

**National Aerosol-Climate Interaction Program (NACIP)
La Jolla Workshop, January 9-11, 2002**

Report from Working Group 2

Participants: *Clarke, Mangis, Melville, Novakov, Prather, K., Schwartz (SC), Thiemens, Valero*

Rapporteurs: *Guazzotti, Li*

Working Group 2 was asked to focus discussion on the following topics:

“What key information is lacking regarding atmospheric aerosol processes, optical and hygroscopic properties, mixing, chemistry, and the direct effect? What research is required to provide this information?”

The Working Group initiated discussion by indicating that the first things that should be defined are the requirements, i.e., the information that must be sought about aerosols and processes which are relevant to climate issues. Then, prioritization of the research needs should be attempted. Also, the type of activities which need to be carried out in order to achieve these requirements should be defined, including laboratory experiments, field campaigns, and modeling efforts. It was agreed that for this Program, field campaigns together with lab experiments that will help in the chemical characterization of aerosols should be considered.

In addition to their charge of addressing direct forcing, two other key issues which were discussed in the Plenary Session were brought to attention by the group: 1) the fact that processing in clouds is unrestricted in models (as indicated by Jacob) and 2) the fact that surface tension due to changes in aerosols chemical composition is not constrained in models (as indicated by Seinfeld). It was recommended that these two issues deserve attention under the Program.

The anthropogenic forcing should be considered a focus for the Program. In particular, efforts should be taken to help understand/ characterize/ quantify which species contribute the most to the anthropogenic forcing while recognizing their often mixed character that can include natural contribution.

Due to the strong influence of mixed aerosol chemistry on ambient aerosol optical properties and on the $f(\text{RH})$, both were identified as key issues. It was indicated that it is necessary to characterize the species responsible of the changes in $f(\text{RH})$ (e.g., vertical changes in $f(\text{RH})$, changes in $f(\text{RH})$ as a function of time due to the evolution of a particular air mass, etc.). It was also felt necessary to establish demonstrated capabilities to model $f(\text{RH})$ based on the physicochemical properties of the aerosol, since hygroscopic properties are very much dependent on the aerosol type and mixing status.

Other issues which need to be considered and were discussed include the characterization of natural aerosols (clean air) taking into consideration that the characteristics of the background aerosol need to be understood for proper interpretation of remote sensing and evaluation of anthropogenic influences. A special emphasis was

placed on sea-salt aerosols, their fluxes, and their dependence on wind speed and other factors such as properties of oceanic surfaces (kinematics of breaking). It was indicated that the current large uncertainties in the wind dependence should be reduced since this is a parameterization generally employed in models. Also, removal mechanisms should be studied with detail. Chemical characterization of carbonaceous aerosols (BC, OC, TC) should be considered important, including efforts directed towards the study of their influence on heterogeneous surface reactions and hygroscopicity. Also, proper artifact evaluations for the techniques currently employed should be considered through development of standards and engaging in comparison exercises. It was stated that attention should be given to the evaluation of how the measured aerosol size and mass are affected by the measurements (e.g., presence of semi-volatile compounds – sampling techniques which use denuders vs. those that don't; temperature effect in aircraft sampling; filter-based measurements which provide only second hand information on aerosol morphology and composition). Other issues discussed included the following:

- the necessity for improved understanding to close the gaps between measurements and models;
- influence of aerosol microphysics on adsorption properties; in particular attention should be given to absorption cross section properties of internally mixed absorbing aerosol and wet absorbing aerosols, since no experimental work has proven the modeled enhancement effects yet ;
- modification of the aerosol properties by the collection techniques employed (which makes the collected aerosol different from the suspended one present in the atmosphere);

Other general considerations raised included:

- the necessity for more real time measurements of vertical profiles;
- more characterization of size distribution source functions ;
- representativeness of emission factors;
- chemical composition and complexity of natural and anthropogenic dust, and their effect on the optical properties of the dust aerosol (the existing dust aerosol model employed generally uses a fixed dust composition (refractive index) and effective size parameters. More information needs to be added to dust archives for various geophysical sources);
- mixing status of the aerosol (real mixing status remains uncertain (WG2 & WG4 discussion));
- source characterization (WG2 & WG4 discussion).

In addition to strategies currently employed in the study of the direct effect, it was stated that turbulent mixing and entrainment should be considered when planning future experiments. This is important since it would help understand the nature of “plume” evolution seen by satellite and parameterized in models, but this probably should not be included in a priority list for this Working Group. It was also felt important to assess the direct effect in both cloud free and regions of broken cloud due to possible cloud enhancements.

After much consideration the fundamental need was defined as follow:
“To improve understanding of link between aerosol optical effects (3-D, time) to the associated aerosol physical and chemical properties and sources (natural and anthropogenic); and to integrate these understandings into CTM models in realistic way and evaluate these models in conjunction with remote sensing measurements.”

Key issues in terms of the fundamental need stated above were discussed and grouped in four main categories: 1) Aerosol Processes, 2) Optical and hyroscopic properties, 3) Chemistry, and 4) Mixing issues. The main issues addressed in each one of these categories are presented in the summary at the end of this report (see Annex I).

Following contemplation of the main topics indicated above, discussion focused on the research needed to accomplish the stated issues. There was consensus among the members of the group on the necessity for longer-term coordinated studies, which will help in evaluating the representativeness of the acquired data sets. These longer-term studies will help in systematic evaluation and augmentation of model and satellite products. Also, this strategy will permit optimization of measurements over time since it will allow for feedback during the campaign/s. The favored region/s for such a study should be considered taking into consideration input from the models, satellite and measurements communities. The site/s should be adequate for the targeted questions. Also, it will be necessary to assure funding for sustained evaluation of acquired data sets beyond the data acquisition phase. Laboratory studies for the characterization of aerosol properties were also discussed. The main points addressed concerning the research needed are summarized in Annex II of this report.

Subsequent discussion focused on the tools required for accomplishing the stated objectives. Emphasis was given on the necessity for fast aerosol chemistry measurements, and improved mini-lidar systems. Discussions on the necessity for smaller aircraft and UAV developments for these experiments were also presented. Developments of smaller mass spectrometer capabilities for single particle analysis were also discussed. Summary of the key issues considered is presented in Annex III.

Joint discussions among the meeting participants who constituted part of Working Groups 2 and 4 were carried out as well. One of the main issues considered during the integration was the fact that, although Working Group 2 was asked to focus on the direct effect, the indirect effect should of course be considered when planning the Program. It was indicated that the Program should stay focused, with its scientific questions clearly stated. Also, it would be important to define the magnitude of the uncertainties that would be acceptable (define “how good is good enough?”). Measurement intercalibration should be considered as an important aspect for the success of the Program. Discussions were presented in terms of the methodology to be used in order to select the site/s for the field campaign/s. It was suggested to choose the region/location where the aerosol constitutes a problem, making it the target for long-term observations . The necessity for long-term global measurements of aerosol single scattering albedo was presented as well.

Both Working Group 2 and 4 suggested integrating the above efforts to reduce uncertainties in the linking of measured aerosol properties to both satellite retrievals and

model capabilities. Specific suggestions included augmenting the historical approach of “intensive campaigns” with more limited but longer-term sustained measurements. These call for refinement of current new-technology light-aircraft instrument packages that can be flown frequently and cost-effectively at carefully selected sites throughout the year. These would provide more robust statistics through improved vertical and temporal assessments that are difficult to achieve in intensive campaigns. Study regions would be selected to best characterize aerosol properties of particular interest (dust, biomass burning, pollution, sea-salt, etc.) and to provide maximum opportunity to combine and test satellite and model data associated with these sources. Several characteristics were identified as being important for these study regions:

Sources representative of a target aerosol type(s)

Location favorable for well constrained meteorological data (*essential for model uncertainty*), model suitability (*minimized uncertainty from small scale variability or perturbations etc.*) and satellite retrieval (*multiple satellites, good passes, times, sunglint issues etc.*)

Identify Representative Measurement Strategies to link to Satellite/Models (*appropriate scales needed to best link measurements, models and satellite data*)

Ground station(s) measurements for long-term context (*needed for both model data and for interpreting episodic aircraft data – upgrade of existing station preferred*)

Routine vertical sampling with smaller instrument packages on light aircraft (*scattering, absorption, $f(RH)$, Size-distributions, radiative fluxes, chemistry*)

Effective range of operation (*all necessary components should be within useful range of light aircraft flights (say 200km radius??)*)

Logistical support in region for possible intensive campaigns (*resources available so that intensive campaigns add to the data base and can be carried out in the context of this longer term 3-D data sets*)

ANNEX I

AEROSOL PROCESSES

Source Related

- Combustion processes and primary condensation sites for heterogenous chemistry (Size)
- Distribution emission assessment – soot, fly ash, high temp condensates)
- Dust Production (Natural and Anthropogenic?)
- Sea-Salt Concentrations – Fluxes – Wind Speed (reduce current large uncertainty)

Transport/Removal

- Wet Removal Rates for Various Precipitation Types/Regions
- Nucleation and nuclei evolution in aerosol populations

Studies should include strategies to improve understanding of air mass evolution and dynamics (eg.Turbulent mixing, Entrainment)

OPTICAL and HYGROSCOPIC

- Absorption and SSA (reduce PSAP Artifacts-pressure, humidity, scattering)
- Increased emphasis on Flux Divergence and gradients as direct forcing strategy
- Establish size and composition dependent scattering, absorption, hygroscopicity-refractive index [at least coarse and fine aerosol]
- Establish demonstrated capability to model $f(RH)$ based on physiochemical properties
- Structure of aerosol fields - Remote Sensing (Small/Cheap effective Lidar)
- Shape effects??

CHEMISTRY

- BC/OC/TC –Need for Standards, Evaluations, Comparisons of Methods
- Relative Mass fractions
- OC and influence on heterogeneous surface reactions
- OC Tracers for Surface sources
- OC and hygroscopicity
- Semi-volatile issues (samples vs. ambient aerosol)
- More rapid Size-resolved chemistry and mixing state to resolve spatial structure [PILS – IC, flash-desorption, Single Particle Mass Spec]
- Natural/Anthropogenic Contributions to each aerosol component for associated direct effects

MIXING ISSUES and How well do we need to know these?

- In-Situ* Sampling Strategies for establishing mixed components
- Absorption X-section properties in internally mixed and wet aerosol – [actual measurements vs. model]
- Natural vs. Anthropogenic relative contributions for mixed species
- Influence of mixtures on $f(RH)$
- Heterogeneous surface reactions and modified aerosol physical chemistry (dust, sea-salt)

ANNEX II

RESEARCH NEEDED

-Need longer term In-Situ measurements in selected environments for systematic evaluation and augmentation of model and satellite products and to allow optimizing measurement strategies over time. Instrument, Modelers, and Satellite representatives should establish favored regions to evaluate certain aerosol types (biomass, anthropogenic pollution, dust, sea-salt, etc.).

- Identify Target aerosol type(s)

- Location favorable for meteorology, model suitability and satellite retrieval

- Identify Appropriate Measurement Strategies to link to Satellite/Models

- Ground station(s) measurements for long-term context

- Routine vertical sampling with smaller instrument packages on light aircraft

 - (scattering, absorption, $f(RH)$, Size, radiative fluxes, chemistry??)

- Logistical support in region for possible intensive campaigns

-Also, explore commercial platforms for long term monitoring program.

-Include more experiments that include studies of direct forcing for absorbing aerosol in cloudy vs. cloud free atmospheres.

ANNEX III

IMPROVED RESEARCH TOOLS

- New artifact free (scatter, pressure, RH) airborne light absorption measurement
- Fast Aerosol Chemistry (ions and organics)
- Support making existing instrumentation smaller, faster, better
- Improved/cheaper mini-lidar for airborne or ground based aerosol structure
- Small aircraft and UAV developments

OTHER LAB AND FIELD STUDIES:

- Improved understanding of $f(\text{RH})$ in terms of real aerosol physicochemistry
- OC/EC/TC and artifact evaluations through Standards and Comparisons
- Efforts to catalog and archive older relevant data sets