Recent advances in the characterization of aerosol vertical distribution on a global scale

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1. Introduction

Understanding the climate of our planet and how human activities are changing it is one of the greatest scientific challenges facing humanity nowadays. The rise of global average surface temperature by 0.6 °C since the late 19th century, attributed to radiative forcing (i.e. perturbation of the Earth’s energy balance) by anthropogenic greenhouse gases and aerosols, underline the current concern about consequences of climate system modification. A quantitative assessment of the human contribution to climate change requires not only to establish possible future emission scenarios, but also a synthesis of uncertainties along the cause-effect chain from emissions to temperatures. The up-to-date scientific understanding of the topic, summarized in the fourth assessment report of the Intergovernmental Panel on Climate Change, establish the very likely human contribution to the climate change, but it also points out the wide range of uncertainty inherent in current model prediction associated with the radiative effect of aerosols, still labelled in the report as low level of scientific understanding.

The three known effects of aerosols on climatic processes, namely the direct effect caused by scattering and absorption of solar radiation by aerosols, the indirect effect that produce the modification of cloud properties, such as size distribution of droplets, by aerosols acting as cloud condensation nuclei, and finally the recently investigated semi-direct effect due to evaporation of cloud droplets in aerosol-rich layers because of the rise in temperature caused by absorption of solar radiation by aerosols, still requires a better constraining estimates of their magnitude.

The characterization of these effects, taken into account the large variability in geographical and seasonal distributions of the different types of aerosols (marine, anthropogenic pollution, desert dust), requires the use of long-term, detailed global measurements from distributed networks of ground-based instruments and satellites, and also comprehensive regional experiments in clean and polluted environments.

The long-term monitoring of aerosol microphysical properties on a global scale will improve the current knowledge of the influence of aerosols on climate, allowing a more accurate understanding of how the climate system will respond to the anthropogenic forcing. This chapter will comment about recent advances in the characterization of aerosol vertical...
distribution by means of optical remote sensing systems coordinated in networks of continental scales and onboard satellites.

2. Effects of aerosols and clouds on climate change

The increasing concentration of greenhouse gases in the atmosphere has caused most of the warming observed worldwide over the last century. The fourth assessment report of the Intergovernmental Panel on Climate Change (Solomon et al., 2007) established the very likely human contribution to the climate change, but it also points out the wide range of uncertainty inherent in current model prediction associated with the radiative effect of aerosols and clouds, still labelled in the report as low level of scientific understanding. The complexity of aerosol processes and their interaction with clouds in our environment yields large uncertainties in quantitative understanding of their role in many of the major environmental issues. Not only an appropriate quantification of aerosol and clouds on a global scale, including vertical distribution, is required, but also, further studies are needed to tackle the difficulties to adequately representing the radiative properties of the aerosol-cloud interaction.

Suspended particulate matter in the atmosphere, commonly known as aerosol, have many sources ranging from sea spray and mineral dust, that are mechanically generated by wind at the Earth’s surface, or biogenic aerosols such as pollen, mold spores, and airborne bacteria and viruses, to secondary aerosols like sulphates, nitrates and organics produced primarily by chemical reaction of gases in the atmosphere that condense to form particulate mass. Aerosols typically range in size from around 10 nanometres to around 100 micrometers and have limited lifetimes in the atmosphere. Aerosols in the lower part of the atmosphere normally last for only several days before being washed out by rain or settled by gravity. In the upper parts of the atmosphere, aerosols can persist much longer. Volcanic eruptions, that release tons of aerosols into the atmosphere, can propel these particles into the stratosphere, where they can persist for several years. These aerosols are created as a result of natural processes, and their sources and sinks have remained fairly stable in the last century, apart from the influence of human activities such as soil use modification. In addition to these sources, human activities can generate concentrations of aerosols far in excess of natural sources. In fact, there is compelling evidence that anthropogenic activity is increasing the concentration of tropospheric aerosols. Major cities produce large amounts of pollution aerosols as a result of industrial activity and automobile emissions. The burning of fossil fuels (primarily oil and coal) to produce energy emits large quantities of aerosols into the atmosphere. Fires set by humans around the world for agricultural purposes, such as clearing cropland, are also major sources of biomass burning aerosols. Unlike the long-lived greenhouse gases, which are distributed uniformly over the globe, aerosol distribution have substantial spatial and temporal variations, with largest concentration of pollution aerosols found near industrial regions in the northern hemisphere. The effects of aerosols tend to be localized and larger near their source regions, which makes it difficult to estimate the net global impact of aerosols on climate. Scientific evidence indicates that in regions with high anthropogenic aerosol concentrations, aerosol forcing is of the same magnitude, but opposite in sign, to the combined effect of all greenhouse gases. Also, this regionally-concentrated distribution can directly alter the general circulation patterns.
Estimating the effects of aerosols on climate is particularly challenging, because the radiative response to aerosol particles varies with size and chemical composition of the particles relative to the wavelength of the incident radiation (Dubovik, 2002). Experimental evidences has shown that all of these properties can and do change with time, such as when mineral-based desert dust moves over an urban area and black-carbon based aerosols attach to the mineral core. Aerosols influence the atmospheric energy budget through direct, semi-direct and indirect radiative effects. Direct effect is caused by the scattering and absorption of incoming solar radiation. Scattering of radiation reduces solar heating at the surface immediately below, causing regional “solar dimming” (Liepert, 2002), that somehow counteract some greenhouse-gas warming. The amount of cooling depends on the above mentioned aerosol parameters, but also on the type of underlying surface. Another likely consequence of the aerosol surface cooling is a reduction of evaporation and precipitation. To further complicate matters, the forcing can switch from negative values (cooling) in clear-sky conditions to positive values (warming) at a cloud cover of about 25%, because the energy distribution due to the presence of the cloud is different than a clear sky (Ramanathan et al. 2001).

The semi-direct effect is related with the absorption of solar radiation by aerosols, such as black-carbon and some mineral-based aerosols, that heats the surrounding atmosphere and can actually suppress the formation of clouds by elevating the atmospheric temperature, preventing the condensation of water vapor. This forcing can strengthen the low-level thermal inversion of the boundary layer, which can perturb low-level clouds, enhance aerosol lifetimes, and alter the boundary layer moisture. This has been observed in events related with the South Asian haze, where the warming in the aerosol layer can nearly totally desiccate stratuscumulus cloud layers and alter the properties of the trade wind cumulus layer (Ackerman et al., 2000).

Finally, the indirect effect involves the influence of aerosols on the properties of clouds, such as the microstructure, dynamics, coverage and stability of cloud layers. An increase in aerosols, acting as cloud-condensation nuclei or ice nuclei, creates larger concentrations of clouds droplets, which leads to increased cloud lifetime and albedo. This is because normally the droplets grow, collide and coagulate until they grow large enough to fall as raindrops, but as the amount of aerosols in the cloud is relatively low, the cloud will consist of relatively fewer but larger droplets. If the amount of aerosols is increased, the cloud droplets that form tend to be smaller and more numerous; it takes longer for raindrops to form, and the clouds last longer. Clouds consisting of smaller droplets also reflect more sunlight back to space and contribute to increased cooling of the underlying surface (Twomey, 1977). As anthropogenic aerosols are most highly concentrated in the lower atmosphere, the indirect effect is expected to be most important in low-level clouds. The most obvious impact of clouds on the hydrological cycle is that of precipitation. In removing water from the atmosphere, precipitation modifies cloudiness and cloud structure. Moreover, the latent heating associated with precipitation is a driving force for atmospheric circulations. Precipitation frequencies can also be affected by aerosols, for instance by the suppression of raindrops in shallow and deep convective clouds, which are the major sources of thermodynamic forcing of the general atmospheric circulation (Levin, 2009).

The effect of clouds on the global radiative balance depends on the competition between the reflection of incoming solar radiation and the absorption of Earth's outgoing infrared radiation. The overall impact of high-altitude clouds is to warm the planet while the low-
altitude clouds tend to cool it. Research to date suggests that, globally averaged, the overall cooling caused by clouds is more powerful than the warming they cause. Anyhow, these behaviours are strongly linked to microphysical processes in clouds because changes in those processes can modify the spatial extent, spatial distribution, and lifetimes of clouds, the water vapor distribution outside of clouds, and the fluxes of water and radiation through the atmosphere. Another reason why modelling clouds is difficult is that clouds

Figs. 1. Evidence of the indirect effect of aerosols: The image from the Moderate Resolution Imaging Spectroradiometer captured on January 27, 2003 and processed to create a true-color image using bands 1 (650 nm), 4 (555 nm), and 3 (469 nm) shows an unusually high number of ship tracks (thin white lines), visible in the clouds off of the coasts of France (Left) and Spain (Bottom). (Image courtesy of J. Descloitres, MODIS rapid response team, NASA/GSFC, Greenbelt, Maryland)

change almost instantaneously compared to the rest of the climate system and on very small scales. Most models are not capable of representing phenomena that change so rapidly or impact such a small portion of Earth at any given time (Baker, 1997). Owing to the typically high altitude and often remote location, the nucleation of ice is the less well-understood process. Atmospheric ice formation at temperatures warmer than about 40 °K below the equilibrium freezing temperature, requires the presence of a special solid particle that acts as an ice nucleus, enhancing the stability of an ice embryo due to the
presence of a surface. Materials that can act as ice nuclei include, but are not limited to, mineral dust, anthropogenic metal oxides, pollen and bacteria.

The clearest observational evidence for an indirect aerosol effect is provided by ship tracks, which are trails in ambient low-level clouds that result from the effluent from ships. Although ships are not significant sources of pollution themselves, they release enough aerosols from their smokestacks to modify overlying clouds. Those aerosols act as cloud condensation nuclei, which may either produce new cloud particles where none existed before, or may attract water from existing cloud particles, creating brighter clouds due to the enhancement of liquid-water content, possibly caused by suppression of drizzle, and smaller droplets. An example of this is shown in figure 1; in this satellite image of clouds over the Atlantic Ocean, the thin white lines are bright clouds consisting in small droplets that form due to aerosols emitted by ships. Another example is the pollution tracks as viewed by the Advanced Very High Resolution Radiometer satellite imagery (Rosenfeld, 2000). Perhaps the most significant aspect of the analysis of these pollution tracks is the conspicuous absence of them over the United States and Europe, implying that these regions are so heavily polluted that local sources can not be distinguished from the widespread pollution-induced narrow droplet spectra in those regions. A striking example is provided by anthropogenic lead-containing particles, one of the most efficient ice-forming substances commonly found in the atmosphere. Post-industrial emissions of particulate lead have been estimated to offset a proportion of the warming attributed to greenhouse gases, by “supercharging” pre-existing particles, making them highly efficient ice nuclei that allow clouds to form at lower altitudes (Cziczo, 2009). But after the regulation of tetraethyl lead, an additive to automotive petrol, in the mid-1980, total lead has dropped significantly, with a 20-fold decrease reported in the continental United States in the two decades since 1980. This might imply a drastic reduction in the offset, although a proper estimation of the climatic effect is still pending. Like the ship tracks and contrails produced by aircraft, the impact of those pollution tracks on regional and global climate is not yet known, but recent satellite observations of these phenomena are providing new information.

The main uncertainties in climate model simulations are due to the difficulties in adequately representing the interdependent microphysical, chemical, and dynamic processes that characterize the aerosol-cloud system in the atmosphere, that must be better understood in order to quantify the effects of anthropogenic aerosols on the albedos, emissivities, cloud-top temperatures and extent of clouds. Present models do not have sufficient sophistication in cloud microphysics to include those aerosol influences (Grabowski, 2009). As global climate models have not included all these complexities, the simulation of impact of aerosols on global climate is not possible yet. First and foremost, a reliable global inventory of aerosol emission rates, lifetimes, global distribution and concentrations is urgently needed. The aerosol vertical distribution depends on the distributions of the emissions, on chemical production for secondary aerosols, on the distribution of clouds, precipitation and wet deposition processes, and finally, on the transport characteristics determined by the flow field. On the theoretical and experimental side, further investigations regarding the microphysical processes and radiative effects in clouds and aerosols are required. Another large uncertainty in climate studies is the effect of cloud multilayering. It has been observed that the largest variations in predictions of climate warming are due to the different ways the models specify how clouds are vertically distributed and overlap, which influence both the magnitude and vertical
profile of heating in the atmosphere, and also the predicted precipitation. However, the interaction between aerosols and clouds is sufficiently complex that even cloud-resolving models have difficulty in accurately simulating their physics and dynamics. Therefore, detailed vertical profiles of aerosol and clouds will be relevant parameters to evaluate aerosol forcing, understand the effect of aerosol on cloud microphysics and precipitation, assess the degree of interaction of aerosols with the cloud layer and also the effect of clouds on aerosols, due to the major role of water in determining aerosol optical properties. The vertical profiles will also help to better understand aerosol lifetimes, their source regions through backtrajectory analysis and long-range transport events, which occur at elevated layers, decoupled from the ground.

3. Characterization of the aerosol vertical distribution on a global scale

As above-mentioned, altitude-resolved information on aerosols, including long-range transport in the free troposphere, and cloud observations including cirrus clouds, are essential for understanding the climate role of atmospheric aerosols. The most promising source of routine information on the vertical distribution of aerosols are lidar systems. Other possible options to provide altitude-resolved information are aerosol balloon sondes and aircraft measurements, but they are not yet widely available and they are also less economical. The lidar technique, which stands for Light Detection And Ranging, operates in a similar way as radar, but using light instead of microwaves. Lidars have been used for several years to determine the planetary boundary layer height because of the large gradient in aerosol concentration that occurs between the top of the boundary layer and the free troposphere. Therefore, ground-based lidars may fill the ongoing need for insight into the structure of the atmosphere and its variability with time. There are only a few lidar instruments planned for deployment on satellites, as it will be discussed below. In contrast, there are several research lidar measurement stations and networks that are well organized with high standards regarding quality control, ongoing development of new controlling measures, and data archival. The integration of aerosol lidar observations with other measurements by radiosonde, ozone sonde, sunphotometer and satellite is most useful allowing for a maximum synergy of information. Also, the informational content of lidar observations is greatly enhanced by air parcel trajectory analysis.

3.1 Light Detection And Ranging

Remote sensing by lidar has received wide application in investigation of atmospheric trace constituents, clouds, wind and temperature since its invention a few decades ago. The lidar technique provides information on several aerosol and clouds properties with high spatial and temporal resolution, working in a similar way as radar. The system typically consists of a laser transmitter and an optical receiver in parallel or collinear arrangement. Figure 2 shows a schematic depict of a lidar system with three emitting wavelengths, a typical configuration when Nd:YAG lasers, the most reliable and widely used type of laser, are used, that typically emits at 1064, 532 and 355 nm. The system transmits intense, short-duration light pulses of linear polarization at a high repetition rate into the atmosphere within the receiver field of view. The intensity of the light elastically backscattered by molecules and aerosols is measured versus time through the telescope receiver, collimating
optics, a narrow bandpass filter for daylight suppression, and an appropriate detector. In the figure, several detection channels are shown, in a typical configuration of advanced lidar systems for measuring backscatter intensity, molecular or Raman signals and polarization components of the signal, as it will be discussed below. The signal profile will be stored by a fast analog-to-digital converter or by a photon counting device. Relative intensity data are accumulated separately from all altitude intervals for a selected averaging period, which may include thousands of individual laser shots.

The lidar of lowest complexity measures the aerosol backscatter signal at one wavelength. This lidar allows the retrieval of the aerosol backscatter coefficient, although critical assumptions have to be made in the inversion of the lidar signal in order to obtain aerosol optical properties. The procedure, with all its subsequent modifications and improvements, simply suffers from the fact that from only one observable (the energy returned as a function of time), two unknowns (the aerosol backscatter coefficient of the aerosol and the two-way transmission losses through the atmosphere due to light extinction by molecules and aerosols between transmitter, backscattering volume at a certain range and the receiver) must be determined, therefore, the system is underdetermined. Many techniques have been discussed in the literature to work around this difficulty (the slope method, Collis and

Fig. 2. Schematic of a lidar system showing the laser transmitter on the left, the optical receiver in biaxial configuration on the right. Several different returns are presented as photons. On the right, a photograph of the actual implementation of the Madrid station lidar, showing the laser on first term, three mirrors for the three different Nd:YAG wavelengths send to the atmosphere through the ceiling window, and the telescope and detection line behind.

Russell, 1976; the Bernoulli solution to the equation, Klett, 1981, Fernald, 1984; and column closure by the use of ancillary optical depth information, Welton et al., 2001). Nevertheless, the measurement remains only an estimate of either the backscatter or the extinction
coefficient as long as elastic lidar-only data is available, because the relation between these magnitudes, that must be assumed constant in the above mentioned techniques, actually depends on the microphysical, chemical, and morphological properties of the aerosols, which, in turn, depend on relative humidity. Thus, even in the well-mixed layer, the relation might not be constant with height because the relative humidity usually increases with height. There is value, however, in the backscatter lidar, providing range to a target and structural information on the atmosphere once corrected for detector non-linearity, background and range. Basic lidar products are mixed-layer and cloud-base heights and qualitative information of aerosol layers in the free troposphere, visible and subvisible cirrus and stratospheric aerosol layers after major volcanic eruptions. Figure 3 shows the so-called quicklook or colour plot obtained by representing the range corrected signals taken along a certain period of time in a height-time display. It provides an overview of the atmospheric situation in terms of evolution of the boundary layer, lofted aerosol layer, like the Saharan dust intrusion at 5 Km, and cloud distribution, such as the cirrus clouds detected as a thick layer at the beginning of the measurements, 03:20 UTC, on the left part of the image, and disappears an hour later, right part.

Fig. 3. Lidar profile and Quicklook at 532 nm, obtained over Madrid (40.45°N, 3.73°W) on the 21st of June, 2009, between 3:20 and 4:40 am. The background substracted and range-corrected signals shows relevant features at 5 Km, caused by a Saharan dust intrusion over the site, and between 8 and 12 Km, due to cirrus, that dissapear along the measuring time.

In the near-range, the signal is affected by losses due to the incomplete overlap of laser beam and receiver field of view. A good knowledge of this overlap is an important prerequisite for a proper retrieval of the backscatter coefficient in the lowest and usually most polluted part of the atmosphere. Due to this limitation, the reference range required in the inversion of the lidar signal is chosen at altitudes where the aerosol backscatter coefficient is negligible compared to the known molecular backscatter value, the so-called aerosol-free layer, and the equation is solved using the backward integration solution proposed by Klett (Klett, 1981).
This far-end calibration also leads to more stable solution than the near-end calibration, but clear air conditions are needed at some range to calibrate the signal. They are normally found in the free troposphere for laser wavelengths < 700 nm, but it might be critical for longer wavelengths (e.g., 1064 nm or the eyesafe wavelength of 1550 nm) because of weak Rayleigh scattering. The inversion procedure also includes the calculation of the molecular backscatter profile. Standard-atmosphere assumptions, nearby radiosonde data of temperature and pressure, or weather prediction model outputs for the lidar site are used to compute the entire Rayleigh scattering profile along the laser beam.

### 3.2 Advanced lidar systems

Accurate retrieval of extinction and backscatter profiles without making assumptions about the aerosol is only possible when measurements of two independent signals are performed. Recent developments of the technique, such as the use of the atmospheric nitrogen Raman return (Ansmann et al., 1990), or the broadening of the lidar return by molecules in a technique called High Spectral Resolution Lidar (HSRL) (Shipley et al., 1983), allow the independent determination of aerosol backscattering and extinction coefficient profiles. Both methods are based in the direct determination of the extinction coefficient profile by means of a channel that detects a pure molecular signal, and a second channel that provides the backscatter coefficient from a signal detecting aerosols and molecules. These advanced lidar systems determine the aerosol optical properties in a quantitative way and permit the estimation of main microphysical properties. Rapid progress in laser technology and data acquisition is supporting an increasing specialization in the design of lidar systems, leading to the point where systems can be built or certain purposes with high reliability and durability. Even several lidar systems for some specific applications are now becoming available commercially.

#### 3.2.1. Raman lidar technique

The independent determination of backscatter and extinction coefficient profiles can be achieved by the measurement of pure molecular backscatter signal, because the molecular extinction profile and the backscatter coefficient can be calculated a priori with sufficient accuracy so that the aerosol extinction, the only remaining unknown, can be retrieved from the molecular signal. The use of the vibrational Raman scattering signal from nitrogen (or oxygen) offers the technologically easiest implementation, due to the large Raman shift (2331 cm\(^{-1}\)) that allows a reliable separation from the elastic aerosol signal with standard filters. Due to the low values of the nitrogen backscattering coefficient, that produce weak Raman signals, approximately by a factor of 500 compared to Rayleigh signals, this technique works best in the absence of the strong daylight sky background, therefore most Raman lidars operate only at nighttime in this mode. Anyhow, high power laser (>250 mJ/pulse) Raman lidars equipped with 0.3-nm interference filters to block sunlight, allow daytime operation at least throughout the convective boundary layer. For nighttime operation the filter bandwidth can be broad and the technical implementation is quite straightforward and it has been widely used, mainly at 355nm and 532nm, the second and third harmonics of the Nd:YAG laser. Recent applications of the technique provide the water vapor profile using its Raman signal (3652 cm\(^{-1}\)), which may be very interesting for studies of the aerosol-cloud interaction.
Pure rotational Raman scattering by nitrogen and oxygen offers a scattering cross section that is about a factor of 30 higher than vibrational Raman scattering. The drawback is that the Raman shift is quite small, about 30 cm\(^{-1}\) only, so that separation from the elastic aerosol backscatter is more challenging, keeping in mind that out-of-band blocking has to be on the order of \(10^{-6}\). Both filter techniques and double grating polychromators have been demonstrated for this approach. In particular the combination with a Fabry-Perot comb filter can suppress daylight sufficiently to allow daytime operation. A more sophisticated setup also allows one to retrieve the temperature profile simultaneously.

### 3.2.2 High Spectral Resolution Lidar Technique

High Spectral Resolution Lidar (HSRL) is based on the Doppler broadening of the Rayleigh line, leading to an about 0.01cm\(^{-1}\) wide line surrounding the much narrower peak from aerosol scattering. In the HSRL technique, one channel detects the molecular signal by suppressing the centre part of the backscatter spectrum, containing the aerosol return, with an ultra-narrowband filter, generally an iodine vapour cell. A second channel records the total signal from aerosol plus molecular scattering. The combination of both signals allows determining extinction and backscatter profiles independently. The advantage of this technique is that it suitable for daytime operation because the Rayleigh scattering cross section of air is more than three orders of magnitude greater than that for vibrational Raman scattering. The drawback is high system complexity and high demands on system adjustment as well as on performance control.

Both techniques have been operated successfully for aerosol profiling. Data analysis schemes have been developed to retrieve vertical profiles of aerosol optical properties. The algorithms are well-tested and are nowadays almost routinely applied. Another important advantage of Raman lidars and HSRLs is that the profile of the backscatter coefficient is determined from a signal-ratio profile so that the overlap effect cancels out, regarding that the two channels are well adjusted and show the same overlap characteristics. As a consequence, the retrieval of the backscatter coefficient is possible down to heights rather close to the surface. In combination with Sun photometers, a comprehensive set of vertically and spectrally resolved optical properties can be determined. Anyhow, the requirements of expertise of the operating personnel with optical systems and inversion algorithms are still a limitation for the automation and network operation of these systems.

Once reliable extinction and backscattering coefficient profiles are obtained, the extension of the technique to multiple wavelengths offers the opportunity to determine vertically-resolved microphysical properties, such as size distribution parameters, volume concentrations and refractive index (Böckmann et al. 2005). During the past decade sophisticated inversion techniques have been developed and successfully tested that permit the retrieval of microphysical properties of aerosols from their optical properties provided by advanced multiwavelength lidar observations. For aerosol sizes in the typical range of the accumulation mode, measurements of the backscatter and extinction coefficients at the Nd:YAG wavelength (1064, 532 and 355nm) are necessary and sufficient to estimate the aerosol volume and surface density as well as the refractive index (Müller et al., 1999). The retrieval procedure is ill-posed and requires sophisticated regularization methods, so that presently the procedures are still experimental and applied for selected cases only. The low number of measured aerosol optical properties requires introducing physical and mathematical constraints in the inversion algorithms in order to come up with sensible...
microphysical aerosol parameters. These algorithms do not attempt to accurately derive the exact shape of aerosol size distributions, which might not be achievable even in the near future due to the low number of measured optical information and the lack of appropriate mathematical tools. However it is possible to derive mean parameters such as the effective radius of the aerosol size distribution with comparably high accuracy. At present it does not seem possible to fully retrieve aerosols in the so-called coarse mode of aerosol size distributions which is largely determined by aerosols from natural sources such as mineral dust. However, aerosols from anthropogenic activities are mainly present in the fine mode fraction which is accessible to the inversion algorithms (Müller et al., 2007).

The combination of advanced lidar products with Sun photometer data offers another approach, as the potential of Sun photometry to derive optical, microphysical, and radiative properties of aerosols is already well-documented (Eck et al., 1999). Also, depolarization observations can be used to improve the identification of aerosol types, such as desert dust. The emitted laser light is linearly polarized and the return signals are measured in two polarization channels which are parallel- and perpendicular-oriented to the laser polarization. From the linear total (aerosol + molecular) depolarization ratio of the scattering volume that is obtained from the ratio of the perpendicular- to the parallel-polarized signal component, the aerosol depolarization ratio can be calculated if the aerosol backscatter coefficient and the respective linear molecular depolarization ratio are known. Spherical aerosols as water droplets produce an aerosol depolarization ratio of almost zero in the case of 180° scattering. Dust aerosols cause a depolarization ratio of 25%-35%. Smoke, urban haze, and maritime aerosols show depolarization ratios of <10%. Ice aerosols (ice clouds) lead to depolarization ratios typically >40%-50% (at off-zenith laser beam angles). Present inversion algorithms assume spherical shape of the aerosols. Only recently efforts have been undertaken to introduce methods that allow for a characterization of aerosols of non-spherical shape, such as mineral dust. However, the underlying theoretical aspects of light-scattering by irregularly shaped aerosols still are in a rather exploratory status. Last but not least, profiles of microphysical aerosol properties can be derived with the available algorithms, however with an extreme consumption of computer and human operator time. Thus it is desirable to extend the available algorithms toward an efficient processing of profiles of optical data, which in turn delivers profiles of microphysical aerosol properties. Schemes with higher degree of automation are under development.

4. Networks of lidar stations

Following the example of AERONET, a global network of systematic column-integrated aerosol optical depth observations using surface-based sun-tracking photometers, different attempts exist to characterize the vertical distribution of aerosols on continental scales and further extent to global scale within the initiative of the Global Atmosphere Watch (GAW) aerosol programme (Bosenberg & Hoff, 2007). It is the goal of the GAW programme to coordinate and homogenize the different existing network in order to determine the spatio-temporal distribution of aerosol properties related to climate forcing and air quality on multi-decadal time scales and on regional, hemispheric and global spatial scales. Such initiative is being discussed nowadays under the frame of GAW Aerosol Lidar Observation Network (GALION).
Presently, it is not feasible to implement a global aerosol lidar network by installing a homogeneous set of systems at a number of stations selected for optimal coverage because advanced lidars are still relatively complex and delicate instruments requiring substantial efforts for operation. Instead, it is important to make use of existing systems at established stations, of the experienced operators of these systems, and of existing network structures. Nowadays, several lidar networks perform regular measurements on continental scale to establish a comprehensive dataset of the aerosol vertical distribution and also assess volcanic, dust, fires or pollution events. Those networks include the Micro-pulse lidar network (MPLNet), the European Aerosol research lidar network (EARLINET), the Asian Dust Network (AD-Net), the Commonwealth of independent states lidar network (CIS-LiNet), Regional East Aerosol Lidar Mesonet (REALM) and the American Lidar Network (ALINE). A brief description of each of them follows:

The only tropospheric profiling network which can claim global coverage is the NASA MPLNET (http://mplnet.gsfc.nasa.gov/), designed for satellite validation and co-located with AERONET sites in order to produce quantitative aerosol and cloud products by synergy with sunphotometer measurements. The Micro-Pulse Lidar (MPL) was developed at NASA Goddard Space Flight Centre in the early 1990s and it consists of single-wavelength (523 nm), high repetition, low power, eye safe commercially available backscatter lidar capable of determining the range of aerosols and clouds in unattended operation mode. At present, MPLs are operated at 22 sites around the world. The combination of MPLNET consisting of low-cost, eyesafe, automated 532-nm backscatter lidars with AERONET (Holben et al., 1998), NASA’s global network of more than 200 continuously running Sun photometers, is an example for a successful application of the lidar-photometer technique.

The European Aerosol Research Lidar Network, (EARLINET), (http://www.earlinet.org) is a voluntary association of institutions with an interest in aerosol science and a long-term commitment in vertical profiling of aerosol properties with advanced laser remote sensing. Presently EARLINET comprises 25 stations distributed over Europe. Instrumentation is rather inhomogeneous because most lidars existed before the network was established in 2000, but most systems are now equipped with at least one Raman channel for independent determination of extinction and backscatter. The main goal is to establish a climatology for the aerosol vertical distribution by building a quantitative comprehensive statistical database of the aerosol, therefore regular operation at three times per week has highest priority for all stations. Special studies of, for instance, Saharan dust outbreaks across the Mediterranean, distribution of smoke from wildfires, volcanic eruptions, air mass modification across Europe, diurnal cycle, or CALIPSO validation required numerous additional observations which were organized as necessary through corresponding alerting schemes. Quality assurance for hardware and software was performed through direct intercomparisons, tools for routine performance checks are under test.

The Asian Dust Network (AD-Net) (http://www-lidar.nies.go.jp/AD-Net/) is an international virtual community designed originally to track outbreaks of dust from China, Mongolia and Russia. Different instruments are involved, such as multi-wavelength Raman lidars in Tokyo and Gwanngju (Korea), HSRL system in Tsukuba (Japan) and automated two-wavelength polarization lidars in Japan (8), Korea (1), China (1), and Thailand (1), which are coordinated by the National Institute for environmental Studies (NIES) lidar
network, most of them co-located with skyradiometer from SKYNET. Those automated small and compact lidars are recently being upgraded with Raman channels.

Fig. 4. Distribution of the existing networks stations on the globe. The different networks are indicated by the symbol and color: MPL-Net: Blue circles, EARLINET: green squares, AD-Net: red crosses, NDACC and REALM: red triangles, CisLiNet. Brown squares and ALiNE: orange circles.

The Commonwealth of Independent States lidar network (CIS-LiNet) has been established by lidar teams from Belarus, Russia and Kyrgyz Republic. Its objective is carrying out lidar observation coordinated at the territory from Minsk to Vladivostok in cooperation with EARLINET and AD-Net. There is an aim to provide the lidar stations with sun radiometers, and include them in the global radiometric network AERONET. The Regional East Aerosol Lidar Mesonet (REALM), is a network of lidar research groups on the east coast of the United States (http://alg.umbc.edu/REALM) operative since 2002, a collaboration of existing lidar facilities has attempted a network operation. But to date only two groups and three lidars have voluntarily contributed consistent data to the network with two other groups contributing campaign style activities. Finally, the American Lidar Network (ALiNE) is an informal agreement among the existing lidar groups in Latin America. It includes also research teams working to host lidar instruments in the near future.

Another network that employ lidars, but with a different research aim, is the Network for the Detection of Atmospheric Composition Change (NDACC, previously NDSC), has been monitoring the stratosphere and upper troposphere for at least 15 years. NDACC consists of more than 70 high-quality, remote-sensing research sites for observing and understanding the physical/chemical state of the stratosphere and upper troposphere and assessing the impact of stratospheric changes on the underlying troposphere and on global climate. Only
a subset of the stations actually contains lidars. In this subset, the lidars are designed primarily to profile \( \text{O}_3 \) in the stratosphere and stratospheric aerosols. As it can be gathered, the operation of a global lidar network will initially depend completely on voluntary contributions from the various existing networks, most of which, themselves, are based on voluntary cooperation, plus contributions from individual stations. Therefore, the consistency of data across the network and its quality assurance might be compromised.

5. Satellite missions with lidar instruments onboard

![Image of satellite constellation](image_url)

**Fig. 5.** The “A-train” constellation of satellites, with CALIPSO on the third place from the left. Sinergy with data provided by the radar onboard CloudSat, the polarization sensitive radiometer POLDER onboard Parasol and MODIS spectra onboard Aqua satellite will provide the most complete characterization of clouds and aerosols on global scale ever. Crédits: CNES/III. P. Carril

Lidars have been used as ground-based systems, on airborne platforms, and also from space, as a different strategy for aerosol vertical distribution characterization on a global scale. Unlike lidars that profile the atmosphere above at a single geographic location, spaceborne lidars allows the study of aerosols in regions that are difficult or impossible to
explore by other means. The era of spaceborne lidar started with the Lidar In-space Technology Experiment, or LITE, (McCormick, 1997), a backscattering lidar system that flown in the cargo bay of the space shuttle “Discovery” in September 1994, to evaluate technological requirements of a spaceborne lidar and its scientific capability. The mission convincingly demonstrated the value of spaceborne lidar in retrieving the vertical structure of aerosols and clouds on a global scale and provided the backbone for more recent satellite-based lidars. In 2006, the French Centre National d’Etudes Spatiales and US National Aeronautics and Space Administration collaborate to launch the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) (Winker et al, 2007) spaceborne lidar, that collects profiles of the lidar attenuated backscattering coefficients of aerosols and clouds at 532 and 1064 nm from -2 to +40 kilometres above ground level using the CALIOP instrument. The CALIPSO satellite mission objective is to determine precisely the altitudes of clouds, aerosol layers and their overlap; identify the composition of clouds and the presence of "subvisible," or invisible, clouds; and estimate the abundance and sources of aerosols. It will provide the first global survey of cloud and aerosol profiles from space, with seasonal and geographical variations. CALIPSO will fly in formation with CloudSat and in concert with the other satellites of the "A-Train," a constellation of several Earth-observing satellites depicted in figure 5. The combination of data from the CloudSat radar, CALIPSO lidar, PARASOL radiometer and MODIS on the Aqua satellite observing the same spot on the ground within minutes, provides a rich source of information that can be used to assess the role of aerosols and clouds in climate, as well as reduce in some extent the ambiguity in deriving the aerosol profile from the lidar measurements (Kaufman et al. 2003).

CALIPSO will provide the first statistics on the global vertical distribution of aerosols and aerosol types and "subvisible" cirrus clouds (very thin clouds invisible to the naked eye). The synergy of CloudSat and CALIPSO measurements will provide information on the vertical structure of clouds and daily coverage of global cloud characteristics. It is expected to provide the first indirect but validated estimate of the contribution of clouds and aerosols to the vertical distribution of atmospheric warming, from which the first operationally based estimates of direct aerosol properties and uncertainties can be made.

Several examples of the global transport of aerosol were evident from spaceborne data. Aerosol from natural sources, such as Saharan dust, was measured on several orbits of LITE and CALIPSO. Although it has been known for quite sometime that large quantities of Saharan dust are transported across the Atlantic towards the Caribbean, the unique capabilities of spaceborne lidars proved ideal for tracking and quantifying the magnitude of these events, showing enormous plumes of hundreds of kilometres reaching altitudes above five kilometres or aerosol plumes generated by biomass burning in South America extending hundreds of kilometres from the source region, that make possible to assess their impact and quantify their contribution to long-range transport. Another example would be the measurement of anthropogenic aerosol leaving the Eastern United States and riding the “gulfstream highway” towards Europe (Hoff & Strawbridge, 1996).

The validation of the spaceborne instrument is required to assess the quality of the measurements, especially for aerosol types which have poorly or unknown lidar ratios which must be assumed in the CALIPSO aerosol retrieval since it is an elastic-type instrument. Network of ground-based lidar stations can support the aerosol observations from space by making targeted observations on CALIPSO overpass times. The better sensitivity of ground-based lidar systems can be used to confirm the sensitivity limits of the
satellite instruments characterize the atmospheric features missed by the satellite instruments and determine additional parameters. In the near future, the European Space Agency will launch the Earth Explorer Atmospheric Dynamics Mission (ADM-Aeolus) (Reitebuch et al. 2004), that will provide global observations of wind profiles from space using an active Doppler Wind Lidar, and also The Earth Clouds Aerosols and Radiation Explorer (EarthCARE) Mission, with the ATLID instrument onboard, which will be used to map global distributions of aerosols. Both satellite platforms will be equipped with HSRLs, with very different characteristics than CALIPSO. That will not provide a homogeneous set of aerosol measurements. A global ground-based lidar network providing stable, long-term measurements will be necessary to provide a benchmark against which to reference multiple satellite instruments.

6. Conclusion

In summary, while there is considerable evidence supporting the hypothesis that human activity is modifying climate, further research is required to strengthen the physical understanding. The lack of precise knowledge about all the processes of importance to climatic change appears to be limiting progress in furthering our quantitative understanding of human impact of climate. The largest uncertainty in current model predictions is related with the radiative effects of aerosols. There is a need for long-term monitoring of radiative properties of the aerosols-clouds system with high temporal and vertical resolution, as it plays a crucial role in climate. Recent advances in lidar technology allow to foresee a system capable of monitor the aerosol-cloud interaction with high temporal and spatial resolution taking advance of the HSRL for daytime measurements and Raman signals to derive aerosol and water vapor properties. Employing this advanced technology to probe Earth’s atmosphere is expected to reduce the uncertainties in the climate forecasts. Regarding networks of instruments, advanced aerosol lidar systems are still relatively complex and delicate instruments to operate, therefore substantial engineering effort is still required towards increased reliability and automated operation. As a future perspective, further developments in algorithms that simultaneously invert complementary data acquired with lidars, satellites and Sun photometers, allowing for a maximum synergy of information, will provide better retrievals of aerosol and cloud microphysical properties. Integration of innovative new satellite observations such as CALIPSO, ADM-Aeolus or EARTHCARE, field experiments, and laboratory studies with models will pave the way for breakthroughs in our understanding of how aerosols are modifying the environment.

7. References

Ackerman, A.S., Toon, O.B., Stevens, D.E., Heymsfield, A.J., Ramanathan, V. & Welton, E.J. (2000). Reduction of tropical cloudiness by soot. Science, 288, 1042-1047
Ansmann, A., Riebesell, M. & Weitkamp, C. (1990). Measurement of atmospheric aerosol extinction profiles with a Raman lidar, Opt. Lett., 15, 746-748.
Baker, M. B. (1997). Cloud Microphysics and Climate. Science, 276, 1072-1078.
Böckmann, C., Mironova, I., Müller, D., Scheidenbach, L., Nessler, R. (2005) Microphysical aerosol parameters from multiwavelength lidar. J. Opt. Soc. Amer. A, 22, 518-528
Bosenberg, J & Hoff, R. (2007) Plan for the implementation of the GAW Aerosol Lidar Observation Network, GALION. WMO GAW Report No 178, Hamburg, March 2007

Collis, R. T. H. and Russell, P. B. (1976). Lidar measurement of particles and gases by elastic backscattering and differential absorption. Laser Monitoring of the Atmosphere, Hinkley, E. D., pp. 71-152, Springer-Verlag, New York.

Cziczo, D. J. et al. (2009). Inadvertent climate modification due to anthropogenic lead. Nature geoscience 2, 333-336

Dubovik, O., Holben, B. N., Eck, T. F., Smirnov, A., Kaufman, Y. J., King, M. D., Tanr’e, D., & Slutsker, I. (2002). Variability of absorption and optical properties of key aerosol types observed in worldwide locations, J. Atm. Sci., 59, 590-608

Eck, T. F., Holben, B. N., Reid, J. S., Dubovik, O., Smirnov, A., O’Neill, N. T., Slutsker, I., and Kinne, S. (1999). Wavelength dependence of the optical depth of biomass burning, urban, and desert dust aerosols, J. Geophys. Res., 104, 31 333–31 349

Fernald, F.G. (1984). Analysis of atmospheric lidar observations: some comments, Appl. Opt. 23, 652-653

Grabowski, W. W. & Wang, L. (2009). Diffusional and accretional growth of water drops in a rising adiabatic parcel: effects of the turbulent collision kernel. Atmospheric Chemistry and Physics 9, 2335-2353

Hoff, R. M., & Strawbridge, K. B. (1996). LITE observations of anthropogenically produced aerosols, Advances in Atmospheric Remote Sensing with Lidar, pp 145-148. 18th International Laser Radar Conference (ILRC), Springer-Verlag, Berlin, Germany

Holben, B. N., Eck, T. F., Slutsker, I., Tanre, D., Buis, J P., Setzer, A, Vermote, E., Reagan, J. A., Kaufman, Y. J., Nakajima, T. Lavenu, F., Jankowiak, & Smirnov, A. (1998). AERONET - A federated instrument network and data archive for aerosol characterization, Remote. Sens. Environ., 66, 1-16.

Kaufman et al. (2003). Retrievals of profiles of fine and coarse aerosols using lidar and radiometric space measurements, IEEE Transactions on geoscience and remote science, 41, 1743-1754

Klett, J.D.(1981). Stable analytic inversion solution for processing lidar returns, Appl. Opt. 20, 211-220.

Levin, Z & Cotton, W. R. (2009). Aerosol Pollution impact on precipitation: A scientific review, Springer ISBN: 140208689X 9781402086892.

Liepert, B.G. (2002). Observed reduction of surface solar radiation at sites in the United States and worldwide from 1961 to 1990. Geophys. Res. Lett. 29, 61.1-61.4

McCormick, M.P. (1997). The flight of the lidar In-Space Technology Experiment (LITE). Advances in Atmospheric Remote Sensing with lidar. Selected papers of the 18th International Laser Radar Conference (ILRC), 141-144, Berlin, July 1996, Springer, Berlin.

Müller, D., Wandinger, U. and Ansmann, A. (1999). Microphysical particle parameters from extinction and backscatter lidar data by inversion with regularization: Simulation. Appl. Optics, 38, 2358-2368

Müller, D., Mattis, I., Ansmann, A., Wandinger, U., Ritter, C. & Kaiser, D. (2007). Multiwavelength Raman lidar observations of particle growth during long-range transport of forest-fire smoke in the free troposphere, Geophys. Res. Letts., 34, 2006GL027936
Ramanathan, V., Crutzen, P. J., Kiehl, J. T., Rosenfeld, D. (2001). Aerosols, Climate, and the Hydrological Cycle, Science 294, 2119-2124
Reitebuch, O., Chinal, E., Durand, Y., Endemann, M., Meynart, R., Morancais, D., Paffrath, U. (2004). Development of an airborne demonstrator for ADMAeolus and campaign activities. 22nd Int. Laser Radar Conference (ILRC), pp. 1007-1010. Matera, Italy, ESA SP-561,
Rosenfeld, D. (2000). Suppression of rain and snow by urban and industrial air pollution. Science 287,1793-1796
Shipley, S.T., Tracy, D.H., Eloranta, E.W., Trauger, J.T., Sroga, J.T., Roesler, F.L. & Weinman, J.A. (1983) A High Spectral Resolution Lidar to Measure Optical Scattering Properties of Atmospheric Aerosols, Part I: Instrumentation and Theory, Applied Optics, 23, 3716-3724
Solomon, S. (2007). Climate Change 2007: The physical Science Basis. Contribution of Working Group I to the Fourth assessment Report of the Intergovernmental Panel on Climate Change, Cambridge Univ. Press, Cambridge, UK
Twomey, S. (1977). Influence of pollution on the short-wave albedo of clouds. J. Atmos. Sci. 34, 1149-1152
Welton, E. J., Campbell, J. R. & Spinhirne, J. D. (2001) Global monitoring of clouds and aerosols using a network of micropulse LIDAR systems, Proc. LIDAR Remote Sensing for Industry and Environmental Monitoring, 4153, 151–158.
Winker, D. M., Hunt, W. H., and McGill, M. J. (2007). Initial performance assessment of CALIOP, Geophys. Res. Lett., 34, L19803, doi:10.1029/2007GL030135.
Our planet is nowadays continuously monitored by powerful remote sensors operating in wide portions of the electromagnetic spectrum. Our capability of acquiring detailed information on the environment has been revolutionized by revealing its inner structure, morphology and dynamical changes. The way we now observe and study the evolution of the Earth’s status has even radically influenced our perception and conception of the world we live in. The aim of this book is to bring together contributions from experts to present new research results and prospects of the future developments in the area of geosciences and remote sensing; emerging research directions are discussed. The volume consists of twenty-six chapters, encompassing both theoretical aspects and application-oriented studies. An unfolding perspective on various current trends in this extremely rich area is offered. The book chapters can be categorized along different perspectives, among others, use of active or passive sensors, employed technologies and configurations, considered scenario on the Earth, scientific research area involved in the studies.

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