

Preface

The Southern Hemisphere marine Aerosol Characterization Experiment (ACE 1) was the first of a series of experiments organized through the International Global Atmospheric Chemistry (IGAC) Program designed to quantify the chemical and physical processes controlling the evolution and properties of the atmospheric aerosol relevant to radiative forcing and climate. ACE 1, which was conducted from November 15 to December 14, 1995, over the southwest Pacific Ocean, south of Australia, quantified the chemical, physical, radiative, and cloud nucleating properties and controlling processes of the aerosol in this minimally polluted marine atmosphere. The experiment involved the efforts of scientists from 45 research institutes in 11 countries. Measurements were made from the National Center for Atmospheric Research (NCAR) C-130 aircraft, the National Oceanic and Atmospheric Administration (NOAA) Research Vessel *Discoverer*, the Australian Fisheries Research Vessel *Southern Surveyor*, and ground stations at Macquarie Island and Cape Grim, Tasmania.

The first collection of 33 ACE 1 papers appeared in the *Journal of Geophysical Research*, 103(D13), 1998. This second collection of 12 papers further expands our understanding of marine atmospheric chemistry and the atmospheric aerosol in this remote part of the planet. *Griffiths et al.* [this issue] describe the characteristics of the four different water masses in the ACE 1 study area and how the biological, physical, and chemical properties of these water masses affected the atmosphere. Biologically, the ocean was a source of dimethylsulfide (DMS) [*Bates et al.*, 1998b; *Curran et al.*, 1998; *Jones et al.*, 1998], hydrocarbons [*Pszenny et al.*, this issue], methyl nitrate [*Blake et al.*, this issue], and methyl halides [*Moore et al.*, 1996; *Murphy et al.*, 1997; *Groszko and Moore*, 1998; *Blake et al.*, this issue] to the atmosphere. The ocean may also have been a source of biogenic calcium carbonate which enhanced the alkalinity of sea-salt aerosol water and in turn enhanced ozone oxidation of sulfur dioxide in sea-salt aerosol water [*Gurciullo et al.*, this issue; *Sievering et al.*, this issue]. Physically, the different water masses affected the structure [*Wang et al.*, this issue (a)] and turbulence [*Wang et al.*, this issue (b)] of the atmospheric boundary layer. Physically generated sea-salt particles dominated the mass of both submicron and supermicron marine boundary layer (MBL) aerosol particles in this region during ACE 1 [*Quinn et al.*, 1998; *Huebert et al.*, 1998]. Single particle analysis revealed that over

90% of the aerosol particles with diameters >130 nm [*Murphy et al.*, 1998] and up to 70% of the particles with diameters >80 nm [*Kreidenweis et al.*, 1998] contained sea salt. Organic species were detected in over 50% of the particles with diameters >160 nm and were associated with sea salt [*Middlebrook et al.*, 1998]. The dominance of sea-salt aerosol in this region clearly shows the need to include sea salt in climate models. In the ACE 1 study area, sea salt controlled the magnitude of aerosol light scattering [*Quinn et al.*, 1998; *Carrico et al.*, 1998; *Murphy et al.*, 1998] and the number of cloud condensation nuclei [*Covert et al.*, 1998].

Although the intent of ACE 1 was to study the background marine atmosphere, gas and aerosol measurements showed that even this remote region is impacted by anthropogenic activities. Layers containing continental air characteristic of aged biomass burning emissions were observed above 3 km over the remote southern Ocean [*Blake et al.*, this issue]. Transmission electron microscopy of samples throughout the troposphere in the ACE 1 study area showed that between 11 and 46% of the sulfate particles with diameters >100 nm contained soot [*Posfai et al.*, this issue]. The source of this soot was most likely South African biomass burning and aircraft emissions [*Posfai et al.*, this issue].

A major emphasis of the IGAC Aerosol Characterization Experiments is to identify and quantify processes controlling aerosol distributions. Measurements in the vicinity of Macquarie Island during ACE 1 provided a unique opportunity to study, in situ, the role of biogenic species on the formation and growth of newly formed particles [*Weber et al.*, 1998]. The photochemical production of new particles from sulfuric acid gas also was clearly observed in cloud outflows [*Clarke et al.*, 1998]. The growth and evolution of these particles aloft was evident over periods of hours to a day [*Clarke et al.*, 1998]. In regions of post-frontal subsidence, these ultra-fine and Aitken mode particles were mixed into the MBL [*Bates et al.*, 1998a, b; *Brechtel et al.*, 1998].

The evolution of particles in the MBL was studied using both Eulerian and Lagrangian strategies. Diurnal cycles and photochemical box model calculations were used to show that 30–50% of the DMS was converted to SO₂ [*De Bruyn et al.*, 1998]. A major sink (approximately 35%) for this SO₂ appeared to be through ozone oxidation in sea-salt aerosol water [*Sievering et al.*, this issue]. In summer, 35 ± 10% of the non-sea-salt (nss) sulfate was associated with supermicron sea salt, while in winter this fraction increased to 58 ± 22% [*Andreae et al.*, this issue].

The Lagrangian measurement strategy continued to evolve during ACE 1 [*Bates et al.*, 1998a; *Huebert and Lenschow*, 1999]. The balloons tracking the air masses were improved with the capability to adjust their own buoyancy in response to drizzle [*Businger et al.*, 1999], and new techniques were implemented to measure en-

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Paper number 1999JD900365.

trainment velocities [Russell et al., 1998; Lenschow et al., 1999; Wang et al., this issue (b)]. Measurements of OH [Mauldin et al., 1998], CO [Kok et al., 1998], and hydrocarbons [Pszenny et al., this issue; Wingen-ter et al., this issue] were used to constrain the oxidative capacity of atmosphere. Repeated measurements of aerosols and gas phase precursors over a 30-hour period made it possible to follow the evolution of gas phase photochemistry [Mari et al., 1998; Suhre et al., 1998] and the formation of methane sulfonate and nss sulfate [Huebert et al., 1998; Mari et al., this issue]. The repeated measurements combined with a one-dimensional (1-D) model provided sufficient constraints to better quantify the processes controlling the sulfur budget in this area [Mari et al., this issue].

ACE 1 (<http://saga.pmel.noaa.gov/ace1.html>) was the first in a series of IGAC aerosol experiments. The data are available in a central archive (<http://www.joss.ucar.edu>) and on CD (contact bates@pmel.noaa.gov). The understanding gained in this experiment is being used to study progressively more complex environments. The Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX) focused on clear-column closure experiments off the East Coast of the United States in July 1996 [Russell et al., 1999]. ACE 2 extended the process and closure studies to the eastern North Atlantic Ocean in June/July 1997 and focused on the anthropogenic aerosols from the European continent and mineral dust from the African continent (<http://rea.ei.jrc.it/~vandinge/ace2/ace2main.html>). Planning is now underway for the next ACE that will focus on the region downwind of the rapidly increasing pollution sources in eastern Asia (<http://saga.pmel.noaa.gov/aceasia/>). We hope the papers presented in this special section will serve as a further stimulus to join both in the process of interpreting the unique ACE 1 data set and in future ACE.

Acknowledgments. This research was funded by the Atmospheric Chemistry Program of the U.S. National Science Foundation (NSF), the Office of Global Programs and the Office of Oceanic and Atmospheric Research of the United States National Oceanic and Atmospheric Administration (NOAA), the U.S. National Aeronautical and Space Administration (NASA), the Australian Commonwealth Scientific and Industrial Research Organization (CSIRO), the Australian Bureau of Meteorology (BoM), the Antarctic Science Advisory Committee Project 829 of the Australian Antarctic Division (ANARE), and the New Zealand National Institute for Water and Atmospheric Research (NIWA). This research 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). This is NOAA/PMEL contribution 2102.

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References

- Andreae, M. O., W. Elbert, Y. Cai, T. W. Andreae, and J. L. Gras, Non-sea-salt sulfate, methanesulfonate, and nitrate aerosol concentrations and size distributions at Cape Grim, Tasmania, *J. Geophys. Res.*, this issue.
- Bates, T. S., B. J. Huebert, J. L. Gras, F. B. Griffiths, and P. A. Durkee, International Global Atmospheric Chemistry (IGAC) Project's First Aerosol Characterization Experiment (ACE 1): Overview, *J. Geophys. Res.*, *103*, 16,297–16,318, 1998a.
- Bates, T. S., V. N. Kapustin, P. K. Quinn, D. S. Covert, D. J. Coffman, C. Mari, P. A. Durkee, W. De Bruyn, and E. Saltzman, Processes controlling the distribution of aerosol particles in the lower marine boundary layer during the First Aerosol Characterization Experiment (ACE 1), *J. Geophys. Res.*, *103*, 16,369–16,383, 1998b.
- Blake, N. J., et al., Aircraft measurements of the latitudinal, vertical, and seasonal variations of NMHCs, methyl-nitrate, methyl halides, and DMS during ACE 1, *J. Geophys. Res.*, this issue.
- Brechtel, F. J., S. M. Kreidenweis, and H. B. Swan, Air mass characteristics, aerosol particle number concentration, and number size distributions at Macquarie Island during the First Aerosol Characterization Experiment (ACE 1), *J. Geophys. Res.*, *103*, 16,351–16,367, 1998.
- Businger, S., R. Johnson, J. Katzfey, S. Siems, and Q. Wang, Smart tetroons for Lagrangian air mass tracking during ACE 1, *J. Geophys. Res.*, *104*, 11,709–11,722, 1999.
- Carrico, C. M., M. J. Rood, and J. A. Ogren, Aerosol light scattering properties at Cape Grim, Tasmania, during the First Aerosol Characterization Experiment (ACE 1), *J. Geophys. Res.*, *103*, 16,565–16,574, 1998.
- Clarke, A. D., J. L. Varner, F. Eisele, R. L. Mauldin, D. Tanner, and M. Litchy, Particle production in the remote marine atmosphere: Cloud outflow and subsidence during ACE 1, *J. Geophys. Res.*, *103*, 16,397–16,409, 1998.
- Covert, D. S., J. L. Gras, A. Wiedensohler, and F. Stratmann, Comparison of directly measured CCN with CCN modeled from the number-size distribution in the marine boundary layer during ACE 1 at Cape Grim, Tasmania, *J. Geophys. Res.*, *103*, 16,597–16,608, 1998.
- Curran, M. A. J., G. B. Jones, and H. Burton, Spatial distribution of dimethylsulfide and dimethylsulfoniopropionate in the Australasian sector of the Southern Ocean. *J. Geophys. Res.*, *103*, 16,677–16,689, 1998.
- De Bruyn, W. J., T. S. Bates, J. M. Cainey, and E. S. Saltzman, Shipboard measurements of dimethyl sulfide and SO₂ southwest of Tasmania during the First Aerosol Characterization Experiment (ACE 1), *J. Geophys. Res.*, *103*, 16,703–16,711, 1998.
- Griffiths, F. B., T. S. Bates, P. K. Quinn, L. A. Clementson, and J. S. Parslow, The oceanographic context of the First Aerosol Characterization Experiment (ACE 1): A physical, chemical, and biological overview, *J. Geophys. Res.*, this issue.
- Groszko, W., and R. M. Moore, Ocean-atmosphere exchange of methyl bromide: NW Atlantic and Pacific Ocean studies, *J. Geophys. Res.*, *103*, 16,737–16,741, 1998.
- Gurciullo, C., B. Lerner, H. Sievering, and S. N. Pandis, Heterogeneous sulfate production in the remote marine environment: Cloud processing and sea-salt particle contributions, *J. Geophys. Res.*, this issue.
- Huebert, B. J., and D. H. Lenschow, What have Lagrangian experiments accomplished?, *IGACTiv. Newsl.*, *17*, 2–3, May 1999.
- Huebert, B. J., S. G. Howell, L. Zhuang, J. A. Heath, M. R. Litchy, D. J. Wylie, J. L. Kreidler-Moss, S. Coppicus, and J. E. Pfeiffer, Filter and impactor measurements of anions

- and cations during the First Aerosol Characterization Experiment (ACE 1), *J. Geophys. Res.*, *103*, 16,493–16,509, 1998.
- Jones, G. B., M. A. J. Curran, H. B. Swan, R. M. Greene, F. B. Griffiths and L. A. Clementson, Influence of different water masses and biological activity on dimethylsulphide and dimethylsulphoniopropionate in the subantarctic zone of the Southern Ocean during ACE 1, *J. Geophys. Res.*, *103*, 16,691–16,701, 1998.
- Kok, G. L., A. S. H. Prévôt, R. D. Schillawski, and J. E. Johnson, Carbon monoxide measurements from 76°N to 59°S and over the South Tasman Sea, *J. Geophys. Res.*, *103*, 16,731–16,736, 1998.
- Kreidenweis, S., L. M. McInnes, and F. J. Brechtel, Observations of aerosol volatility and elemental composition at Macquarie Island during the First Aerosol Characterization Experiment (ACE 1), *J. Geophys. Res.*, *103*, 16,511–16,524, 1998.
- Lenschow, D. H., P. B. Krummel, and S. T. Siems, Measuring entrainment, divergence and vorticity on the mesoscale from an aircraft, *J. Atmos. Oceanic Technol.*, in press, 1999.
- Mari, C., K. Suhre, T. S. Bates, J. E. Johnson, R. Rosset, A. R. Bandy, F. L. Eisele, R. L. Mauldin III, and D. C. Thornton, Physicochemical modeling of the First Aerosol Characterization Experiment (ACE 1) Lagrangian B, 2, DMS emission, transport, and oxidation at the mesoscale, *J. Geophys. Res.*, *103*, 16,457–16,473, 1998.
- Mari, C., K. Suhre, R. Rosset, T. S. Bates, B. J. Huebert, A. R. Bandy, D. C. Thornton, and S. Businger, One-dimensional modeling of sulfur species during the First Aerosol Characterization Experiment (ACE 1) Lagrangian B, *J. Geophys. Res.*, this issue.
- Mauldin, R. L., III, G. J. Frost, G. Chen, D. J. Tanner, A. S. H. Prévôt, D. D. Davis, and F. L. Eisele, OH measurements during the First Aerosol Characterization Experiment (ACE 1): Observations and model comparisons, *J. Geophys. Res.*, *103*, 16,713–16,729, 1998.
- Middlebrook, A. M., D. M. Murphy, and D. S. Thomson, Observations of organic material in individual marine particles at Cape Grim during the First Aerosol Characterization Experiment (ACE 1), *J. Geophys. Res.*, *103*, 16,475–16,483, 1998.
- Moore, R. M., W. Groszko, and S. J. Niven, Ocean-atmosphere exchange of methyl chloride: Results from the NW Atlantic and Pacific Ocean studies, *J. Geophys. Res.*, *101*, 28,529–28,538, 1996.
- Murphy, D. M., D. S. Thomson, and A. M. Middlebrook, Bromine, iodine, and chlorine in single aerosol particles at Cape Grim, *Geophys. Res. Lett.*, *24*, 3197–3200, 1997.
- Murphy, D. M., J. R. Anderson, P. K. Quinn, L. M. McInnes, F. J. Brechtel, S. M. Kreidenweis, A. M. Middlebrook, M. Posfai, and D. S. Thomson, Submicron sea salt particles and aerosol radiative properties in the remote Southern Ocean marine boundary layer, *Nature*, *392*, 62–65, 1998.
- Posfai, M., J. R. Anderson, P. R. Buseck, and H. Sievering, Soot and sulfate aerosol particles in the remote marine troposphere, *J. Geophys. Res.*, this issue.
- Pszenny, A. A. P., R. G. Prinn, G. L. Kleiman, X. Shi, and T. S. Bates, Nonmethane hydrocarbons in surface waters, their sea-air fluxes, and impact on OH in the marine boundary layer during ACE 1, *J. Geophys. Res.*, this issue.
- Quinn, P. K., D. J. Coffman, V. N. Kapustin, T. S. Bates, and D. S. Covert, Aerosol optical properties in the marine boundary layer during the First Aerosol Characterization Experiment (ACE 1) and the underlying chemical and physical aerosol properties, *J. Geophys. Res.*, *103*, 16,547–16,563, 1998.
- Russell, L. M., D. H. Lenschow, K. K. Laursen, P. B. Krummel, S. T. Siems, A. R. Bandy, D. C. Thornton, and T. S. Bates, Bidirectional mixing in an ACE 1 marine boundary layer overlain by a second turbulent layer, *J. Geophys. Res.*, *103*, 16,411–16,432, 1998.
- Russell, P. B., P. V. Hobbs, and L. L. Stowe, Aerosol properties and radiative effects in the United States East Coast haze plume: An overview of the Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX), *J. Geophys. Res.*, *104*, 2213–2222, 1999.
- Sievering, H., B. Lerner, J. Slavich, J. Anderson, and J. Cainey, O₃ oxidation of SO₂ in sea-salt aerosol water: The size distribution of non-sea-salt sulfate during ACE 1, *J. Geophys. Res.*, this issue.
- Suhre, K., et al., Physicochemical modeling of the First Aerosol Characterization Experiment (ACE 1) Lagrangian B, 1, A moving column approach, *J. Geophys. Res.*, *103*, 16,433–16,455, 1998.
- Wang, Q., et al., Characteristics of the marine boundary layers during two Lagrangian measurement periods, 1, General conditions and mean characteristics, *J. Geophys. Res.*, this issue (a).
- Wang, Q., et al., Characteristics of the marine boundary layers during two Lagrangian measurement periods, 2, Turbulence structure, *J. Geophys. Res.*, this issue (b).
- Weber, R. J., P. H. McMurry, L. Mauldin, D. J. Tanner, F. L. Eisele, F. J. Brechtel, S. M. Kreidenweis, G. L. Kok, R. D. Schillawski, and D. Baumgardner, A study of new particle formation and growth involving biogenic and trace gas species measured during ACE 1, *J. Geophys. Res.*, *103*, 16,385–16,396, 1998.
- Wingenter, O. W., D. R. Blake, N. J. Blake, B. C. Sive, E. Atlas, F. Flockner, and F. S. Rowland, Tropospheric hydroxyl and atomic chlorine concentrations, and mixing timescales determined from hydrocarbon and halocarbon measurements made over the Southern Oceans, *J. Geophys. Res.*, this issue.