

DOE RESEARCH ON ATMOSPHERIC AEROSOLS



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The Department of Energy has a statutory obligation "to assure incorporation of national environmental protection goals in the formulation and implementation of energy programs and to advance the goal of restoring, protecting, and enhancing environmental quality and assuring public health and safety" and to conduct a "comprehensive program of research and development on the environmental effects of energy technology." In fulfillment of these obligations the Department supports environmental research through grants and contracts administered within its Environmental Processes and Effects Research Program. Key components of that Program are an programs dealing with atmospheric science, subsurface science, environmental radon, ocean margins, and ecosystem research.

Atmospheric aerosols are the subject of a significant component of research within DOE's environmental research activities, mainly under two programs within the Department's Environmental Sciences Division, the Atmospheric Radiation Measurement (ARM) Program and the Atmospheric Chemistry Program (ACP). Research activities conducted under these programs include laboratory experiments, field measurements, and theoretical and modeling studies. The objectives and scope of these programs are briefly summarized here:

ARM--Atmospheric Radiation Measurement Program. The ARM Program is the Department's major research activity focusing on atmospheric processes pertinent to understanding global climate and developing the capability of predicting global climate change in response to energy related activities. The objectives of this program are:

- 1) To relate observed radiative fluxes and radiances in the atmosphere, spectrally resolved and as a function of position and time, to the temperature and composition of the atmosphere, specifically including water vapor and clouds, and to surface properties, and sample sufficient variety of situations so as to span the range of climatologically relevant possibilities; and
- 2) To develop and test parameterizations that can be used to accurately predict the radiative properties and to model the radiative interactions involving water vapor and clouds within the atmosphere, with the objective of incorporating these parameterizations into general circulation models (GCMs) and other models capable of describing and predicting climate change.

The ARM approach consists mainly of testing and improving models using long-term measurements of atmospheric radiation and controlling variables at highly instrumented sites in north central Oklahoma, in the Tropical Western Pacific, and on the North Slope of Alaska. Although characterization of atmospheric aerosols or their direct and indirect radiative influence are not specific objectives of ARM, the role of aerosols in influencing atmospheric radiation has been broadly recognized in formulating the program, and several research projects within the ARM program are addressing these influences, in some instances as a major component of their activity. It must be conceded, however, that atmospheric aerosols are a relatively minor focus of the overall ARM program.

ARM activities dealing with atmospheric aerosols include:

- Measurement of aerosol properties at the ARM site in Oklahoma, modeling the radiative effects of these aerosols, and comparison of modeled and measured radiation.

- Description of aerosol loadings and properties in subhemispheric to global scale chemical models and model evaluation by comparison with in-situ and remote sensing.
- Examination of aerosol climatic influences in studies with GCMs and in examination of spatial and temporal patterns of change in climatic records vis-a-vis aerosol loadings.

Further information on the ARM program is presented in ARM (1995) and Stokes and Schwartz (1994).

ACP--Atmospheric Chemistry Program. Atmospheric chemistry research within DOE addresses primarily the issue of atmospheric response to emissions from energy-generation sources. As such this program deals with the broad topic known commonly as the atmospheric source-receptor sequence. This sequence consists of all aspects of energy-related pollutants from the time they are emitted from their sources to the time they are redeposited at the Earth's surface. The objective of this program is:

To quantify the physical, chemical, geological, and biological processes governing transport, dispersion, and transformation of energy-related materials in the atmosphere, with emphasis on continental and oceanic fates of energy-related air pollutants.

Atmospheric processes governing source-receptor relations include air-surface exchange, heterogeneous and homogeneous reactions, and precipitation scavenging. A substantial component of the ACP research activity is directed to atmospheric aerosols. Specific aerosol issues include:

- Attainment/non-attainment of standards for PM-10 (particulate matter having aerodynamic diameter less than 10 μm).
- Transformation and deposition processes and their influences on pollutant lifetimes and fates.
- Direct aerosol influence on the atmospheric radiation balance.
- Effects of aerosols and aerosol precursors on cloud formation and properties.

More detail on the Atmospheric Chemistry Program and on projects being conducted within this program is given in DOE (1995).

The accompanying material briefly reports on some 25 research projects dealing with atmospheric aerosols that are conducted under the ARM or ACP Programs, outlining their objectives and key findings, and providing an e-mail address as a contact for further information on each project.

References

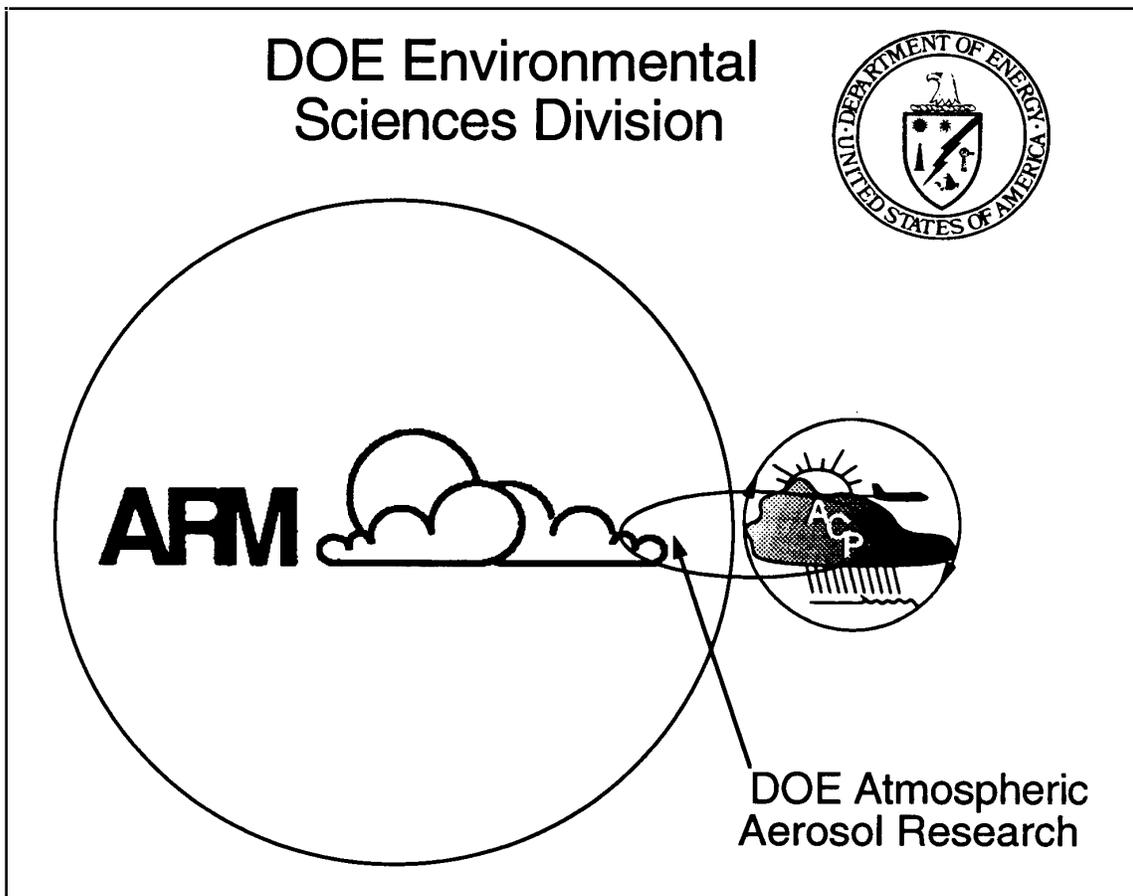
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KEY DOE PROGRAMS RESPONSIBLE FOR ATMOSPHERIC AEROSOL RESEARCH

- Atmospheric Radiation Measurement (ARM).
- Atmospheric Chemistry Program (ACP).



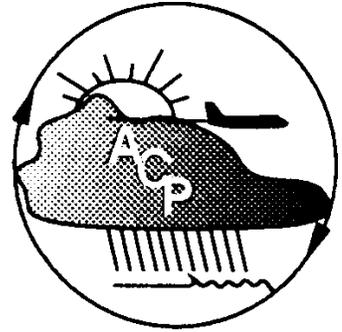


ARM OBJECTIVES

- Relate observed radiative fluxes and radiances in the atmosphere, spectrally resolved and as a function of position and time, to the temperature and composition of the atmosphere, specifically including water vapor and clouds, and to surface properties, and sample sufficient variety of situations so as to span the range of climatologically relevant possibilities.
- Develop and test parameterizations that can be used to accurately predict the radiative properties and to model the radiative interactions involving water vapor and clouds within the atmosphere, with the objective of incorporating these parameterizations into general circulation models.

APPROACH

- ARM approach consists mainly of testing and improving models using long-term measurements of atmospheric radiation and controlling variables at highly instrumented sites in Oklahoma, the Tropical Western Pacific, and the North Slope of Alaska.



ACP OBJECTIVES

- Quantify the physical, chemical, geological, and biological processes governing transport, dispersion, and transformation of energy-related materials in the atmosphere, with emphasis on continental and oceanic fates of energy-related air pollutants.
- Atmospheric processes governing source-receptor relations include air-surface exchange, heterogeneous and homogeneous reactions, and precipitation scavenging.
- Specific aerosol issues include:
 - Attainment/non-attainment of PM-10 standards.
 - Transformation and deposition processes and their influences on pollutant lifetimes and fates.
 - Direct aerosol influence on the atmospheric radiation balance.
 - Effects of aerosols and aerosol precursors on cloud formation and properties.

AEROSOL CHARACTERIZATION AT THE ARM SGP SITE IN LAMONT, OK

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- Develop instrumentation for continuous measurement of the physical properties of aerosols at a height of 10 m at SGP site of ARM.
- Measured properties include aerosol size distribution (optical particle counter), number concentration (CN), scattering coefficient (nephelometer), and absorption coefficient (filter transmission).
- Develop instrumentation for measurement of the RH dependence of the aerosol scattering coefficient.
- The above measurements are in support of radiative transfer calculations.

MFRSR TOTAL COLUMN AEROSOL MEASUREMENTS AT FIVE WAVELENGTHS

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- Develop and test compact, rugged, automated instrument for measuring downwelling direct and diffuse shortwave radiation at seven selected wavelength bands: MultiFilter Rotating Shadowband Radiometer (MFRSR).
- Calculate column aerosol optical depth using spectral irradiance data from continuous measurements made by the MFRSR. Daily averages are available for about 200 days per year.
- Measurements are clearly influenced by Mt. Pinatubo aerosols through the summer of 1994.
- Wavelength information can be inverted for estimate of size distribution. Usually the atmospheric aerosol is well described by a power law (i.e. "Junge distribution").
- Occasionally (most particularly for well aged Pinatubo aerosol seen from very clean sites such as MLO) one can infer significant contribution from large particles, but without measurements at wavelengths to several μm only the total cross-section can be estimated, not the mean size.

SOLAR IRRADIANCE AND AEROSOL OPTICAL DEPTH MEASUREMENTS IN THE QUANTITATIVE LINKS NETWORK

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- Nine-station radiometric network was operated in the northeastern US from late 1991 through 1995 to measure solar irradiance components at 6 narrowband wavelengths.
- Measurements of the direct beam component and the direct-to-diffuse irradiance provide a means for estimating aerosol optical depth, cloud optical depths, surface albedo, and an aerosol size parameter.
- Data can be used to investigate how clouds and aerosols interact with long and shortwave radiation to regulate the heating of the planet.
- Results at each site show the effect of a decaying Mt. Pinatubo stratospheric perturbation superimposed on a seasonal boundary layer aerosol loading pattern.

SCANNING RAMAN LIDAR MEASUREMENTS OF ATMOSPHERIC WATER VAPOR AND AEROSOLS

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- A scanning Raman, Rayleigh/Mie lidar system is used to simultaneously measure atmospheric aerosols and water vapor.
- Both aerosol extinction and backscattering are directly and simultaneously measured so that the aerosol backscatter phase function can be derived.
- Lidar scanning ability permits aerosol and water vapor measurements to be made adjacent to coincident tower-based instrumentation.
- Lidar data are being used in conjunction with sun photometer aerosol optical thickness and sky radiance data to study relationships between aerosol optical and physical properties and water vapor.

AEROSOL DIRECT SHORTWAVE RADIATIVE FORCING AT THE OKLAHOMA ARM SITE

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- Examination of the dependence of direct radiative forcing by sulfate aerosols on particle size, composition, relative humidity and solar zenith angle.
 - Normalized forcing, $W\ m^{-2}$ per $g(SO_4^{2-})\ m^{-2}$, or $W/g(SO_4^{2-})$, is highly sensitive to particle size and RH but is relatively insensitive to sulfate species.
 - Zenith angle dependence shows maximum at high angle, $\sim 75^\circ$.
- Closure experiments will relate vertical distribution of aerosol and measured aerosol microphysical properties to radiative forcing at the ARM site.

CLOUD PARAMETERIZATION IN GLOBAL CLIMATE MODELS: THE ROLE OF AEROSOLS

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- Develop parameterization for the effect of aerosols on cloud droplet size distribution.
- Use ARM data to test method of parameterization.
- Initial results were developed using NCAR CCM1 model; the method is being implemented in the ECHAM climate model.
- Estimated forcing by anthropogenic sulfate aerosols is highly dependent on the relative importance of aqueous and homogeneous transformation of SO₂ to sulfate as well as on the background aerosol levels.

DEVELOPMENT OF A STRATIFORM CLOUD PARAMETERIZATION FOR GCMs

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- CSU RAMS cloud microphysics has been incorporated in NCAR CCM2.
- Droplet number is introduced as a prognostic variable.
- Aerosol activation is parameterized in terms of vertical velocity, aerosol concentration, composition, and size distribution.
- Subgrid variability of activation is treated by integrating over PDF of vertical velocity.
- Realistic droplet number concentrations are simulated given prescribed aerosol number concentrations.

GLOBAL AEROSOL MODEL EVALUATION

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- Objective: to perform a comprehensive evaluation of a detailed global tropospheric aerosol model using satellite and surface-based aerosol optical depth data, and surface concentration and deposition data.
- Model consists of PNL version of CCM2 and PNL global aerosol/chemistry model linked in core.
- CCM2 will use simple 4-dimensional data assimilation (nudging) to reproduce observed meteorology. CCM2 has a detailed cloud microphysics parameterization.
- Aerosol/chemistry model treats aerosol as a set of log-normally distributed modes including sulfate, dust, sea salt, and (eventually) smoke.
- Project focuses on model evaluation, using AVHRR satellite optical depth data over oceans, surface-based optical depth data (limited spatial coverage), and surface concentration and deposition data.

HEMISPHERIC SCALE CHEMICAL AND MICROPHYSICAL AEROSOL MODEL

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- Develop and exercise model for mass loading and size distribution of sulfate and other aerosol species.
- Model is driven by observation-derived meteorology permitting comparison with observed aerosol loadings at specific dates and locations.
- Model accurately reproduces temporal and geographical distribution of sulfate loadings over North Atlantic and adjacent continents.
- Aerosol microphysics will be represented by the moments of the size distribution, whose evolution can be represented similarly to that of chemical species.

ALBEDO PERTURBATIONS BY ANTHROPOGENIC AEROSOLS

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- Evaluation of direct and indirect (cloud) forcing by anthropogenic aerosols as a function of aerosol loading.
 - Identification of sources of uncertainties in these forcings.
 - Estimation of the magnitudes of these uncertainties.
- Examination of fractional particle activation in clouds.
 - Activation fraction decreases with increasing accumulation mode particle concentration $> 600 \text{ cm}^{-3}$.
 - Activation fraction increases with increasing instability as inferred from temperature lapse rate.
- Model calculations of geographical distribution of forcing and comparison to clear-sky and cloud albedo perturbations evaluated from AVHRR radiance measurements.
- Examination of climate response (CCM1) shows equal response to greenhouse and aerosol forcing in global annual mean, but strong interhemispheric and seasonal patterns.
- Examination of seasonal and latitudinal patterns of temperature anomaly trend shows lower rate of warming in midlatitude summer consistent with anthropogenic sulfate influence.

CONFIRMATORY ANALYSIS OF SAGE I AND II DATA

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- Develop a new inversion approach to the multi-wavelength SAGE data.
- Confirm or modify the ozone densities measured by the SAGE instruments.
- Provide a new assessment of ozone and aerosol trends in the stratosphere.
- Our results show that the SAGE ozone data in the lower stratosphere are affected by the atmospheric aerosol content.
- SAGE gives an ozone/aerosol correlation which is not substantiated by coincident ozone measurements.

ULTRAFINE AEROSOL SIZE DISTRIBUTIONS: NEW PARTICLE FORMATION IN THE ATMOSPHERE

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- Vapor pressures of sulfuric acid above aqueous solutions of sulfuric acid and sulfuric acid/ammonium sulfate mixtures have been measured at temperatures and relative humidities relevant to the atmosphere.
- A pulse-height analysis method has been implemented with an ultrafine condensation nucleus counter to measure concentrations of particles in the 3 to 4 nm diameter range.
- An inversion algorithm has been developed to obtain size distributions of particles in the 3 to 10 nm diameter range.
- Field measurements indicate that nucleation occurs at sulfuric acid concentrations 1 to 2 orders of magnitude lower than values predicted by the binary theory of $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ nucleation. We speculate that additional species (possibly ammonia) may be involved in the nucleation process.
- Gas phase H_2SO_4 concentrations at Mauna Loa typically agree well with calculated steady state values based on production by the reaction of SO_2 with OH and removal by condensation on preexisting particles.

AEROSOL CHEMISTRY AND SPECTROSCOPY

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- Experimental and theoretical investigations of deliquescence and growth of hygroscopic aerosols as a function of chemical composition, relative humidity, and temperature.
- Measure thermodynamic and optical properties of concentrated sulfate and nitrate solutions, using a single-particle levitation technique.
- Identify the chemical and physical state of the species in single suspended particles using laser Raman and Mie resonance spectroscopic methods.
- Elucidate metastable states in microparticles.
 - Identify previously unknown metastable solid states.
 - These metastable states form only from highly supersaturated solution droplets.
 - The presence of solid metastable states is not predicted from bulk-phase solution thermodynamics
 - Metastable states exhibit deliquescence properties different from those of the expected stable states.

ATMOSPHERIC AEROSOL MICROPHYSICS

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- Develop computational simulation description of collisional growth of irregular particles including their compositional dependences.
 - Incorporates numerous advances in theory of long-range intermolecular forces between condensed bodies.
- Develop description of equilibrium vapor pressure over irregular particles.
 - Paired insoluble spheres with sulfuric acid on surface will effectively serve as CCN, whereas single insoluble sphere with sulfuric acid on surface will not activate at realistic cloud supersaturations.
- Develop fully-coupled computational model of initial stage of cloud condensation nucleation including interactive effects of vapor depletion, background gas heating, polydispersity in aerosol size and composition, and stochastic supersaturation as occurs as aerosols enter clouds.
- Develop description of small cluster reaction rates.
 - Two different Ni potentials utilized to give first accounting for approach of cluster collision description to aerosol collision description. Utility of simple (L-J) vs. complex (many-body) potentials shown.

CLOUD DYNAMICS AND AEROSOL-CCN EFFECTS ON SHIP-TRAIL CLOUDS

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Meteorological, CN, CCN, nephelometer, and filter measurements during the Monterey Area Ship Trail (MAST) experiment in June, 1994, showed features not represented in current models of aerosol-CCN-cloud phenomena:

- Ship-trail clouds can rise above background clouds, and bottoms of ship trails are often lower than bottoms of background clouds.
- The edges of ship trails are often associated with absence or reduction of cloud, and Doppler radar often identifies these regions as areas of increased subsidence.
- Ship trails do not form when the boundary layer is well mixed, when the marine boundary layer is greater than about 750 m . These observations are more consistent with nonlinear heat turbulence than with aerosol injection.
- Ship trails are usually associated with greatly increased CN and CCN concentrations.
- Ship trails were rarely observed when background CCN levels were relatively high.
- Small ships with low heat output but high smoke generation were observed to produce ship trails.

AEROSOL MEASUREMENTS IN THE NORTH ATLANTIC REGIONAL EXPERIMENT (NARE)

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- Aerosol measurements over the North Atlantic south and east of Halifax, Nova Scotia, from the surface to 5 km, for four weeks, August 1993.
- Advected continental aerosol, often of fairly high concentration, was typically present in warm air isolated from the relatively cold air close to the ocean surface by an inversion at the top of the marine boundary layer.
- Dominant anion was sulfate: $\text{NO}_3^-/\text{SO}_4^{2-}$ equivalence ratio 0.15. Aerosol was acidic: $\text{NH}_4^+/\text{SO}_4^{2-}$ equivalence ratio 0.6. Sea salt component small, probably because of offshore transport in a very stable atmosphere.
- High correlation, $r = 0.8$, between SO_4^{2-} mass concentration and number concentration of accumulation mode aerosol particles ($0.15 < d < 3.0 \mu\text{m}$).
- Layers of transported aerosol were frequently above altitudes of marine boundary layer clouds, suggesting minimal impact on cloud radiative properties, at least for the period of the measurements.

ATMOSPHERIC CHEMISTRY OF ORGANIC OXIDANTS AND ALDEHYDES: AEROSOL STUDIES

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- Cylindrical internal reflectance FTIR spectroscopy has been evaluated for use in studies of aqueous reactions of organic oxidants and aldehydes.
- The method has been used to remeasure the absorption cross-sections for water and dissolved inorganic (sulfate, nitrate, etc.) and organic species (formaldehyde, phenol, nitrophenol, etc.).
- The dissolved pollutants present in aerosols lead to radiative warming from longwave absorption, counterbalancing the cooling due to incoming light scattering.
- Further studies are underway using this method to examine the aqueous reactions of organic oxidants particularly peroxides, peroxyacids, and PANs.

REACTIVE UPTAKE OF GASEOUS Cl₂ AND Br₂ BY AQUEOUS SURFACES

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D. R. Worsnop, M. S. Zahniser and C. E. Kolb
Aerodyne Research, Inc.

- The uptake of gas-phase Cl₂ and Br₂ by aqueous NaBr and NaI solutions has been studied as a function of concentration (2.5x10⁻⁴ to 0.5 M), temperature (263-293 K), and gas-liquid interaction time (2-15 ms).
- The magnitude of the measured halogen uptake and its functional dependence on ion concentration are not in accord with a simple bulk phase reaction mechanism:
 - Reactions at the gas-liquid interface are significant.
 - The reactions of the halogen molecules at the interface proceed via a complex, for example
$$\text{Cl}_2(\text{g}) + \text{Br}^- \longrightarrow (\text{Cl}_2\text{Br})_{\text{interface}}^- \longrightarrow \text{Cl}^-(\text{aq}) + \text{products}$$
 - Reactions of ions with the halogen molecules at the interface are more facile than in the bulk liquid.
 - The Cl₂/Br⁻ and Cl₂/I⁻ surface reactions become significant for bulk ion concentrations greater than about 0.05 M.

ROLE OF SEA SALT AEROSOL REACTIONS IN GLOBAL TROPOSPHERIC OZONE

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- Determination of reaction probabilities for the reactions of a variety of gases with NaCl and NaBr, components of sea salt aerosols, has elucidated which reactions are most likely to be important in the troposphere.
- Measurement of the kinetics and mechanisms of atomic chlorine produced by photolysis of the gas phase products of the NaCl reactions with organics, including biogenics and dimethyl sulfide.
- Elucidation of the role of water adsorbed on solid NaCl in promoting the reactions and generating new particles.
- Measurement of the kinetics and mechanisms of hydrolysis of the halogen-containing gas phase products of the NaCl and NaBr reactions.

AEROSOL CHARACTERIZATION ON THE PNL GULFSTREAM G-1 AIRCRAFT

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- Develop a fully automated aircraft sampling package to collect aerosol for chemical characterization using SEM/X-ray techniques.
- To investigate the chlorine loss from sea salt collected in the marine boundary as a function of size and composition.
- Collaborate with Battelle Columbus Lab in measuring trace gases over the ocean (DMS, Cl₂) and with UC Irvine in reduction and analysis of the gaseous data.
- To study aerosol chemistry interactions in the boundary layer .

MEASUREMENTS OF RADON USING THE PNL GULFSTREAM G-1 AIRCRAFT

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- A compact, near real time radon/radon progeny monitor, the "Radgrabber", was recently tested during the 1995 ACP study on Long Island, NY.
- Preliminary data analyzer operated successfully and will provide the G-1 aircraft with a new tool for boundary layer and atmospheric transport studies.

ENVIRONMENTAL RADON, THORON, AND RELATED AEROSOLS

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- Objective is to improve on the data used for estimating the health impact from indoor radon and radon progeny.
- A computer code for simulation studies of indoor air quality has been extended to include radon and radon progeny.
- Aerosol samples have been taken in a former uranium mine to better reconstruct the radiation dose incurred by 1960's miners.
- The aerosol in residential housing has been measured so that health data from miners can be extrapolated to the home environment.