FORM A: GACP ACCOMPLISHMENT REPORT

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Institution: University of Washington

TITLE: Validation of the Lidar Retrieval of Aerosol Extinction.

ABSTRACT:
Understanding potential climate change depends upon improved quantification of climate forcing by anthropogenic aerosols, including clear-sky backscattering of solar radiation to space. Because of its exquisite precision, vertical resolution, and the relative ease of data acquisition, lidar technology has a great potential to contribute to this research. However, this potential remains largely unrealized due to calibration and retrieval uncertainties, especially with regard to an optical property known as the lidar ratio - the ratio of light extinction to 180-degree backscatter. We have modified an integrating nephelometer to allow direct and highly accurate measurements of 180-degree backscatter and will deploy this device at a coastal station under existing funding. We request, herein, logistical support for a small but crucial field experiment and salary support for a subsequent data analysis project intended to enable quantitative use of lidar data (past and future) in the development of a global climatology of aerosol forcing. Under the field project our 180° nephelometer will be deployed at the NOAA/CMDL aerosol research station at Bondville, IL in order to obtain systematic, empirical data on the lidar ratio and to carry out a rigorous lidar validation experiment in conjunction with a horizontally-pointing Micro-Pulse Lidar (MPL). Subsequent data analysis efforts, based on these results, will include the development of an empirically-based climatology of lidar ratios for lower tropospheric aerosols and a general assessment of uncertainties for lidar retrievals of aerosol properties. Lidar data sets that could potentially be brought to bear on the aerosol forcing issue include ground-based monitoring with MPL (e.g. at the DoE ARM site), extensive airborne campaigns with NASA's DIAL system, the space shuttle lidar mission (LITE), and possible future satellite lidars (GLAS and PICASSO).

GOALS:

Objective 1 (technological)

Compare aerosol extinction and aerosol lidar ratio derived independently from horizontally-pointing lidar and calibrated in-situ measurements at a site in the Midwest (Bondville, IL) that provides the logistical support, historical data base and supporting aerosol optics, chemistry and physics measurements and has a relatively uniform (spatially and temporally) urban/industrial aerosol. Our hypothesis is that these independent methods respond to the same physical phenomena and accurately quantify those phenomena within well-understood experimental uncertainties. Our scientific methods will be capable of disproving this hypothesis if it is false. This experiment will constitute the first rigorous test of a hypothesis that is fundamental to the PICASSO-CENA strategy of using spaceborne lidar to better quantify direct aerosol radiative forcing.

Logical uses of data from this comparison:

1. Test for large, unexpected errors in one or both methods.
2. Test validity of those assumptions for which uncertainties were poorly quantified.
3. If unacceptable errors are encountered (in part or all of the data set), suggest strategies for identifying and overcoming the sources of error.
4. If discrepancies are within estimated experimental uncertainties, provide a robust quantification of uncertainty for both methods.
5. Identify the major sources of uncertainty for each method - such information will help to optimize the use of each method in future experiments and will suggest where technical improvements would be most valuable.
6. Build toward the series of planned experiments using in-situ measurements beneath the PICASSO-CENA aircraft validation unit.

Objective 2 (integrative science)

Use the full data set acquired under objective one to address a variety of scientific issues related to aerosol radiative forcing at a polluted, regionally representative, mid-latitude site. While the data set will be limited in time, it will also be highly pedigreed in quality. Some specific issues that could be addressed include:

1. Characterize the aerosol lidar ratio, its variability, and controlling factors.
2. Characterize the horizontal, vertical, and temporal variations of aerosol extinction, especially with regard to spatial and temporal autocorrelation scales.
3. Examine co-variances among aerosol optical, physical, and chemical properties as well as atmospheric state parameters (especially T and RH).
4. Examine the feasibility of using combined lidar and in-situ measurements (assuming large variations in ambient RH) to constrain the hydration effect on aerosol light absorption

APPROACH:

The data set shall have these features:

1. Same air volume will be measured by both methods
2. Comparison will be made at ambient T, P, and RH
3. A statistically significant number of samples shall be collected for a range of conditions.
4. Uncertainties shall be quantified
5. All assumptions and tunable parameters shall be clearly identified, including those with poorly quantified uncertainties

Major sources of uncertainty for the MPL are spatial inhomogeneities and interference from sunlight. It is possible that farming operations, road traffic, and/or gusty winds will create significant inhomogeneities near the surface, especially during the day. Thus, best data is likely to be obtained just after sunset or before sunrise.

Major sources of uncertainty for the in-situ measurements are inlet losses of coarse particles, nephelometer truncation error for coarse particles, calibration uncertainty for the absorption measurement, hydration/RH effect on absorption, and hydration/RH effect on light scattering. Thus, the most tightly constrained comparison data will be obtained under conditions of low coarse mode extinction (relative to fine mode), low relative humidity, and low absorption (relative to scattering - i.e. high single scattering albedo.) The best scenario is to obtain (1) a statistically significant number of samples under these conditions plus (2) samples exhibiting a large range of values for coarse mode fraction, ambient RH, and single scattering albedo.

Data will be analyzed in near real-time for rapid identification of methodological errors and to facilitate integrative thinking about the science objectives while we are still in the field.

In the case that unexpected difficulties require a reduction in the overall scope of activities, it will be important to know which activities can be eliminated or scaled back with the least affect on the scientific goals. To assist in this process, I have attempted here to list the measurements in order of importance, taking Objective One as the fundamental task to be accomplished.

For objective one (comparing lidar and in-situ determinations of extinction and lidar ratio), the following measurements are critical and constitute the "minimum mission":

1. horizontal MPL located several km from site
2. in-situ scattering and absorption at near-ambient RH
3. in-situ 180-backscatter at near-ambient RH
4. ambient T, P, RH at top of stack.

The following measurements will improve the comparison by reducing the number of required assumptions and the resulting uncertainties:

5. scattering as a function of RH over 50-90%
6. RH variation along path and in vertical
7. low-RH size distributions
8. low-RH scattering and absorption
9. aerosol mass and chemistry (preferably at 4-hour or better time resolution).

For objective two, the following measurements will improve the scope and quality of scientific issues that can be investigated:

10. vertical MPL at BND station
11. column optical depth by sun-photometer
12. meteorological context: winds, precipitation, regional observations, regional weather maps, soundings.

TASKS COMPLETED:

Since the funding award on 15 March 1999 we have:

1. Established a site near Bondville, Illinois where the combined in-situ and MPL field experiment can be performed in collaboration with Dr. Mark Rood of the University of Illinois Champaign Urbana and Dr. John Ogren of NOAA CMDL.
2. Established a scientific plan and subcontract for operating MPL at the site in cooperation with Dr. John Reagan and coworkers at the U. of Arizona.
2. Made preliminary measurements of physical and optical properties at the site.
3. Established a protocol for gathering regional meteorological data during the experiment.
4. Established a protocol for enhanced aerosol chemistry sampling and analysis.
5. Operated the β-180° nephelometer in a field experiment (INDOEX) to test its capabilities.

FUTURE PLANS:

1. The highlight of this project is a field experiment that will be conducted in August and September of 1999 to compare aerosol extinction and aerosol lidar ratio derived independently from horizontally-pointing lidar and calibrated in-situ measurements.
2. The remainder of the first year and the entire second year will be devoted to data analysis, interpretation of the results and application of the results to future lidar measurements and publication of the results.

RESULTS:

Pending the field experiment in August and September 1999, there are no significant results to date.

FORM B: GACP SIGNIFICANT HIGHLIGHTS

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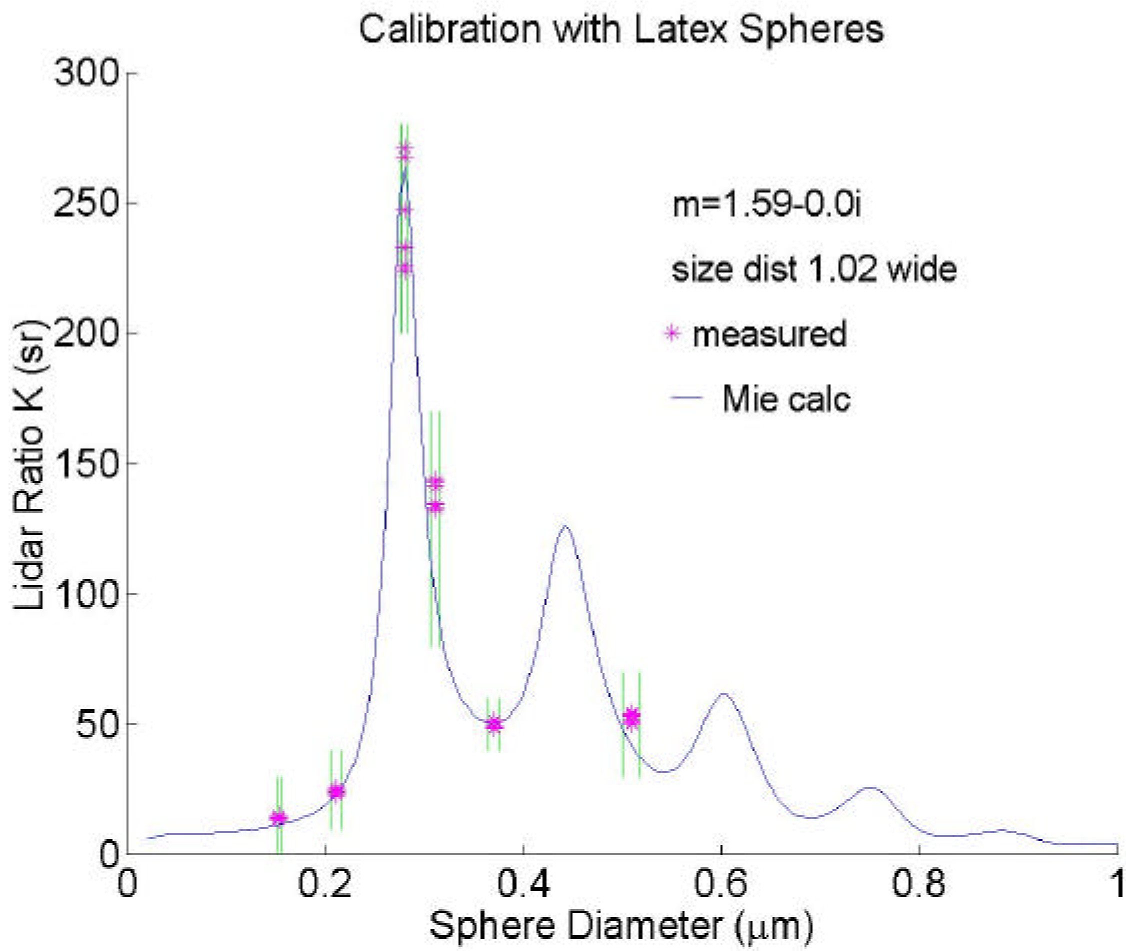
SIGNIFICANT HIGHLIGHTS:

During the summer of 1998 the design, calibration and lab testing of a new instrument, the 180-degree backscatter nephelometer was done. This β-180 nephelometer is a modified version of the commercially available integrating nephelometer. A patent application was submitted by Sarah Doherty, Dr. Tad Anderson and Dr. Robert Charlson for the design. In August, 1998, we submitted a paper to Applied Optics presenting the instrument design and performance; the paper was published in March, 1999.

Laboratory tests with monodisperse latex spheres demonstrated that the lidar ratio could be measured with the β-180 nephelometer and the integrating nephelometer to within ±15% of that predicted by Mie theory including size and refractive index uncertainty.



After the instrument had been tested in the lab and at our marine boundary layer field station, it was prepared for mounting on the National Center for Atmospheric Research (NCAR) Research Aviation Facility (RAF) C-130 aircraft for participation in the INDian Ocean EXperiment (INDOEX). It flew on the C-130 for the ferry flights from Colorado to the Maldive Islands and flew on most of the research flights during the field campaign, which ran from Feb. 12 – March 26, 1999.

The results from the INDOEX experiment are being analyzed now. The lidar ratio survey and humidity dependence of lidar ratio present new data not determined before. The comparison with ground and aircraft based lidar should be possible from this data set to enable closure experiments and accurate determination of optical depth with the lidar measurements.

The combination of these results, verification of laboratory calibration and rigorous field tests, gives us confidence that we will be able to perform the lidar comparison field test in late Summer of 1999 and will be able to measure the aerosol lidar ratio to within ±20% by two independent techniques. Thus, our hypothesis that these independent methods respond to the same physical phenomena and that they accurately quantify those phenomena within well-understood experimental uncertainties can be tested.

FORM C: FUTURE PLANS

Name: David S. Covert and Theodore L. Anderson

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Field deployment and operations plan for Bondville, IL August-September 1999 experiment

Lidar

Two MPL's will be brought to the experiment :

1. Horizontally sensing MPL in weather-sealed, environmentally controlled container that can be manually positioned to make occasional vertical measurements for calibration purposes. MPL provided on loan from NASA/LaRC, container box to be provided by Jim Spinhirne, NASA/GSFC. MPL to be operated on as nearly a continuous basis as possible.
2. Slant-path MPL in a small weather-sealed building with air-conditioning. Slant-path positioning mirror system to be provided on loan by Jim Spinhirne, NASA/GSFC. Building, air-conditioning, electrical wiring, and pointing mirror mounting fixture all to be purchased/fabricated. MPL slant-path experiments to be conducted a few times daily, weather permitting. Otherwise, this MPL will be operated vertically pointing on as nearly a continuous basis as possible.

The horizontal MPL will be located approximately 4 km from the station and pointed at the sample inlet. This unit will provide data for direct comparison with in-situ measurements at the station. In addition, it will provide information on the horizontal variability of extinction in the vicinity of the site.

The slant-path MPL will be located at the station. This unit will provide information on the vertical variation of extinction at the site. It will thus help to establish the extent to which surface measurements are representative of the boundary-layer integral and of the column integral.

In-situ extinction and 180° backscatter at near-ambient RH

UW will bring to the BND site:

- A 3-wavelength, total scatter/hemispheric backscatter nephelometer (450, 550, 700nm)
- A 180° backscatter neph (532 nm wavelength)
- An absorption photometer (550 nm wavelength)
- A particle counter (0.01-1.0 µm diameter detection)
- 1- and 10-µm impactors

The purpose will be to measure extinction and 180° backscatter at as near to ambient RH as possible. To achieve ambient RH, the shed will have large holes and a large fan to ventilate it continuously with ambient air. We hope to operate the 180-neph within 1 degree of ambient temperature and the integrating neph within degrees of ambient temperature. If so, then the 180-neph will run at 5-10% RH below ambient and the regular neph will run at 10-20%RH below ambient.

At times when the MPL is not operating (primarily, early AM when sunlight interference with MPL is maximum) the RH of the 180° neph will be reduced via heating to provide a measure of the RH-dependence of 180° backscattering.

The errors of the scattering and absorption measurements are rather well known. We have made a preliminary uncertainty analysis for this experiment, based on what we estimate to be the possible ranges of five key factors:

1. ambient RH (tested range 50-90%)
2. delta-RH between ambient and the 180-neph (tested range 5-10%)
3. delta-RH between ambient and the regular neph (tested range 10-20%)
4. uncertainty of the 180-backscatter measurement (tested range 10-20%)
5. fraction of extinction due to coarse mode particles (tested range 5-50%)

The result of this analysis are as follows:

	Extinction Uncertainty	180-Backscatter Uncertainty	Lidar Ratio Uncertainty
Base Case (most optimistic)	+/-10%	+/-11%	+/-14%
High RH and high delta-RH	+/-15%	+/-20%	+/-25%
High uncertainty in 180 meas	+/-10%	+/-20%	+/-22%
High coarse mode	+/-13%	+/-14%	+/-19%
Worst Case	+/-15%	+/-32%	+/-35%

This uncertainty analysis indicates several key diagnostic parameters to be monitored during the campaign. We are developing data processing software that will provide estimates of the above three quantities and their uncertainties in near-real-time during the campaign.

<u>Atmospheric state: temperature, pressure, RH</u>
At a minimum, T, P, and RH need to be measured in the vicinity of the aerosol sample inlet at the top of the stack. If possible, T and RH measurements will also be implemented to determine the vertical profile at the site and the horizontal profile between the off-site, horizontally-pointing MPL and the site.

<u>In-situ scattering at low RH and scanning RH</u>
Year-round measurements of scattering and absorption at low RH and of scattering at scanning RH are made at BND. These will of course continue during the fall intensive. Scattering is measured using TSI nephelometers (model 3563) and absorption using a differential transmission absorption photometer (model PSAP, Radiance Research.) A multi-orifice impactor upstream with a low-RH aerodynamic cut at 1 µm diameter is alternated into the sample stream so that alternate 10-minute periods sample Daero < 10 µm and Daero < 1 µm. The scanning RH system is in parallel to the low-RH system. Water is added to the airstream in a controlled fashion. RH is scanned from about 50% to about 90%. Due to its complexity, this system has many challenging aspects to successful operation. Note that it is desirable to operate such that the minimum RH experienced by the aerosol anywhere in the plumbing is >40%; this will minimize the chance of irreversible efflorescence (crystallization) of ammonium sulfate.

<u>In-situ size distributions at low RH</u>
Measured by Differential Mobility Particle Spectrometer (DMPS) and Aerodynamic Particle Sizer (APS) at low relative humidity with 10-minute time resolution. One scan takes 10 minutes, so homogeneity over the 10-minute period must be assumed. (This assumption can be checked from the continuous neph and particle count data. At present there is no plan or protocol for flagging or removing data during highly variable periods.)

DMPS inversion and recording of all data is on Macintosh computer. Data disks are downloaded daily (at present) and archived by UIUC personnel. The data included a quick calculation of 550nm total scatter, hemispheric backscatter, and 180° backscatter using Mie look-up tables which assume: real refractive index is 1.45, imaginary refractive index is 0.00, particle density (needed to convert APS aerodynamic sizes to geometric) is 1.8 g/cc. The calculated scattering and backscattering are thus available within a day for direct comparison to the dry neph measurements.

Size distribution measurements were made during two previous intensive campaigns at BND, that were intended to build toward the fall intensive. These were Nov 18 - Dec 3, 1998 and April 28 - June 18, 1999 (the latter is ongoing at present). Daily plots of the comparison between measured and predicted light scattering and backscattering can be viewed at a website developed by UIUC, http://cenpc111.ce.uiuc.edu//index.html

<u>In-situ sampling for mass and chemical composition</u>
Normal chemical sampling at BND is based on 24-hour filter samples, collected in an automated mode using an 8-port carousel (7 daily samples and 1 blank). Filters are teflon fiber (Millipore Fluropore, 1 µm pore size). They are weighed for gravimetric mass, extracted with water/methanol and analyzed for major ionic constituents. The sample flow rate is 30 lpm (about 43 cubic meters per day). The filter carousel is

preceded by a 1-µm aerodynamic diameter impactor, operated at low RH. Super-µm mass and chemistry is also determined using the Tedlar substrates from this impactor; however, the time resolution is much less, since one super-µm sample represents the integral over all 7 sub-µm filters. Filter sampling is done by UIUC; mass and chemical analysis are done by NOAA/PMEL.

During the LINC campaign, high time resolution (4-hour) filter samples will be collected for at least 14 and preferably 21 days. This will provide information on temporal variations and short term correlations between aerosol mass, aerosol chemistry, and the other measured aerosol properties. In addition, a small portion of the liquid extracts will be analyzed for organic speciation via spray ionization mass spectrometry at the UW Chemistry Department. While not central to LINC objectives, this organic analysis is a research direction we want to take.

To accomplish 4-hour sampling:

1. Trish Quinn of NOAA/PMEL has shown that analyzable total mass and sulfate mass should be present, based on previous experience with 24-hour samples at BND.
2. John Ogren of NOAA/CMDL has agreed to alter the program controlling the automatic sampling. This turns out to be a simple change to one variable. Following the daily change of the carousel, the 4-hour increments will begin at the top of the hour in which the sampling is restarted. This should be on even times, such that the 4-hour sample periods will begin at: 00, 04, 08, 12, 16, 20 (UTC). For example, sampling could be stopped at 15:50 UTC (10:50 PDT) each day for changing of the carousel and restarted at 16:05 UTC (11:05 PDT). Note that present carousel changing is done at 00 UTC (21:00 PDT), but making the change in the late morning would be much better for this campaign, since the evening is a heavily used portion of the day.
3. To facilitate daily carousel changing, a second carousel will be brought to the site by UW.
4. UW will provide a graduate student (Uli Steidl) during and after the campaign to help with the addition filter-changing work and to perform the additional filter handling, pre-weighing, and post-weighing according to the established PMEL protocol.
5. Filter extraction, IC analysis, and data reduction will be done by NOAA/PMEL (Trish Quinn). The 4-hour filters will be averaged to 24-hours in order to maintain a consistent, long-term record for the BND station.
6. Subsequent data analysis/integration and chemical analysis for organic compounds will be performed by UW (Uli Steidl).
7. Each carousel will normally have two blanks, since 4-hour sampling uses only six of the 8 filters per day. Filter samples will also be taken during the station zero checks (HEPA filter placed on stack inlet, common to all in-situ instruments.) These will constitute blanks with flow.

<u>Meteorological context</u>
Hourly observations from the 5 nearest NWS stations are being recorded by UW (Dave Covert) via an automated archiving program. This data set also includes the rawinsonde observations (vertical profiles of T, P, RH, winds) from the nearest station, Lincoln, IL.

<u>Column optical depth by sun-photometer</u>
UA will provide and operate a 10-channel auto sun-tracking solar radiometer (UA design). It will be operated when clear skies permit and will obtain spectral optical depths over the wavelength range from 380 nm to 1030 nm.

<u>Integration strategy</u>
The first step of "integration" is to merge data from the various instruments and groups into a common data base. Achieving that in a timely fashion is the goal of this section. Requirements are agreed data products and formats, an agreed data delivery schedule, and an archiving and distribution plan.

FORM D: GACP BIBLIOGRAPHY

Name: David S. Covert and Theodore L. Anderson

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c. List of oral presentations or posters at professional society meetings and conferences:

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