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Introduction: European Integrated project on Aerosol Cloud Climate and Air Quality interactions (EUCAARI) – integrating aerosol research from nano to global scales

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Abstract

The European Aerosol Cloud Climate and Air Quality Interactions project EUCAARI is an EU Research Framework 6 integrated project focusing on understanding the interactions of climate and air pollution. EUCAARI works in an integrative and multidisciplinary way from nano- to global scale. EUCAARI brings together several leading European research groups, state-of-the-art infrastructure and some key scientists from third countries to investigate the role of aerosol on climate and air quality. Altogether 48 partners from 25 countries are participating in EUCAARI. During the first 16 months EUCAARI has built operational systems e.g. established pan-European measurement network for Lagrangian studies and four stations in developing countries. Also an improved understanding of nanoscale processes (like nucleation) has been implemented in global models. Here we present the research methods, organisation, operations and first results of EUCAARI.

1 Background

In polluted urban environments atmospheric aerosol particles and trace gases 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 can change climate patterns and the hydrological cycle (Chung et al., 2005; Lohmann and Feichter, 2005; IPCC, 2007). Aerosol particles also influence the radiation intensity and distribution at the surface of the Earth and thus directly affect the terrestrial carbon sink (Gu et al., 2002). Quantifying the effect of aerosols on the planet's radiative balance is an urgent task to understand climate change. The uncertainty in aerosol radiative forcing is typically greater than 100%, and for some aerosol components, it is more than 200%, which makes the evaluation of climate sensitivity difficult. Furthermore, the regional scale forcing can be significantly greater than the global average value, as can the associated uncertainties. The contributions of the various aerosol sources, the

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role of long-range transport, and the contribution of primary and secondary particulate matter to the ambient aerosol concentrations over Europe are not well known. Furthermore, increasing primary emissions and unknown secondary aerosol formation over rapidly developing regions in Asia, Africa and South-America adds a great uncertainty to the global aerosol forcing.

To understand better the various effects in the atmosphere, detailed information is required on how different sources and transformation processes modify the properties of aerosol particles and trace gases. Trace gases and atmospheric particles are tightly connected with each other via physical, chemical, meteorological and biological processes occurring in the atmosphere and at the atmosphere-biosphere and atmosphere-ocean interfaces. For example, atmospheric aerosol formation, which involves the production of nanometre-size particles by nucleation and their growth to detectable sizes (Kulmala, 2003) is an important phenomenon. Emission changes, forest management and land use change, and various feedback mechanisms between the biosphere and the atmosphere change 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 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. One of the important issues to resolve is how the different components affecting radiative forcing interact with one another. The reduction of some uncertainties related to aerosol radiative forcing since the IPCC (2001) report is a promising development, while e.g. black carbon may turn out to be more important in a climate context than hitherto thought (Ramanathan and Carmichael, 2008).

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In this paper we present the objectives of the European Union funded project EUCAARI, describe the research methods and provide examples of results obtained during the first 16 months of EUCAARI. In this paper we will describe how this large integrated project is organized and how it operates to meet its scientific objectives. Practically, besides foreseen and totally new results EUCAARI will built its integration also on existing knowledge like big international field campaign e.g. ACE-1, ACE-2 and ACE-Asia (see Griffiths et al., 1999; Raes et al., 2000; Huebert et al., 2003).

2 Mission and objectives

The EUCAARI mission: The EUCAARI project brings together several key European research groups in the fields of atmospheric science and global change to investigate the effects of aerosol particles on climate and air quality. Quantifying the effect of aerosols on the planet's radiative balance is one of the most urgent tasks in our efforts to understand future climate change. The other tasks are e.g. the role of biosphere as sinks and sources in carbon and nitrogen cycles. The very large uncertainty in aerosol radiative forcing over the industrial period makes the evaluation of climate sensitivity difficult (Andreae et al., 2005). As a whole, the contributions of the various aerosol sources, the role of long-range transport, and the contribution of primary and secondary particulate matter to the ambient aerosol concentrations over Europe are still not well known. Particularly uncertain are parameters influencing the radiative balance and properties of clouds, such as the number size distribution, chemical composition and particle mixing state. Furthermore, the interactions between air quality and climate are largely unknown, although some links has been identified (e.g. Swart, 2004) or even quantified (Dentener et al., 2005)

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The objectives of the EUCAARI project are the following (see Fig. 1):

Objective 1: Reduction of the current uncertainty of the impact of aerosol particles on climate by 50% and quantification of the relationship between anthropogenic aerosol particles and regional air quality. To achieve this objective EUCAARI will concentrate on the areas of greatest uncertainty and will:

1. Identify and quantify the processes and sources governing global and regional aerosol concentrations
2. Quantify the physico-chemical properties of atmospheric aerosols
3. Quantify the feedback processes that link climate change and atmospheric aerosol concentrations with emphasis on the production and loading of natural aerosols and their precursors

Objective 2: Quantification of the side effects of European air quality directives on global and regional climate, and provide tools for future quantifications for different stakeholders.

EUCAARI also contributes to technological developments by improving aerosol and ion measurement techniques. This is 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 may have spin-offs for industrial applications. EUCAARI will also produce advanced aerosol and cloud models that can be used outside the project by global and regional climate and air quality modelling communities.

EUCAARI Objective 1 will be achieved by improving our understanding of atmospheric aerosol physics and chemistry. This improvement will enhance our ability to forecast short-term regional air quality as well as to estimate the long-term aerosol effects on current and future global climate. Objective 2 will facilitate the linking of the air quality and global climate change problems and will provide the necessary tools for their quantification for use by different stakeholders.

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3 Research methods

The EUCAARI objectives are relevant to end-user communities ranging from private companies to policy-makers and including local, national, and international authorities. The objectives are very ambitious, but achievable by focusing on the key scientific problems in the aerosol-cloud-climate-air quality field. During project planning the consortium identified 12 key scientific problems from nanometre scale processes to the overall aerosol-cloud effect on climate. These were

1. In-situ formation (nucleation) of aerosols.
2. Number and mass emissions of primary aerosol from natural and anthropogenic sources at urban, regional, and global scales.
3. Formation of secondary organic aerosol and the partitioning of semi-volatile compounds between the gas and aerosol phases.
4. Ageing of aerosols and evolution of their properties during their atmospheric lifetime.
5. Attribution of the different aerosol mass components in Europe to specific sources.
6. Current and future contributions of natural versus anthropogenic, and primary versus secondary sources to particle number concentrations.
7. Long-range transport of aerosol particles and their precursors from and to Europe as well as their transport within Europe.
8. Seasonal behaviour of aerosol particles in four developing countries.
9. Air quality and local climate interactions inside and outside Europe.
10. The impact of aerosols and trace gases on cloud droplet activation, cloud lifetime, and extent (the aerosol indirect effects).

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11. Interactions between the aerosol cycle, the water cycle, and the biosphere.

12. Climatic feedbacks related to anthropogenic/biosphere-aerosol-cloud-climate interactions.

Both experimental and theoretical methods are used. Laboratory and field experiments, including development of novel instrumental techniques, provide new information about aerosol and cloud properties and processes, while developments in basic theories, simulations, and models give us a way to integrate and compare the results in a broader context. EUCAARI has also established long-term ground-based aerosol measurements in economically growing countries in Asia, Africa, and South-America to plug significant gaps in our global observation capability.

As shown in Fig. 2 EUCAARI contains a research chain beginning at the molecular scale and extending to the regional and global scale. The scientific approach starts from basic theories of nucleation and chemical processes followed by models of detailed aerosol dynamic/atmospheric chemistry and vegetation-atmosphere exchange, laboratory experiments with continuous field measurements at several research stations, and global-scale modelling. Understanding the highly non-linear processes related to earth system at different spatial and time scales will give insight to meet our objectives. However, also the connections to society via emission inventories and policy oriented models are needed. The main disciplines include aerosol and environmental physics and technology, atmospheric chemistry and physics, analytical chemistry, micrometeorology, climate modeling and forest ecology. This multidisciplinarity provides an opportunity to add value and gain synergy.

We focus on those topics in the research chain where the uncertainties are largest. At small scales, we use 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 are required to understand aerosol dynamics, particle concentrations and composition. Significant advances in laboratory data and modeling techniques are needed for a num-

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ber of important aerosol systems. Fundamental aerosol processes, especially those connected to biogeochemical cycles – like carbon cycle via formation and growth of organic aerosol particles (see e.g. Kulmala et al., 2004) – need to be understood in order to quantify the 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 are needed to understand atmospheric aerosol transport, trace gas (e.g. CO_2, methane, N_2O, O_3, SO_2, NO_x, VOCs) and water vapor exchange and deposition processes. Boundary layer studies form a link to regional-scale and global-scale processes. To simulate global climate and air quality, the most recent progress in this chain of processes must be compiled, integrated and implemented into climate change (CC) and air quality (AQ) numerical models.

The understanding of different processes and their inclusion in climate models is crucial. For example, if only the aerosol mass loading in the atmosphere is simulated based on emissions of precursor gases (like SO_2) and primary emissions, the number concentration of aerosol particles and further cloud condensation nuclei (CCN) concentration might be seriously under- or sometimes overestimated unless the size and number of particles is considered (see e.g. Spracklen et al., 2006, 2008).

EUCAARI is building on the available data from previous field campaigns and long-term measurements in order to establish global datasets. The consortium has started to collect the available data inside and outside Europe and use them for the benefit of the EUCAARI objectives and scientific questions.

The data integration within EUCAARI will involve a combination of data analysis (accuracy, consistency and representativeness), modelling and field experiments. The experimental and model data is collected in a web-based platform located at partner NILU in Norway. The EUCAARI observation system combines long-term and spatially extensive surface-based measurements both in Europe and developing countries (China, India, Brazil, South-Africa), including the European network of supersites for aerosol research (EUSAAR), with satellite retrievals of key parameters. EUCAARI will

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use west-east and north-south station-to-station networks together with Lagrangian and Eulerian airborne measurements and the Cabauw supersite to quantify the effects on regional aerosol properties of emissions, aerosol formation, transformation, transport and deposition. These measurements will include parameters relevant for climate change (the radiative fields in clear and cloudy skies, and their susceptibility to aerosol fields) and air quality (particulate matter (PM) mass, size-resolved aerosol chemical composition, ozone, and NO_x). In May 2008 the intensive campaign of airborne measurements was performed. In 2008–2009 a sub-network of supersites will provide higher resolution data and detailed chemical composition and will extend the observations into the free troposphere (see Table 1).

4 Organization

EUCAARI is organized around 4 science elements: (1) Emissions and Formation, (2) Transport and Transformation, (3) Climate and Air Quality Effects, and (5) Impacts and Integration. These elements concentrate on atmospheric and climate processes and are structured to maximize the integration of different methodologies and scales. These scientific elements, in which the major scientific advances will be made, are supported by element (4) – Infrastructure. Element 4 provides the necessary tools and services for creating the deliverables in elements 1–3 and 5. The 21 work packages are described in Appendix A and their interactions are given in Table 2 and Fig. 3.

5 Operations and first results

During the first 12 months EUCAARI produced ca 50 deliverables. The results range from laboratory experiments to establishment of global measurement network and model development. The reports of the deliverables are very different and include:

– creation of project infrastructure (e.g. management, communication, web-pages).

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The project is coordinated by the University of Helsinki, but a significant part of the project management on the element and WP level is done by specialists of relevant fields. The development of email-lists, web pages and other forms of communication have been crucial for a multidisciplinary project of this kind;

- assessment reports on the level of scientific understanding in different WP areas thus informing the whole EUCAARI consortium;
- detailed revision of plans for EUCAARI progress (especially for aircraft and Lagrangian experiments); During the first year extensive air mass (trajectory) analysis was performed to identify the major transport paths over Europe to select the ground based sites for the Lagrangian experiment (see Fig. 4);
- installation of new measurement equipment for field experiments, training of experimental scientists and installation of new stations outside of Europe. The weight of these activities was on the developing countries training and station-building;
- first benchmark results of climate modelling against which the project progress will be determined;
- first relevant scientific reports in many fields, especially on nucleation and organic aerosol studies.

EUCAARI partners have published peer-reviewed journal articles on their work in EUCAARI, especially in a special issue in journal Atmospheric Chemistry and Physics. The articles range from new aerosol modules (Kokkola et al., 2008) in GCMs to quantum chemical calculations of effect of ammonia to nucleation (Ortega et al., 2008) and field measurements of long-range transport (Birmili et al., 2008) .

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Element 1:

New instruments capable of detecting and estimating the number concentrations of neutral clusters in different atmospheric environments have been developed (Kulmala et al., 2007a). One of these is the Neutral-Cluster and Air Ion Spectrometer (NAIS) which has been developed and constructed mainly by our SME partner Airel and the University of Tartu. With this instrument the atmospheric nucleation rate can be determined at 2 nm size. Moreover, nucleation measurements in water-sulfuric acid systems were carried out to unravel the first steps of new particle formation in the atmosphere.

Reaction pathways, chemical kinetics and thermodynamics of organic aerosol formation and aging from biogenic and anthropogenic hydrocarbons and oxygenated derivatives are determined experimentally. A kinetic model framework for aerosol surface chemistry and gas particle interactions has been established (Pöschl et al., 2007; Ammann and Pöschl, 2007), and a chemical master mechanism of polycyclic aromatic hydrocarbon oxidation on aerosol particles is under development.

A plant chamber system is used to investigate the formation of biogenic secondary organic aerosols. In the first experiments no particles were observed when only ozonolysis of VOCs was taking place; only the initiation of OH reactions started particle formation. The event consisted of a sharp increase of nucleation mode particles which grew at a quite stable rate. The observed formation rate of 3-nm particles varied from 1 to 140 cm^{-3} s^{-1}, and the growth rates of the particles depended linearly on the total carbon concentration [ppbC] of the VOC classes in the reaction chamber.

Literature surveys and parameterisations of aerosol particle emissions from forests, sea spray, and biomass burning (wildfires) have been compiled. An anthropogenic particle number emission inventory and a method for parameterizing the effect of subgrid scale aerosol dynamics on aerosol number emission rates have been established.

A model for multicomponent partitioning of semi-volatile aerosol components including water vapor suitable for use in large scale models has been developed. The partitioning model has been coupled with the Master Chemical Mechanism (MCM) of atmo-

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spheric gas phase oxidation chemistry to identify and describe particularly important chemical species and reaction pathways of aerosol formation.

Element 2:

We have refined the methods for source apportionment of organic aerosol, based on four complementary methods: i) GC-MS molecular tracers; ii) H-NMR spectral fingerprints (Decesari et al., 2007; Moretti et al., 2008) ; iii) AMS spectral fingerprints (Lanz et al., 2007, 2008); and iv) ^{14}C analysis (Szidat et al., 2007).

Cloud Condensation Nuclei (CCN) activation experiments have been performed for a variety of aerosol systems. CCN activity and droplet growth kinetics of fresh and aged monoterpene and sesquiterpene secondary organic aerosols (SOA) have been performed (Engelhart et al., 2008; Asa-Awuku et al., 2008; Duplissy et al., 2008). Asa-Awuku et al. (2008) reported that monoterpene SOA had a slightly reduced surface tension as compared to water and was not 100% soluble. On the other hand, Duplissy et al. (2008) were able to perform a closure for α-pinene SOA between sub- and supersaturated conditions with the surface tension of water. Two different CCN counters compared well for monoterpene CCN activity but with significant differences for sesquiterpene SOA (Asa-Awuku et al., 2008). Biogenic secondary organic particles were found to be reasonably active CCN. If hydroxyl radicals were present the activation diameter shifted by 1–2 nm/h. Kinetic effects were found to be negligible. The monoterpenes system could be parameterized assuming complete solubility, a surface tension of water and a molecular weight of 180 g/mol. This shows the potential for 3-D models to capture the activation of monoterpene SOA.

Within the EU Integrated Infrastructure Initiative EUSAAR (European Supersites for Atmospheric Aerosol Research) a series of new hygroscopicity tandem differential mobility analyzers were built and intercompared. These instruments measure the water uptake at enhanced relative humidity and will be operated during a full year at different sites (see Table 1).

Furthermore, the 4-D distribution of aerosol and cloud properties on regional and

global scales using combinations of satellite instruments will be provided. In the first 16 months, en extensive review of the state of the art of aerosol and cloud retrieval using satellite data, for the instruments used in EUCAARI, has been performed.

Finally, an assessment of the relative contribution of natural and anthropogenic sources as well as primary and secondary sources to particle number concentrations at climate-relevant sizes will be performed; these activities will be started in the second phase of the project.

Element 3:

A new instrument to study ice nucleation, the Zurich Ice Nucleation Chamber (ZINC), has been built (Stetzer et al., 2008). It is a continuous flow diffusion chamber following the design by Rogers (1988) but has a flat parallel plate geometry. The instrument can operate at temperatures as low as 236 K with the current setup with ice supersaturations of up to 50%. Activation experiments with silver iodide particles were used to validate the instrument against literature data. The data exhibit an almost linear trend for the onset of freezing for an activated fraction of 2% of all particles between 257 K (111.5% RHi) and 237 K (119% RHi) with very good agreement with literature data.

The aerosol indirect effect (aerosol impacting cloud dynamics) is the main source of uncertainty in the estimation of the cloud feedback on climate (Sandu et al., 2008). Recent large-eddy simulations (LES) of nocturnal stratocumulus exposed to various aerosol loadings have shown that the response of the cloud liquid water path can be either an increase or a decrease, depending on the large scale conditions. In preparation to the EUCAARI Intensive Observation Period, LES simulations have been coupled for the first time with the diurnal cycle. They reveal that, during the day, increased CCN concentrations lead to a decrease of the liquid water path (LWP), irrespective of the large scale conditions.

The intensive year of measurement (March 2008–March/May 2009) in Europe has been prepared. Some intensive campaigns are ongoing: aerosol nucleation and organic aerosol formation measurements in polluted, clean and marine environments, and chemical and physical measurements at the ground during the aircraft campaign (May 2008).

Four measurement sites in developing countries have been selected. In India the sampling site chosen is located at the outskirts of New Delhi. It is about 30 km south of the centre of the city. The site is urban background surrounded by farms and fields with no major local pollutant sources. Prevailing winds are from north-east, during the monsoon season southerly winds are more common. The measurements are done using a transportable measurement container, which has been built, equipped and tested in Finland. Extra precautions have been taken toward the extreme heat and humidity during summer seasons.

In China the sampling site is a regional GAW site north of Beijing at the foot of the mountains. The site receives polluted air from the north and clean air from the north. This site has an existing infrastructure for aerosol measurements including aerosol mass concentration, light scattering and absorption measurements. During summer, the dew point temperature can reach values well above 22°C. For this reason, an automatic diffusion dryer is presently being built and will be in operation by the end of January 2009 together with instruments to measure number size distributions. The Raman lidar measurements and the filter sampling will also start in 2009.

In Brazil a sampling site for the in-situ measurements is located in the rain forest north of Manaus, Amazonia. The aerosol inlet will be installed above the canopy (at approximately 45 m). Through a pipe the aerosol will be brought into a newly installed container. To avoid condensation of water in the sampling pipe below the canopy, the whole inlet system will be kept at 32°C. The relative humidity is high and the aerosol will be dried with an automatic diffusion dryer before it enters the measurement con-

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tainer. The sampling system and the dryer have been built. The inlet system, aerosol dryer, absorption photometer, and mobility size spectrometer will be installed in January 2009. The Raman lidar will be operated a few kilometres away and is expected to be operational in January 2009 as well.

In South-Africa the sampling site is located on a hill east of Johannesburg in the main region of coal-fired power plants and biomass burning. The existing station is upgraded for the in-situ aerosol measurements. As for the other sites, due to the high dew point temperatures in summer time, an automated aerosol dryer has to be installed. The dryer as well as a mobility size spectrometer are being built and will be installed together with a nephelometer in January 2009. The Raman lidar measurements and the filter sampling will also start in 2009.

Element 5: impacts and integration

The majority of the results reported in the fourth assessment report of the Intergovernmental Panel on Climate Change (IPCC) (Forster et al., 2007), suggest that anthropogenic aerosol on a global annual average exerts a cooling effect on the Earth-Atmosphere-System (see discussion in Haywood and Schulz, 2007). However, there was a discrepancy between observation-based estimates at or near $-1.0\,W/m^2$ and model-based estimates near $-0.4\,W/m^2$. Later, several independent studies of EUCAARI partners have explored details responsible for this discrepancy. These studies demonstrate that the more negative estimates of observation-based approaches are most likely incorrect. The use of quasi-global sun-/sky photometer data of AERONET, which define not only aerosol optical depth (AOD) but also yield simultaneous information on aerosol absorption and size (without making a-priori assumptions), suggests a $-0.2\,W/m^2$ aerosol direct cooling (Kinne, 2008). This value is identical to the average from AeroCom models (Schulz et al., 2006), in which, however, contributions from nitrate and anthropogenic dust are missing. Altogether, this may suggest that the discrepancy between model based and observations' based estimates have been reduced.

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Scenario Modelling is carried out at the Meteorological Synthesizing Centre West (MSC-W) of EMEP at the Norwegian Meteorological Institute (met.no). MSC-W builds its contribution to EUCAARI on the experience gained through supporting policy work within both the UN-ECE and EU frameworks both through direct model calculations and scenario analysis and through the production of source-receptor matrices, which are used as input to integrated assessment models such as the IIASA-RAINS model. Recent applications of the EMEP model for policy have supported the revision of the NEC Directive under the CAFE (Clear Air for Europe) programme of the European Commission.

In a EUCAARI-related study Solberg et al. (2008), argued based on atmospheric trace species measurements in the boundary layer over Europe during the exceptionally warm summer of 2003, that a number of positive feedbacks between the weather conditions and atmospheric composition contributed to the elevated surface ozone observed. The findings are also relevant for the PM concentrations. Regional-scale model calculations indicate that enhanced levels of biogenic isoprene could have contributed up to 20% of the peak ozone concentrations. Measurements indicate elevated concentrations of isoprene compared to previous years. Sensitivity runs with a global chemical transport model showed that a reduction in the surface dry deposition due to drought and the elevated air temperature both could have contributed significantly to the enhanced ozone concentrations. Because of climate change, such heat waves may occur more frequently in the future and may gradually overshadow the effect of reduced emissions from anthropogenic sources of VOC and NO_x in controlling surface ozone.

The EUCAARI database for observations is building on previous developments at NILU and the project utilises the EMEP database and the modifications made for the CREATE project. The EMEP database (EBAS) was until recently residing inside the NILU firewall and data were exported to a static web interface on an annual basis in order to make observations available to a larger audience. The ambition in ongoing projects such as EUSAAR and EUCAARI has been to develop a web interface for

EBAS and to allow for a direct searching in the index database. In order to allow this, it has been necessary to move EBAS to a new server, which can be accessed from outside NILU. This was accomplished in June 2007 and required a new installation of the database engine (SYBASE) and recompilation of all associated software used for data manipulation and extraction. The system now works satisfactorily on the new server.

6 Conclusions

The interactions and feedbacks between aerosols and clouds, aerosols/clouds and climate, as well as air pollution and climate are many and intricate. The study of them requires a multidisciplinary approach. The research chain concept has to be followed to develop a deeper science understanding.

During the first 16 months EUCAARI has developed and constructed a prototype of a neutral cluster spectrometer (Kulmala et al., 2007a), planned the network for the intensive operational year, started intensive measurements and installed and compared different models. As a summary we can state that during the first year the tools for successful future operation and to find answers to the major research questions were developed.

During the first 16 months the implementations of our basic strategy to utilize recent results obtained in nanoscale in global scale have started. As an example the recently developed ion and cluster spectrometers will be used in all ground base stations (see Table 1). A clearer example is the use of the recent atmospheric nucleation scheme (Kulmala et al., 2006) in global models (Spracklen et al., 2006, 2008; Makkonen et al., 2008).

Our multidisciplinary approach from nano to global scale will be further developed during the next years in the effort to meet the EUCAARI objectives.

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Appendix A

Work package structure

Work Package Structure: Each of the four science elements is subdivided into several work packages (WPs) to address specific questions. Each element covers all scales from microscopic properties to global scale climate-aerosol and aerosol-air quality – climate interactions.

1. Emissions and Formation Element (leader: U. Pöschl, MPI-C)

   1.1 Nucleation: Determination of formation mechanisms of atmospheric aerosol particles and clusters. Work will focus on the most probable nucleation pathways and investigation of the existence of neutral and ion clusters in the atmosphere. Theoretical studies, microscale modelling and nucleation chamber studies. Tests and development of cluster spectrometers. Leader: V.-M. Kerminen, FMI

   1.2 Formation and growth of organic aerosols: Evaluation of secondary organic aerosol formation from a variety of anthropogenic and biogenic precursors. Will involve experiments on aerosol yield, chemical composition, physical and optical properties, as well as the identification of marker compounds. Construction and application of a detailed model framework for aerosol formation with improved oxidation schemes of the relevant precursors. Evaluation of the near-source production of semi-volatile organic products. Sensitivity analysis of aerosol yields under different atmospheric conditions. Leader: T. Mentel, FZJ

   1.3 Anthropogenic and biogenic emissions of aerosols and precursors: Development of scale-independent emissions inventories of aerosols (including size distribution, mixing state, and composition) and their precursors. The emissions will be suitable for use in urban, regional, and global scale models

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with uncertainties due to scale differences quantified. Emission fluxes will be measured using detailed micro-meteorological measurements under varying environmental conditions. Up-scaling by process models evaluated by field measurements to develop a number emissions pre-processor for Chemical Transport Models. Leader: H. D. van der Gon, TNO

1.4 Multicomponent gas-aerosol partitioning and thermodynamics: Development of a modelling framework to treat the partitioning of complex organic compounds to the aerosol phase. Will include formation of gaseous precursors grouped according to their volatility, partitioning of compounds to representative aerosol products, and condensed phase reactions. Currently available thermodynamic models will be evaluated against a benchmark model and results of laboratory experiments. Leader: G. McFiggans, UMAN

2. Transport and Transformation Element (leader: U. Baltensperger, PSI)

2.1 Evolution of aerosol properties during transport: Quantification of changes in aerosol physical, chemical and hygroscopic properties as anthropogenic air masses are transported and mix with clean air. Will include studies under clear and cloudy conditions. Continuous measurements from the ground-station network will be combined with airborne measurements to follow the evolution of aerosol properties on a time-scale of a few days. Leader: P. Laj, CNRS

2.2 Aerosol Characterization in developing countries: Measurements and modelling of representative physical, chemical and optical properties of aerosol under heavily pollution conditions in Europe and economically developing countries (China, India, South Africa, and Brazil). Annual and seasonal variations of the observed aerosol properties. Measurements of physical (number size distribution), chemical (size-resolved chemical composition) and optical properties (light scattering, light absorption, vertical profile of the extinction coefficient, aerosol optical depth). Leader: A. Wiedensohler, IFT

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2.3 Satellite retrievals: Use of satellites to understand regional aerosol variations and aerosol-cloud interactions. Will provide the 4-D distribution of aerosol and cloud properties on regional and global scales, using combinations of satellite instruments (multi-spectral, multi-angle and polarisation), complemented by lidar (CALIPSO) data. Improvement of retrieval algorithms. Validation vs. ground based data. Leader: G. de Leeuw, FMI

2.4 Regional aerosol source apportionment and long range transport: Apportionment of anthropogenic and natural, as well as primary and secondary aerosol components. Use of data from existing monitoring networks and archived data. Development and implementation of new methodologies for source apportionment particularly for organic aerosols. Use of backward transport model calculations for spatial source apportionment and geographic link to emission inventories. Determination of the aerosol concentration in Europe due to long-range transport. Influence on AQ/PM of transnational transport within Europe. Analysis of satellite information on the aerosol distribution in the northern hemisphere in combination with ground based aerosol measurements in Europe (EUCAARI and monitoring networks) using regional and global CTM, including data assimilation. Leader: E. Swietlicki, LU

2.5 Global/Regional Scale Particle Number: primary versus secondary, natural versus anthropogenic: Assessment of the relative contribution of natural and anthropogenic sources as well as primary and secondary sources to particle number concentrations at climate-relevant sizes. Sensitivity of the particle concentrations to changes in anthropogenic and natural emissions using global and regional offline model simulations using best available primary emissions inventories (WP 1.3) and new particle formation mechanisms. Leader: I. Riipinen, UHEL

3. Climate and Air Quality Effects Element (leader: K. Carslaw, ULEEDS)

3.1 CCN/IN activation and optical properties: Laboratory measurements of cloud

activation in warm, mixed-phase and ice clouds. Ability of aerosols to act as cloud condensation (CCN) and ice nuclei (IN). Combines long-term observations with modelling of aerosol activation and cloud processing. Will include the effects of light-absorbing particles within clouds on the radiation budget. Leader: U. Lohmann, ETHZ

3.2 Assessment and quantification of the aerosol indirect climatic effects: Experimental studies to quantify the aerosol direct, semi-direct, first indirect, and second indirect effects (i.e. the impact of aerosol on boundary layer cloud life cycle and extent). Will involve experiments at the Cabauw supersite to measure turbulent fluxes, aerosol light-absorption, hygroscopic properties, cloud microphysical and radiative properties, and precipitation. LES simulations of boundary layer clouds forced by observations with diverse aerosol types. The effect of pollution plumes (tracked by models and satellite observations) on cloud radiative properties will be quantified. Leader: R. Boers, KNMI

3.3 Parameterizations for global and regional models: Development and testing of parameterizations of aerosol formation, transformation, optical properties, and interaction with clouds developed in EUCAARI. More realistic representation of aerosols and aerosol-cloud interaction in climate models, leading to better quantification of the direct and indirect effects. Evaluation using benchmark datasets. Leader: E. Vignatti, Ispra

3.4 Regional and global air quality: Quantification of the spatial distribution and variability of particulate air pollution over Europe in terms of particle number, mass concentration, composition, and visibility. Effects of urban centres, interactions with the chemistry of gas phase air pollutants, and changes with different emissions. Effect of climate change on aerosol concentrations. Leader: C. Pilinis, UAEG

3.5 Aerosol forcing and climate response: Quantification of the aerosol optical depth, absorption optical depth, and the direct and indirect radiative forcing using GCMs. The sensitivity to changes in emissions, specific processes and

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new model approaches will be determined. Will use climate-equilibrium simulations to quantify the response of temperature and precipitation to changes in emissions of greenhouse gases and aerosols from specific economic sectors. Effect of aerosol composition and concentrations on the climate sensitivity. Leader: J. Feichter, MPI-M

3.6 Feedback processes and interactions: The impact of a changing climate on natural aerosol emissions and feedbacks between the aerosol cycle, water cycle and biosphere. Quantification of the magnitude of anthropogenic aerosol effects consistent with temperature changes over the last 100 years. Regional climate sensitivities to the aerosol-climate interaction. Time-slice experiments for the recent past and for future projections using earth-system models. Time-slice simulations will be set up on transient climate simulations done for the IPCC AR4. Leader: O. Boucher, METO

4. Project Infrastructure Element (Leader: H.-C. Hansson, SU)

4.1 Ground sites development outside Europe: Knowledge transfer of aerosol measurement techniques and data analysis. Building and maintaining the measurement stations outside European networks. Leader: H. Lihavainen, FMI

4.2 Field site preparation for Lagrangian experiments and advanced studies: Deployment of instruments at ground-based sites to provide all required parameters for analyses of aerosol transformation during Lagrangian experiments and continuous measurements. Includes field site selection based on continuous monitoring of aerosol properties performed within existing networks. Leader: H.-C. Hansson, SU

4.3 Airborne infrastructures: Preparation, planning and operation of aircraft during intensive observation periods. This task will be carried out in close cooperation with the EUCAARI intensive ground based sites and the North-South and East-West European transects. The unique comprehensive airborne

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measurements on large spatial scales will be used in the majority of work packages. Leader: R. Krejci, SU

5. Integration and Impacts Element (leader: Ø. Hov, Norwegian Meteorological Institute – met.no)

5.1 Aerosol impacts on climate and air quality: Integrating into policy: The overall effect of current, past and future aerosol loadings on regional and global climate. The effect of aerosols on air quality across Europe. Climate change effect on air quality across Europe. Modelling studies on regional and global using GCMs and CTMs. Effect of reductions in emissions of precursors and aerosols in and outside Europe on the air quality and climate of Europe. Determination of best medium-term economic and technical strategies and policy instruments to protect the atmosphere so that the economic burden is minimized. Modelling of economic costs of air pollution abatement strategies with an emphasis on aerosols in Europe. Leader: Ø. Hov, NMI

5.2 EUCAARI Platform – Data: A web-accessible database will be created that contains measurement data and model output, suitable as a testbed for aerosol models. The development of user interfaces and dissemination of project results to the end-users and stakeholders. The EUCAARI database integrates all data collected within the project and relevant data from other sources. Data will be made accessible to the project partners in a common format via an internet interface that allows for data searching and production of simple plots (e.g., of time series, or of the data geo-location). A Lagrangian dispersion model will be run 20 days backward in time from all measurement platforms collecting in situ data during EUCAARI for source-region identification. Results will be linked directly to the measurement data. Leader: A. Stohl, NILU

5.3 EUCAARI Platform – Models: Significant improvement of the evaluation process of regional and global aerosol models to document modelling progress

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made during EUCAARI. Uncertainties in our prediction of aerosol impact on climate and air quality will be estimated by comparing different models and processes. In this work package benchmark tests will be developed to analyze model biases and the effect of different physico-chemical properties of the aerosol in the models. Leader: M. Schulz, LSCE-CEA

Appendix B

EUCAARI partners

Participant number	Participant name	Participant short name	Country
1	University of Helsinki	UHEL	Finland
2	The French National Center for Scientific Research	CNRS	France
3	Max-Planck-Gesellschaft zur Förderung der Wissenschaften e.V.	MPI	Germany
4	Leibniz-Institute for Tropospheric Research	IFT	Germany
5	Institute of Atmospheric Sciences and Climate	CNR-ISAC	France
6	Eidgenössiche Technische Hochschule Zürich	ETZH	Switzerland
7	Netherlands Organisation for Applied Scientific Research	TNO	Netherlands
8	Netherlands Royal Meteorological Institute	KNMI	Netherlands
10	University of Leeds	ULEEDS	UK
11	University of Lund	LU	Sweden
12	University of Pannonia	ACUV	Hungary
13	Finnish Meteorological Institute	FMI	Finland
14	European Commission – Directorate General Joint Research Centre Commission	EC-DG JRC	European
15	UK Met Office	METO	UK
16	Norwegian Institute for Air Research	NILU	Norway
17	Norwegian Meteorological institute	met.no	Norway
18	National University of Ireland, Galway	NUIG	Ireland

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Participant number	Participant name	Participant short name	Country
19	Paul Scherrer Institut	PSI	Switzerland
20	University of Oslo	UO	Norway
21	Foundation for Research and Technology Hellas	FORTH	Greece
22	Academy of Sciences of the Czech Republic	ASCR	Czech Republic
23	French Meteorological Service	CNRM	France
24	Forschungszentrum Jülich GmbH	FZJ	Germany
25	North- West University, South-Africa	NWU	South Africa
26	The Energy and Resources Institute, India	TERI	India
27	University of Copenhagen	UKBH	Denmark
28	University of East Anglia	UEA	UK
29	University of Kuopio	UKU	Finland
30	University of Manchester	UMAN	UK
31	APLBA	APLBA	Brazil
32	Airel AS (Ltd.)	AIREL	Estonia
33	University of Birmingham	UNI BHAM	UK
34	German Aerospace Center	DLR	Germany
35	University of Crete	UoC	Greece
36	Hebrew University of Jerusalem	HUJ	Israel
37	The International Institute for Applied Systems Analysis	IIASA	Austria
38	University of Stockholm	SU	Sweden
39	Warsaw University	IGFUW	Poland
40	University of Aveiro	UAV	Portugal
41	University of Tartu	UTAR	Estonia
42	University of Mainz	JGUM	Germany
43	Peking University	PKU	China
44	Chinese Academy of Meteorological Sciences	CAMS	China
45	PANEPISTIMIO AIGAIOU	UAEG	Greece
46	Laboratory of Climate and Environmental Sciences	CEA	France
47	Risø National Laboratory	RISØ	Denmark
48	German Meteorological Service	DWD	Germany

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Table 1. EUCAARI Long-term campaign; ground-based activities March 2008–March 2009.

Station	Operator	LIDAR	AIS/NAIS	HTDMA	CCNC	EUSAAR station
Mace Head, IR	NUIG		x	x UHEL	x	x
Vavihill, SE	SU		x	x		x
Melpitz, DE	IFT	x	x	x UMAS		x
Hyytiala, FI	UHEL		x	x		x
K-Puzta, HU	ACUV		x UHEL			x
Jungfraujoch, CH	PSI		x CNRS	x	x	x
Pallas, FI	UKU		x UHEL	x SU		x
Puy de Dome, FR	CNRS	x	x	x		x
Cabauw, NL	KNMI	x	x UHEL	x UHEL	x	x
Finokalia, GR	UoC					x
Kosetice, CZ	ICPF AS		 CR	x		x
Hohen Peissen-berg, DE	DWD		x UHEL			
San Pietro Capofiume, IT	CNR-ISAC		x UHEL			

x: Partner responsibility only indicated when different than site responsibility. Partner acronyms are explained in Appendix B.
LIDAR: Light Detection and Ranging
AIS/NAIS: Air Ion Spectrometer/Neutral Cluster and Air Ion Spectrometer
HTDMA: Hygroscopicity tandem differential mobility analyzer
CCNC: Cloud condensation nuclei counter
EUSAAR: EC Framework 6 Integrated Infrastructure Initiative project European Supersites for Atmospheric Aerosol Research

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Table 2. Inputs and outputs related to work packages (WPs).

WP	Inputs from other WPs (*critical inputs*)	Outputs to other WPs (*critical outputs*)
1.1 Nucleation	2.2 *Measurements outside EU* 4.2 Field site preparation for Lagrangian experiments and advanced studies 4.3 Airborne infrastructures	2.1 Regional scale aerosol formation and evolution of properties during transport 2.5 *Global particle number* 3.3 *Development of parameterizations* 3.4 Regional and global air quality 4.2 Ground field measurements
1.2 Formation and growth of organic aerosols		1.4 Multicomponent gas-aerosol partitioning and thermodynamics 2.1 Regional scale aerosol formation and evolution of properties during transport 2.4 *Long-term characterization of aerosols* 3.1 CCN/IN activation and optical properties 3.4 Regional and global air quality
1.3 Anthropogenic and biogenic emissions of aerosols and precursors	2.4 *Long-term characterization aerosols* 2.5 *Primary versus secondary, natural versus anthropogenic particle number concentration*	2.4 Long-term characterization aerosols 2.5 Primary versus secondary, natural versus anthropogenic particle number concentration
1.4 Multicomponent gas-aerosol partitioning and thermodynamics	1.2 Formation and growth of organic aerosols	3.4 *Regional and global air quality* 3.5 *Modelling of direct and indirect forcing and effects* 3.3 Developments of parameterizations
2.1 Regional scale aerosol formation and evolution of of properties during	2.2 *Measurements outside EU* 4.2 *Field site preparation for Lagrangian experiments and advanced studies* 4.3 *Airborne infrastructures* 1.1 Nucleation 1.2 Formation and growth of organic aerosols 2.4 Long-term characterization of aerosols	3.4 *Regional and global air quality* 3.5 *Modelling of direct and indirect forcing and effects* 2.3 Satellite retrievals 2.4 Long-term characterization of aerosols

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Table 2. Continued.

WP	Inputs from other WPs (*critical inputs*)	Outputs to other WPs (*critical outputs*)
2.2 Aerosol Characterization and Modelling in developing countries	4.1 *Ground site development outside Europe*	1.1 *Nucleation*
	2.3 Satellite retrievals	2.1 *Regional scale aerosol formation and evolution of properties during transport* 2.5 *Primary versus secondary, natural versus anthropogenic particle number concentration* 2.3 Satellite retrievals 5.2 EUCAARI Platform – Data
2.3 Satellite retrievals	2.1 Regional scale aerosol formation and evolution of properties during transport 2.2 Aerosol Characterization and Modelling in developing countries 4.3 Airborne infrastructures	3.1 *CCN/IN activation and optical properties* 3.2 *Assessment and quantification of the aerosol climatic effects* 3.3 *Development of Parameterizations* 3.4 *Regional and global air quality* 3.5 *Modelling of direct and indirect forcing and effects* 2.1 Regional scale aerosol formation and evolution of properties during transport 2.2 Aerosol Characterization and Modelling in developing countries 5.2 EUCAARI Platform – Data
2.4 Long-term characterization of aerosols	1.2 *Formation and growth of organic aerosols* 1.3 *Anthropogenic and biogenic emissions of aerosols and precursors* 4.2 Field site preparation for Lagrangian experiments and advanced studies 4.3 Airborne infrastructures	1.3 *Anthropogenic and biogenic emissions of aerosols and precursors* 3.4 *Regional and global air quality* 2.1 Regional scale aerosol formation and evolution of properties during transport 5.2 EUCAARI Platform – Data

Table 2. Continued.

WP	Inputs from other WPs (*critical inputs*)	Outputs to other WPs (*critical outputs*)
2.5 Primary versus secondary, natural versus anthropogenic particle number concentration	1.1 *Nucleation*	1.3 *Anthropogenic and biogenic emissions of aerosols and precursors*
	1.3 *Anthropogenic and biogenic emissions of aerosols and precursors* 2.2 *Aerosol Characterization and Modelling in developing countries* 4.3 Airborne infrastructures	3.4 *Regional and global air quality* 2.1 Regional scale aerosol formation and evolution of properties during transport 5.2 EUCAARI Platform – Data
3.1 CCN/IN activation and optical properties	2.3 *Satellite retrievals* 4.3 *Airborne infrastructures* 1.2 Formation and growth of organic aerosols	3.2 *Assessment and quantification of the aerosol climatic effects* 3.3 *Development of parameterizations* 3.2 Assessment and quantification of the aerosol climatic effects
3.2 Assessment and quantification of the aerosol climatic effects	2.3 *Satellite retrievals*	3.5 *Aerosol forcing and climate response*
	3.1 *CCN/IN activation and optical properties* 3.4 *Airborne infrastructures*	3.6 *Feedbacks* 3.1 CNN/IN activation and optical properties 3.3 Development of parameterizations 5.2 EUCAARI Platform – Data
3.3 Development of parameterizations	1.1 *Nucleation* 2.3 *Satellite retrievals* 3.1 *CCN/IN activation and optical properties* 3.6 *Feedback processes and interactions* 1.4 Multicomponent Gas-aerosol partitioning and Thermodynamics 2.1 Regional scale aerosol formation and evolution of properties during transport 3.2 Assessment and quantification of the aerosol climatic effects	3.5 *Modelling of direct and indirect forcing and effects* 5.1 *Aerosol impacts on climate and air quality: Integrating into policy* 5.3 EUCAARI Platform – Models

Table 2. Continued.

WP	Inputs from other WPs (*critical inputs*)	Outputs to other WPs (*critical outputs*)
3.4 Regional and global air quality	1.3 *Anthropogenic and biogenic emissions of aerosols and precursors* 1.4 *Multicomponent gas-aerosol partitioning and thermodynamics* 1.1 Nucleation-Models 1.2 Formation and growth of organic aerosols 2.1 Regional scale aerosol formation and evolution of properties during transport 2.3 Satellite retrievals 2.4 Long-term characterization of aerosols 4.2 Ground field measurements	5.1 *Aerosol impacts on climate and air quality: Integrating into policy* 5.3 EUCAARI Platform
3.5 Aerosol forcing and climate response	1.3 *Anthropogenic and biogenic emissions of aerosols and precursors* 1.4 *Multicomponent gas-aerosol partitioning and thermodynamics* 2.1 *Regional scale aerosol formation and evolution of properties during transport* 2.3 *Satellite retrievals* 3.2 *Assessment and quantification of the aerosol climatic effects* 3.3 *Development of Parameterizations* 2.5 Primary versus secondary, natural versus anthropogenic particle number concentration	5.1 *Aerosol impacts on climate and air quality: Integrating into policy* 5.3 *EUCAARI Platform – Models* 3.6 Feedbacks
3.6 Feedback Processes and Interactions	3.2 *Assessment and quantification of the aerosol climatic effects* 3.3 *Parameterization* 1.3 Anthropogenic and biogenic emissions of aerosols and precursors 2.5 Primary versus secondary, natural versus anthropogenic particle number concentration 3.5 Modelling of direct and indirect forcing and effects	5.3 *EUCAARI Platform – Models*
4.1 Ground site development outside Europe		2.2 *Aerosol Characterization and modelling in developing countries*

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Table 2. Continued.

WP	Inputs from other WPs (*critical inputs*)	Outputs to other WPs (*critical outputs*)
4.2 Field site preparation for Lagrangian experiments and advanced studies	5.2 *EUCAARI Platform – Data* 1.1 Nucleation	2.1 *Regional scale aerosol formation and evolution of properties during transport* 2.4 *Long-term characterization of aerosols* 1.1 Nucleation 3.4 Regional and global Air quality
4.3 Airborne infrastructures		2.1 *Regional scale aerosol formation and evolution of properties during transport* 3.1 *CCN/IN activation and optical properties* 3.2 *Assessment and quantification of the aerosol climatic effects* 1.1 Nucleation 2.3 Satellite retrievals 2.4 Long-term characterization of aerosols
5.1 Aerosol impacts on climate and air quality: Integrating into policy	1.3 *Anthropogenic and biogenic emissions of aerosols and precursors* 3.3 *Parameterization* 3.4 *Regional and global air quality* 3.5 *Aerosol forcing and climate response* 3.6 *Feedback processes and interactions*	5.3 EUCAARI Platform-Models
5.2 EUCAARI Platform – Data	2.1 Regional scale aerosol formation and evolution of properties during transport 2.2 Aerosol Characterization and Modelling in developing countries 2.3 Satellite retrievals 2.4 Long-term characterization of aerosols	4.2 *Ground field measurements*
5.3 EUCAARI Platform – Models	3.5 *Aerosol forcing and climate response* 3.6 *Feedback processes and interactions* 5.2 *EUCAARI Platform – Data* 3.3 Development of parameterizations	

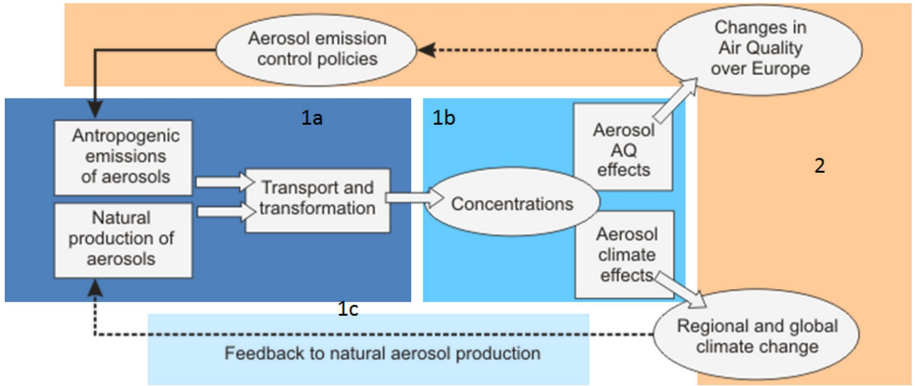

Fig. 1. Schematic of EUCAARI Objectives (Kulmala et al., 2007a).

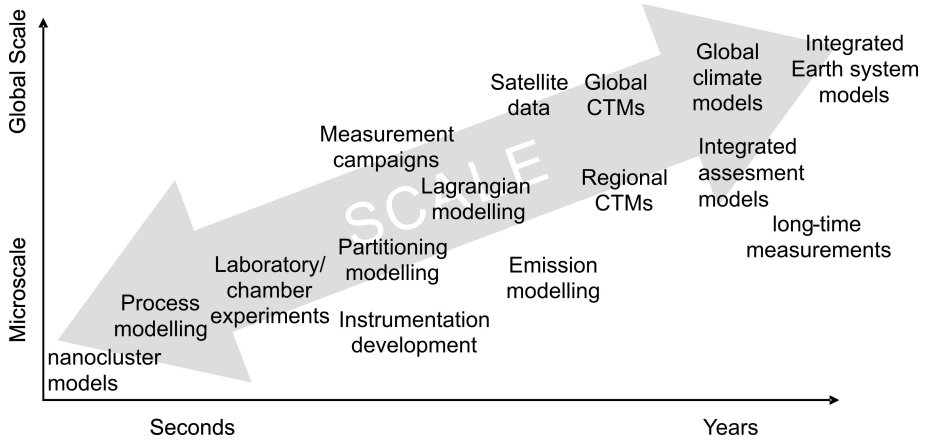

Fig. 2. Model and data integration philosophy over spatio-temporal scales. CTM is Chemical Transport Model (Kulmala et al., 2007a).

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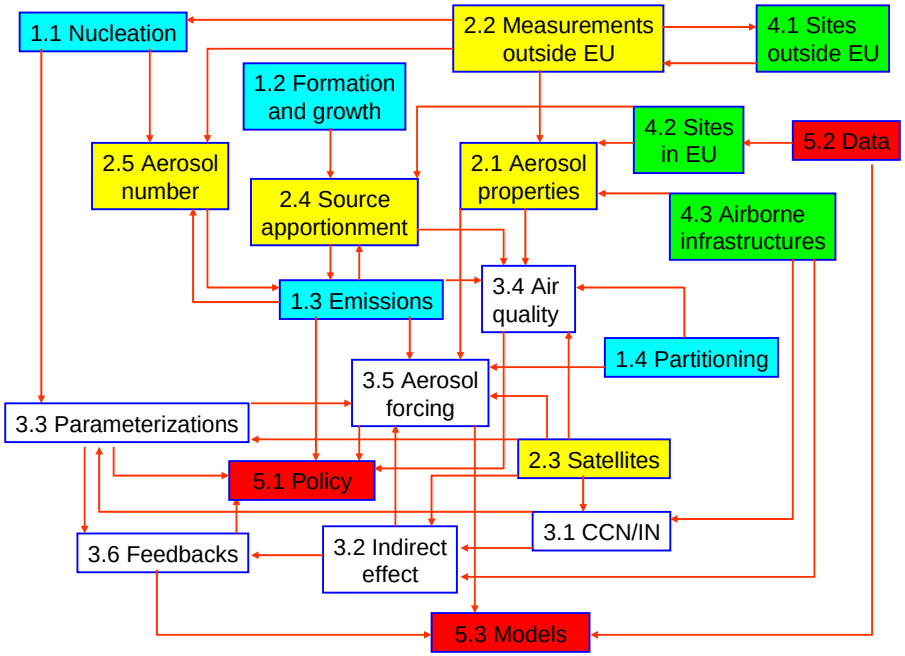

Fig. 3. Major connections between work packages, the names of the workpackages are shortened for compactness.

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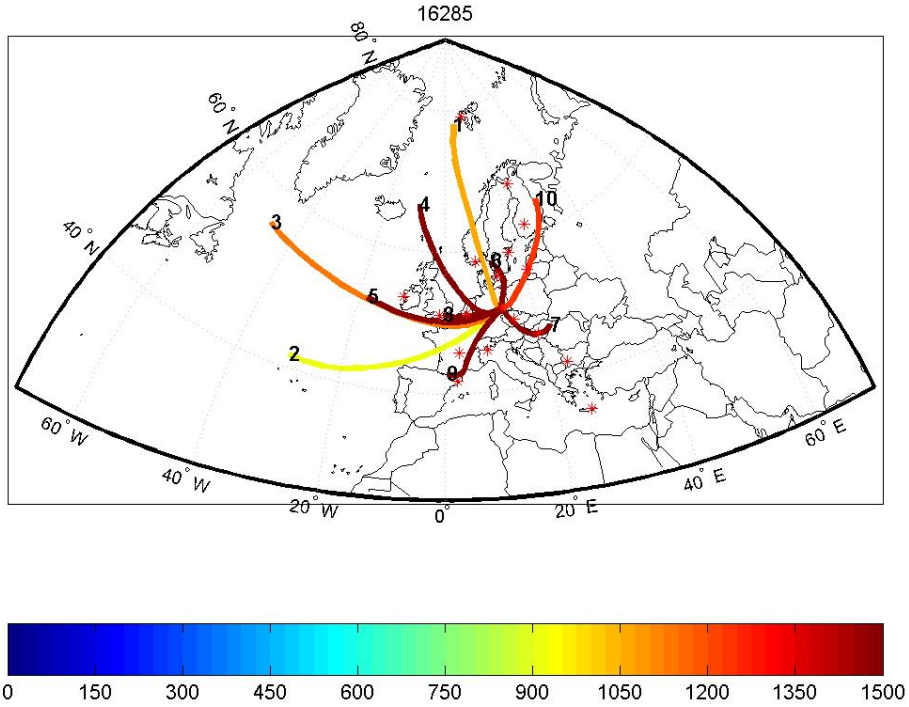

Fig. 4. Example of trajectory analysis showing the frequencies of air mass transport directions arriving at the Melpitz station during summer.

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