

BACCI: Research Unit on Biosphere - Aerosol - Cloud - Climate Interactions

1. Background

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. Stieb *et al.*, 2002). In regional and global scales, aerosol particles and trace gases have a potential to change climate patterns and hydrological cycle (Lohmann and Feichter, 2005; IPCC, 2007). Aerosol particles also influence the radiation intensity distribution that reaches the earth surface having a direct influence on the terrestrial carbon sink (Gu *et al.* 2003). Better understanding of the various effects in the atmosphere requires detailed information on how different sources (including those of 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 as an example is atmospheric aerosol formation, which involves the production of nanometer-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 and 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 one another.

We have recently proposed a mechanism that couples the effect of CO₂ and aerosol particles on climate (Kulmala *et al.* 2004b, Figure 1). 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 (GPP) drives plant gross primary production (GPP), the difference between net ecosystem exchange of CO₂ (NEE) and total ecosystem respiration (TER). Photosynthetic efficiency is correlated with the light availability, and in the boreal zone it is inhibited in winter (Hari and Mäkelä, 2003). Actively growing forest ecosystems are usually sinks for CO₂, and a direct negative feedback (the higher the CO₂ concentration, the higher the rate of photosynthesis) exists between increasing atmospheric CO₂ concentrations and photosynthesis, whereas a positive feedback lies between ecosystem respiration and temperature. With higher temperatures water availability becomes 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), which are synthesized in processes closely linked with photosynthesis (the biological functions of BVOC emissions are still under debate, and may vary with times and between plant species). Newly formed particles in forested areas have been found to contain large amounts of organic material originating from BVOC emissions (O'Dowd *et al.*, 2002a; Tunved *et al.*, 2006).

The ratio of BVOC emissions to carbon assimilation is generally a few percent (Grace and Rayment, 2000), but under stress it can be significantly higher. If the increased CO₂ concentrations enhances photosynthesis, then the biosynthesis and emissions of several BVOCs may increase and possibly modify the aerosol particle formation routes.

The mechanism described above, linking photosynthesis and aerosol forcing in a novel way, is only one among many connections and feedbacks. The climatic and other effects of aerosols and trace gases are coupled together both via human actions, such as emission policy and land use change, and via various natural feedback mechanisms involving the biosphere and atmosphere. 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 (e.g. plant-emitted methane; Kepler et al., 2006) may appear in the future. We have recognized that the socio-economic challenge to BACCI is to react quickly to new research needs and findings from the scientific community, by assimilating the new knowledge and by testing and validating its significance from the point of view of our own aims and objectives.

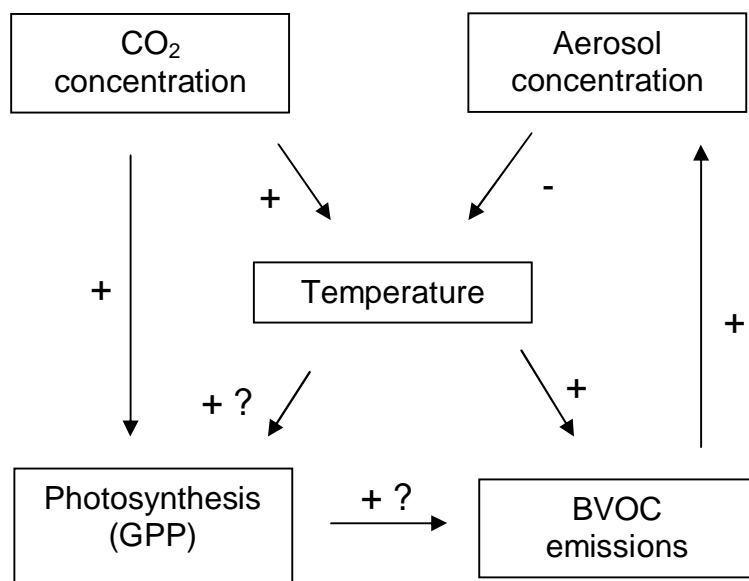


Figure 1. Schematic figure of the coupling between atmospheric CO₂ concentration, assimilation of carbon by photosynthesis (ecosystem gross primary production GPP), emissions of biogenic volatile organic compounds (BVOCs), aerosol particle concentrations, and atmospheric temperature (Kulmala et al., 2004b). Elevated CO₂ concentration will increase temperature (+) and photosynthesis (+). Increased temperature will enhance BVOC emissions (+) and probably also photosynthesis (+?). Increased photosynthesis may amplify BVOC emissions (+?). Increased BVOC emissions will enhance aerosol formation and growth, and therefore also the CCN concentrations (+). Enhanced aerosol and CCN concentrations will decrease temperature (-) due to increased reflection of sunlight from low clouds back to space (note that they also amplify diffuse radiation, which has a positive influence on photosynthesis (Gu et al., 2003)).

Apart from the formation routes for atmospheric particles, it is also important to follow their subsequent growth and fate. The newly formed aerosol particles created in atmospheric nucleation events 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 to the particle size, since the size determines whether the particles are able to penetrate to the lungs and further to the blood circulation. We have studied 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, and shown that nucleation can produce a significant fraction of cloud condensation nuclei regardless of the regional emission strength of primary particulate matter.

Several chemical effects on cloud drop formation that may have influence on the global radiation budget of the Earth were identified by Charlson et al (2001) and Nenes et al. (2002). These effects include the surface tension lowering of water droplets due to various organic aerosol species, and the 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 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., JGR 2005) and parameterized the effect in such a way that it can now be explored using climate models (Romakkaniemi et al., ACP 2005).

2. Objectives

The main scientific objective for BACCI is to study the life cycle of aerosol particles and their importance on climate change and on human health. Particularly, we have focused on a) the effect of biogenic aerosols on global aerosol load; b) aerosol-cloud-climate interaction and c) relationships between atmosphere and different ecosystems (ocean and boreal forest).

The measurable objectives were established to promote close co-operation, joint projects and scientific carriers of young scientists.

As a result of the measurable objectives we have (until 30.08.2007):

- a) published over 660 research articles in highly ranked, scientific journals
- b) educated 44 doctors (PhD or equivalent) and ca 60 MSc's in the field of atmosphere-biosphere science
- c) established several new, European level collaborative projects like EUCAARI, EUSAAR and iLEAPS

3. The consortium

In order to be able to meet our objectives, and to answer our research questions we aim at inter-, multi and cross-disciplinary research with a high level of technological and scientific innovation. 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 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. Within BACCI we have overcome these barriers and formed a multinational group of scientists from various field of science.

3.1. The Research Unit (Senior Scientists)

Finland: Prof. Markku Kulmala, Prof. Pertti Hari, Prof. Ari Laaksonen, Prof. Yrjö Viisanen, Prof. Marja-Liisa Riekkola, Prof. Kaarle Hämeri, Prof. Eero Nikinmaa, Prof. Kari Lehtinen, Doc. Hanna Vehkamäki, Doc. Üllar Rannik, Doc. Kari Hartonen, Doc. Tuulia Hyötyläinen, Prof. Veli-Matti Kerminen, Prof. Risto Hillamo, Dr. Tuomo Pakkanen, Dr. Jorma Joutsensaari, Doc. Jaana Bäck, Doc. Hannele Hakola, Doc. Janne Rinne, Doc. Ismo Napari
Sweden: Prof. Hans-Christen Hansson, Doc. Johan Ström, Doc. Robert Jansson, Doc. Christer Johansson, Prof. Henning Rodhe, Doc. Douglas Nilsson, Prof. Kevin Noone, Prof. Bengt Martinsson, Prof. Erik Swietlicki, Dr. Peter Tunved, Dr. Radovan Krejci, Dr. Birgitta Svenningsson
Denmark: Prof. Søren Larsen, Prof. Sara Pryor, Prof. Merete Bilde, Dr. Jakob Man, Dr. Niels Morten Nielsen, Dr. Lise Lotte Sørensen
Norway: Prof. Øystein Hov, Prof. Frode Stordal, Prof. Ivar Isaksen, Dr. Kjetil Tørseth, Dr. Gunnar Myhre, Dr. Mihalis Lazaridis, Dr. Josef Pacyna, Dr. Christopher Hoyle

3.2. Sites of the research

Finland: University of Helsinki (UHel), Department of Physical Sciences
University of Helsinki (UHel), Department of Forest Ecology
University of Helsinki (UHel), Department of Chemistry
University of Kuopio (UKu), Department of Applied Physics
Finnish Meteorological Institute (FMI)
Sweden: University of Stockholm, Institute of Applied Environmental Research (ITM)
University of Stockholm, Department of Meteorology
University of Lund, Department of Nuclear Physics
Denmark: Risø National Laboratory
University of Copenhagen. Department of Chemistry
Norway: Norwegian Institute of Air Research (NILU)
University of Oslo, Department of Geophysics

3.3. Key research questions

Our research consortium has addressed the following research questions related to our objectives:

1. How often and in how large areas does new particle formation occur?
2. What happens to clusters below 3 nm?
3. Are there neutral clusters present?
4. What is the chemical composition of nucleation mode aerosol particles?
5. What are their thermodynamical properties?
6. What are the chemical precursors for fresh aerosol particles?

7. How much do biogenic aerosols contribute to the tropospheric aerosol load?

To answer these questions we need both **experimental** (laboratory and field experiments, including development of novel instrumental techniques) and **theoretical** (basic theories, simulations, model development) methodologies. The questions are extending from molecular to global scale, using multidisciplinary methods and approaches.

3.4. Research Methods

The BACCI scientific approach starts from basic nucleation theories, followed by models of detailed aerosol dynamic/atmospheric chemistry and vegetation–atmosphere exchange. Crucial topics are well-defined laboratory experiments with versatile continuous field measurements in our research stations. Eventually these methodologies and their results can be extended to global-scale modelling. Our approach thus covers both experimental (laboratory and field experiments) and theoretical (basic theories, simulations, model development, parameterizations) aspects. The main disciplines used cover aerosol and environmental physics, environmental technology, atmospheric chemistry and physics, analytical chemistry, micrometeorology, forest ecology, plant physiology and ecophysiology. The effective interaction and use of knowledge on different areas under the common framework creates the added value and synergy benefits of co-operating research teams.

The approach and the scientific achievements of BACCI can be described using a research chain 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 have focused on those aspects of the research chain where the uncertainties are largest. In practice, we have started from molecular simulations (Monte Carlo and Molecular Dynamics) to understand nucleation and aerosol thermodynamic processes. These microscopic processes of nucleation together with condensation/evaporation and coagulation were required to understand aerosol dynamics, particle concentrations and composition. Significant advances in laboratory data and modelling techniques were also needed for a number of important aerosol systems. Similarly, we have modeled photosynthesis, autotrophic respiration and BVOC synthesis at cellular, stomatal and leaf (or shoot for conifers) scales, and tested the models with chamber measurements in laboratory and field.

Several fundamental aerosol and carbon cycle processes were recognized to be necessary to understand, in order to be able 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. At larger scales, advances in our understanding of boundary layer meteorology were needed to understand atmospheric aerosol transport, trace gas (e.g. CO₂, methane, N₂O, O₃, SO₂, NO_x, VOCs) and water vapor exchange and deposition processes. Boundary layer studies form a link to regional-scale processes and further to global-scale phenomena. In order 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.

Figure 2 shows the general philosophy for our All-Scales-Modelling and the approach from nanoclusters to integrated Earth System Models (EMS).

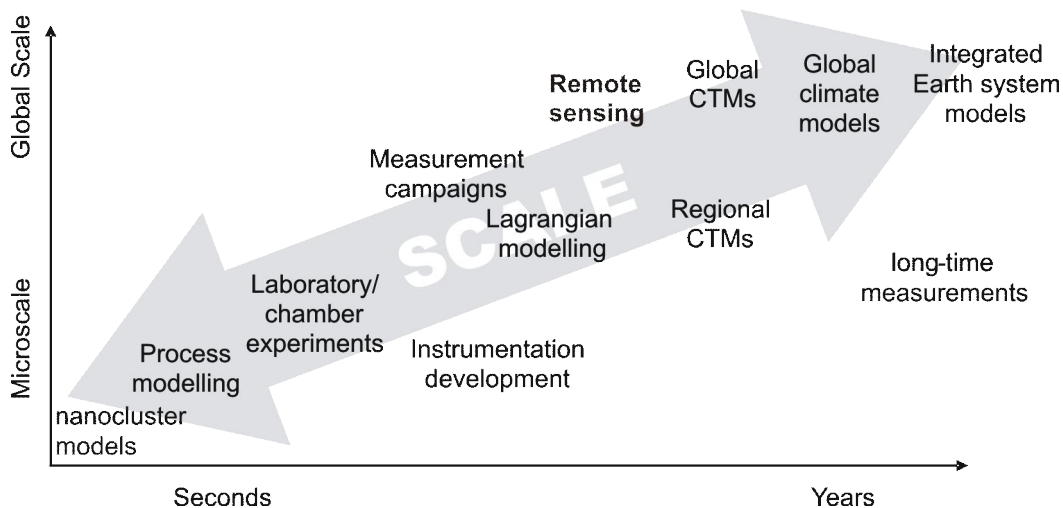


Figure 2. Model and Data Integration Philosophy over spatio-temporal scales. CTM is Chemical Transport Model

In practice the work in BACCI was divided into 10 work packages (WP's), all of which have been operated in a well-defined manner to meet the primary objectives. Members of several participants have been involved in all work packages and have collaborated to reach the overall goals. A schematic figure showing the WPs and their connections is given in Figure 3.

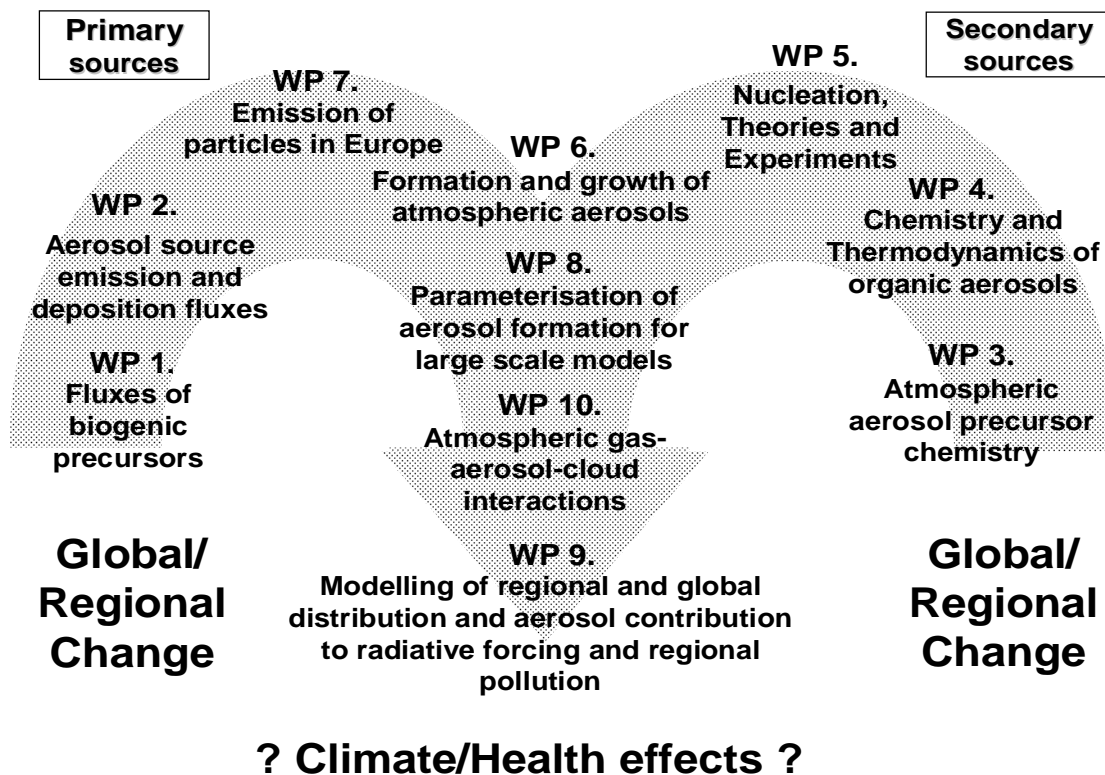


Figure 3. BACCI work packages and their connections.

3.5. Long-term field measurements

Long-term field measurements are a core activity in BACCI, which to the same extent have not been performed anywhere in the world. The continuous measurements are accompanied by 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). The aerosol number density, particle number size distribution, trace gas concentrations and basic meteorological measurements form a core of the observations. During the intensive periods, very detailed measurements of trace gases, aerosol chemistry and microphysic and various boundary layer parameters have been conducted. Separate field campaigns in different locations all over the world have been completed, either as BACCI co-operative basis or as part of other international projects.

The processes and properties of vegetation and atmosphere are strongly coupled with each other. Therefore performing simultaneous measurements of several phenomena in a forest ecosystem is essential for an in-depth analysis of the connections between the processes and components of the system. A good example is the multiple couplings between atmospheric processes and vegetation (Figure 1): Carbon fluxes primarily connect the atmosphere, trees and forest soil with each other. However, also rainfall, transpiration, evaporation and run off are important components in the atmosphere-ecosystem coupling. Finally, the volatile carbon compounds emitted by trees or other forest ecosystem components are evidently very important for the formation and growth of aerosol particles. This emphasizes the importance of running simultaneous, parallel measurements of many interlinked components of the ecosystem-atmosphere continuum.

The rapid development of measuring techniques has enabled versatile field measurements. New trace compounds can now be measured in field conditions, the measuring accuracy and precision is increasing and the required time resolution is decreasing. For example, more than ten gases can be simultaneously monitored in order of minutes with the present instrumentation using dynamic plant chamber technique, together with the novel proton-transfer mass spectrometry (PTR-MS). In addition, large measuring systems can be automated with digital technique and sophisticated software. The BACCI groups have pioneered in developing flexible and comprehensive measurement systems that can be operated throughout the seasons under harsh field conditions.

The goal of intensive long-term observations, similar to the SMEAR II type research is an improved understanding on how physical, chemical, and biological processes interact with each other, and how the energy and matter are transported and transformed through the land-atmosphere interface. Particularly emphasis is on interactions and feedbacks at all scales; from past to future and from local to global scale. The goals are very much similar than the scientific aims of the international iLEAPS Programme (Integrated Land Ecosystem Atmospheric Processes Study) within the framework of the second phase of IGBP, the International Geosphere – Biosphere Programme, and both programmes, BACCI and iLEAPS, work coherently and in synergy.

3.6. Joint field campaigns in BACCI

Besides continuous measurements we have had several joint field activities during 2003-7:

- 1) QUEST campaigns in Hyytiälä in spring 2003 and spring 2005, and in San Pietro Capofiume (PoValley) in spring 2004. During these campaigns we explored the effectiveness of multiple instrumentations set up, in order to be able to understand atmospheric aerosol formation. We have simultaneously measured sulphuric acid, ammonia, terpenes, cluster ions, aerosols, meteorology, photosynthetic activity etc.
- 2) The Pallas Cloud Experiments (PACE) in the Pallas station during October-November, 2004 and September-October, 2005. The measured quantities included “in-cloud” and “out-of-cloud” particle number size distributions, CCN spectrum, cloud droplet number concentration and size distribution, PM_{2.5} and size-segregated inorganic composition of the aerosol. In October 2006, a cloud experiment was conducted at the Puijo tower in Kuopio, employing aerosol and cloud droplet measuring instrumentation
- 3) Intensive field measurements of biogenic and anthropogenic VOC concentrations, their oxidation product concentrations, photochemically relevant species and meteorology in both a clean forest environment as well as anthropogenically polluted environments
- 4) Activities on joint flux and micrometeorological investigations throughout the years at several field sites.
- 5) Air-sea fluxes of aerosols in the marine atmosphere during the Galathea3 global cruise track (August 18th 2006 to April 25th 2007). We focused on evaluation of the particle fluxes, composition and size distribution of particle in the marine atmospheric boundary layer as well as on some of the nucleation processes.
- 6) Continuous aerosol and ozone measurements at the Akrotiri station at Chania, Crete, Greece. We focused on the influence of African dust outbreaks, on the particulate matter ambient concentration and their chemical composition. Number size distribution data and nucleation outbreaks are under study. Continuous PM₁₀ and PM_{2.5} and ozone measurements were performed in May 2003 - June 2007, specific nucleation measurements during December 2006, February 2007 - May 2007, and measurements are running also during the whole 2007 period for nucleation events.
- 7) Measurement campaigns in July 2006-February 2007 in a tall open eucalypt forest in Southeastern Australia provided new insights on biogenic aerosol formation mechanisms in semi-arid climate, where daytime new aerosol formation took place in more than 50% of days. Also nocturnal aerosol formation events were frequently observed in summer and autumn.
- 8) Campaigns at Mace Head, South Africa, India and China. In these campaigns we have found out aerosol formation at different sites, both urban and rural and thus we have shown that new particle formation is a truly worldwide phenomenon following similar pattern all over the globe.
- 9) During the 2004 and 2007 intensive field campaigns in the Arctic we studied Arctic haze phenomenon and transition from polluted spring period towards the clean summer using combined effort of ground-based, airborne and satellite observations. Understanding of this change propagating through the whole tropospheric column is a significant contribution to correct assessment of the future climate evolution in the Arctic.
- 10) Intensive ground-based and airborne aerosol observations in Antarctica during austral summer 2006-2007 delivered unique data set from one of the possibly last pristine environments on the Earth.
- 11) EUCAARI spring 2007 campaign. In this campaign we have proved the earlier predicted existence of neutral clusters in a forest atmosphere.

4. The quality and novelty of the results

The main focus of BACCI has been in the following topics (for all topics we have published several papers either in *Nature* or *Science*):

- 1) **formation and growth mechanisms of atmospheric aerosols, aerosol dynamics:** (Kulmala et al., *Nature*, 2000; Kulmala, *Science*, 2003; Berndt et al., *Science*, 2005; Tunved et al., *Science*, 2006; Kulmala et al., 2007),
- 2) **the effect of secondary biogenic aerosols on global aerosol load:** (O'Dowd et al., *Nature*, 2002a; O'Dowd et al., *Nature*, 2002b; O'Dowd et al., *Nature*, 2005),
- 3) **aerosol-cloud-climate interaction:** (Kulmala et al., *Nature*, 1997; Charlson et al., *Science*, 2001), and
- 4) **the relationships between the atmosphere and different ecosystems, particularly boreal forest:** (Valentini et al., *Nature*, 2000; Hari et al., *Nature*, 2003; Ciais et al., *Nature*, 2005; Magnani et al., *Nature*, 2007).

One of the most significant recent results obtained is the observation that the formation of new particles and their subsequent growth seem to be ubiquitous (Kulmala et al. 2004a). Such observations have been made in very different lower-tropospheric environments, including Antarctica and surrounding oceans (Koponen et al., 2003), boreal forests in Northern Europe (Mäkelä et al., 1997; Held et al., 2004; Vehkamäki et al., 2004; Dal Maso et al., 2005), other remote and rural continental areas (Weber et al., 1997; Birmili et al., 2003), coastal environments around Europe and United States (O'Dowd et al., 1999, 2002b; Wen et al., 2006), various urban centers (Shi, 2003; Jeong et al., 2004; Stanier et al., 2004; Stolzenburg et al., 2005), and even heavily-polluted regions and megacities (Dunn et al., 2004; Wehner et al., 2004; Mönkkönen et al., 2005; Laaksonen et al., 2005). Aerosol formation has also been observed in the free troposphere, being a frequent phenomenon in the upper troposphere and in cloud-outflow regions (Twohy et al., 2002; Singh et al., 2002, Kulmala et al., 2006).

While biogenic aerosol formation mechanisms are known for the most part because of the research carried out during recent years, there is an urge for applying them in the global scale in the future. This calls for wide understanding and ability to observe how the biogenic formation mechanisms are linked to underlying ecosystem processes. The BACCI community has made an integrated attempt to understand various, but interlinked, biosphere-atmosphere interactions by applying inter- and multidisciplinary approaches in a coherent manner. Due to the emergence of new measurement techniques for detection of neutral and ion clusters and the multitude of organic compounds, together with improved chemical analysis methods of nm-scale particles, and the joint usage of these techniques, we have recently been able to show the linkages between the biogenic precursor formation and occurrence of aerosol formation events in boreal areas (Tunved et al. 2006). Further, we have collected exceptionally long data series on biosphere-atmosphere interactions, especially for carbon dioxide, methane and water vapour over different ecosystems (e.g. Dal Maso et al., 2005; 2007). Currently we are in the stage where the amount and representativeness of data can for the first time be used for developing realistic parameterizations for large-scale, global models.

The BACCI collaboration has brought a lot of new information on the global climatic effects of aerosol particles and trace gases and their interactions with chemical and biological processes, as well as on the indirect feedbacks. 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.

5. BACCI contribution to addressing key scientific and technological issues

During the BACCI project we were able to find answers to our Key Research questions:

1) How often and in how large areas does new particle formation occur?

- It seems to occur in large areas, and to be an important phenomenon (e.g. Kulmala et al., 2004a). We have measured more than 1000 new particle nucleation events at SMEAR II station since 1996. In boreal forests, the event numbers appear to have a maximum between March and May.

2) What happens to ions below 3 nm?

- Ion spectrometers can show some part of the evolution and dynamics of small ion clusters. Therefore we have developed several ion spectrometers (see section 2).

3) Are there neutral clusters present?

- Ion clusters (both positive and negative) are always around, in practice everywhere.
- Model calculations show that there are also neutral clusters (Kulmala et al., 2000).
- There is increasing evidence on their existence in vivo (e.g. Kulmala et al., 2005a), and we have developed new instrumentation to detect neutral clusters in field conditions.

4) What is the composition of nucleation mode aerosol particles?

- Recently we have developed new in situ methods like CPC battery to study water solubility of those small particles (Kulmala et al., 2007)

5) What is the related thermodynamics?

- Although we know that saturation concentration of condensing vapours seems to be around $3 \times 10^6 \text{ cm}^{-3}$, the deep insight on thermodynamical properties of atmospheric clusters is still missing.

6) What are the chemical precursors of fresh aerosol particles?

- There is clear evidence that sulphuric acid will participate in the formation process (Kulmala et al., 2006, Sihto et al., 2006)
- The first ever laboratory study of 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).
- There is also clear evidence from the atmosphere that organic vapours are participating on new particle formation, or at least on their subsequent growth (Tunved et al., 2006, Allan et al., 2006, Laaksonen et al., 2007).

7) How much do biogenic aerosols contribute to the tropospheric aerosol load?

- They seem to be significant, especially in the forested boreal areas (Spracklen et al., 2006, Tunved et al., 2006)

The BACCI work contributes directly to research on climate change and to resolving the contributions of both 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, in order to be able to reduce the uncertainties in predictions on climate change. We have recognized that climate change is very wide and global problem, tightly connected to other global problems such as air pollution, and that all these uncertainties 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 for assessments by IPCC, for verification of the Kyoto protocol and for measures in controlling and mitigating emissions of gaseous and particulate pollutants.

From a technological point of view, new aerosol, cloud droplet and trace gas instruments have been developed and the environmental information technology have been promoted.

There is no individual group, discipline, institute, or country able to solve the complex and interlinked issues on 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.

Internationally, the BACCI research groups are in a leading position in the research area of formation of atmospheric aerosols. Our approaches involve e.g. basic nucleation theories, detailed aerosol dynamic and atmospheric chemistry models, well defined laboratory experiments, wide continuous field measurements and 3D modelling in combination with satellite data. We also have a leading position in the research area of micrometeorological fluxes of aerosol particles and their precursors. Part of the BACCI research teams have been evaluated in the recent national Center of Excellence calls e.g. in Finland with great success (funded among the 15 candidates out of 113 including all disciplines).

BACCI has a unique possibility to study biosphere – aerosol – cloud –climate interactions, since we have a truly interdisciplinary team and efficient cooperation between the Nordic research groups. BACCI has organized 25 international scientific workshops and conferences all over North Europe, including Estonia, Lithuania and North-West Russia between 2002-2007. The working links with other units and teams are an essential part of the implementation of the research program; this has been accomplished in the form of research visits, common field campaigns and training of the students at all levels.

BACCI has established a dedicated educational programme, including both masters and doctoral level studies, and also education for post-docs and teachers in universities, including e.g. web-based teaching. BACCI is a founding partner in a Nordic Graduate School: CBACCI (Biosphere-Carbon-Aerosol-Cloud-Climate Interactions, <http://www.atm.helsinki.fi/index.php?action=getfile&data=CBACCI&file=index.html#>), which consists of 19 universities (25 research groups) in 10 European countries, and has organized 16 international summer schools and workshops in 2005-2007. CBACCI offers courses in atmospheric sciences, biogeochemical cycles, and forestry, and collaborates closely with the International Master of Science Programme in Atmosphere-Biosphere Studies (ABS, <http://www.atm.helsinki.fi/ABS/>), operating in 6 universities in Finland, Sweden and Denmark. In addition, a Finnish graduate school 'Physics, chemistry, biology and meteorology of atmospheric composition and climate change' is participating the BACCI activities.

The BACCI scientists have hosted or performed over 170 research visits longer than two weeks between 2003-2007, involving institutions in France, Germany, Ireland, South Africa, Italy, Switzerland, US, UK, Greece, China, Australia, Russia, Antarctica, India, Argentina and Chile. The BACCI partners have direct working connections to more than 60 international laboratories. We have participated in more than 35 International projects (5 as coordinator), and are closely connected to the IPCC and IGAC programmes. We are participating e.g. in global Fluxnet (flux tower network) and GAW (Global Atmospheric Watch) programs and international field campaigns to investigate atmospheric aerosols and tropospheric chemistry. We are also co-operating with several companies like Vaisala, Nokia and Dekati Ltds. Our research also belongs to all host organisations' main strategic areas.

BACCI forms the core in iLEAPS, whose project office is located in Helsinki and mainly based on BACCI activities. iLEAPS (integrated Land Ecosystem Atmosphere Processes Study, <http://www.atm.helsinki.fi/ILEAPS/>) is the land-atmosphere project within the framework of the second phase of IGBP (International Geosphere-Biosphere Programme). It aims to improve understanding of processes, linkages and feedbacks, and to clarifying the changes due to human activities in the land-atmosphere interface affecting the Earth System. The iLEAPS research ranges from cell level to global scale, from diurnal to centennial and from past to future. It strongly encourages international and cross- and multi-disciplinary collaboration. The main themes in iLEAPS are land-atmosphere exchange, feedbacks between land biota, aerosols and atmospheric composition, teleconnections in the land surface-atmosphere-water-system, transfer of material and energy in the soil/canopy/boundary-layer system. Both measurements and modelling of interaction processes are applied.

During the Nordic Centre of Excellence period we have been significantly collaborating with the European Network of Excellence (ACCENT). BACCI research has directly led to several EU-level research programmes, and thus boosted the European research in the field of atmospheric sciences into the world's frontier level. BACCI forms the core of European Union Integrated project EUCAARI (2006-2010, coordinator M. Kulmala), which works towards integrating these diverse communities on the pan-European scale and also on the global scale, by involving leading US scientists as members of the advisory board and Partners from four developing countries. EUCAARI therefore creates the critical mass required to adequately support European policy-making at the international level, relevant to climate change and environmental issues. Besides coordinating EUCAARI, BACCI also co-operates closely with the European infrastructure projects including the experimental facilities for laboratory studies (EUROCHAMP), ground sites (EUSAAR), airborne measurements (EUFAR) and the satellites. BACCI groups act as coordinators of EU-Marie-Curie MOD-OBS project on modelling and observation of the atmosphere.

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 models stresses the importance of developing 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 dependence of photosynthesis and respiration processes and evapo-transpiration on environmental factors and climate are required for large-scale models. The BACCI group has a code of a complex Earth System Model (COSMOS project, Max Planck Institute in Hamburg) and can modify and run the model by itself. All results of BACCI concerning new aerosol and biogeochemical parameterizations are readily available and used in COSMOS project.

BACCI has also contributed to technological developments by improving aerosol and ion measurement techniques. 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 great commercial potential in the research community market and potential spin-offs for industrial applications.

BACCI partners are also co-operating with EMEP. The EMEP operational model is the major tool used in Europe for air quality policy design. The EMEP model focuses on regional particulate matter (PM) levels and trans-boundary transport of pollutants. The model results and the available field measurements strongly suggest that regional trans-boundary contribution dominates the observed PM₁₀ concentrations in most areas (Accent Workshop 2006). An important focus in EMEP is to calculate the source-receptor relationships of air pollutants between the member countries of the Convention (Europe including Eastern Europe, Caucasian and Central Asian countries). The pollutants derive from emissions of SO₂, NO_x, VOC, NH₃, HM, POP and primary PM, and the emphasis is on ecosystem damage, human health impacts and intercontinental transport. The characterization of PM across Europe (mass, chemical composition, size distribution and eventually number densities) is a high priority. EMEP runs an extensive network of rural observation sites (coordinated by NILU).

6. Dissemination of the results

The scientists working in BACCI have published over 660 scientific papers in more than 100 different peer reviewed journals during the last five years.

6.1. 50 top publications by BACCI research groups

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