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Optical methods appear to be a desirable technology for detection, primarily because such technology is rapid, the components are inexpensive, and the techniques can be automated, requiring no chemical reagents. Such methods are based on the fundamental principles of electromagnetic interaction with small particles, i.e., measurement and analysis of the scattered light.

The meeting aim was to bring together the experts working in different fields of science to discuss the current state of the art and to formulate new research directions. The conference aim was to facilitate a more intensive development in these topics:

- 1. Aerosol Composition
- 2. Instrumentation
- 3. Aerosol Optical Modeling
- 4. Remote sensing

The meeting took place at Congress Centre Smolenice of the Slovak Academy of Sciences in Smolenice, Slovak Republic.

For further information see the conference home page http://www.ocaa2013.sav.sk/

ABSTRACTS

Aerosol cloud interaction measured with MODIS and in situ data

Irshad Ahmad¹, Mika Komppula², Harri Portin², and Sami Romakkaniemi¹

¹ Department of Applied Physics, University of Eastern Finland, P.O. Box 1627, 70211, Kuopio (Finland) ² Finnish meteorological institute Kuopio unit, P.O. Box 1627, 70211, Kuopio (Finland) tel: +358 40 355 3922, e-mail: irshad.ahmad@uef.fi

Abstract

Aerosol induced changes in cloud droplet number concentration (CDNC) was analyzed using direct ground based in situ measurements from Puijo measurement station Kuopio (Finland), and Moderate Resolution Imaging Spectroradiometer (MODIS) instrument onboard the Aqua and Terra satellites. Both the in situ measured and MODIS retrieved CDNC are clearly dependent on the number concentration of aerosol particles for low level clouds observed at the station. On average in situ measured and MODIS retrieved CDNC was 217 cm⁻³ and 171 cm⁻³, respectively. Both CDNC observations depend on accumulation mode aerosol number concentration (N_{acc}) in a similar fashion. In both cases CDNC is clearly dependent on N_{acc} values lower than 400 cm⁻³.

1 Introduction

Low level boundary layer clouds are playing an essential role in earth's radiation budget. Beyond local meteorology, their radiative properties are also dependent on cloud droplet microphysical properties. CDNC depends on aerosol particle size distribution and chemical composition, temperature, and on the updraft velocity prevailing at the cloud base [1]. For a given liquid water content the increase in Cloud Condensation Nuclei (CCN) results in a decrease of cloud droplet size leading to increased reflection of solar radiation back to space (First indirect effect (1st AIE)) [2].

Aerosol cloud interactions have been directly measured in several studies. However, most of the work done used small temporal scale which provides small data sets using airborne techniques. Although the aircraft flights provide exact in clouds measurements of cloud properties, the cost of such flights is a limiting factor for long term observation. The ground based measurements suffer from fixed location and thus the measurements can usually be considered to be representative for limited area only. To overcome these problems we can use remote sensing, and especially satellite observations. However, such techniques still suffer from severe limitations, but together with ground based measurements satellites can provide valuable information on aerosol cloud interactions.

In this work we have analyzed low level continental boundary layer clouds observed at Puijo measurement station (62°54'34" N, 27°39'19" E), Kuopio (Finland). The station is providing continuous aerosol, clouds, and weather data since June, 2006. Cloud and aerosol properties are measured using a Cloud Droplet Probe (CDP) and Differential Mobility Particle Sizer (DPMS), respectively [3]. Puijo station (Puijo tower) is a good place for the long term observation of aerosol-cloud interactions as the tower is surrounded by clouds during 15% of the days. We have investigated the aerosol and cloud droplet data collected between the years 2006 and 2012. We have analyzed dependence of cloud droplet size distribution parameters on the aerosol size distribution; and we will also continue the work for studying the effect of aerosol composition, CCN, origin of air masses, and meteorological parameters on cloud properties.

2 **Results**

We have used one hour averages of N_{acc} (particles having diameter > 80 nm) measured with DMPS, and CDNC from CDP. These instruments are located in Puijo measurement station. For remote sensing technique we used MODIS onboard Aqua and Terra satellites for the retrieval of 1°x1° spatial

scale cloud droplet effective radius and cloud optical thickness, which were used to calculate CDNC. The mean value of CDNC observed at Puijo was 217 cm⁻³, while MODIS retrieved CDNC was 171 cm⁻³. These values are not far apart from each other and the dependence of CDNC on N_{acc} is similar from both measurements, as shown in Fig. 1. A running mean of 30 data points is also shown in Fig. 1 to make the trend visible for readers. As expected the scatter in case of MODIS data is more as compared to direct ground based data. The effect of aerosol on CDNC is clear for smaller values of N_{acc} . The dependence of CDNC on N_{acc} is insensitive after CDNC exceeds 215cm⁻³. Although the dependence of CDNC on aerosol is clear using only aerosol number concentration, the inclusion of information on chemical composition could reduce the scatter seen in Fig 1. This will be the topic of our further studies.



Fig 1: Validation of the MODIS retrieved aerosol cloud interaction using Puijo measurement station. Top panel is for MODIS data while bottom panel shows ground based in situ data. Blue lines represent 30 data points running mean.

Acknowledgments

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Characterization of aerosol particles with digital holography

Matthew J. Berg

Department of Physics & Astronomy, Mississippi State University, 355 Lee Boulevard, Mississippi State, Mississippi 39762 USA tel: +001 785-317 3378, fax: +001 662 325-8898, e-mail: mberg81@gmail.com

Abstract

This work will present several original ideas involving digital holography that, when combined, could characterize flowing aerosol particles *in situ* including size, shape, and composition information. Such characterization is possible because the measured hologram contains the magnitude *and* phase information of a particle's scattered wave, and thus, unambiguous reconstruction of particle images can be done. To extend this imaging to include particle-material information, a photothermal technique is described that may allow the infrared absorption of a particle to be measured.

1 Introduction

In past work by this group, the digital holographic imaging of aerosol particles is achieved with an experimental apparatus is shown in Fig. 1, which involves two subsystems: particle sensing and hologram recording. An aerosol stream is delivered via a nozzle to a measurement volume where an optical trigger senses the presence of a particle [1,2]. When one is sensed, a pulsed laser is activated, illuminating the particle and forming a hologram on an optoelectronic sensor such as a Charged Coupled Device (CCD) camera. The light is then brought to a focus several mm from the nozzle outlet. In this way, a particle is illuminated by what is approximately a diverging spherical wave originating from the beam waist. The beam continues to expand, along with the particle's forward-scattered light, until reaching the CCD sensor. Here, the unscattered and scattered light interfere and the resulting pattern is recorded. This interference pattern constitutes the digital hologram, t^{holo} .



Figure 1: Digital holographic imaging of aerosol particles in situ. See text for further explanation.

To generate an image of a particle from a measured hologram, the Fresnel-Kirchhoff (FK) diffraction theory can be used to simulate the diffraction of light through the digital hologram. This diffraction reconstructs the near-field wave of the particle, the absolute value of which yields an image analogous to that obtained with conventional optical microscopy. An example of the holographic imaging of a ragweed-pollen cluster is shown in Fig. 1, where the top plot displays the hologram, the middle is the reconstructed particle image, and the bottom is an optical microscope image of the *same* cluster for validation.

2 Photothermal perturbation

To extend this holographic-imaging concept to include information related to material composition, a particle is illuminated by UV laser pulses twice in rapid succession while also being exposed to light from a pulsed infrared IR laser. This forms two holograms on the CCD sensor. Performing the computational reconstruction, i.e., the FK diffraction integral, on this double-exposed hologram yields an image of the particle superimposed with interference fringes. These fringes originate from any change in the particle's physical state that occurs between the two UV pulses. Absorption of the IR light by the particle will cause a photothermal perturbation, which will register as the fringes in the reconstructed image. Since the degree of perturbation varies with composition, the fringes will contain information related to the particle material.

3 Conclusion

Preliminary work to simulate the digital holograms of a particle undergoing photothermal expansion will be presented. Such results provide a sense for the feasibility of the basic concept. In addition, progress toward the construction of a proof-of-principle experiment to demonstrate this concept will be given.

Acknowledgments

This work is supported by the U.S. Army Research Office.

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The use of Optical particle counter as sizing instrument and as reference instrument for PM regulated monitoring

B. Bergmans¹, F. Lenartz¹, L. Spanu¹ and G. Gerard¹

¹ ISSeP, 200 rue du chera. 4000 Liege, Belgium tel: +32 472 76 13 96, fax: +32 42 52 46 65, e-mail: b.bergmans@issep.be

Abstract

Optical particle counter (OPC) could be used for determining the level of particle in ambient air. This paper presents two different utilizations of the instrument. The use of an OPC as equivalence method for PM regulated monitoring in a network configuration and the use of an OPC as sizing instrument for particle granulometry determination of the coarse fraction. In the first part of the paper the complete equivalent procedure conducted at ISSeP to prove the equivalence of OPC Grimm EDM180 is presented. The equipment has been tested in 4 different locations following all requirements of the guide of equivalence. Calibration equation obtained and uncertainties of the different measurements performed are also discussed. The second part of the paper presents typical correlation between SMPS and OPC measurements in their commune size range. Combination of both instruments results to obtain the full granulometric pattern is also conversed.

1 Introduction

1.1 OPC as candidate equivalence method for PM monitoring

To fulfill with European regulation (European Air Quality Directive 2008/50/EC), PM10 and PM2.5 level should be monitored by all member states. The reference method consists in collecting particles on filter after separation by impaction and gravimetric determination of the collected mass as described in EN12341 [1] and EN14907 [2]. This method requires a lot of work and is not really practice in a network configuration. That's the reason why another method could be used if equivalence has been proved following a specific guide (GFE) [3] approved by the Directive. Optical particle counters (OPC) are really good candidates. These instruments are relatively cheap, robust and require low maintenance. They also provide data for both class sizes with a low time resolution which allows peak measurement and the possibility to fix alert level. Nevertheless only few networks use this type of instruments because of the difficulty to prove the equivalence in all locations and configurations. Indeed, OPC is a counting device and the particle count need to be converted into mass by calculation using a range of assumptions the validity of which may vary due to the nature of the particles and the ambient condition during sampling. This paper presents the comparison of OPC Grimm EDM180 with the reference method both for PM10 and PM2.5 and the obtained equivalence calibration equations.

1.2 Correlation between OPC and SMPS

OPC provides information in number in a range from around 0.3 μ m up to 30 μ m. Due to the high diminishing of the Mie scattering light with decrease of particle size, these instruments could not be used for ultrafine particles. For this purpose a SMPS is typically used and provides information in the range from a few nm up to 1 μ m. Nevertheless, OPC can be used as a complement of the SMPS to obtain information on the coarse fraction which could be interesting in some specific health studies. OPC and SMPS can also be compared in their commune size range and both data combined to have a continuous pattern of the granulometry of the sample.

2 Fascinating results

2.1 OPC as candidate equivalence method for PM monitoring

2.1.1. Equivalence calibration equation

Comparison of Grimm EDM180 with the reference method has been performed at 4 different locations (2 urban backgrounds, 1 rural and 1 industrial sites). These sites are representative of the entire Walloon network. 4 campaigns have been conducted at each location and include some days where high concentrations are observed. High variation in atmospheric conditions (humidity and wind) is also observed in the selected period. The final aim was to cover all conditions and be representative of the whole network all over the year. The number of paired measurements is minimum 40 per site with a total of around 200 for the complete study. All the applied procedure fulfill with the recommendations of the guide for equivalence. The campaign initially performed in 2009 and 2010 has been done again in 2011 and 2012 and gives equivalent conclusions.



Figure 1: Grimm EDM180 calibration curve for PM2.5 and PM10.

2.1.2. Uncertainty of the measurement during the equivalence procedure

The procedure for equivalence describes the maximum uncertainty which is allowed. Uncertainty should be first tested intra-method to insure that both the candidate and the reference method are under control. Every instruments have thus been doubled in the measurement campaign to achieve a good time recovery, but also to allow this intra-comparison. Uncertainty obtained for the reference method was 0.93 and 0.89 μ g/m³ respectively for PM10 and PM2.5 which is below the limit value of 2 μ g/m³ fixed in the GFE. The limits for candidates are 2.5 μ g/m³ and the value obtained were respectively 1.56 and 1.44 μ g/m³. The expanded combined uncertainties of the candidate method have been calculated at the limit concentration value of 50 μ g/m³ for PM10 and 30 μ g/m³ for PM2.5. These uncertainties should remain below 25% to fulfill with the Directive. 13% and 18% was respectively achieved for PM10 and PM2.5. We can thus conclude that Grimm EDM180 is equivalent to the reference method when using the calibration equation determined during this equivalence campaign (see 2.1.1).

2.2. Correlation between OPC and SMPS

Total particles number in the range 250 - 800 nm has been measured using an OPC Grimm 1.109 and a SMPS. Typical response is showed in Figure 2. Several measurements were performed at different locations and seasons to highlight possible effect on the result, but no significant variation has been observed.



Figure 2: Typical pattern obtained with both instruments in their commune size range

Correlation between instruments is quite good. A Pearson coefficient of correlation R = 0.94 has been obtained for the whole set of data collected during 9 months of sampling. Uncertainties of both methods are estimated around 10% and data from both instruments can be combined to obtain the full granulometric spectrum.



Figure 3: Correlation between OPC and SMPS in their commune size range

3 Conclusion

Equivalence between OPC Grimm EDM180 and reference method has been established both for PM10 and PM2.5 in different localizations and configurations. The obtained calibration equations are presented. The results obtained have been confirmed by two other campaigns and similar results have been obtained.

For PM10 the average slope of the equivalence curve is really close to 1 with a very low variation from site to site which proves that OPC is a good candidate method.

OPC could also be used in conjunction with an SMPS as a sizing instrument if information on coarse particle is interesting. A Pearson correlation coefficient R = 0.94 has been observed between OPC and SMPS for their commune sizing range and this value is not much influenced by the site or

the ambient conditions. An uncertainty around 10% is expected for both instrument and data can be combined to obtain the full granulometric spectrum.

Acknowledgments

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Iasi aerosol mixtures, one year time-scale overview: AERONET data

Mihai Marius Cazacu¹, Adrian Timofte², Florin Unga¹ and Silviu Octavian Gurlui¹

¹ "Alexandru Ioan Cuza" University of Iasi, Faculty of Physics, Atmosphere Optics, Spectroscopy and Lasers Laboratory, 11 Carol I Blvd., 700506 Iasi, Romania, sgurlui@uaic.ro ² National Meteorological Administration, Regional Forecast Center Bacau, 1 Cuza Voda Str., 600274 Bacau, Romania

Abstract

Starting from 7th of May 2012 a CIMEL sun-photometer (CE-318) implemented within AERONET (AErosol RObotic NETwork) program was installed in Iasi County, under the management of LOA-SL department (Atmosphere Optics, Spectroscopy and Lasers Laboratory of Faculty of Physics, "Alexandru Ioan Cuza" University of Iasi). This paper presents optical proprieties analysis of different types of aerosols and the season variability of them in 1 year of measurements, data correlated with meteorological factors, HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) models, MODIS-FIRMS models (Moderate Resolution Imaging Spectroradiometer - Fire Information for Resource Management System) and LIDAR data. The major categories of aerosol types are evidenced, such as urban/industrial aerosol, biomass burning and dust.

1 Introduction

Iasi city is one of the largest cities and a municipality from North-Eastern part of Romania. The city being in continuous development, from this springer several urban projects have started to improve the streets, the alleys and rehabilitation of some cultural buildings and parks [1].

The instrument used for this study is a Cimel Automatic Sun Tracking Photometer CE 318, solarpowered, weather-hardy, robotically-pointed sun and sky spectral sun photometer, [1, 2], located at the Astronomical Observatory from Iasi (Latitude: 47.19306° North, Longitude: 27.55556° East, Elevation: 175.0 meters) [2].

Complementary research to the sunphotometer measurements obtained data has been performing by means of LIDAR technique. The LIDAR data was used to monitor the atmospheric aerosols and clouds in the troposphere [4, 5].

2 Results

Preliminary results are shown in Figure 1. It can be observed that urban/ industrial aerosols have a major presence with big influences of biomass burning aerosol type. The recorded data are selected at Level 1.5 with zenith angle larger than 50° and retrieval error less than 5% during 1 year.

As the literature says in Gilles et all., 2012, the Figure 2: Single Scattering Albedo at 440 nm dependence of wavelength and absorption (often expressed as Absorption Ångström Exponent – AAE) dependence of Extinction Ångström Exponent (EAE), which is an indicator of particle size, confirm the influences of biomass burning over urban/industrial aerosols load [3].



Figure 1: Single Scattering Albedo (440 nm) dependence of Extinction Ångström Exponent (440 – 870 nm).

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Figure 2: AERONET LOA-SL IASI Site – Single Scattering Albedo (SSA) Monthly Average (left), Absorbing Ångström Exponent (AAE) 440 – 870 nm dependence of Extinction Ångström Exponent (EAE) 440 – 870 nm, 1 YEAR of DATA (may 2012 – may 2013)

3 Conclusion

One year data of sunphotometer measurements were analyzed. Data show that over Iasi area, Romania, urban/industrial aerosol and biomass burning have strong influence over entire aerosols load. Complementary data will taking into account using several meteorological factors, HYSPLIT's, LIDAR data and MODIS – FIRMS investigations, too.

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Fast laser imaging optical emission spectroscopy: aerosols types detection and ranging

Silviu Octavian Gurlui¹, Adrian Timofte², Florin Unga¹ and Mihai Marius Cazacu¹

¹ "Alexandru Ioan Cuza" University of Iasi, Faculty of Physics, Atmosphere Optics, Spectroscopy and Lasers Laboratory, 11 Carol I Blvd., 700506 Iasi, Romania, sgurlui@uaic.ro ²National Meteorological Administration, Regional Forecast Center Bacau, 1 Cuza Voda Str., 600274 Bacau, Romania

Abstract

Depending on the space-time chemical composition, sources type activity or process, meteorological conditions, air pollutants (both organic and inorganic compounds), generally the air pollution causes significant damage to human health and environment and dramatically influencing the climate changes. These pollutants may include some of the following well known chemical compounds: sulphur dioxide, ozone, carbon dioxide, volatile organic compounds, hydrogen sulphide, particulate matter, carbon monoxide, toxic air contaminants (mercury, lead, etc) but their contributions in the time-scale atmosphere dynamics are far from fully understood as following discussed details.

1 Introduction

Aerosols have a significant impact on severe convective storms, electric phenomena, e.g. their intensity, location or type (rain or hail). Therefore, atmospheric electric phenomena were demonstrated to be sensitive to convective cloud, fog, rain and snow. Complex self-organization tropospheric behaviour included a lot of chemical organic and inorganic compounds aerosols, variable sources type activity, meteorological conditions, budget radiations, clouds liquid droplet and ice nuclei formation. [1-2]. The impacts of urban-enhanced aerosol concentrations is the research subject of high interests that include models approach because they may induce a significant turbulence on the dynamics, microphysics, convective but also upon the storm development and precipitations. A complementary characterisation of aerosol (Rayleigh, Raman scattering, and fluorescence) included the chemical composition, size particle space-time distribution, temperature and humidity profiles, etc is generally hampered by some limitation of remote sensing techniques. [3-5]. As a complementary approach to the satellite measurement spectral data, ground based instruments as Sunphotometer, Radiometer, DOAS, LIDAR & DIAL, mass-spectroscopy & shadowgraphy – laser matter desorption, in a variety of configurations are extensively developed and some of these must be improved even to obtain UV-IR image – remote investigated ice nucleation laver-atmospheric pattern, as following will be proposed in this paper [2].

2 Results

Moreover, in order to better understanding the fundamental of some critical physico-chemical transformation of the atmosphere compounds but also for applications point of view, we propose a new LIDAR power instrument as is shown in Fig. 1. This instrument is able to capture fast plume airborne image (2ns gate time) and may used to investigate in real time several chemical compound behaviour at a given point of the free atmosphere. Some advantage of the proposed instrument may summarized as following: time-space resolved temperature and humidity profiles (Raman scattering of oxygen 353 nm & 354 nm; real-time monitoring tool to measure concentration of sulphur dioxide injected in the troposphere from the volcanic eruption ash, water profiles and their dynamics, acid rain, chemical transformation/physical properties; DIAL H₂O and SO₂; UV fluorescence using the two stronger absorption emission lines of 218.9 nm and 220.8 nm, respectively; water vibrational Raman scattering at 407 nm; ice nucleation variability, physical and chemical impact, the influence upon the

mixed-phase cloud; dust mineralogical composition dynamics influences upon the ice nucleation; Raman scattering of oxygen – 353 nm & 354nm; fast ICCD imagery (IR spectrum) of ice nucleation pattern clouds; studies of the ice nucleation and propagation in plants; detection and characterization of carcinogenic BaP or PAH compounds adsorbed on the soot in a given smoke plume, bacterial and fungal spores, biogenic aerosols generally that have the potential to fluorescence; UV fluorescence of PAH {Benzo(ghi)perylene Excitation (Ex-301 nm/ Emission Em-420 nm); Naphthalene (Ex-275 nm/ Em-325 nm); Anthracene (Ex-255 nm/Em-375 nm); Pyrene (Ex-270 nm/ Em-420 nm), etc).



Figure 1: Experimental set-up. Optical emission spectroscopy: laser desorption, detection and ranging.

3 Conclusion

In this paper we imaged several laser-spectroscopy techniques in order to improve remote & detection, resolved optical emission spectroscopy, coupled with laboratory simulation, in order to study various atmospheric phenomena: storms, lighting and plasma gas mixture discharge, laser-condensed matter interaction, etc.

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Past aerosol campaigns and what we must improve

Helmuth Horvath¹

¹ University of Vienna, Faculty of Physics, Aerosol and Environmental Physics 1090 Vienna, Austria tel: +431 4277 51177, fax: +431 4277 9511, e-mail: Helmuth.Horvath@univie.ac.at

Abstract

Aerosol campaigns have already a long tradition. An early and very successful campaign was ACHEX (1970-1972). The advantage of a campaign is the concentration of measuring capabilities during the study, and the possibility for extensive modeling; the disadvantage are the limitations in time and space, which might not be representative for the aerosol on a regional or global scale. Besides expertise on instrumentation, the campaign needs a careful planning, professional management, and sufficient time for post-processing, to be successful. Examples for campaigns are given.

1 Introduction

One of the wise sayings of the Greek philosopher Aristoteles (384 to 322 BC) is: *The whole is greater than the sum of its parts*. This can be applied without any restriction to aerosol campaigns. Many campaigns have taken place so far and the outcome has been always positive.

In nowadays experimental science, so much specialization has taken place, that practically no person will be able to have all technologies on hand to characterize the aerosol considering all aspects. Therefore it is natural, that a group of scientists intensely studies the problem, usually in an intensive campaign lasting weeks to months. The campaign needs a careful planning, careful management and also funding. Sufficient time should be available for the evaluation of the data, to draw and conclusions which are beyond the details of the measurements.

In the following, a few campaigns will be described, obviously the list cannot not be exhaustive, rather it should show, what is possible.

2 Examples for campaign

ACHEX (California Aerosol Characterization Experiment). In the mid 1960s Aerosol science has developed to a point that particle size distributions between 0.05 and 30 µm could be measured continuously, size segregated sampling with subsequent chemical analysis was possible, optical measurement of the atmosphere were available. At the same time the smog in Los Angeles was a nuisance. The groups being able to characterize the aerosol got together for a short joint campaign in 1969 with promising results, triggering the ACHEX experiment 1970 to 1972. The outcome of the experiment was among others the multimodal mass size distribution, a successful chemical mass balance, mechanisms of aerosol formation in the atmosphere and optical properties derived from mechanical/chemical properties.

VISTTA (Visibility Impairment from Sulfur Transformation and Transport in the Atmosphere, 1979). The SW of the US is known for its good visibility. Both ground based and airplane measurements characterized the aerosol and possible influences on visibility by power plants. The outcome of this study is: The power plant plume appearance is dominated by NO₂, secondary aerosol formation can be neglected up to 100 km from the plant.

ACE (Aerosol Characterization Experiments 1995 – 2004; ACE-1, ACE-2, ACE-Asia) integrates a network of ground stations, intensive field measurements, satellite observations and models. The main purpose of this set of experiments is the study of the aerosol, its transport and its effect on the radiative balance, which e.g. is 30 W/m^2 for Asia for the clear sky. Investigations on the indirect effect have also been performed.

Besides these large projects many smaller investigations, limited to a specific geographic region or special scientific questions have been preformed: Just to name a few: LACE 98 (Lindenberg Aerosol Characterization Experiment, 1998, Germany), VELETA 2002 (eValuation of the Effects of eLevation and aErosols on the ultravioleT rAdiation, Granada, Spain); Special land based campaign on aerosols, India, February 2004; CARES (Carbonaceous Aerosols and Radiative Effect Study) Calfornia, June 2010.

An interesting project is the Aerosol Campaign for schools 2013/14, which is part of the GLOBE (Global Learning and Observation to Benefit the Environment).

3 Conclusion

The concentration of scientific power of many participants in an aerosol campaign in a usually gives broader insight into the properties, transformation, evolution, effects of the aerosol. Due to the special and temporal variation of the aerosol still only snapshots of the aerosol can be obtained. But campaigns can be used for supplementing or validating other observation methods such as satellite observation. The spatial variation can be coped with by using a large number of observations, which can be made possible with simple observation in schools.

A large campaign needs a professional management and sufficient time for the evaluation of the data.

Lab scale set-up for a behavioral study of iodine aerosols

Hee-Jung Im*, Jei-Won Yeon, and Kyuseok Song

Nuclear Chemistry Research Division, Korea Atomic Energy Research Institute, 150 Deokjin-dong, Yuseong-gu, Daejeon 305-353, Republic of Korea tel: +82 42 8684740, fax: +82 42 8688148, e-mail: imhj@kaeri.re.kr

Abstract

Iodine is responsible for the main short-term radiological impact on health through the ¹³¹I isotope and its organic chemical form over long-term releases. However, iodine and the retention of gaseous organic iodine are seldom discussed owing to a lack of knowledge and experimental data. They are commonly known to move in gaseous or several types of aerosol forms. Therefore, to study how iodine-related aerosols are formed and act in the containment of a nuclear power plant and further in the environment, a lab-scale set-up including iodine generating and introducing equipment, a water droplet generator, and an aerosol collector or a sorbent testing tub was installed as a single system with a steady control.

1 Introduction

Iodine is a relatively high fission-yield product of spent fuel, and the monitoring of radionuclides is important in keeping a close watch on nuclear facilities, not only during their operation but also during their decommissioning. Volatile nuclides can readily interact with steam and water droplets in a nuclear reactor, and the contaminated aerosols formed can be exhausted in air during emergency venting or after nuclear power plant damage [1]. The released aerosols can further interact both homogeneously and heterogeneously with tropospheric particulate matter, such as smoke, sea salt aerosols, or cloud droplets.

2 **Results and Discussions**

Since iodine can be easily released into the environment and accumulated in the human body owing to its high volatility, an evaluation of the formation and behavior of the contaminated aerosols and a stabilization and determination technique of the aerosols are essential.

The methods to generate iodine species (such as I_2 and CH_3I) that are typically determined in contaminated exhaust air were properly chosen, and water droplets with constant particle sizes were generated and evaporated using the Model 3450 Vibrating Orifice Aerosol Generator (TSI Incorporated, USA) [2]. The generated volatile iodine gases were introduced into the water droplet to look into all

aerosol related behavior, and several spectroscopic methods were applied to analyze the aerosols qualitatively and quantitatively. Moreover, for the efficiency testing of the sorbents [3], which are designed and synthesized in our laboratory for the localization of radioactive aerosols and volatile radionuclides, a capillary tube filled with a synthesized material was also added to a venting tube of the aerosol particle output. The experimental setup shown in Figure 1 was first tested using iodine gases with water droplets of well-characterized aerosol particles.



Figure 1: Schematic of set-up for a behavioral study of iodine aerosols.

Acknowledgments

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Extracting the aspect ratio of atmospheric aerosols from sun and sky photometry data

Miroslav Kocifaj¹

¹ ICA, Slovak Academy of Sciences, Dúbravská Road 9, 845 03 Bratislava, Slovak Republic tel: +421 5930 9267, fax: +421 2 54773548, e-mail: kocifaj@savba.sk

Abstract

The sun and sky photometry are valuable tools that are almost routinely used in collecting the information on aerosol particles. Most typically the size distribution or refractive index of aerosol particles can be inferred from multispectral and/or multiangle optical data. To determine the aspect ratio as a function of the particle radii is highly non-trivial task since the kernel of a particular integral equation is a non-linear function of sought solution. The methodological as well as numerical solution concepts to this problem are introduced in this paper showing that aspect ratio can be determined iteratively.

1 Introduction

The long-term observations in the Earth's atmosphere have proven that aerosol particles of different origin coexist in complex multicomponent populations. When interacting with the solar radiation these particles influence significantly the spectral and angular behaviors of the radiative field near the ground. There is no doubt that the microphysical properties of aerosols play important role in many physical and optical processes undergoing in the local atmosphere.

It is well recognized that aerosol particles of different sizes have different residence times. For instance, sub-micrometer sized aerosols can remain suspended for up to several weeks in the air [1] while other particles can be readily removed by rain. For instance, the urban pollution is increasingly dependent upon motor vehicles that emit particles with sizes between 50 and 200 nm which could have a residence time of about 1 week in the atmosphere. Due to processes of formation these particles can become highly non-spherical. In metropolitan areas, the dust load is directly related to changes in particle size distribution [2]. However, the non-sphericity as a function of particle size is completely unknown quantity even if the composition of these particles is known or inferred indirectly. Therefore there is a high demand for any direct or indirect methods that can be used to identify the characteristic sizes of oblate/prolate aerosol particles.

Neither natural nor anthropogenic processes can favour production of ideally spherical and homogeneous particles. To avoid large complexities with characterization of particle shapes, the concept of aspect ratio is often taken into consideration. Here the aspect ratio relates the largest and smallest characteristic size of arbitrarily shaped particle. One of easiest ways how to simulate the nonspherical particles with different aspect ratios is to employ mathematically well-defined geometries, like ellipsoids. In this study we introduced a solution method for extracting the aspect ratio of atmospheric aerosols from sun and sky photometry data. The aspect ratio is determined as a function of particle size.

2 Iterative technique in solving the inverse problem

As found by other authors, the aspect ratio may become dependent on particle radius [3]. In such a case the optical quantity O_i (e.g. the aerosol extinction coefficient or the scattering phase function) can be formulated as follows

$$O_{i}(x) = \int_{0}^{\infty} K_{i}(r, x, a) f(r) a(r) dr, \qquad (1)$$

where K_i is the kernel of the integral equation, r is the particle radius, f(r) is the function interpreted as modified size distribution, and a(r) is used to characterize the size-dependent aspect

ratio. Assuming O_i is the aerosol extinction coefficient, the parameter x is usually the wavelength. For O_i being the scattering phase function, the parameter x is either the scattering angle or the wavelength. If the mean refractive index of aerosol particles is *a-priori* known, both f(r) and a(r) can be recovered from two data-functions $O_i(x)$ and $O_j(x)$. The iterative technique we used starts with zero approximation for $a(r) = a_0(r) \equiv 1$ that is used in retrieval of $f_0(r)$ based on the measured aerosol extinction data. The product of $f_0(r)$ and $K_i(r, x, a_0)$ forms the kernel of the integral equation for scattering phase function. The first approximation to the a(r), i.e. $a_1(r)$, is determined using the kernel that is a non-linear function of aspect ratio. However, the inversion of this kind of Fredholm integral equation is possible after applying a special mathematical concept (not shown here). The iterations are completed when $||a_n - a_{n-1}|| < \varepsilon_a$ as well as $||f_n - f_{n-1}|| < \varepsilon_f$. Here ε_a and ε_f are predefined error margins. It has to be noticed that there is no guaranty for successful convergence of a(r) and f(r), especially when the measurement errors are large enough or when chosen particle model doesn't fit the real conditions during the experiment.

3 Conclusion

The information on size-dependent aspect ratio a(r) is highly important in understanding the aerosol production, interaction, evolution, and also in understanding the underlying physics (including physical and chemical processes occurring in the local atmosphere). The iterative approach we applied in retrieval of a(r) and f(r) can be extended to other quantities, such as mean dielectric function of aerosol particles.

Acknowledgments

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Optical efficiency of tubular light guide under different aerosol scattering phase functions

Ladislav Kómar¹, Igor Kohút², Martin Bednárik², and Miroslav Kocifaj¹

 ¹ ICA, Slovak Academy of Sciences, Dúbravská Road 9, 845 03 Bratislava, Slovak Republic
² Geophysical Institute, Slovak Academy of Sciences, Dúbravská cesta 9, 845 28 Bratislava, Slovak Republic

tel: +421 5930 9267, fax: +421 2 54773548, e-mail: kocifaj@savba.sk

Abstract

The optical efficiency of a straight light tube is analyzed accepting various models for aerosol scattering phase functions. The angular distribution of light beams entering the light pipe is determined by means of modified method of successive orders of scattering. Both first and second scattering orders are taken into consideration when computing the light field near the ground. The numerical computations we made are advantageous in designing the light-tube installations and other competitive solutions that utilize light as an alternative source of energy.

1 Introduction

Monitoring, quantifying, and interpreting the sky states become increasingly important in the last decades, especially in metropolitan areas where the daylight is considered as an alternative source of illumination in interior spaces. It is well recognized that the natural light is well tolerated by humans and therefore the systems for delivery of daylight to the interiors appear attractive in both public and private buildings. A hollow light pipe can collect the natural light, guide the beams through the straight pipe from its top to bottom, and distribute the light to the room depending on the optical interface mounted at the ceiling. The amount of light and its temporal variability mimic the state of the light field outdoors.

The large effort is therefore expended to search for a more universal and scalable daylight model that accepts the actual meteorological situation. There is no doubt that omission of parameters characterizing the atmospheric and surrounding environments causes a large uncertainty in classification of skies. Bellia et al. [1] considered the surface albedo, cloud index and turbidity factors as environmental parameters. In addition, Perez et al. [2] introduced sky clearness and brightness indices and statistical functions to simulate random luminance patterns for cloudy skies. In spite of evident temporal and spatial variability of environmental characteristics, many of existing models are rather based on empirical formulae.

Traditionally, the illumination engineers model the sky luminance as a product of two functions – indicatrix and gradation [3]. Such a split has its origin in former solution of light scattering problem for a very clean atmosphere [4]. It is well-known that angular behavior of scattered light in a non-turbid molecular atmosphere is formulated in terms of Rayleigh theory, which dictates the indicatrix to be proportional to the single scattering phase function $p(\mathcal{G}) \propto 1 + \cos^2 \mathcal{G}$. Here \mathcal{G} is so-called scattering angle. The formal gradation function and $p(\mathcal{G})$ can be then simply separated, implying that $p(\mathcal{G})$ works as a function modulating the total sky radiance/luminance. In principle, the theoretical luminance depends on transmission coefficients measured toward sun and sky element [5]. However, the real optical state of the atmosphere can neither be simulated by molecular scattering nor by single scattering approximation. Even if we confine ourselves to cloudless conditions, the diffuse component of ground reaching radiation can alter quite markedly [6]. One of the primary reasons for such large variability is heterogeneity of airborne aerosol particles [7]. The aerosols belong to most unstable atmospheric constituent influencing the electromagnetic radiation in visible spectrum. Therefore, the information on aerosol particles is important for reasonable estimations of radiance/luminance patterns and for modeling the light tube at its entrance.

2 Solution method

To predict the angular distribution of scattered light in a turbid atmosphere, the radiative transfer equation has to be solved subject to boundary conditions. However, a presence of broken clouds makes this problem extremely difficult because of 3D geometry. Note that 3D radiative transfer schemes are difficult to solve because of theoretical and numerical complexities. In light of this fact, an exact 3D solution appears to be unattractive for illumination engineering applications. To overcome these difficulties we have developed a theoretical solution based on the method of successive orders of scattering thus enabling CPU non-intensive computations.

3 Conclusion

The angular distribution of light beams entering the light tube at its top has been computed based on the method of successive orders of scattering. Consequently, the relation between optical efficiency of a hollow light pipe and the aerosol phase function was analyzed showing that aerosol particles can alter the light field in interiors depending on diameter, length and optical properties of the light tube. The results of our case-study can contribute to designing the light-tube installations through a more appropriate prediction of daylight availability in interior spaces.

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Retrieving radial inhomogeneities in particle composition of single, levitated aerosol particles using Mie resonance spectroscopy

Ulrich K. Krieger¹, Daniel M. Lienhard¹, Sarah Steimer^{1,2} and Yiea-Funk Te¹

¹ Institute for Atmospheric and Climate Science, ETH Zürich, Zurich, Switzerland ² Laboratory of Radiochemistry and Environmental Chemistry, Paul Scherrer-Institute, Villigen, Switzerland tel: +41 44 633 4007, fax: +41 44 633 1058, e-mail: ulrich.krieger@env.ethz.ch

Abstract

We use elastic Mie resonance spectroscopy to experimentally investigate radial inhomogeneities in water concentration of model organic aerosol particles. When the timescale of aerosol particle equilibration with respect to water vapor is limited by the diffusion of water in the condensed aerosol phase, strong gradients in water concentration will develop. Inverting kinetic Mie resonance spectra to temporal concentration gradient data may yield a better understanding of the underlying diffusion process.

1 Introduction

Recent observations have indicated that organic aerosol particles in the atmosphere may exist in an amorphous semi-solid or even solid (i.e. glassy) state, e.g. [1]. The influence of highly viscous and glassy states on the timescale of aerosol particle equilibration with respect to water vapor have been investigated for some model systems of atmospheric aerosol, e.g. [2,3]. In particular, it has been shown that the kinetics of the water absorption/desorption process is controlled entirely by liquid-phase diffusion of water molecules for a highly viscous aerosol particle. A liquid phase diffusion model based on numerically solving the non-linear diffusion equation predicts strong internal gradients in water concentration when condensed phase diffusion impedes the water uptake from the gas phase. Here we study the internal concentration gradients in single, levitated, micron size aerosol particles of aqueous shikimic acid using elastic Mie resonance spectroscopy.

2 Experimental

A single aqueous shikimic acid particle is levitated in an electro-dynamic balance (for details see [2]), dried for several days at room temperature, cooled to the target temperature and exposed to a rapid change in relative humidity. In addition to measuring the elastically backscattered light of a "white light" LED source and recording the full spectrum with a spectrograph as in [2], we use a tunable diode laser (TDL) to scan high resolution TE- and TM spectra. Since we perform the experiment at -10° C the changes in the Mie-spectra due to water uptake are sufficiently slow to resolve the kinetics of water uptake.

3 Results

Fig. 1 shows the two exemplary Mie spectra (LED based, panel (a) and TDL based, panel (b) and the temporal shift of different Mie resonance modes upon humidification (panel (d) and (e)). Upon water uptake all resonances shift towards higher wavelength (red shift) in accordance with the growth of the particle. However all modes show a blue shift at a particular time, which is distinctly different for different mode orders (see panel (c)). Modes of lower order (narrow width) exhibit the blue shift earlier compared to the modes of higher order number (larger resonance width).



Figure 1. (a) High resolution Mie resonance spectrum (TM-polarization) measured with TDL scanning, three different modes are labeled. (b) Low resolution Mie resonance spectrum measured with LED source and spectrograph OMA detection. (c) RH vs time (left scale), Onset time of blue shift of the various modes labeled in (a) and (b) (right scale). (d) Resonance wavelength vs time: mode (1) black, mode (2) gray, mode (3) light gray. (e) Resonance wavelength vs time: mode (4) black, mode (5) gray.

3 Discussion

We will compare the observed Mie spectra with Mie calculations [4] for the concentration profile originating from the diffusion model and various assumptive concentration profiles and discuss the inversion of temporal Mie resonance spectra to concentration profiles.

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Influence of Saharan dust events on scattering phase function

Marie Laborde^{1,2}, Grant Kassell², Anne Kasper-Giebl³, Ingrid Meran^{3,4}, Ulrike Nickus⁵, Regina Hitzenberger⁵, Anna Wonaschütz⁵, Grisa Mocnik⁶, Luka Drinovec⁶, Gerhard Wotawa⁷, Gerhard Schauer⁷

> ¹AerosolConsultingML,Ennetbaden, Switzerland ²Ecotech Pty Ltd., Knoxfield, Australia

³Institute of Chemical Technologies and Analytics, TU-Vienna, Vienna

⁴Institute of Meteorology and Geophysics, University Innsbruck, Innsbruck

⁵Aerosol Physics and Environmental Physics, University Vienna, Vienna

⁶Aerosol d.o.o., Kamniska ulica 41, Slovenia

⁷Central Institute for Meteorology and Geodynamics, Wetterdienststelle Salzburg, Salzburg tel: +41767559648 e-mail: marie.laborde@aerosolconsulting.com

Abstract

The atmospheric aerosol is a complex mixture of particles of different chemical composition and physical properties. Both characteristics affect the way particles interact with the sun's radiation. The asymmetry parameter, "g", is often used in climate change models to parameterize the relative amount of light scattered towards the earth's surface as opposed to scattered back towards the light source. This parameter is often approximated using the backscattering measurement of a nephelometer and the Heyey approximation [1]. A more accurate parameter (although more demanding during computation) to use would be the aerosol function, defined as the amount of light scattered as a function of the scattering angle θ . Measuring the particles phase function in ambient conditions is a challenge due to the rather low intensity of the light scattered by the particles within the defined solid angle. It is however important since little is known about the influence of the particles' properties (especially their shape) on the phase function. Highly non-spherical particles such as Saharan dusts are therefore interesting to study in this sense. The dataset presented here contains data from the Sonnblick Observatory, Austria and aims at investigating the influence of particle shape on the aerosol phase function by looking at several Saharan dust events.

1 Introduction

The atmospheric aerosol is a complex mixture of particles of different chemical composition and physical properties. Both characteristics affect the way particles interact with the sun's radiation. As an example, their shape, size and refractive indices determine the amount and the direction of the light scattered, also known as one of the aerosol first effect. The asymmetry parameter, "g", is often used in climate change models to parameterize the relative amount of light scattered towards the earth's surface as opposed to scattered back towards space. This parameter is often approximated using the Heyey approximation [1] although a better parameter (although more demanding during computation) to use would be the aerosol function. The aerosol function $P(\theta)$ is defined as the amount of light scattered as a function of the scattering angle θ .

Measuring the particles phase function in ambient conditions is a challenge due to the rather low intensity of the light scattered by the particles within the defined solid angle. It is however important since little is known about the influence of the particles' properties (especially their shape) on their phase function. Highly non-spherical particles such as Saharan dusts are therefore interesting to study in this sense. The dataset presented here contains data from the Sonnblick Observatory, Austria and aims at investigating the influence of particles shape on the aerosol phase function by looking at several Saharan dust events.

2 Instrumentation and setup

The aerosol's physical properties were characterized at the Sonnblick Observatory using several instruments. A Scanning Mobility Particle Sizer (Vienna Type DMA) measured the aerosol size distribution below 1 μ m while an APS (TSI 3321) monitored particles in the size range of 0.5 to 20 μ m diameter. The total scattering, integrated scattering over a different range of angles and the backscattering coefficients were measured by an Aurora 4000, Ecotech nephelometer. This measurement allowed the calculation (by subtraction) of the particles' phase function. The aerosol light absorption was measured using an Aethalometer (Magee Scientific AE 33).

3 Results

The time periods featuring Saharan dust events (SDE) were first isolated from the rest of the dataset by looking at the wavelength dependence of the aerosol single scattering albedo [2]. For each time period (during SDE and outside SDE), the aerosol asymmetry parameter g and the aerosol function were calculated. The asymmetry parameter was calculated using two different methods (i) the abovementioned Andrews's approximation (Eq. 1) and (ii) using the polar nephelometer measurement and its definition (Eq.2).

$$g = -7,143889b^3 + 7.464439b^2 - 3.96356b + 0.9893,$$
 (1)

Equation 1: Asymmetry parameter as calculated using the Henyey-Greenstein approximation [1]

$$g = \frac{\sum_{n=1}^{N-1} \left(\sigma_{neph}^{\alpha_n} - \sigma_{neph}^{\alpha_{n+1}}\right) \cdot \cos\left(\frac{\alpha_n + \alpha_{n+1}}{2}\right) \cdot \sin\left(\frac{\alpha_n + \alpha_{n+1}}{2}\right)}{\sum_{n=1}^{N-1} \left(\sigma_{neph}^{\alpha_n} - \sigma_{neph}^{\alpha_{n+1}}\right) \cdot \sin\left(\frac{\alpha_n + \alpha_{n+1}}{2}\right)}$$
(2)

Equation 2: Asymmetry parameter calculated using the measured scattered light at different angles α_n by the Aurora 4000 [3].

Firstly, it can be seen in Fig. 1 that the Henyey-Greenstein approximation is not evenly distributed and gives significantly different values than the one obtained using Eq. 2.



Figure 1: Histogram of the asymmetry parameter calculated using two different methods. The error bars represent the statistical significance: $1/\sqrt{number of points}$.

The asymmetry parameter (calculated using Eq. 2) and the aerosol phase function were then calculated for the time periods when SDE were observed and for the rest of the dataset (Fig. 2). A clear influence of the Saharan dust events on the asymmetry parameter (Fig.2) and the aerosol phase function can be seen.

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Figure 2: Asymmetry parameter measured during Saharan dust event (orange trace) and outside Saharan dust events (black trace). The error bars represent the statistical significance: $1/\sqrt{n}$ number of points.



Figure 3: Median of the aerosol phase function during Saharan dust event (orange trace) and outside Saharan dust events (black trace).

Conclusion

These results are important to consider for climate change models and will be compared to theoretical values using Mie theory and M-matrix calculations.

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A measuring system for retrieval of urban emission function from nightsky radiance data

Héctor Antonio Solano Lamphar¹, František Kundracik², Miroslav Kocifaj¹

¹ ICA, Slovak academy of sciences, Dúbravská cesta 9, 845 04 Bratislava, Slovak Republic tel: +421 91 8224259, e-mail: lamphar@gmail.com ²Department of experimental physics, 842 48 Bratislava 4, Mlynská dolina - pavilón F2

Abstract

The brightness of the night sky is a common global problem characterized by luminous flux emitted to the *upper hemisphere* and mostly produced by ground-based light sources. Light emitted is generally absorbed and scattered by the atmospheric constituents before it is recorded at the ground. Thus, the remote sensing of emission sources usually requires the information on the optical properties of the atmospheric environment. The authors propose a new technique with intention to retrieve urban emission function from sky glow data. Specifically, the experiments are being performed using a reliable low-cost microcontroller-based system that controls and automates the measuring device.

1 Introduction

It is well-known that the uplight emissions from the ground-based light sources are subject to absorption and scattering processes depending on the optical properties of the atmosphere – in particular seasonally variable particulates suspended in the atmospheric boundary layer. The turbid atmosphere is a heterogeneous medium composed of polydisperse aerosol system which is fairly the greatest modulator of down welling radiation. In such environment, the aerosol optics and the physical properties of the ground-based light sources appear to be decisive factors influencing the sky glow during nightime. Normally, the characterization of the urban emission function is possible through a semi-empirical approach. Nevertheless the empirical formulae have no theoretical foundation and thus cannot mimic real situations.

The main intention of this work is to present a measuring device and system for retrieval of urban emission function from night sky luminance data. The concept of data processing follows the recently developed theoretical model that is based on the experimentally determined ratios of zenith luminance to horizontal illuminance. These ratios have to be measured at a set of discrete distances from the light source. The data recorded then form a base for obtaining the mean emission function and aerosol optical depth concurrently. The light meter device we proposed can operate accurately at various distances even if the radiant intensity varies over several orders of magnitude. The ability to operate the device over such a wide dynamical range is guaranteed thanks to the optical system -using a telephoto lens on the one hand, and mounting with wide opening angle on the other hand- which both together enable the same chip to measure the radiance and irradiance simultaneously. This approach is advantageous for experimentalists who can determine the mean emission function of a ground-based light source using routine low-cost equipment or even using available data archives.

2 Experimental device

The experimental device is composed of a sensor from a DSLR camera and a microcontroller-based system as described below.

Basically, the PC-controlled single-board microcontroller operates a *monolithic photodiode with on-chip transimpedance amplifier* to make the measurements of ambient light possible. A single robotic arm is used to close and open the lens of the optical equipment, implying that the system can be easily adapted to long time exposure avoiding the thermal dependence of the sensor and extracting

any noise with a dark frame picture. The exposure time of every measurement is settled depending of the reading of the photo-detector. The measurements without lens provide the irradiance data, while the system with fixed lens enables to measure the radiance. For the numerical purposes RAW images are taken, so the digitized scans are obtained in a preferred data format. For each scan the average brightness is computed and consequently normalized to the fixed exposure time.

3 Experiments vs. theory

In principle, a set of field campaigns is necessary to verify if what we measured in the atmosphere is the same as what is obtained numerically. The targeted experiments have to be made in the same night but at different distances from a city. In this phase of system tests we considered the emission pattern to satisfy Garstang's function (1986) [1], so the retrieval method we implemented reduces to the determination of both Garstang's scaling parameters and AOD.

4 Conclusions

The illumination engineers as well as the night-sky modellers constantly require adequate data on the radiant or luminous intensity distribution (usually the data of zenith luminance/radiance and horizontal illuminance/irradiance); therefore, the research focused on the emission function is pertinent since the sky glow problem can be better understood and counteracted. Nowadays, various approximations to the night-sky radiances (or empirical models of night-sky luminance patterns) are in use, but they are more-or-less inapplicable in retrieval of emission function.

The main purpose of the present work is to implement and verify the theoretical method for inversion of the sky luminance in order to obtain the urban emission function. To do this a dedicated measuring device has been designed and used in routine measurements.

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Particle-to-particle variation in scattering by mineral dust: From simplified models to realistic, inhomogeneous particles

Hannakaisa Lindqvist¹, Timo Nousiainen^{1,2}, and Karri Muinonen^{1,3}

 ¹ Department of Physics, University of Helsinki, P.O. Box 48, FI-00014 Helsinki, Finland
² Finnish Meteorological Institute, P.O. Box 503, FI-00101 Helsinki, Finland
³ Finnish Geodetic Institute, Geodeetinrinne 2, P.O. Box 15, FI-02431 Masala, Finland tel: +358 40 7255 651, e-mail: hannakaisa.lindqvist@helsinki.fi

Abstract

The performance of simplified models in modelling scattering by single mineral dust particles is quantified using the stereogrammetrically characterized inhomogeneous model shapes as the most accurate available representation of mineral dust. A comparison reveals that neither the averaged scattering properties nor the particleto-particle variation are realistically reproduced by any simplified shape, although the Gaussian random spheres model depolarization considerably well.

1 Introduction

Light scattering by mineral dust has been successfully modelled using distributions of simplified shapes, such as spheroids or ellipsoids. However, recent studies [1] show that simplified models may perform inconsistently; for example, the best-fitting shape distribution at one wavelength may perform poorly on another wavelength. Indeed, it appears that the good performance of the simplified model particles is artificial, due to the versatile base they offer for fitting as a distribution, rather than a close resemblance in scattering on individual particle level [2].

A unique approach to realistic modelling of scattering by mineral dust was presented by [3]: they retrieved the shapes of four dust particles directly from a stereo pair of electron microscope images using stereogrammetry. The inhomogeneous composition of the particles was derived by energy-dispersive spectroscopy. Light scattering properties for these wavelength-scale particles were computed using the discrete-dipole approximation. As these results are very accurate representations of scattering by single mineral dust particles, they are ideal for testing the performance of simplified models at single-particle level. As simplified models, we have selected here an equal-volume sphere, a spheroid for which the aspect ratio has been retrieved from that of the stereogrammetric shape, and single realizations of the Gaussian random sphere [4], whose shape statistics have been retrieved from the stereogrammetric shapes.

2 Results

Scattering by the simplified models was compared to that of the inhomogeneous stereogrammetric shapes of a calcite, a dolomite, a silicate, and an aggregate particle. The results were averaged over a lognormal size distribution, where the largest particle size considered had a volume equivalent diameter of 2.8 μ m at the simulation wavelength of 550 nm. Intensity, the degree of linear polarization, and the depolarization-connected S_{22}/S_{11} for the particle models are presented in Fig. 1. As can be seen, the simplified shapes do not succeed in producing either the particle-averaged or the natural particle-to-particle variation of scattering by mineral dust particles. As an exception, however, the Gaussian random spheres appear to model depolarization successfully. Spheres performed quite poorly and have been left out for the clarity of the plots. The impact of inhomogeneity was also tested for the stereogrammetric shapes with an effective refractive index and found small compared to the differences seen in Fig.1.



Figure 1: Intensity (left), the degree of linear polarization (middle), and depolarization-connected S_{22}/S_{11} (right) for light scattered by a lognormal size distribution of wavelength-scale model particles. The averaged scattering by inhomogeneous stereogrammetric shapes is depicted with a black line, enveloped by the particle-to-particle variation in gray. The envelope and the vertical bars are drawn from minimal to maximal values obtained.

3 Conclusion

The stereogrammetric characterization of mineral dust particles utilized in conjunction with the discrete-dipole approximation computations provides a unique opportunity to study the variation of scattering between single wavelength-scale dust particles. A descriptive simplified model should mimic not only scattering by an ensemble of dust particles but also produce realistic particle-to-particle variation, which was tested through comparisons with scattering by these stereogrammetric shapes. It was found that, even though the simplified shapes were retrieved from the dust particle shapes, their scattering did not respond to that of dust either as single particles or averaged over a small ensemble.

Acknowledgments

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Volumetric current integral equation formulation for modeling scattering by atmospheric aerosol particles

J. Markkanen¹, H. Lindqvist¹, T. Nousiainen², K. Muinonen^{1,3}, and S. Järvenpää⁴

 ¹ University of Helsinki, Department of Physics, Finland
² Finnish Meteorological Institute, Earth Observation Unit, Finland
³ Finnish Geodetic Institute, Masala, Finland
⁴ Aalto University, Department of Radio Science and Engineering, Finland tel: +358 504160605, e-mail: johannes.markkanen@helsinki.fi

Abstract

The aim of this presentation is to introduce a numerical method for modeling light scattering by irregular particles such as atmospheric aerosol particles. The method is based on the electric current volume integral equation formulation (J-VIE) which is accelerated by a multilevel fast multipole algorithm (MLFMA) giving O(N log N) type of scalability for the operation count as well as memory consumption. The accuracy of the method is compared with the accuracy of the discrete dipole approximation (DDA) based code in the case of scattering by a single aerosol particle.

1 Introduction

In the optical community, the most widely used numerical method for solving scattering by irregularly shaped particles is the discrete dipole approximation (DDA). The DDA is based on an integral equation formulation in which the volumetric dipole moments are approximated with finite elements, and the point matching technique is applied to convert the continuous equation into a discrete one. This, however, is not the most optimal scheme, and it has been argued that the convergence in the norm of the solution cannot be guaranteed with the employed testing scheme [1]. A robust discretization of the volume integral equation (VIE) is known as a J-formulation in the engineering community, and it has proven to be very effective for modeling complicated materials. In this presentation, we compare the solutions produced by the standard DDA code (ADDA) [2] and the J-VIE formulation accelerated by the MLFMA [3].

2 J-VIE formulation

Consider scattering by an inhomogeneous and anisotropic dielectric object bounded by a domain D with permittivity ε located in free space. The electric current volume integral equation can be written as

$$J^{inc} = J - (\epsilon - 1)(\nabla \nabla + k^2 I) \int_{V} GJ \, dV, \tag{1}$$

in which G is the free space Green's function, I is the unity dyad, and J^{inc} is the incident volumetric current. The above equation is a Fredholm integral equation of index zero from $L_2(D)$ (function space of square integrable functions in D) into itself if the permittivity ε is different from -1 and 0 [4]. This means that a stable numerical method can be obtained if the domain of the operator and the range of the operator are forced to be in $L_2(D)$. Choosing the testing functions from the L_2 -dual space of the range of the operator (the L_2 is dual to itself), Galerkin's method leads to a discrete system which approaches the original equation as the discretization density increases [1]. It is worth noting that the DDA uses point matching, i.e., the testing functions are Dirac delta distributions, hence it is not clear that the discrete system preserves the mapping properties of the original integral equation. This motivates us to study whether there is a significant difference in the solution accuracies between J-VIE and ADDA.

3 Numerical example and discussion

Our numerical example is an aerosol particle consisting of calcite, ammonium sulfate, hematite, and kaolinite with the volume fractions of 98%, 0.7%, 0.7%, and 0.7%, respectively [5]. The wavelength is 550 nm, and the volume equivalent size parameter is 4.



Figure 1: Comparison of the J-VIE MLFMA solution and the ADDA solution.

Computations were carried out with a desktop computer, and in the ADDA the particle was discretized with 800k dipoles, and in the J-VIE around 100k tetrahedral elements were used. An excellent agreement in far-scattering quantities is observed when the discretization is dense enough. It is notable that tetrahedral elements provide more flexible geometrical modeling than uniform cubical elements, and therefore, less elements can be used in the J-VIE. However, the FFT-based acceleration in the ADDA consumes less memory than the MLFMA acceleration in the J-VIE. Thus, a systemic analysis is needed to understand the actual error behavior with respect to the discretization density and computational demand.

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Multiple Scattering of Light by Large Complex Particles

Karri Muinonen^{1,2}, Antti Penttilä¹, and Gorden Videen³

 ¹ Department of Physics, University of Helsinki, P.O. Box 64, FI-00014 Helsinki, Finland
² Finnish Geodetic Institute, Geodeetinrinne 2, P.O. Box 15, FI-02431 Masala, Finland
² Space Science Institute, 4750 Walnut Street, Suite 205, Boulder, Colorado 80301, United States tel: +358 9 19122941, e-mail: karri.muinonen@helsinki.fi

Large complex atmospheric particles show two ubiquitous effects near backscattering: linear polarization parallel to the scattering plane (negative polarization) and a nonlinear increase of brightness toward the backscattering direction (opposition effect). The phenomena are modeled using a radiativetransfer coherent-backscattering method (RT-CB) applied to a realistic aggregate model for a large particle. The direct modeling utilizes a large number of polydisperse Mie scatterers randomly distributed within a spherical volume of a pre-defined packing density. For the comparison of the RT-CB and superposition T-matrix methods, the reader is referred to the recent work by Muinonen et al. (ApJ 760, 118, 2012). The inverse modeling makes use of the so-called phenomenological fundamental single scatterers (Muinonen & Videen, JQSRT 113, 2385, 2012). When applied to interpret the direct computations described above, the inverse modeling allows us to put constraints on the single-scattering albedo, phase function, and polarization characteristics as well as the mean free path length between successive scatterings. The present computational exercise sheds light on the predictive power of the inverse analysis using the phenomenological scatterers. With the help of the Discrete-Dipole Approximation, the T-matrix method, and experimental measurements, the modeling may allow us to constrain the size, shape, and refractive index of the fundamental scatterers within remotely sensed atmospheric particles.

Characterization of mineral dust samples from measurements of scattering matrix elements at two different wavelengths in the visible

Olga Muñoz¹, Fernando Moreno¹, and José Luis Ramos¹

¹ Instituto de Astrofísica de Andalucía, CSIC. Glorieta de la Astronomía, sn. Granada 18008, Spain. tel: +34 958 121311 fax: +34 958 814530, e-mail: olga@iaa.es

Abstract

We present measurements of the complete scattering matrix as a function of the scattering angle of four desert dust samples. The measurements are performed at two different wavelengths (448 and 647 nm) in the scattering angle range from 3 to 177 degrees.

1 Introduction

Desert dust aerosols play an important role in the Earth's radiative balance. Computational characterization of small desert dust particles from the observed scattered light remains a difficult task due to their complicated morphology. Therefore, laboratory measurements are valuable references. In this work we experimentally study the complete scattering matrices as functions of the scattering angle of four desert dust samples. The measurements are performed at the IAA Cosmic Dust laboratory [1] at two different wavelengths namely, 448 and of 647 nm, in the scattering angle range from 3 to 177 degrees.

2 Discussion

The desert dust samples studied in this work correspond to two different deserts, Sahara and Gobi desert. Two of the samples, *Sahara Tunisia1* and *Sahara Tunisia2*, were collected directly from the ground in two locations in south Tunisia. The samples were collected in recesses that are able to trap airborne fine clay-fraction particles [2]. The third sample, *Sahara OSN*, was collected during a Sahara sand event at the Observatory of Sierra Nevada (OSN) in Granada, Spain. The OSN is located at 2896 m above sea level in Sierra Nevada Mountains at a distance of more than 1500 km from the source. The fourth sample, *Gobi Beijing*, was collected in Beijing during a desert dust storm. The *Sahara OSN* and *Gobi Beijing* samples can be considered as airborne samples since they were collected at long distances from the source.

The color of the samples ranges from whitish for the *Sahara Tunisian1* to dark brown for the *Gobi Beijing* sample including reddish and light brown for the *Sahara Tunisian2* and *Sahara OSN* samples, respectively. The volume distribution of the four desert dust samples is measured with a Mastersizer 2000 from Malvern instruments. The Mastersizer uses either Mie theory or Fraunhofer theory for spheres to retrieve the volume distribution of the sample under study. The main differences between the airborne samples and the samples directly collected from the ground as far as the measured size distribution is concerned is that the airborne samples show narrower size distributions.

By means of comparison of the measured scattering matrix elements of the four desert samples with previous measurements presented in the Amsterdam-Gramada Light Scattering Database [3], we will constrain which elements of the scattering matrix can help in distinguishing different type of aerosols in the atmosphere of the Earth. In particular, we will see whether it is possible to distinguish between volcanic ash and desert dust from observations of different elements of the scattering matrix in a certain scattering angle range. The measurements will be available in digital form in the Amsterdam-Granada Light Scattering Database (www.iaa.es/scattering) [3].

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Polarization resolved angular optical scattering of aerosol particles

Brandon Redding¹, Yong-Le Pan², and Hui Cao¹

¹ Department of Applied Physics, Yale University, New Haven, CT, USA ² Army Research Laboratory, Adelphi, MD, USA e-mail: brandon.redding@yale.edu

Abstract

Real-time detection and identification of bio-aerosol particles are crucial for the protection of soldiers against chemical and biological agents that may be intentionally released on the battle fields [1]. The strong elastic light scattering properties of airborne particles provides a natural means for rapid, non-invasive aerosol characterization [2]. Recent theoretical predictions suggested that variations in the polarization dependent angular scattering cross section could provide an efficient means of identifying different airborne particles [3]. In order to experimentally validate this prediction, we built a high throughput, sampling system, capable of measuring the polarization resolved angular scattering cross section of bio-aerosol particles and aggregates. The system records the polarization resolved scattering cross section of a particle or aggregate flowing through an aerosol jet stream in a single shot. We calibrated the system by comparing the polarization dependent scattering cross section of individual polystyrene spheres with the scattering cross section predicted by Mie theory. We then used the system to study five particles types: Polystyrene aggregates composed of 1 µm and 500 nm spheres, BG (bacillus subtilis) aggregates, arizona road dust aggregates, and tryptophan molecules. We extracted the polarization aspect ratio as a function of scattering angle for all five particle types.

1 Introduction

We developed an experimental apparatus to record the polarization resolved angular optical scattering cross section of bio-aerosol particles in real-time. The experimental setup enables a single-shot measurement of the angle resolved scattering cross section for both polarizations from an individual aggregate flowing through the detection setup in an aerosol jet stream. The setup was calibrated by recording the scattering cross-section of individual polystyrene spheres with a diameter of 3 μ m, as shown in Fig. 1. The experimental scattering patterns matched the patterns predicted by Mie theory [Fig. 1(c,d)].

2 Scattering cross section of aggregates

We then recorded the scattering cross section from a series of particle types, including aggregates composed of polystyrene spheres (PS) and aggregates composed of BG. Since the scattering cross section depends on the orientation and position of the particles in the aggregates, we recorded the scattered intensity from many (~200) aggregates of each type and calculated the average scattering pattern. Due to the different shape of the PS and BG, we expect that the scattering cross sections of aggregates composed of these materials will exhibit different polarization aspect ratios, defined as $P=(I_{\perp}-I_{\parallel})/(I_{\perp}+I_{\parallel})$, where $I_{\perp}(I_{\parallel})$ is the scattered intensity in the perpendicular (parallel) polarization. In Fig. 2, we show the polarization aspect ratio for PS and BG aggregates.

3 Conclusion

We have developed a real-time, high-throughput system capable of measuring the polarization resolved angular scattering pattern from individual aggregate flowing through the detection setup in an

aerosol jet stream. We measured the polarization aspect ratio for several aggregate types and found that the polarization ratio depended on the shape of the individual particles within the aggregate.



Figure 1 (a,b) Scattering cross section for parallel (a) and perpendicular (b) polarizations from 3 μ m diameter polystyrene spheres. Each row shows the cross-section extracted from an individual sphere. The average cross section over 20 particles is shown in (c,d) for the two polarizations along with the scattering cross section predicted by Mie theory.



Figure 2: Comparison between the polarization aspect ratio of BG and polystyrene (PS) aggregates. SEM images of the BG and PS aggregates are shown in the inset.

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Hygroscopicity and mixing state of aerosols in the planetary boundary layer

B. Rosati¹, E. Weingartner¹, P. Zieger^{1,2}, M. Gysel¹, and U. Baltensperger¹

¹ Laboratory of Atmospheric Chemistry, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland ² Department of Applied Environmental Science, Stockholm University, Svante Arrhenius väg 8, SE-10691 Stockholm, Sweden tel: + 41 56310 4204 - e mgil: hermadatte regati@psi.eh

tel: +41 56310 4204, e-mail: bernadette.rosati@psi.ch

Abstract

Aerosol particles experience hygroscopic growth at enhanced relative humidities (RH) which leads to changes in their optical properties. The white-light humidified optical particle spectrometer (WHOPS) is a newly developed instrument to specifically investigate the dependence of aerosol light scattering with regard to hygroscopic growth. Moreover, the mixing state of the particles can be inferred by analyzing their water uptake. The airborne instrument was operated onboard of a Zeppelin during the PEGASOS campaigns where vertical profiles were flown within the planetary boundary layer (PBL). First results of a campaign at the Po-Valley show that both, internally and externally mixed particles are present. Besides, the hygroscopic growth is strongly dependent on the origin of the air mass.

1 Introduction

Aerosol particles interact with the incident solar radiation by scattering or absorbing the light. Depending on the chemical composition and size of the particles, this effect is more or less pronounced. If the particles are hygroscopic and a high relative humidity (RH) is present, they will grow and their index of refraction will be altered, which means a significant change in their optical properties [1]. Since the RH is known to change with height [2], vertical profiles are necessary to investigate changes in the optical properties of aerosols.

Another crucial aspect is the mixing state of particles. The common separation is made between internal and external mixtures. The internal mixture describes particles of the same size and same chemical composition, while externally mixed particles are composed by different substances with different physical and chemical properties. Particles that are internally mixed will grow homogeneously with increasing RH, on the other hand the different compounds in externally mixed particles will take up water in different amounts leading to a polymodal growth distributions at elevated RH.

1.1 Development of new instrumentation

The Pan-European Gas-Aerosols-climate interaction Study (PEGASOS) employed a Zeppelin to explore the PBL. Within this project, the white-light humidified optical particle spectrometer (WHOPS) was developed to investigate hygroscopic and optical properties of aerosol particles. This instrument is set up in a way that particles are first dried and then reach a differential mobility analyzer (DMA) where a specific mobility diameter d_{mob} is selected. In the next step these particles are exposed to a defined RH (~ 95%) and are further analyzed with a white-light optical particle spectrometer (WELAS). This instrument measures the scattering cross section (S) of individual particles which can further be converted to an optical diameter. Finally the ratio of the humidified optical diameter to the dry mobility diameter allows the calculation of the hygroscopic growth factor (GF) at a defined RH. In addition, the mixing state can be inferred from the measured GF distributions. As a second option the humidification step can be bypassed, feeding the dry monodisperse particles directly into the WELAS. The comparison between dry d_{mob} and dry optical diameter allows retrieving information on the effective index of refraction of the dry particles.

2 First results of mixing state and growth

The hygroscopic growth and mixing state of aerosols recorded during the three main campaigns in the Netherlands, in the Po-Valley in Italy (both 2012) and in Finland (2013) will be shown. Figure 1 depicts an example of a height profile recorded with the WHOPS (for $d_{mob} = 300$ nm) during the Po-Valley campaign in 2012. No clear differences between the two heights (i.e. at 100 m and 700 m above ground) are visible but it is obvious that there exists a distinct hygroscopic fraction. The broad humidified distributions in this example indicate that the aerosol is to some extent externally mixed. Throughout the campaign internal and external mixtures were recorded.



Figure 1: Data from a flight at San Pietro Capofiume (Italy) on 20.6.2012; a dry d_{mob} of 300 nm was selected; solid and dashed lines: flights at 100 m and 700 m above ground, respectively; upper and lower graph show the dry and humidified distributions, respectively.

3 Conclusions

The newly developed airborne instrument WHOPS characterizes the optical and hygroscopic properties of aerosol particles. The white light particle spectrometer allows the characterization of larger particles (d > 250 nm). As the size of individual particles is measured, the time resolution is high enough for airborne applications. Besides vertical and horizontal profiles of the hygroscopic growth factor, information on the mixing state and the particles' refractive index can be inferred. First results recorded at the Italian Po valley show externally and internally mixed particles with a substantial ability of hygroscopic growth. GF up to approximately 2.2 (RH~95%) were recorded.

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Miniature, low-cost optical particle counters

Z. Ulanowski¹, P. H. Kaye¹, E. Hirst¹, A. Wieser², W. R. Stanley¹

¹ Centre for Atmospheric and Instrumentation Research, University of Hertfordshire, Hatfield AL10 9AB, United Kingdom

² Karlsruher Institut für Technologie, Institut für Meteorologie und Klimaforschung, Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany tel: +44 1707 284604, fax: +44 1707 284185, e-mail: z.ulanowski@herts.ac.uk

Abstract

A range of miniature, low-cost optical particle counters for characterizing atmospheric particles has been developed. They are intended for use with disposable balloon-borne radiosondes, dropsondes or in dense ground-based sensor networks. Versions exist that are suitable for determining the size distributions and number concentrations of cloud particles or atmospheric aerosols such as mineral dust or volcanic ash.

1 Introduction

Routine meteorological data is obtained by profiling the atmosphere using disposable "radiosondes", thousands of which are launched daily on meteorological balloons. These give temperature, pressure, humidity and wind speed. Additional measurements are obtained from so-called "dropsondes", released from research aircraft. However, a crucial property not yet measured for lack of suitable equipment is the size and concentration of atmospheric particulates: cloud particles and aerosols. Instead, indirect, measurements are employed, relying on remote sensing, to meet the demands from not only weather forecasting, but many other areas such as climate research, air quality monitoring, civil contingencies etc. In addition, research aircraft can be used *in situ*, but airborne measurements are expensive. Moreover, aircraft use is restricted to near-horizontal profiling, which is a limitation, as cloud properties are driven by processes taking place in the vertical direction. Therefore, vertical profiling with sondes, either upwards or downwards, is often preferable. There are also many instances when aircraft cannot be used for *in situ* measurements, ranging from common occurrences, such as vigorous storm systems, to the unusual but far-reaching events, such as volcanic eruptions.

The Centre for Atmospheric and Instrumentation Research at the University of Hertfordshire specializes in the development of light-scattering instruments for the characterization of aerosols and cloud particles. In the last few years a range of miniature particle counters for characterizing atmospheric particles has been developed. They are intended for use together with systems such as disposable balloon-borne radiosondes, aircraft dropsondes or in dense ground-based sensor networks. Versions exist that are suitable for determining the size distributions and number concentrations of particles with different size ranges. They have been used for vertical profiling of atmospheric aerosols such as mineral dust, volcanic ash, as well as clouds, and for ground based air-quality monitoring [5,6].

2 Selected results

An early counter was developed in 2009 for profiling atmospheric mineral dust. It was designed for attaching to a standard Vaisala RS92 radiosonde and launching on a small meteorological balloon. It provided a size distribution in five bins and could also be interfaced to an electric charge sensor to detect aerosol charging [1-3]. In 2010 this counter was used to profile ash layers from the Eyjafjallajökull eruption [4]. A number of these units are currently held by the UK Met Office for use in the event of further volcanic ash episodes.

A disadvantage of conventional optical particle counters that draw ambient air in through a narrow inlet is that they can become blocked, which is almost inevitable during measurements in the presence

of supercooled cloud droplets, for example. Therefore, a different counter version was developed, which can have more open-path geometry, as the sensing zone is defined optically rather than being delimited by the flow system. This counter is currently used in a network of about 50 multi-parameter sensor stations for air quality monitoring around Heathrow airport, and for fog characterization. These counters size particles into bins in the range from 0.35 to 17 μ m [5,6]. The counter has now been adapted for use with radiosondes or dropsondes. The dropsonde version has been successfully tested by launching it from research aircraft together with the so-called *KITsonde*, developed at the Karlsruhe Institute of Technology, which determines standard meteorological parameters and GPS position for transmission back to the aircraft [7].

3 Conclusion

Low cost optical particle counters can be used in conjunction with meteorological radiosondes or dropsondes to obtain vertical profiles of the size distributions and concentrations of atmospheric aerosols and cloud particles on a routine basis. They also allow measurements where research aircraft cannot fly safely, as was demonstrated during the Eyjafjallajökull volcanic eruption. Open-path particle counters are especially valuable, as they permit measurements in the presence of liquid water.

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Cavity ringdown spectroscopy for characterization of single aerosol particles

Chuji Wang^{1,2}, and Yongle Pan²

 ¹ Mississippi State University, Starkville, MS, 39759
² US Army Research Laboratory, 2800 Power Mill Rd, Adelphi, MD, 20783 tel: +001 662 325 9455 e-mail: cw175@msstate.edu

Abstract

Cavity ringdown spectroscopy (CRDS) is a high sensitivity and high selectivity laser absorption spectral technique. CRDS can be applied in atmospheric aerosol characterization by measuring absolute optical absorbance, providing supplementary information to those extract from aerosol scattering measurements. Combining the optical trapping technique (OT), CRDS can be implemented to characterize single aerosol particles by measuring wavelength dependent absorption coefficients. We propose to study single aerosol particles using the OT-CRDS technique. Here we describe designs and their configurations of the proposed experimental system. Technical advantages and challenges of the new technique are discussed.

1 Introduction

Most research efforts in aerosol characterization focus on physical properties, such as size distributions. Comparatively, study of chemical properties of aerosol, such as chemical composition, thermodynamics, surface properties, spectral fingerprints, refractive index (the imaginary part), etc. has been traditionally left behind. To achieve a better understanding of aforementioned chemical properties of aerosol particles is ultimately reliant upon a fundamental study of *single* aerosol particles in different sizes, compositions, optical properties (absorbing or nonabsorbing), and surrounding environments using a highly capable technique. We propose to investigate physical and chemical properties of single aerosol particles *at single particle level* using optical trapping-cavity ringdown spectroscopy (OT-CRDS) in combination with conventional aerosol characterization methods/techniques [1-4].

2 Experimental system

Figure 1 shows schematic of the OT-CRDS system. Polystyrene beads $(0.1 - 5 \mu m)$, water droplets, and decane droplets $(5 - 10 \mu m)$ will be used initially to validate the OT-CRDS system. A trapped single particle will be viewed by CCD imaging through illumination of the particle by a laser beam at 532 nm. A single ringdown measurement will takes less than 1 second. A dye laser scanning will be set at 0.02 nm/step to obtain ringdown spectra with a medial resolution. If a ringdown data point is generated from averaging over 100 ringdown events and repetition rate of the YAG laser is 20 Hz, a spectral scan over the coated wavelength range of one set of ringdown mirrors ($\sim 10\%$ of the central wavelength) will take 2-3 hours. This estimate indicates that the needed trapping time (the particle is stabilized spatially) for a wide wavelength scan is as long as a few hours [2]. In this way the single particle in a fixed position (relative to the ringdown beam) will be interrogated continuously in a wavelength range of 30 - 40 nm without changing the system's setting. The wavelength range should be wide enough to cover different wavelength-dependent absorption structures of the particle while scattering is relatively flat (total optical loss due to scattering is a constant). In this way, absorption can be separated from scattering. For particles whose absorption consists of broad bands, ringdown measurements of the particles will be performed by using different sets of mirrors to cover a broader spectral range, i.e. hundreds of nm.



Figure 1. Schematic of the proposed OT system for CRDS measurements of a single aerosol particle. (a) Optical configuration of the dual Bessel beams trapping. (b) Arrangements of the trapping beams and the CRDS beams with respect to the reaction chamber.

3 Discussion and summary

Advantages of the proposed technique include: 1) Total optical scattering loss in CRDS is insensitive to laser wavelength in a narrow spectral band while absorption features are highly wavelength dependent. By scanning the laser wavelength in a wide spectral range, CRDS measurements will yield information on extinction, scattering, and absorption of a single particle simultaneously. 2) Detection limit of extinction coefficients of the proposed OT-CRDS system will be 6×10^{-9} cm⁻¹ in the UV and 1.2×10^{-10} cm⁻¹ in the NIR, corresponding to detection limits for extinction coefficients of 6×10^{-5} cm⁻¹ and 1.2×10^{-6} cm⁻¹ for a single aerosol particle of 5 µm in diameter in the respective spectral regions. For instance, water droplets have a minimum complex part of RI of 1×10⁻⁸ at 515 and 532 nm (minimum to no absorption); this number corresponds to the absorption coefficient of 4.3×10^{-4} cm⁻¹ for a water particle of 5 µm in diameter. The estimated detection limits of the proposed OT-CRDS system will be sensitive enough for the water absorption coefficient, 4.3×10^{-4} cm⁻¹, at 515 and 532 nm. 3) The OT-CRDS technique is capable of measuring wavelength-dependent complex RI (n_r and n_i) using a broad-band OT-CRDS. 4) The technique allows ringdown scans in a wide wavelength range, from which absorption features of single particles will be characterized. 5) The technique enables us to study surface properties of aerosol particles, i.e. effect of relative humidity on the size and RI of particles.

The challenges of the proposed work include the sophisticated optical configurations and limited database in absorption measurements of single aerosol particles.

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Characterization of dust particles with the degree of linear polarization

Evgenij Zubko¹, Yuriy Shkuratov¹, and Gorden Videen²

 ¹ Astronomical Institute of V.N. Karazin Kharkov National University, 35 Sumskaya St., Kharkov, 61022, Ukraine, e-mail ezubko@rambler.ru
² Space Science Institute, 4750 Walnut Street, Suite 205, Boulder, CO 80301, USA

Abstract

Using the discrete dipole approximation we study light scattering from agglomerated debris particles and demonstrate that the degree of linear polarization can be utilized for remote sensing of chemical composition of atmospheric aerosols.

1 Introduction

Passive remote sensing implies the retrieval of physical and chemical properties in a target from characteristics of the scattered solar electromagnetic radiation. While the incident solar radiation is unpolarized, the scattered light acquires partial linear polarization that is often quantified with the degree of linear polarization P. If the sunlight interacts with randomly oriented irregularly shaped particles, such as atmospheric aerosol or cosmic dust, the degree of linear polarization is defined as follows: P = -M21/M11, where M21 and M11 are elements in the 4×4 Mueller matrix [e.g., 1].

The degree of linear polarization significantly varies with the geometry of light scattering. For instance, in Figure 1, it is shown the degree of linear polarization as function of the scattering angle θ that was measured in feldspar particles [2]. However, the angular profile shown in Figure 1 is qualitatively representative for other samples studied in [2]. Namely, the angular profile typically reveals two features: the negative polarization branch near backscattering ($\theta = 150 - 180^{\circ}$) and the positive polarization branch at intermediate scattering angles; whereas, location and amplitude of these maximum and minimum polarization are characterized with P_{max} , θ_{max} , P_{min} , and θ_{min} as shown in Figure 1. Using the discrete dipole approximation (DDA), we study light scattering from the so-called *agglomerated debris particles* [3] and show that these parameters can be utilized for retrieval of physical properties in target particles.



Figure 1: The degree of linear polarization as function of the scattering angle in feldspar sample measured at wavelength $\lambda = 0.633 \ \mu m$ [2].

2 **Results and Discussion**

In order to make a realistic modeling, we average light-scattering properties of the agglomerated debris particles with a power law size distribution r^{-a} ; where, r is the radius of particles and power

index *a* is varied from 1 to 4. Four characteristics of the angular profile of linear polarization $|P_{\min}|$, θ_{\min} , P_{\max} , and θ_{\max} are shown in Figure 2.



Figure 2: $|P_{\min}|$, θ_{\min} , P_{\max} , and θ_{\max} in agglomerated debris particles as function of the power index *a*.

As one can see in the left panel in Figure 2, the amplitude of the negative polarization unambiguously decreases while material absorption increases. Note that the refractive index m = 1.855 + 0.45i is representative for carbonaceous materials; whereas, Im(m) = 0 - 0.02 can be attributed to mineral dust. Thus, when measuring the negative polarization, one can discriminate mineral dust from carbonaceous materials. Moreover, location of minimum of negative polarization monotonically decreases while Re(m) grows (see second panel in Figure 2). It makes it possible to put further constraints on chemical composition of target particles. Amplitude of positive polarization P_{max} appears to be even more sensitive to the material absorption compared to P_{min} . It especially holds for little variations in Im(m) in the range from 0 to 0.02 (see third panel in Figure 2). Location of the maximum of positive polarization noticeably shifts towards forward scattering while the constituent material changes from dielectric to conductor (fourth panel in Figure 2).

Finally, it is important to stress that simultaneous fit of the negative polarization branch and the positive polarization branch also yields a reliable estimation of the reflectance in target particles. However, it makes it possible to retrieve number density of target particles from photometric measurements.

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