

# MEASUREMENTS OF CONTRIBUTORS TO ATMOSPHERIC CLIMATE CHANGE

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## ABSTRACT

Our goal of better understanding the changes in Earth's climate requires improved atmospheric sensors and techniques for measuring the primary causal factors: aerosol optical scattering and greenhouse gas absorption. We describe extensions of our lidar sensor developments that use a satellite based receiver and ground based lasers to provide a major advance in our ability to measure the concentrations of chemical species and the aerosol properties. The aerosol instrument proposed for these studies uses a multi- $\lambda$  laser pointed ahead of the satellite to measure the optical scattering on segments of arcs along the orbit to characterize aerosol properties. A ground-based MWIR supercontinuum laser is the source for measuring the path-integrated absorption of chemical species to determine concentrations of the greenhouse gases in the 3-5  $\mu\text{m}$  region using a spectrometer on the satellite.

## 1. INTRODUCTION

The fact that man's use of Earth's resources is changing the global atmospheric properties cannot be denied. Many scientific investigations, both measurements and simulations, show that surface temperatures have increased because of increases in infrared optical absorption that traps the Earth's thermal radiation before it radiates back into space. The increase in greenhouse gas is primarily due to our use of anthropogenic fuels for energy and transportation. The combination of burgeoning population, with individual needs for energy and transportation is creating a major problem in our future, both in available resources and causing further changes to our climate. An additional factor, which is even less well understood, is the effect of aerosols and their influences on the environmental system. The aerosol content is increased as particles are injected into the atmosphere, and as photochemical processes operate to generate smog aerosol in a complex chemical soup of trace species. The increase in planetary albedo generally has a cooling effect; however, the overall effect is very complex when the factors of radiative transport of both visible and infrared wavelengths through the atmosphere are considered. The difference in transmission due to scattering by different size particles complicates it further. Also, the effective residence time depends upon the growth rate of particles to sizes that are easily separated by gravity,

and that rate depends on their hygroscopic properties. The current consensus on the relative contributions to radiative forcing by greenhouse species and aerosol scattering are indicated in Fig. 1 [1]. The complex non-linear coupling of the environmental system requires more attention to provide an understanding of this epoch changing experiment in atmospheric climate change that we are conducting.

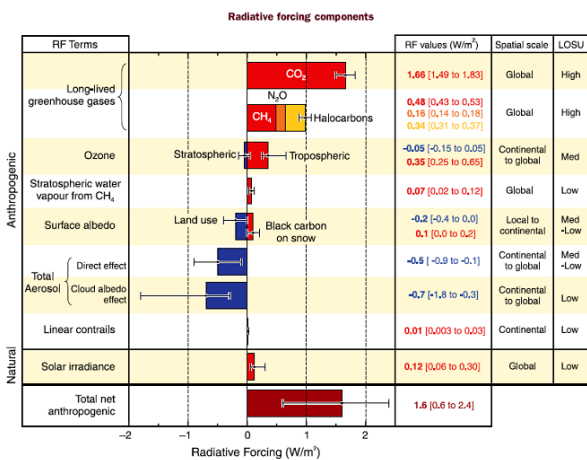


Figure 1. The estimated radiative forcing associated with the several primary sources recognized in the IPCC report [1].

The optical transmission of the atmosphere, shown in Fig. 2, has several windows. The visible and near infrared (NIR) spectrum below  $\sim 2.5 \mu\text{m}$  contains the windows where solar radiation (peaked at 550 nm) is received, and the long wave infrared (LWIR) region 7-20  $\mu\text{m}$  is the region for the return of Earth's radiation (peaked at 15  $\mu\text{m}$ ) back into space.

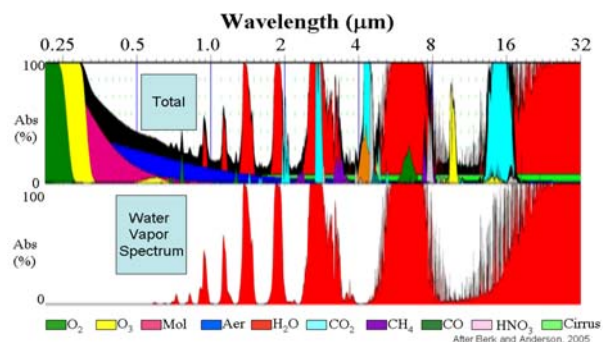


Figure 2. The optical transmission of the atmosphere between ground and space from MODTRAN5<sup>TM</sup> [2].

The optical wavelengths  $>2.5 \mu\text{m}$  contain the optical signatures of the fundamental vibration, rotation, stretching and bending energy states of molecules, and provide unique signatures for measurements of concentrations. The DIAL (Differential Absorption Lidar) and DAS (Differential Absorption Spectroscopy) techniques make use of the range resolved or path-integrated absorption by the molecules to determine concentrations of species. The DIAL technique has been recently developed into its first commercial application to detect and measure natural gas pipeline leaks from an aircraft [3]. The Raman scatter lidar techniques provide another very powerful technique to measure the concentrations of molecular species. The unique wavelengths of the Raman shifted Stokes radiations resulting from the scattering processes are used to measure properties and species concentrations [4-6].

Techniques to measure the characteristic properties of aerosols have been developed that use the information contained in the ratio of the backscattered polarization components of the scattering phase function at angles between 130 and 170 degrees [7-10]. Forming this ratio eliminates the problems caused by different extinction losses along different atmospheric paths. Calculations of the scattering phase functions for the case of single scattering by spherical particles of a known refractive index is rather straight forward from the solution developed by Gustav Mie a century ago, see Fig. 3. We initially focused our efforts on bistatic lidar to measure the aerosol properties during the early 1990's, and this proved satisfactory for uniform aerosol distributions, such as horizontal paths through light fog and haze. Soon after, we developed the multi-static approach for use in vertical profiling of aerosol layers.

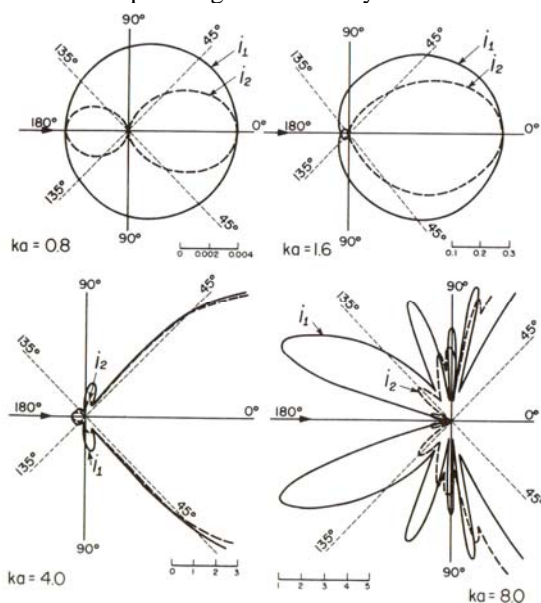


Figure 3. The calculated values of angular scattering intensity of the polarized components viewed parallel ( $i_1$ ) and perpendicular ( $i_2$ ) to polarization plane [11].

## 2. BACKGROUND

Improved understanding of the processes controlling climate change requires new sensors and techniques to measure the concentrations of chemical species and the characteristics/properties of the aerosol distributions. A proposal was submitted to NASA in 2004 to investigate atmospheric aerosols and 'greenhouse' gas distribution on a global scale using the new techniques we were developing. The Species Spectra & Aerosol Scatter Instrument (SSASI) proposal was intended to advance the technology from TRL3 to TRL6 by developing sensors, and by performing a set of scaled experiments using aircraft. That proposal was not accepted; however, the ideas are now presented to the scientific community for consideration and possible future development. Here the original 2004 plan for the NASA Instrument Incubator Program is summarized, along with some added innovations and a couple of recent simulations.

During the last few years, we have worked on the development of the supercontinuum lidar and used it to measure the chemical species present in the atmosphere [12-14]. Experiments have demonstrated the value of using a supercontinuum laser transmitter for DAS measurements in the VIS and NIR, and we continue to work on preparing an MWIR source for measurements in the 3-5  $\mu\text{m}$  spectral window. The multi-static lidar capability now includes multiple scattering conditions and scattering experiments have been extended to include side and forward scattering measurements [15].

## 3. APPROACH TO THE EXPERIMENT

The goal is obtaining measurements of the species concentrations of greenhouse gases and aerosol characteristics at globally distributed locations over a period of two or more years. The elements of the observing system include:

- (1) Well developed data sets obtained at about 15 sites, which are located to represent environmental factors of latitude, urban/rural, coastal/inland, desert/forest, and polluted/clean conditions. The locations near a couple of major cities would allow measurements during times when the urban plume is measured, and also when the background flow is present. Each site would have a moderate size Raman lidar (approximately the capability of our LAPS instrument) for local profiles of the water vapour, temperature, optical extinction and backscatter profiles, and ozone. The Raman lidar would be operated routinely and would have an automated mode for vertical eye-safe ultraviolet operation. Each site would also have an array of six simple imagers, arranged along and across the meridian, to record the multi-static polarization intensity.

- (2) The site would also have an Nd:YAG laser and MWIR laser providing wavelengths at 355, 532, 1064, and 4000 nm that are combined into a multi- $\lambda$  beam, which is accurately pointed and stepped to several points along the arc of the orbit in the sky. At the ground site, the intensities and polarization of the transmitted beams are recorded for analysis of the aerosol size, number density, and shape function.
- (3) A supercontinuum laser is also located at the site, where it points toward and tracks the satellite. It has the capability of tuning to a center frequency between 3 and 5  $\mu\text{m}$  with a bandwidth selection of 100 to 500 nm. The intensity and spectrum of the transmitted beam is measured and used with the satellite absorption spectrum to determine gas species concentration. Spatial distribution of the gas species is determined as the satellite scans along its arc through the sky.
- (4) A small telescope on the satellite is pointed at the ground site using on-board GPS data. The telescope would serve three functions: (a) collect signals for an onboard MWIR spectrometer that measures path integrated chemical species concentration, (b) collect the signals of the 355, 532, 1064, and 4000 nm scattered intensities from the step-to-point-ahead beam as a function of the angle, for both the parallel and perpendicular polarized components of the beam transmitted from the ground, (c) image the scattered beam to describe the vertical profile of aerosol scatters.

The diagram in Fig. 3 illustrates the satellite orbit and shows the small telescope that will point at the ground site to record data. The intensity of the signals measured at the satellite will generally be many times greater than the background signals. The MWIR intensity from the supercontinuum laser will dominate the signal because its wavelength is at the minimum between the solar scatter and Earth emission intensity curves. Also, narrow-band filters are used to select the three wavelengths of the Nd:YAG laser. The diagram in Fig. 4 indicates the station coverage that may be expected during one day. It should be possible to gather data on several orbits each day at each station.

Measurements of extinction between ground and space at the UV, VIS and NIR regions provide results on absorption and scattering as indicated in the simulations shown in Fig. 5. The scattered intensity in the forward direction is not as rich in information as the backscatter signal, but simulations show that results in the forward scatter lobe measured at several wavelengths will provide excellent information on the aerosol properties of size, number, type and some information on shape, as well as the optical extinction.

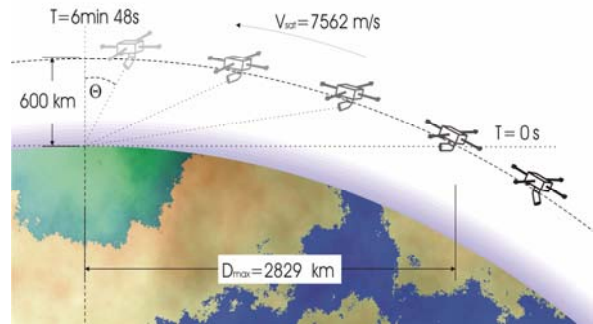


Figure 3. The orbit of the satellite is illustrated to indicate instrument pointing at the ground site.



Figure 4. The typical coverage for a mid-latitude site with the satellite above the horizon indicates a couple of passes/day should be expected.

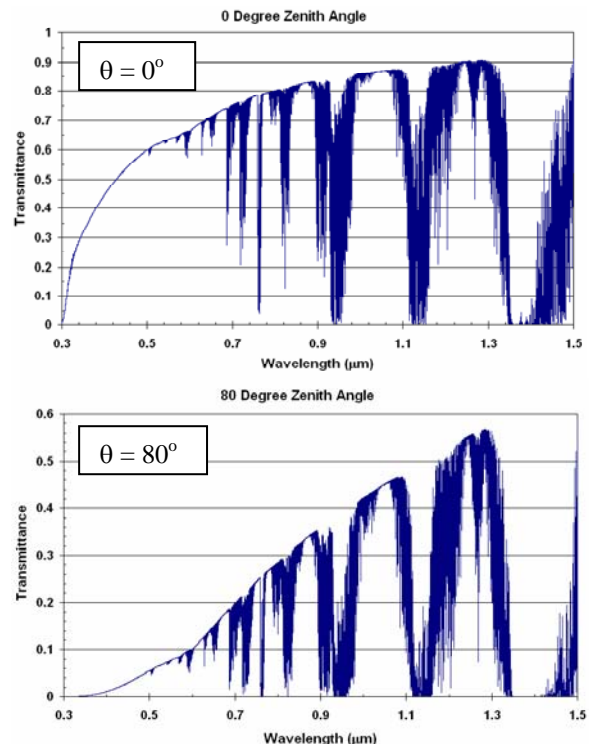


Figure 5. The atmospheric optical transmission between ground and space is shown from a MODTRAN<sup>TM</sup>4 model calculation for atmospheric paths at 0° and 80° zenith angles.

At the MWIR wavelengths, most of the molecular species of interest for research on climate change have unique spectral signatures that are easily measured to determine the species concentrations. Also, this region has the lowest background of solar and thermal radiation. The 2-5  $\mu\text{m}$  transmittance spectra for the path from ground to space path are shown in Fig. 6. Two regions of the spectra are expanded in Fig. 7 to better see the individual spectral lines that are used to analyze results using DIAL or DAS techniques. Many of the analysis advancements made by researchers using hyper-spectral techniques are applicable here.

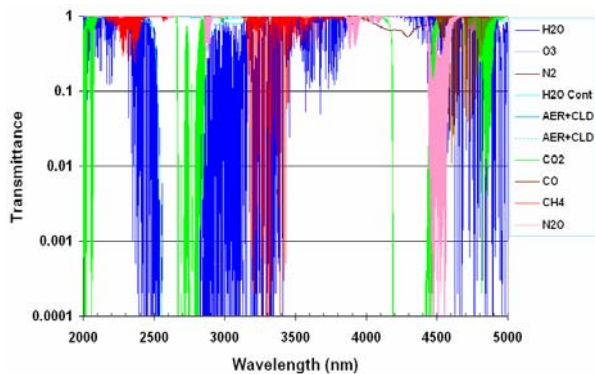


Figure 6. The optical transmittance of the MWIR between ground and space shows absorption lines for use in calculating species densities.

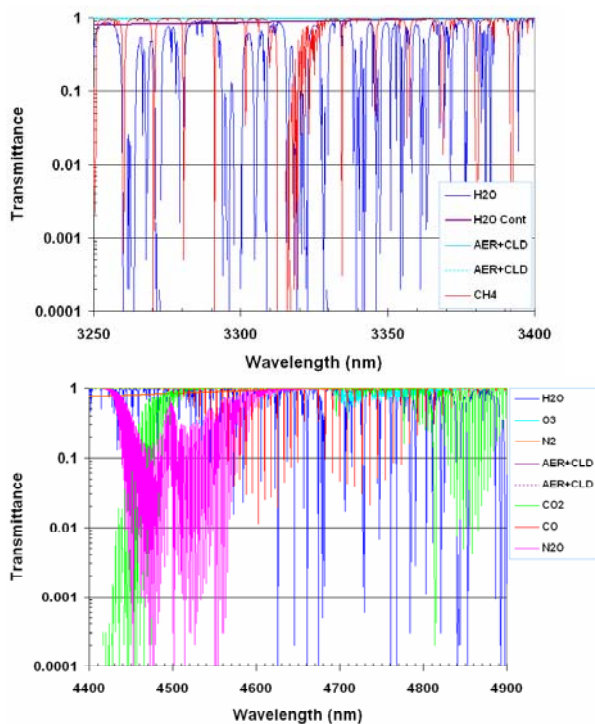


Figure 7. Two of the spectral regions in Fig. 6 are expanded more clearly see the many spectral lines that can absorb the supercontinuum laser emission between ground and space.

The MWIR spectra can be used to measure the atmospheric concentrations of  $\text{H}_2\text{O}$ ,  $\text{CO}_2$ ,  $\text{CO}$ ,  $\text{O}_3$ ,  $\text{CH}_4$  and  $\text{N}_2\text{O}$ . Daily measurements of these species concentrations from globally distributed sites will make a most valuable data set for analyzing the contributions of the greenhouse gases and determining the regions of sources and sinks of the chemical species.

The aerosol experiment uses the angular distribution of the scattered intensity, the polarization phase function measurements, and the path extinction at four wavelengths (355, 532, 1064, 4000 nm) to characterize aerosol properties. The scattering information is most useful when applied to a range of particle size versus wavelength values with ratio between 0.1 and 10. Thus, we expect the selected wavelengths are most useful for characterizing aerosols in the size range between 30 nm and 40  $\mu\text{m}$ . Fig. 8 shows the scattering intensities in the back and forward scatter directions for a 500 nm light beam scattering from 1  $\mu\text{m}$  spherical aerosol particles. Fig. 9 shows the intensity as a function of angle for the several wavelengths between UV and MWIR.

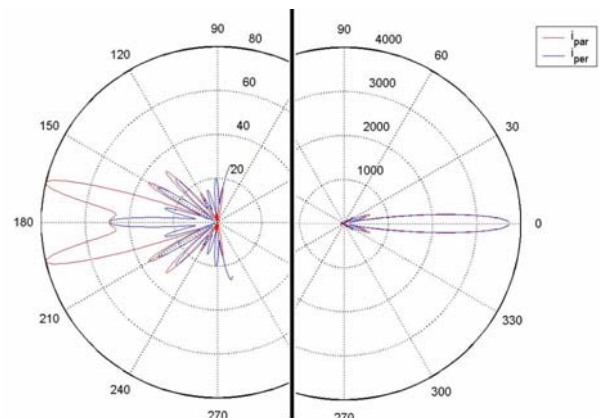


Figure 8. Scattering intensity distributions expected for 1  $\mu\text{m}$  particles scattering 0.5  $\mu\text{m}$  polarized light in directions parallel and perpendicular to the polarization plane (note the change in scale).

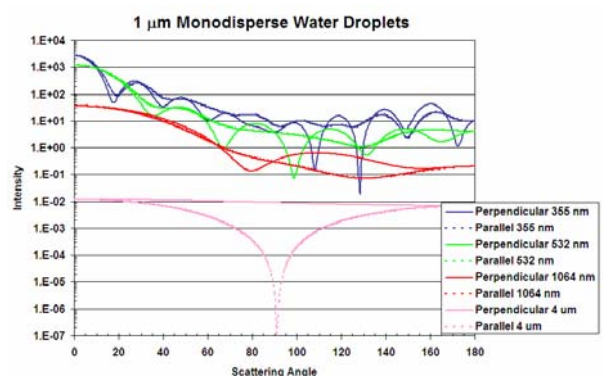


Figure 9. The intensity versus scattering angle is shown for 1  $\mu\text{m}$  aerosols illuminated by 355, 532, 1064 and 4000 nm laser emission.

The calculations shown in Fig. 9 describe the full range of scattering angles for the case of monodispersed water vapor aerosols. In Fig. 10, the scales are expanded to show the forward scatter intensity in the 0 to 20° range for 1 μm and 10 μm aerosols. In the case of satellite measurements, only the forward scattering intensity is measured. Of course the backscatter signal should be simultaneously measured at the ground level for analysis of the near-field aerosol properties. The approach is to point to a location on the orbit path about 15 to 20° ahead of the satellite and make measurements as the satellite motion scans the angles. After the satellite reaches the nearest coincidence point, then the beam is repositioned to a location another 15 to 20° ahead on the satellite path. This procedure should provide six to eight independent measurements during the pass, and thus allow a measurement of spatial variations. The analysis will take into account the changing path and the fact that the aerosols are not monodispersed. We have been able to separate two simultaneously present aerosol size groups while using a single scattering wavelength.

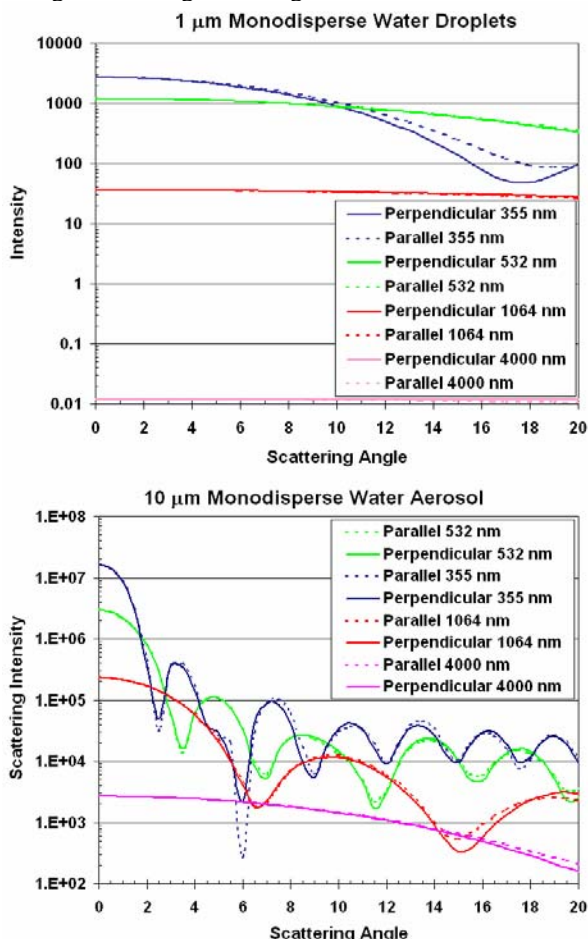


Figure 10. The forward scattering intensity expected for 1 μm and 10 μm aerosols measured at the several wavelengths selected.

Aerosol measurements from satellites are difficult to interpret as evidenced from the fact that different groups have retrieved different aerosol properties from the same data because of different assumptions used in the analysis process. A 2007 workshop in Bremen, Germany has begun the process of addressing these problems [16]. An additional problem that does not appear to have received sufficient attention is the difference in the processes for radiation transfer through different particle size distributions of aerosols. The radiative transfer is quite different through layers of 1 μm and 10 μm aerosols as is quickly evident from examining Fig. 10. Another factor which needs attention is the contribution of the sub-visual aerosols that are present at the edges of clouds, and in the regions where clouds are in their growth and dissipation phases. These small aerosols do participate in scattering of the short wavelength solar radiation; however, they are not detectable with currently used sensors and are not mapped as part of the cloud cover parameter. Haze layers are also difficult to measure, but their effect should be considered as a contribution to the overall radiative forcing. These factors, haze layers and edges of clouds, can be better observed and described by using the wavelength range and sensors described here.

The approach of using a number of ground-based laser transmitter sites that are co-located with the Raman lidar for standard meteorological measurements will result in a valuable data set. The implementation of this approach should provide a cost effective way to measure the optical, chemical, and aerosol scattering properties that are needed for the model and simulation developments. The measurements from space with a well defined reference source have major advantages compared with the hyper-spectral measurements, which rely on reflected sunlight. The complications associated with convolving the variable optical backgrounds affecting source transmission into and through the atmosphere with the surface reflectance and the properties of the backscattered path to the satellite instrument require that several assumptions be made. The type of measurement parameters suggested here provide results that are more directly applicable and should be more useful for developing models and testing simulations used in climate change predictions.

The original work on this project was carried out while one of us (Philbrick) was at Penn State University. We acknowledge co-investigators on the original proposal; Zhiwen Liu, Sven Bilén, and William Brune of Penn State University, Yang Zhang of NC State University, and Philip Hopke of Clarkson University. Also many thanks go to several PSU graduate students who worked on developing experiments described and performing calculations in support of this effort, special thanks to David Brown, Andrea Wyant, and Perry Edwards.

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