*Tellus (2000), 52B, 111–125*
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**TELLUS**
*ISSN 0280–6509*

# The 2nd Aerosol Characterization Experiment (ACE-2): general overview and main results

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(Manuscript received 10 September 1999; in final form 11 October 1999)

ABSTRACT

This overview summarizes the objectives of the Aerosol Characterization Experiments (ACEs) of the International Global Atmospheric Chemistry (IGAC) project and the research strategy implemented in the second of this series of experiments (ACE-2). ACE-2 took place from 16 June to 24 July 1997, over the sub-tropical North-East Atlantic. It provided an opportunity to study the properties, processes and effects of contrasting aerosol types in this region, including background marine and anthropogenic pollution aerosol in the marine boundary layer (MBL), and background aerosol and mineral dust in the overlaying free troposphere (FT). The major achievements of ACE-2 include: (a) identification of entrainment, in-cloud scavenging and coagulation as the major processes transforming a pollution aerosol transported within the MBL; (b) the first documentation of the indirect radiative effect of aerosols at the scale of a cloud ensemble in continental pollution outflow; (c) observation of a wide range in the contribution of organic material to the sub-micron aerosol mass, with possibly the highest contribution in the free tropospheric; (d) improved understanding of the role of condensing HCl, $HNO_3$ and $NH_3$ as a growth mechanism of sub-micron aerosols in polluted air masses advecting over the ocean. A close connection was observed between meteorological factors (such as horizontal and vertical wind speed, boundary layer development, entrainment, humidity fields) and aerosol and cloud characteristics. In the ACE-2 region, these meteorological factors, rather than aerosol microphysics and chemistry, often dominated the shaping of the aerosol size distribution and/or their effect on radiation and clouds. The ACE-2 data presently analyzed provide a qualitative, and in many cases a quantitative understanding of the complex gas/aerosol/cloud system in the sub-tropical marine environment. This will guide future model development. Some major data sets are still to be analyzed.

## 1. IGAC's Aerosol Characterization Experiments

As a result of the first calculations of global atmospheric aerosol fields (Langner and Rodhe, 1991) and the subsequent estimates of their potential large role in the global radiation balance (Charlson et al., 1992), a number of dedicated field experiments were envisioned to improve the understanding of the properties of aerosols and their controlling processes relevant for radiative forcing and climate. A further objective of these experiments was to quantify the radiative effects of the aerosol through observation and modelling.

Three radiative effects have been identified that could result from an increase in atmospheric aerosol particles: (1) the increase of scattering and absorption of incoming solar radiation (referred to as the direct effect), (2) the increase in cloud reflectivity due to more and smaller cloud droplets forming on the aerosol (the indirect "Twomey"

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effect), and (3) the increase in the lifetime of clouds (the indirect "Albrecht" effect) due to reduced precipitation in clouds with more and smaller droplets.

It was clear from the outset that various field experiments were needed to study the aerosol in plumes that originate from different source areas. The existence of regional scale aerosol "plumes" was made visible by space borne AVHRR images (Husar et al., 1997), which show the inferred optical depth of the tropospheric aerosol over ocean areas. Fig. 1, for example, shows the aerosol optical depth for the Northern Hemisphere summer season, based on data from 1989 to 1991. With an a priori knowledge of the location of potential sources, one can distinguish mineral dust from N. Africa and Asia, aerosols from savanna burning in Africa, and pollution aerosols from industrial activities in North America, Europe and Eastern Asia. The clean atmosphere of the Southern Ocean is in sharp contrast with the rest of the world's oceans.

Fig. 1 also shows the areas where coordinated international aerosol experiments were (and are planned to be) organized. The Aerosol Characterization Experiments (ACEs) are part of the International Global Atmospheric Chemistry (IGAC) project. Their general goals and objectives are described by Bates et al. (1998). They differ from the other experiments, in their focus on aerosol properties and processes and their experimental strategy, which is based on *closure experiments*, *Lagrangian experiments* and *modelling*.

In a closure experiment, the measured value of a dependent variable is compared with the value calculated from measured independent variables. The outcome is a direct evaluation of the combined uncertainty of the calculation and the measurements. Hence, closure experiments are a tool for monitoring the performance and improvement of models and measurements. Application of this approach is of particular value in the case of aerosols, since their climate relevant properties (= the dependent variables) must be linked properly to their physical and chemical properties (= the independent variables). The latter are expected to be prognostic variables in 3-D chemical transport models or general circulation models.

In a Lagrangian experiment, an atmospheric air parcel is identified and followed during its transport. This can be performed by equipping several measuring sites along a well-defined air trajectory,

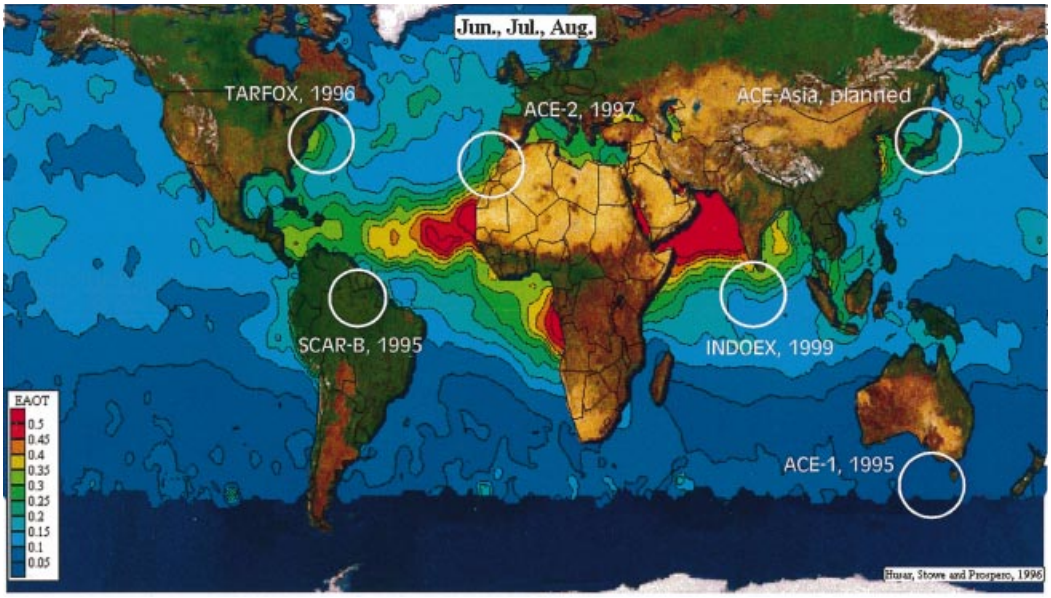


*Fig. 1.* AVHRR picture of aerosol optical depth, with indication of the location of past experiments ACE-1 (Bates et al., 1998), ACE-2, TARFOX (Russell et al., 1999), SCAR-B (Kaufman et al., 1998) and INDOEX, and the planned experiment ACE-Asia.

or by tagging an air mass with a tracer and following it with a movable platform. Changes observed in the chemical composition of the air parcel can thereby be ascribed to processing within the air parcel, rather than to changes of air masses. Changes must be described by process models, which eventually must be integrated in atmospheric models.

Modelling is an integral part of both closure and Lagrangian experiments and is the third ACE strategy element. Microphysical models using Mie theory and Köhler theory are tested as part of the closure experiments. Chemical kinetics and aerosol or cloud dynamics models are tested in Lagrangian experiments. Subsequently these models can be used as a basis for parameterizations in 3-D regional or global models. Finally these larger scale models can be tested against the observed concentration fields.

## 2. ACE-2

The 2nd Aerosol Characterization Experiment (ACE-2) took place from 16 June to 24 July 1997, over the sub-tropical North-East Atlantic (Fig 2).

The ACE-2 objectives were to:

(1) determine the physical, chemical, radiative and cloud nucleating properties of the major aerosol types in the North Atlantic region and investigate the relationships between these properties;

(2) quantify the physical and chemical processes controlling the evolution of the major aerosol types and in particular their physical, chemical, radiative and cloud nucleating processes;

(3) develop procedures to extrapolate aerosol properties and processes from the local to the regional and global scale, and assess the regional direct and indirect radiative forcing by aerosols in the North Atlantic region.

To focus the research towards these objectives several specific questions were formulated, which are repeated in Section 3. The background to these questions is found in the ACE-2 Science and Implementation Plan (Raes and Bates, 1995, http//www.ei.jrc.it/ace2/). 6 experimental activities were defined, each addressing one or more of these questions:

- CLEARCOLUMN: study of the column integrated radiative effect of aerosols, in the cloud-free atmosphere, using surface based stations, aircraft and satellites (Russell and Heintzenberg, 2000).
- CLOUDYCOLUMN: study of the effects of aerosols on the microphysics and radiative properties of MBL clouds using aircraft and satellites (Brenguier et al., 2000).
- LAGRANGIAN: study of aerosol evolution in an air parcel advecting from the European continent within the MBL (Johnson et al., 2000b).
- HILLCLOUD: study of the physical and chemical processing of aerosols when passing through a hill cap cloud, using stations upwind, inside and downwind of the cloud (Bower et al., 2000).
- FREETROPE: study of the free tropospheric aerosol, including mineral dust, from a free tropospheric ground station and aircraft, and its effect on the MBL aerosol and cloud properties.
- LONGTERM: long-term observations throughout the ACE-2 area to provide spatial and temporal context and representativeness of the ACE-2 intensive campaign.

All 6 activities took place simultaneously in the same area between the southern coast of Portugal and the Canary Islands, making use of the same operational logistics. This concentration of personnel, platforms and equipment provided a unique opportunity to implement the often complex experimental strategies required to address the scientific issues.

The choice of the area is explained in detail in the meteorological overview paper (Verver et al., 2000). In summary, the area is affected by contrasting air masses and thus offers opportunities for studying the characteristics of various aerosol types: background marine aerosol and European anthropogenic pollution in the marine boundary layer, and background aerosol and mineral dust in the overlaying free troposphere. In addition, the area offers good opportunities to study the interaction of aerosols with radiation in the cloud-free areas typically found immediately downwind the land masses, and to study the interaction with the low level stratiform clouds developing in the MBL. Bretherton and Pincus (1995) characterized the dynamics of the MBL in this area previously. This aided in the planning of the Lagrangian experiments

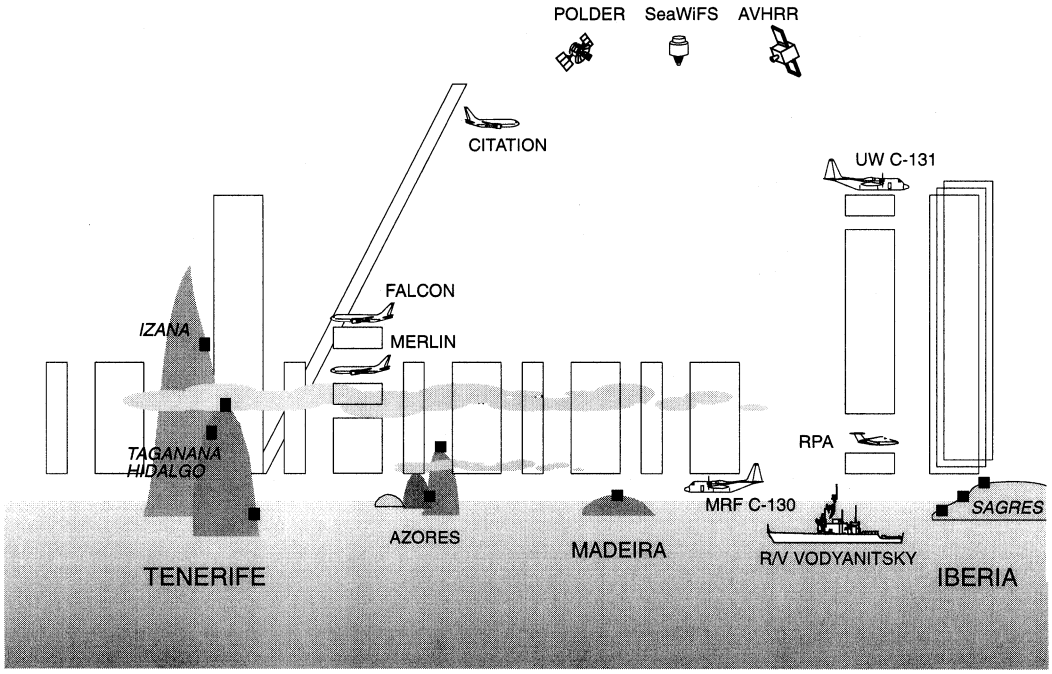


*Fig. 2.* Deployment of the ACE-2 platforms.

The main sites and platforms are listed in Table 1 and shown in Fig. 2. ACE-2 operated from 3 major surface based platforms: *Sagres (Southern Portugal)*, dedicated to aerosol characterization and CLEARCOLUMN studies, *the research vessel Prof. Vodyanitskiy*, dedicated to aerosol characterization, CLEARCOLUMN studies and serving as a platform to initialize the Lagrangian experiments, and *the island of Tenerife (Canary Islands)* which was the measurement center for the

Table 1. *ACE-2 sites and platforms*

| Site/platform | | Operator |
|---|---|---|
| Punta del Hidalgo | Spain | Joint Research Center, Ispra |
| Izaña | Spain | Spanish Meteorological Office, Madrid |
| Taganana sites | Spain | UMIST, Manchester |
| Sagres | Portugal | IFT, Leipzig / JRC, Ispra |
| Madeira | Portugal | Joint Research Centre, Ispra / |
| Azores | Portugal | Portuguese Meteorological Office, Lisbon |
| R/V *Vodyanitskiy* | Ukraine | IBSS, Sebastopol / PMEL, Seattle |
| C-130 Hercules | UK | MRF, Farnborough |
| Dornier | Germany | DLR, Oberpfaffenhofen |
| Merin-IV | France | Meteo France, Toulouse |
| Citation | Netherlands | Delft University, Delft |
| ARAT | France | INSU, Paris |
| Pelican | US | CIRPAS, US Navy |

UMIST: University of Manchester Institute of Science and Technology. IFT: Institute for Tropospheric Research. IBSS: Institute for the Biology of the Southern Seas. PMEL: Pacific Marine Environmental Laboratory, NOAA. MRF: Meteorological Research Flight, UKMO. DLR: Deutsche Forschungsanstalt fur Luft und Raumfahrt. INSU: Institut National des Sciences de l'Univers. CIRPAS: Center for Interdisciplinary Remotely Piloted Aircraft Studies.

HILLCLOUD and FREETROPE experiments. The 6 aircraft operated from the airport "Los Rodeos" on Tenerife. Meteorological forecasts and on-line data management were provided at the ACE-2 operational center, also located at "Los Rodeos".

Quality control in a multi-agency multi-platform experiment is a prerequisite, but also a challenge to implement. Within the context of ACE-2 a number of instrumentation workshops were held before the intensive experiment. Condensation particle counters (44 in total) were intercompared and calibrated, following procedures adopted in ACE-1 (Wiedensohler et al., 1997). All CLEARCOLUMN radiometers were intercompared and calibrated at the German high altitude observatory Zugspitze before the intensive campaign. An intercomparison for ion chromatographic (and related techniques) was performed before and after the intensive campaign (Putaud et al., 2000). During the intenisive campaign measurements of aerosol size distribution (Collins et al., 2000), cloud condensation nuclei (Chuang et al., 2000) and cloud droplet number concentration (Martinsson et al., 2000) were intercompared, as well as many other parameters which are not explicitly described here. These exercises identified simple instrumentation problems, and helped to quantify the uncertainties related to several of the measuring techniques. Many of the instruments were deployed on Tenerife during pre-ACE-2 campaigns in 1994, 1995 and 1996. A part from producing scientific results (Raes et al., 1997; McGovern et al., 1999; Van Dingenen et al., 1999), they helped to optimize the sampling strategy and to resolve logistical problems.

## 3. Main results of ACE-2

This special issue presents a selection of results, without attempting to cover all the observations of all 6 activities. The selection is such that initial answers can be given to the scientific questions formulated in the ACE-2 Science and Implementation Plan (Raes and Bates, 1995). First a summary is given of the main ACE-2 results documented in this special issue. These results are subsequently described in more detail and structured as answers to the original ACE-2 scientific questions.

### *3.1. ACE-2 highlights*

- Lagrangian and Eulerian process studies identified entrainment, in-cloud scavenging of particles and coagulation between particles as the main processes that transform a continental pollution aerosol into a background marine aerosol. A simple model including these processes and tested against the observations, suggests that after about 4.5–6.5 days of transport within a sub-tropical MBL, the contribution of continental pollution to the sub-micron aerosol number and volume might still be 50%. Absolute number concentrations decay slightly faster than absolute volume concentrations (Johnson et al., 2000a; Hoell et al., 2000; Van Dingenen et al., 2000).
- The Lagrangian experiments documented how in the sub-tropics transport of a polluted air mass from the continent to the ocean involves the development of a new MBL within the polluted air mass, with a residual continental layer aloft. Depending on how fast the latter layer is removed by differential advection, entrainment of pollution from that layer into the MBL delays the transition to a clean MBL aerosol (Johnson et al., 2000a; Wood et al., 2000).
- In general, the background sub-micron aerosol mass encountered during ACE-2, remains a factor 3 to 4 higher than that during ACE-1 over the background Southern Ocean. Non seas-salt (nss) $SO_4^=$ concentrations remain about a factor 2 higher. The sulfur cycle in the clean ACE-2 masses can be explained based on emissions of biogenic DMS, and the difference nss $SO_4^=$ in mass between ACE-1 and ACE-2 is explained by the difference in meteorological settings: i.e., the stable conditions in the sub-tropical high over the ACE-2 area resulting in longer residence times than the frequent frontal passages over the ACE-1 area. However, other contributing factors to the higher ACE-2 aerosol mass, such as the slow decay of the anthropogenic contribution from the surrounding continents (including N. America) and ship emissions upwind the ACE-2 area, cannot be ruled out (Bates et al., 2000; Quinn et al., 2000; Putaud et al., 2000; M. O. Andreae, personal communication; Verver et al., 2000).
- In the MBL, physical, chemical and optical aerosol properties were dominated by sub-micron aerosols related to pollution aerosols advecting from the Europe. Seasalt was observed to contrib-

ute significantly to the aerosol properties only in the clean air masses advecting from the open Atlantic. The measured increase in MBL aerosol number concentration with increasing wind speed in the first LAGARNGIAN, will provide a parameterization of the relationship between wind speed and flux of aerosol number (Quinn et al., 2000; Johnson et al., 2000b).

- Chemical mass closure measurements were performed on sub- and super-micron aerosols. Large analytical uncertainties (up to 90%) remain in determining the organic aerosol in individual samples. Values for the average contribution of organic aerosol to the sub-micron aerosol mass are significantly different in the various air masses encountered. The lowest organic contribution was $12 \pm 2\%$, found in the MBL in air masses travelling south along the Western European coast. The highest contribution was $64 \pm 32\%$, found in the FT in air masses originating over N. America. In the latter case, the black carbon contribution was $7 \pm 5\%$. (Errors given are standard errors of the mean, calculated assuming that the uncertainties in a single measurement are random, which in this case gives a lower limit of the uncertainty. The upper limit would be closer to the analytical uncertainty on a single sample (Putaud et al., 2000; Neusüß et al., 2000).)
- Anthropogenic $SO_2$ was observed to react rapidly during the initial transport of a pollution plume over the ocean. Most likely this is due to in-cloud chemistry, as a deepening of the gap between Aitken and accumulation mode and a growth of the accumulation mode particles was observed at the same time. Once at levels of tens of ppt, but still in the pollution plume, $SO_2$ did not have an observable effect on particle growth. In stead, condensation of HCl and $HNO_3$ (some of which is outgassed from super-micron particles during cloud passage) and their fixation by $NH_3$ on the sub-micron particles was observed and modelled. This constitutes a mechanism of aerosol growth in the marine environment, which was previously not accounted for (Osborne et al., 2000; M. O. Andreae, personal communication; Dore et al., 2000; Bower et al., 2000; Flynn et al., 2000).
- The model for adiabatic growth of cloud droplets has been experimentally validated for stratiform clouds. This model predicts the cloud optical thickness to be proportional to (cloud geometrical thickness)$^{5/3}$ and to (cloud droplet number concentration)$^{1/3}$, and offers the means to distinguish between the 2 effects (Brenguier et al., 2000).
- Simultaneous measurements of aerosol physical and chemical properties, CCN, cloud micro- and macrophysics, and cloud reflectance up to the scale of a cloud ensemble ($\sim 100$ km), resulted in the first full documentation of the indirect radiative effect of aerosols in continental pollution outbreaks. The observations pertain mainly to the Twomey effect. Evidence for the Albrecht effect, i.e., the reduction of drizzle in polluted air masses is also present in the data (Brenguier et al., 2000; Pawlowska and Brenguier, 2000; Osborne et al., 2000; Johnson et al., 2000a).
- Application in a hill cap cloud of novel instrumentation for cloud droplet characterization revealed a severe undercounting of cloud droplet number concentrations by standard airborne techniques. The earlier observations that cloud droplet number depends sub-linearly on aerosol number concentration might partially be wrong, and the estimates of the indirect effect based on this dependence too low (Martinsson et al., 2000).
- Aerosol optical depth measurements, using AVHRR imaginary from satellite, were compared with ground based and airborne sun photometry in a wide variety of air masses. In-situ measurements of the properties of various aerosols, including mineral dust, provide the data to optimize the AVHRR AOD retrieval algorithm, and sets the stage for an improved regional quantification of the radiative effect of a variety of aerosols (Durkee et al., 2000; Schmid et al., 2000).

### *3.2. Answers to the ACE-2 scientific questions*

***Question 1***

*Can the measured physical and chemical properties of the atmospheric aerosol be used to predict the radiative and cloud nucleating properties of that same aerosol?*

The consistency in determining the physical and chemical aerosol properties of individual aerosol samples was tested by comparing (1) the sum of the masses of the individual chemical compounds with (2) the gravimetricly determined aerosol mass and with (3) the aerosol mass derived from the aerosol size distribution assuming an aerosol density (Neusüß et al., 2000; Putaud et al., 2000). In the case of sub-micron aerosols, the agreement between (1) and (2) was within $\pm 30\%$ in the

polluted MBL, mainly because of the relatively small contribution of organic aerosol (12% on average), for which the uncertainty in the mass determination can amount to $\pm 90\%$. In the dust-free FT, where the organic fraction can be 64% on average, and the absolute concentrations are typically $10\times$ lower than in the polluted MBL, the agreement between (1) and (2) was within $\pm 90\%$ (Putaud et al., 2000).

Radiative and hygroscopic properties and cloud condensation nuclei (CCN) were measured simultaneously with the physical and chemical properties of the same aerosol at Punta del Hidalgo, Sagres, the ship and Izaña. This offers the opportunity for pursuing *Question 1* in a wide range of conditions.

To date, no thorough radiative closure experiments are presented in which aerosol scattering coefficients, calculated with Mie theory and based on the measured size resolved aerosol chemistry, are compared with the measured scattering coefficients of that same aerosol. Collins et al. (2000) relate airborne measurements of number size distributions with scattering and extinction coefficients using Mie theory, relying on chemical measurements taken at ground in the same general area. This study is informative as it compares results in the clean and polluted MBL, and in the dust free and "dusty" FT. The deviations between measured and calculated scattering coefficients are on average less than 20%, except in the dust layer where they reach 40% on average. The authors attribute the latter deviation to the uncertainty in the phase function of the non-spherical dust particles, and to truncation of the size distribution by the aircraft sampling inlet. More constrained radiative closure experiments on dust aerosols, including in-situ size resolved chemistry, were performed at Izaña during the pre-ACE-2 campaign of 1995 (Hal Maring, personal communication).

Attempts to relate number size distributions and chemical composition with CCN, using Köhler theory, are made by Chuang et al. (2000), Wood et al. (2000) and Snider and Brenguier (2000), using data from the Pelican, C-130 and Merlin respectively. The CCN concentrations, predicted based on the size distributions measured on the Pelican and on the chemical composition measured at ground, were systematically larger than the measured CCN on the Pelican. The CCN spectra measured on the Merlin and C-130 might be explained, if the assumption is made that only a small fraction of the aerosol is soluble. These 3 observations are self-consistent, but seem to be in disagreement with the chemical composition on the aerosol in this area, which implies a highly soluble aerosol (Putaud et al., 2000; Neusüß et al., 2000; Quinn et al., 2000). So far, no attempt has been presented to explain the surface based CCN measurements and the vast amount of hygroscopic growth measurements (Swietlicki et al., 2000) in terms of the physical and chemical aerosol properties. Such a study might shed light on the reasons for the mentioned discrepancy.

With respect to *Question 1*, the ACE-2 data show that in the case of clean marine and polluted continental aerosols, as well as clean FT aerosols "spherical particle" Mie theory can be used to calculate the radiative properties from the physical and chemical aerosol properties to within 20% of the measured value. Further tests of the applicability of Mie theory to dust aerosols are still to be performed with the available data. Preliminary analyses of ACE-2 data indicate major uncertainties in relating physical and chemical properties to the number of CCN. These uncertainties, which are to a large extent instrumental, prevent testing the applicability of the Köhler theory to predict CCN.

***Question 2***

*Are there useful empirical correlations between the mass concentration of individual aerosol components and the radiative and cloud nucleating properties of that same aerosol?*

The many conditions encountered during ACE-2, showed that the chemical composition, and the relative contribution of individual chemical compounds varies widely among the different air masses (Neusüß et al., 2000; Putaud et al., 2000; Quinn et al., 2000; Novakov et al., 2000). For example, the ratio of organic carbon to nss sulfate mass ($\mu gC\ m^{-3}/\mu g\ SO_4^{=}\ m^3$) in the submicron fraction varied from about 0.1 in European pollution outflow, to 0.4 in clean marine Atlantic air masses, to 0.5 in Mediterranean air masses, to 0.8 in the background free troposphere. In spite of the large analytical uncertainties in these ratios, the conclusion that they differ widely between air masses is robust. This shows that a globally valid relationship between the mass concentration of

individual aerosol components and other properties of that same aerosol will not exist.

Many of the analyses of ACE-2 data focused on correlations involving aerosol and/or cloud droplet number concentration, as these are fundamental parameters needed to calculate the indirect radiative effect of aerosols. Van Dingenen et al. (1999) analyzed the correlation between the submicron aerosol volume ($V$) and the number of accumulation mode aerosol particles ($N_{ac}$) of the MBL aerosol, which they found to be linear. This linearity implies a constant $N_{ac}/V$ ratio which might be useful to make global assessments of $N_{ac}$, as fields of $V$ are more easily generated in 3-D models and might also be accessible through remote sensing from space (Hegg and Kaufmann, 1998). However, a further analysis by Van Dingenen et al. (2000), including a larger range of conditions, reveals that $N_{ac}/V$ does not remain constant over that range, but a relationship between $N_{ac}$ and $V$ can still be constructed by considering the source terms of $N_{ac}$ and $V$ and the processes of entrainment and coagulation.

Martinsson et al. (2000) observed that the number of cloud droplets ($N_d$) increases linearly with the number of sub-cloud aerosol particles ($D_{p,dry} > 0.042$ μm), up to $N_d = 3000\ cm^{-3}$. Chuang et al. (2000) on the other hand, observed that $N_d$ increased sub-linearly (i.e., slower) with the number of aerosols ($D_{p,dry} > 0.1$ μm), up to $N_d = 300\ cm^{-3}$. The difference is most likely explained by the different vertical updraft speeds, i.e., up to 2 m/s in the hill cap cloud studied by Martinsson et al. (2000) and $< 0.5$ m/s in the marine stratiform cloud (Chuang et al., 2000).

With respect to *Question 2*, the range of conditions encountered in ACE-2 resulted in the establishment of a number of empirical correlations between relevant aerosol parameters. A large variability generally exists around the best-fit correlations, which make them of limited use as accurate parameterisations in global models. However, the explanation of the observed correlations in terms of processes helped identifying the important processes which should be implemented in models to improve the on-line calculation of aerosol number concentration (see below).

***Question 3***

*Can the measured physical and chemical properties of the aerosol in the vertical be used to accurately predict the column-integrated direct effect of aerosols on radiative transfer?*

The column-integrated direct effect of aerosols on radiative transfer has been studied using sunphotometric measurements of the aerosol optical depth (AOD) from ground stations, the ship, aircraft and satellites. Comparisons of these AOD measurements, when platforms were collocated, are discussed, e.g., by Schmid et al. (2000), and Livingston et al. (2000). AOD retrievals using the spaceborne AVHRR compared well with the ground based sunphotometer measurements, except during the presence of dust where the AVHRR sytematically underestimated the AOD by 10% (Durkee et al., 2000).

This special issue further presents attempts to reconstruct the measured AOD from other quantities such as: (1) altitude resolved aerosol backscatter measured by LIDAR (Welton et al., 2000; Flamant et al., 2000; Livingston et al., 2000), (2) aircraft profiles of aerosol scattering and extinction (Schmid et al., 2000) and (3) aircraft profiles of aerosol number size distributions and (assumed) chemical composition (Collins et al., 2000). The degree of success of these reconstructions is discussed by Russell and Heintzenberg (2000). The agreement is generally within 20% in cases where the aerosol remains undisturbed by both measuring techniques (e.g., LIDAR and sunphotometry). It deteriorates in cases where the aerosol is dried and/or its size distribution truncated during sampling and hygroscopic growth calculations and sampling efficiencies are required to reconstruct the ambient aerosol and hence the AOD. E.g., Schmid et al. (2000) mention that to correct for the size distribution truncation in the Pelican sampling inlet (nominally at $D_p$ 2.5 μm), aerosol scattering values of dust aerosols observed with nephelometers inside the aircraft must be corrected by factors 3.9–4.9! Attempts to reconstruct the AOD of the MBL aerosol, based on point measurements and assuming homogeneity of the aerosol throughout the MBL were unsuccessful (Livingston et al., 2000).

Measurements at Sagres including simultaneous sunphotometer, 6-wavelength LIDAR and airborne in-situ physical, chemical and radiative aerosol properties remain unexploited to date.

Several investigators have inverted the multispectral AOD values to derive column (or layer) integrated number size distributions (Schmid et al.,

2000; Vitale et al., 2000; Formenti et al., 2000). Such methods might be useful to construct global size resolved aerosol climatologies using space borne radiometers, and to calculate the outgoing solar radiation flux due to aerosols, as is done by Vitale et al. (2000). Schmid et al. (2000) and Formenti et al. (2000) present validations of these inversion methods. In the few examples shown by Schmid et al., the volume of MBL aerosol within a confined layer, retrieved with an airborne 14 channel sunphotometer, is within 10% of the in-situ measured values. In a dust layer, however, the discrepancy amounts to a factor of 2.7.

With respect to *Question 3*, the initial analysis of ACE-2 data show that there are remaining problems with measuring aerosol from aircraft, due to poorly defined truncation of the aerosol distribution in the sampling inlet and poor counting statistics. This prevents a straightforward answer to the question. Clearly emerging from the ACE-2 dataset is that, apart from the profiles of aerosol physical and chemical properties, the vertical profile of relative humidity is equally important to reconstruct the vertical profile of aerosol extinction, and from there the AOD and the upwelling radiative solar radiation.

***Question 4***

*Can the measured physical and chemical properties of aerosols and clouds in the vertical column be used to accurately predict the integrated indirect effect of aerosols on radiative transfer?*

The integrated indirect effect of aerosols on radiative transfer is measured as the change in cloud optical depth or cloud reflectivity with changing properties of the aerosol on which the cloud forms. The question thus asks for relating aerosol physical and chemical properties with cloud microphysical properties, and the latter with cloud optical depth.

Although the relationship between aerosols and CCN is problematic in the ACE-2 data presented so far (see above), the data relating (1) aerosol number and cloud droplet number concentration, and (2) CCN and cloud droplet concentration, seem to be in better qualitative agreement with the present understanding. This is shown by the studies of Martinsson et al. (2000) and Flynn et al. (2000), which discuss the first relationship, and by the study of Chuang et al. (2000) which discusses both relationships. The 3 studies are able to reproduce the observations with an adiabatic cloud parcel model using reasonable, though not measured input. Chuang et al. (2000) points to the large uncertainty that remains in relating different measures of aerosol and droplet populations. This is related both to instrumental error and an insufficient physico-chemical characterization of the aerosol/cloud system.

Snider and Brenguier (2000) present the relationship of the second sort. In areas of highest updraft velocity they obtain closure between the measured cloud droplet number concentrations and the calculated values based on the measured CCN spectrum and the measured updraft velocity. All studies conclude that the variability in cloud droplet number concentration is most strongly influenced by the variability in vertical velocity at cloud base. Aerosol chemical composition was less of an issue, most likely because the aerosol was found to be very hygroscopic in all cases studied (Swietlicki et al., 2000). An interesting example of the important rôle of vertical velocity is possibly illustrated in the measurements in the second Lagrangian experiment (Osborne et al., 2000) where the number of accumulation mode particles decreases with time, while the droplet number concentration increased together with the standard variation of the vertical wind speed.

Brenguier et al. (2000) observed that cloud droplets grow adiabaticly in a stratiform cloud. Realizing, therefore, that the optical depth of a cloud is much more dependent on its geometrical depth than on the number of cloud droplets, they implemented an experimental strategy that allowed the deconvolution of the 2 effects.

In clouds of similar geometrical thickness, and at scales ranging from a single cloud to a cloud ensemble (50–100 km), they observed increased cloud reflectances with increasing in-situ measured cloud droplet number concentrations. However, using the adiabatic cloud model (rather than the plane-parallel cloud model), the calculated cloud reflectances based on the in-situ measured cloud parameters still remain systematically lower than those observed.

In their comparison of cloud micro-physical observations in clean and polluted clouds, Pawlowska and Brenguier (2000) show the presence and absence of drizzle in clean and polluted clouds respectively. Similar observations of the indirect Albrecht effect results from the second

Lagrangian experiment, where coalescence of cloud droplets was suppressed as the high CCN concentration kept cloud droplets relatively small (Osborne et al., 2000).

ACE-2 provides the data necessary to test the various steps that relate aerosol physical and chemical properties with cloud reflectances. When moving from clean marine to polluted continental air masses, differences in aerosol physical and chemical properties were observed, together with increases in CCN and droplet number concentration, even in clouds that were otherwise macroscopically similar. Simultaneous with these changes, an increase in cloud reflectance was observed. These observations show for the first time the existence of both indirect effects in polluted continental air masses at scales up to a cloud ensemble. The full ACE-2 data set has not yet been used to quantify all the various links. As an answer to *Question 4*, it must therefore be stated that the indirect effects cannot currently be predicted from the changes in the basic aerosol properties.

***Questions 5, 6***

*What are the rates and efficiencies of the processes in the MBL that change the size distribution and size-dependent chemical composition of a continentally derived aerosol as it advects over the North Atlantic?*

Process studies are presented that are based on (1) Lagrangian experiments in which an aircraft follows a defined air mass (Johnson et al., 2000a), (2) semi-Lagrangian experiments in which differences in atmospheric parameters were measured between stations along a air mass trajectory (Bower et al., 2000), and (3) Eulerian observations from ground based stations, analyzing air masses coming from various areas in Europe and N. America, as well as the clean Atlantic and Arctic (Bates et al., 2000; Van Dingenen, 2000).

Three Lagrangian experiments were performed, during which smart balloons (Johnson and Businger, 2000) were released from the ship near the west coast of Portugal, and followed for $\sim$30 h down to the Canary Islands. Three very different situations were encountered: during the first Lagrangian (Johnson et al., 2000b) a clean marine air mass was followed, during the second (Osborne et al., 2000) a relatively freshly polluted air mass in recent contact with the Iberian peninsula was followed, and during the third (Wood et al., 2000) a relatively old air mass advecting off Northern France was followed. In addition to the evolution of trace gas concentrations, aerosols and cloud parameters, the evolution of the boundary layer structure and entrainment rates (Sollazzo et al., 2000) are presented here.

During the first 2 Lagrangian experiments most changes in the size distribution during $\sim$30 h of evolution were observed in the aerosol number concentration of either the Aitken mode, the accumulation mode or both. The observed changes in size of each of the modes were relatively small. A qualitative analysis (Johnson at al., 2000b) and a more quantitative timescale analysis by Hoell et al. (2000) suggest that the increase in accumulation mode number during the first Lagrangian experiment is due to the production of sea-salt particles at elevated wind speeds. The reduction of the Aitken mode is ascribed to collision of those particles with cloud droplets and with accumulation mode particles. Dilution by entrainment, in particular when the height of the MBL raises as the air mass moves south over a warmer ocean, is the main reason for a general reduction in the aerosol concentration. A rapid decrease of the $SO_2$ concentration, and the development of a greater separation between the Aitken and accumulation mode was observed during the second Lagrangian, and is attributed to aqueous phase $SO_2$ oxidation (Dore et al., 2000).

During the third Lagrangian experiment the aerosol size distribution showed no measurable evolution. This is explained by the fact that the MBL developed within a pollution layer, and for this reason entrainment across the MBL inversion did not result in a dilution of the MBL aerosol (Wood et al., 2000). The constancy of the aerosol further points out that coagulation, condensation and cloud processing rates are ineffective over a time period of $\sim$30 h, in an aged air mass in which the aerosol number concentration and gaseous precursor concentration are low. The insignificant contribution of in-cloud oxidation of $SO_2$ in the polluted air mass, after it traveled for $>$2 days over the ocean, was confirmed in the hill cap cloud experiments (Flynn et al., 2000).

The analysis by Van Dingenen et al. (2000) of the aerosol size distributions taken continuously at Punta Del Hidalgo and on the ship, suggests that entrainment and a general coagulation

process (which includes collisions between aerosol particles and cloud droplets) explains most of the development of the aerosol size distribution. This conclusion is in broad agreement with the conclusions of the Lagrangian studies.

***Question 7***

*How much do aerosols formed or transported in the free troposphere contribute to the direct radiative forcing over the North Atlantic and how do they affect the properties of aerosols and clouds in the MBL.*

The question of aerosol transport is mainly focused on the role of North African dust transported over the (sub-)tropical North Atlantic. During ACE-2, these dust plumes were observed by various space-borne sensors, including AVHRR, Meteosat, POLDER and TOMS. The retrieval of their optical depth from AVHRR has been tested against ground based and airborne sunphotometry (Durkee et al., 2000; Schmid et al., 2000), showing the need to account for the absorption properties of the dust single scattering albedo in the retrieval algorithm. In-situ analysis throughout the dust layers was attempted by some Pelican flights. Öström and Noone (2000) present single scattering albedo's for dust aerosol with diameter $<2.5$ μm, which is the nominal cut-size of Pelican aerosol sampling inlet. These authors point at the large variability ($0.73 \pm 0.12$) that exists within a single dust layer, and between different dust layers. Their values can not be applied to the total dust without introducing large errors (see above). Because of these variabilities and uncertainties an accurate calculation of the radiative effects of dust layers from its macroscopic and basic aerosol properties is not currently possible.

Layers of continental pollution aerosols were also observed within the FT. Based on the data of the Lagrangian experiments, Johnson et al. (2000a) developed a general scenario on how these layers might develop, as the deep (2 to 5 km) convectively driven continental aerosol layer advects over the colder ocean. At the beginning of the second Lagrangian experiment it was observed how the pollution aerosol above the subsidence inversion became entrained and polluted the MBL and lead to an increase in the cloud droplet number of the MBL cloud (Osborne et al., 2000).

Profiles of gaseous and aerosol parameters, up to 6 km from the C-130, and up to 12 km from the Citation have not been analyzed to date.

Aerosol nucleation was not observed in any of the Lagrangian experiments, and was unimportant during 6 weeks of measurements at Punta del Hidalgo and on the ship. There is also no evidence of regular nucleation in the sub-tropical free troposphere up to 12 km above Tenerife (Johan Ström, personal communication). The combined Punta del Hidalgo and Izaña measurements, which include time scales of 5–10 days, support the idea that a steady state between entrainment of FT aerosol, aqueous phase chemistry and precipitation largely explains the size distribution of the clean MBL aerosol (Raes et al., 1997; Van Dingenen et al., 1999).

## 4. Data archiving

The data from the various platforms and activities have been collected and archived in a central data archive at the JRC Ispra. This databank is accessible through the Web (http://ace2.ei.jrc.it) and allows a user to browse, search, compare, visualize and download the available datasets. Each dataset is described by a number of attributes, including the responsible persons and organization, a short and long description, and a number of keywords.

In order to allow uniform data access to and visualization of datasets, a simple ASCII based ACE-2 format has been defined. The format is such that datafiles downloaded by the user are easily read locally by a spreadsheet or other scientific visualization program.

Initial Web access to the system is through a browse and search interface to the catalogue of datasets (around 250), from which different datasets can be selected for further inspection of the various parameters. These parameters can be visualized and compared graphically in a variety of ways (multi parameter time plots, scatter plots, heat maps). One innovative aspect is that parameters which have been sampled with different time resolution (e.g., from different instruments on different platforms) are directly comparable through time- or scatter plot, without the off-line laborious tasks of re-arranging time and data values.

The system is accessible to the public, and a CD-ROM version will be released. The latter will also hold a Java based version of the archive system, allowing the user to use identical off-line browse, search and visualization tools as the on-line version. Further information and request for access can be obtained from http://ace2.ei.jrc.it or aceadmin.dm@jrc.it.

## 5. Conclusion

ACE-2 successfully gathered an extensive data set of aerosol properties and implemented these measurements in experimental strategies that provide a comprehensive view of the complex gas/aerosol/cloud system. This resulted in a qualitative, and in many cases a quantitative understanding of this system in climatologically important environments such as the background and perturbed sub-tropical MBL and FT. A close connection was observed between meteorological factors (such as horizontal and vertical wind speed, boundary layer development, entrainment, humidity fields) and aerosol and cloud characteristics. In the ACE-2 region, these meteorological factors, rather than aerosol microphysics and chemistry, often dominated the shaping of the aerosol size distribution and/or their effect on radiation and clouds. With this understanding the community is better prepared to tackle the challenging task of modelling the complete system, and to judge whether present and future models reproduce observations for the right reasons. Several modelling studies applied to ACE-2 data are in preparation.

ACE-2 made progress in quantifying uncertainties in many of the measurements, finding that in some areas, they remain too large to make firm quantitative assessments. Closure experiments showed that this is the case with something so fundamental as determining the chemical composition of the aerosol, or counting the number of cloud droplets from an aircraft. The mentioned closure experiments are straightforward as they imply simple arithmetic operations. In the case of more complex closure experiments involving, e.g., Mie theory, Köhler theory or process models, the comparison between measurements and model output frequently involves the tuning of some parameter in the model. Much remains to be done in improving measuring techniques and in applying them in experiments which fully constrain the model involved.

## 6. Acknowledgements

ACE-2 is a contribution to the International Global Atmospheric Chemistry (IGAC) Core Project of the International Geosphere-Biosphere Programme (IGBP), and is part of the IGAC Aerosol Characterization Experiments (ACE). ACE-2 was funded primarily by the European Commission, DGXII, Environment and Climate Programme (4th FWP), with significant contributions from the National Science Foundation (US), the National Environment Research Council (UK), the National Oceanographic and Atmospheric Administration (US), Meteorological Research Flight (UK), Meteo France (F), the Office of Naval Research (US), TSI Incorporated (US), and the EC Joint Research Centre. Grant numbers are given in the individual papers. Thanks to all who were involved, directly or indirectly. These were 5 years of pleasant and useful collaboration!

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