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A review of stratospheric aerosol characterization

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Abstract

We present a general overview of the efforts devoted to the characterization of stratospheric aerosols. After recalling the most important parameters used to characterize aerosols, we present an overview of methods used for retrieving these parameters from radiative measurements. We subsequently review the most important climatological studies developed to characterize stratospheric aerosols, and analyse the main contributions and limitations of these works. Finally, we try to identify current topics of interest and perspectives in the field of aerosol characterization.

Keywords: Stratospheric aerosols; Middle atmosphere

1. Introduction

Aerosols are known to play an important role in the physico-chemistry of the stratosphere, e.g. through their role in the formation of polar stratospheric clouds (PSC) and hence in the ozone depletion processes. During important volcanic eruptions, a high amount of sulfate aerosol is injected in the stratosphere (Hofmann, 1988), inducing various changes in the chemistry, thermodynamics and transport in the atmosphere (see Russell et al., 1996 and references therein). For this reason, it is of great importance to be able to characterize aerosol microphysical properties in a reliable way in order to quantify their impact on atmospheric processes and species. Major difficulties in this task are associated with the high variability of the stratospheric aerosol load (some aerosol parameters vary over several orders of magnitude in time and space), and the aerosol distribution in a population of particles with varying shape, composition and size. The present paper reviews the efforts made to describe the microphysical properties of this particle population, focusing on experimental techniques, theoretical description, retrieval methods and the numerical quantification through the build-up of aerosol climatologies.

2. Data sources

Various experimental techniques are used to get information on stratospheric aerosols, through the direct observation of their microphysical features or indirectly through the observation of their radiative properties.

In situ measurements by airborne or balloonborne optical particle counting (Deshler et al., 1992, 1993; Sugita et al., 1999), wire impactors (Sheridan et al., 1992; Goodman et al., 1994; Pueschel et al., 1994) or spectrometers (Berthet et al., 2002; Renard et al., 2002) give direct measurements with a high vertical resolution of local microphysical aerosol properties, such as the shape, size distribution and composition. Their ability to discriminate a wide range of particles, from condensation nuclei (size $\sim$0.1 μm) to large aerosol droplets (size $\sim$1 μm) makes possible a reliable determination of the size distribution, included in the case of multimodal size distributions. When campaigns are planed on a regular time base, time series becomes available over a long period, as in the case of

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the Wyoming data set (Deshler et al., 2003). Such a data set is a precious tool for the study of long-term trends. Specific difficulties concern the quite high uncertainty on the size distribution for large particle sizes, related to the difficulty to detect a statistically representative number of large particles. Pueschel et al. (1994) mentions the problem of evaporation by adiabatic heating of air in the entrance of the impactor, leading to particle losses and to an underestimation of the particle radius. Finally, wire-impactors generally underestimate the particle number with respect to dust sondes, optical filters and laser optical probes (Goodman et al., 1994).

Ground-based instruments typically provide integrated measurements, although lidar also gives an access to vertical extinction profiles with a good resolution. However, the interpretation of the backscattered signal is quite complex (Böckmann et al., 2004) and due to the very reduced number of spectral channels, additional information is required for the derivation of size information (Jäger and Hofmann, 1991; Wandinger et al., 1995; Jäger and Deshler, 2002). The availability of instruments measuring routinely allows the constitution of long time series able to detect long-term trends.

Remote sensing experiments from space allow measurements over a large spatio-temporal range, but with a variable vertical resolution depending on the observation geometry (nadir, occultation or limb) as well as the light source (sun, stars or other celestrial bodies). In this sense, they provide well-suited data sets for the study of large-scale effects, influences such as seasonal effects or long-term variations. But their ability to render fine structures in the vertical aerosol profiles or details about the evolution of air mass motion may be limited and depends on their resolution capabilities and measurement rate.

Remote sensing experiments in the UV–visible range measure the scattering properties of aerosols (the absorption coefficient of sulfate aerosols being negligible in this spectral range), and particle size distributions can be derived from the data (Fussen et al., 2001b; Bauman et al., 2003a; Bingen et al., 2003). However, this derivation is biased due to the impossibility to detect the size of small particles in the Rayleigh limit of scattering, as will be discussed in Section 3. Further, the absorption of UV radiation by stratospheric ozone and the significant influence of Rayleigh scattering at short wavelengths makes these experiments inoperative at low altitude. Following volcanic eruptions, strong scattering of the radiation at the level of the volcanic plume is able to reduce the signal under the sensitivity limit of the detector and to inhibit measurements at lower altitude.

On the other side, remote sounding in the infrared range works in absorption mode, and can give information about the aerosol composition (Grainger et al., 1993; Mergenthaler et al., 1993 and references therein). Difficulties are related to the high dependence of the absorption cross section on the refraction index, which in turn is very sensitive to the temperature. Since they concern large wavelengths with respect to the particle size, IR measurements give only access to integrated microphysical aerosol parameters, but are not able to provide information about the size distribution without additional information (see discussion in Section 3). The retrieval of microphysical aerosol parameters requires a good knowledge of the thermodynamic conditions, and the knowledge of reliable values for the refraction index. Further, IR remote sensing allows observations down to low altitudes, even during periods of high aerosol content. As an illustration, Fig. 1 presents a comparison presented by Grainger et al. (1995) between surface area densities at April 2, 1992 derived from the 12.1 μm ISAMS channel (color map) and corresponding data derived from SAGE II (UV–visible range). ISAMS data show a good resolution of the structures down to the lowest considered altitudes, contrarily to SAGE II derived contours.

Due to their specificity, remote and in situ techniques are very complementary. Combining the kind of information that can be rendered by each of them, allows potentially to derive reliable aerosol characteristics over a large spatio-temporal range.

3. Characterization of stratospheric aerosols

The radiative properties of a population of spherical aerosol droplets are characterized by the extinction coefficient which is given by

$$\beta(\lambda) = \int_0^\infty N \cdot Q_{\text{ext}}(\lambda, m, r) \cdot f(r) \mathrm{d}r, \tag{1}$$

where N is the particle number density expressed in cm^{-3}, $f(r)$ is the normalized particle size distribution, and Q_{ext} is the extinction cross section depending on the wavelength λ and the particle radius r. The spectral dependence of Q_{ext} involves a dependence on the index of refraction m, which in turn depends on the aerosol composition influenced by thermodynamical parameters such as temperature, humidity and pressure (Steele and Hamill, 1981). Stratospheric aerosols consist essentially of a mixture of H_2SO_4 and H_2O. Reference values for the refraction index can be found in the literature (Remsberg et al., 1974; Palmer and Williams, 1975) at some temperatures of interest and for different values of the aerosol composition (also called *aerosol acidity* and having the notation W in the following), expressed as the fraction by weight of H_2SO_4 in the mixture. Q_{ext} can be determined by the Mie scattering theory (van de Hulst, 1995), showing a marked oscillating behaviour in the diffraction regime ($r/\lambda \simeq 1$). For very small particles with respect to the particle size (Rayleigh regime), Q_{ext} becomes only dependent on λ, whereas Q_{ext} tends to a constant value $Q_{\text{ext}} = 2\pi r^2$ for large particles compared to λ (geometrical approximation). In the cases of very small and very large particles, extinction measurements are unable to provide information about particle size.