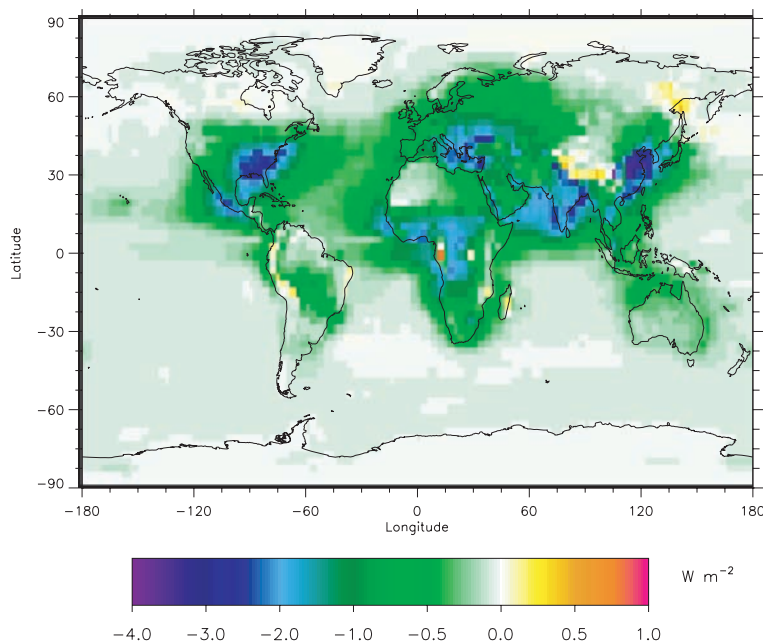


Fig. 12. Radiative forcing of the direct aerosol effect simulated with the Oslo CTM2. The anthropogenic aerosols include sulphate, BC, OC.

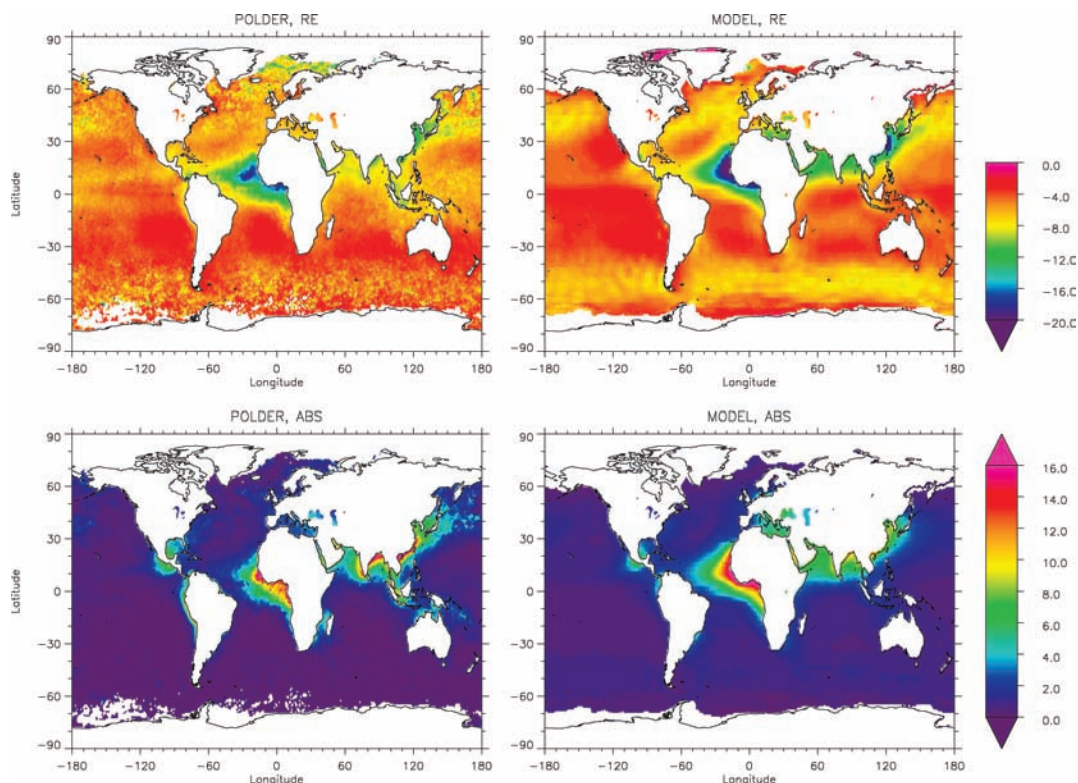


Fig. 13. Radiative effect of aerosols over ocean for clear sky condition derived from the POLDER satellite and from Oslo CTM2 (upper panels) and the atmospheric absorption due to aerosols (lower panels).

retrievals (Myhre et al., 2004; Myhre et al., 2005) the satellite aerosol retrievals have provided important knowledge about the aerosol distribution (Kaufman et al., 2002; Forster et al., 2007). Fig. 13 shows the clear sky radiative effect of aerosols (natural and anthropogenic) and the atmospheric absorption due to aerosols from the model as well as derived from the POLDER satellite (Bellouin et al., 2003; Myhre et al., 2007a). The satellite derived estimate is only available over ocean, and natural and anthropogenic aerosols can not be distinguished (Bellouin et al., 2003). POLDER and Oslo CTM2 show the strongest aerosol radiative effect occur in coastal regions, in particular to the west of Africa, associated with mineral dust and biomass burning particles. The atmospheric absorption due to aerosols is also dominant in coastal regions. This absorption is mainly associated with BC from fossil fuel, bio fuel and biomass burning; however, mineral dust also has some weak absorption and scattering aerosols increase the gas absorption in the atmosphere (Myhre et al., 2007a). The atmospheric absorption reaches almost  $20 \text{ W m}^{-2}$  and the pattern is quite similar in the POLDER derived estimate and the model, with an indication of somewhat weaker values close to South-East Asia in the model.

Surface solar radiation measurements have, over the last 50 yr, shown first a global dimming and later a brightening (Stanhill and Cohen, 2001; Liepert, 2002; Wild et al., 2005). Using the Oslo CTM2 it is shown that atmospheric aerosols (direct aerosol

effect as well as the cloud albedo effect) play a significant role in the global dimming, but that other factors as atmospheric gases and contrails also contribute (Kvalevåg and Myhre, 2007). The dimming is shown to be significant over many land areas; however, changes in the direct and diffuse surface solar radiation of even larger magnitude (Kvalevåg and Myhre, 2007).

Emissions of  $\text{SO}_2$  have been reduced substantially over Europe since 1980 (Vestreng et al., 2007). In a modelling study with the Oslo CTM2, there is a broad consistency between the observed reduction in sulphate and the modelled concentration (Berglen et al., 2007). The largest reduction has been observed over continental and Eastern Europe with slightly smaller reductions in the Oslo CTM2 than observed.

Finally, we have developed a new sectional aerosol model for large-scale applications, termed SALSA (Kokkola et al., 2007). The main purpose of the SALSA model is to find a compromise between computational demand and accurate description of processes. This is accomplished by optimally selecting size classes to represent the particle size distribution and by neglecting processes at sizes where they have negligible effects. When coupled interactively with the climate and other relevant components of the existing Earth system models, SALSA will offer an efficient tool to expand the studies made within the BACCI community up to a global scale, as well as into the future with changing biosphere–atmosphere interactions.

#### 4. Benefits of cooperative research during BACCI

There is no individual group, discipline, institute or country able to solve the complex and interlinked issues of climate change and air quality, alone. Gases and aerosol particles originating from natural and anthropogenic emissions are transported and transformed over geographically large areas. Therefore inter, multi and cross-disciplinary approaches are needed to address these issues. BACCI has had a unique possibility to study biosphere–aerosol–cloud–climate interactions since we have a real interdisciplinary team and efficient cooperation among the Nordic research groups. BACCI has also very intensive connections with the Finnish Centre of Excellence (Research Unit of Physics, Chemistry and Biology of Atmospheric Composition and Climate Change) (see Kulmala et al., 2005b).

The BACCI collaboration has brought a lot of new insight into climatic effects by aerosol particles and trace gases, into interactions between aerosol particles, trace gases and various chemical and biological processes, as well as into indirect feedbacks in the Earth system. The results can be applied in European environmental policy, such as in formulating national and EU-wide air pollution directives and in negotiating the climate policy for EU. The results are also valuable in interpreting the data obtained from studies of biosphere–atmosphere interactions, especially from those estimating the source and sink strengths of particulate and gaseous compounds. Long-term flux measurements, performed jointly with the global scientific community, facilitate the estimation of spatio-temporal changes in carbon sinks and hydrological cycles, leading to more accurate knowledge for multinational negotiations on mitigation of climate change.

The BACCI work contributes directly to research on climate change and on the constraints of biological and anthropogenic factors. The IPCC 2007 report called upon improved knowledge on the contributions and on the integrated effects of various anthropogenic and biogenic sources for aerosols, to be able to reduce the uncertainties in predictions on climate change. Global climate change is tightly connected to other global problems such as air pollution, and we need to recognize that all these problems should be studied simultaneously. Particularly, the BACCI collaboration has contributed to the debate on Arctic and boreal environmental changes. This kind of wide perspective also supports European governmental policy, for example, concerning the Vienna Convention on the Protection of the Ozone Layer and the Convention of Long-range transport of Air Pollutants. The results are relevant to assessments by IPCC, to verification of the Kyoto protocol and to measures for controlling and mitigating emissions of gaseous and particulate pollutants.

BACCI has contributed to technological developments by improving aerosol and ion measurement techniques (e.g. Mirme et al., 2007). This has been achieved by using prototypes of new equipment in field experiments, thus providing the relevant technological trials for product development. The prototype products

have a great commercial potential in the research community market and may have spin-offs for industrial applications.

BACCI has had a substantial impact on integrated Land Ecosystem Atmosphere Processes Study (iLEAPS), the project office of which is located in Helsinki mainly because of the BACCI activities. iLEAPS is the land–atmosphere project within the framework of the second phase of IGBP. It aims to improved understanding of processes, linkages and feedbacks and changes due to human activities in the land–atmosphere interface affecting the Earth-system. The research ranges from cell level to global scale, diurnal to centennial and past to future. It encourages international and cross- and multidisciplinary collaboration. The main themes are land–atmosphere exchange, feedbacks between land biota, aerosols and atmospheric composition, teleconnections in the land surface–atmosphere–water–system and transfer of material and energy in the soil–canopy–boundary-layer system. Both measurements and modelling of interaction processes are applied. BACCI also forms the core of the European Integrated project on Aerosol Cloud Climate and Air Quality interactions (EUCAARI; 2006–2010, coordinator M. Kulmala), which works towards integrating these diverse communities on the pan-European scale and also on the global scale.

Finally, BACCI has established a dedicated educational programme that includes both master and doctoral level studies. BACCI is a founding partner in the Nordic Graduate School biosphere–carbon–aerosol–cloud–climate interactions (CBACCI). Over 20 interdisciplinary intensive courses—field courses and summer and winter schools—have been organized in the context of CBACCI. These courses have gathered students from all Nordic countries and the neighbouring area, including Baltic countries and North-West Russia to work together in a multidisciplinary scientific context. Furthermore, several e-learning courses have been given to CBACCI students. A workshop for teachers has been an annual activity of CBACCI, giving teachers training on teaching, communication and supervising skills, including training on specific skills such as web-based teaching. CBACCI has proved to be very successful in transfer of best practices, knowledge transfer both horizontally and vertically as well as training in general skills—writing of articles and proposals, presentation skills and commercialization of scientific ideas. CBACCI has also enhanced the implementation of the second and third cycles of the Bologna process in Nordic countries. BACCI has been the foundation of a new joint activity of six Nordic universities, the International Master's Degree Programme Atmosphere–Biosphere Studies (ABS), which was launched in 2006.

#### 5. Conclusions and future views

During the BACCI period, the main progress was related to observations and theoretical work related to atmospheric nucleation, particularly on the existence and dynamics of ion (see

e.g. Kulmala and Tammet, 2007) and neutral clusters (Kulmala et al., 2007a). We also focused on investigations on the chemical composition of nucleation mode (Kulmala et al., 2007b) and on the chemical precursors of fresh aerosol particles. One important, although still somewhat open issue is the contribution of biogenic aerosol particles on the global aerosol load. We have also intensively studied the aerosol dynamics; namely transport, transformation and deposition of aerosol particles and their related thermodynamics, as well as microphysics and chemistry of cloud droplet formation.

The formation of new particles and their subsequent growth appears to occur almost everywhere (Kulmala et al. 2004a). The new particles can, depending on their location, increase the concentration of cloud condensation nuclei by a factor more than two over the course of 1 d. We can therefore conclude that atmospheric new-particle production is an important process that must be understood well enough to be described in global climate models. Future work should include continuous observations of aerosol particle size distributions in diverse locations, measurements of gaseous compounds participating in nucleation and growth and determination of the chemical composition and other properties of nucleated particles. Measurements concerning the distribution and composition of nucleating clusters will (Kulmala et al., 2007a, b) significantly improve our understanding of the nucleation process itself. Future work should also include measurements of ion mobility distributions, ion composition and detection of neutral clusters.

From the perspective of global biosphere–atmosphere interactions research, future works should aim at constructing comprehensive stations similar to SMEAR II, investigating the soil–land ecosystem–atmosphere continuum (see also Hari and Kulmala 2005). The importance of inter, multi and cross-disciplinary in environmental and atmospheric research has been fully proved in the work of our Centre of Excellence.

Understanding the spatial distribution of atmospheric pollutants and quantifying their climatic and air quality effects requires mathematical models covering regional and global scales. Comparisons of large-scale model simulations with existing field measurements enhance the possibility to extrapolate results from in situ measurements to regional and global scales. On the other hand, the computational intensiveness of large-scale modelling makes it imperative to develop reliable and efficient parameterizations for aerosol size and composition distributions, particle formation and growth processes and emissions of gaseous pollutants as well as model sub-grid-scale processes. Likewise, for the carbon and water cycle, a better knowledge on dependencies of photosynthesis and respiration processes and evapotranspiration on environmental factors and climate are required for large-scale models.

Our joint efforts within BACCI have shown that working in inter and multidisciplinary way, from molecular to global level, is very efficient and is able to give solutions to scientific questions as well as show new directions to European and even global

science policy in atmospheric sciences. In the future these efforts should continue in coherent manner to be able to give answers related to global climate issues and their interaction to air quality.

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