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DETERMINATION OF PINATUBO-AEROSOL OPTICAL DEPTH AND MICROPHYSICAL PARAMETERS WITHOUT CRITICAL ASSUMPTIONS

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The most important parameters of the Pinatubo aerosol for climate and air-chemistry research are the optical depth and the height profile of the particle surface-area concentration. These essential parameters can be determined from lidar measurements without any critical assumption, if the particle extinction and backscatter height profiles can be measured simultaneously. In addition, the particle volume concentration and the effective radius of the size distribution can be estimated from these measurements. The widely used elastic-backscatter lidar cannot provide the data of interest since this lidar type allows to determine the particle backscatter profile only. Assumptions relating backscattering with surface area and optical depth are needed and lead to large uncertainties in the determined parameters. A simultaneous profiling of the particle extinction and backscatter is possible with a combined Raman elastic-backscatter lidar (Ansmann et al. 1992, Ferrare et al. 1992, Ansmann et al. 1993) or by the use of the HSRL technique (Shipley et al. 1983, Eloranta et al. 1992).

While the optical depth is directly obtained from the extinction profile, the microphysical properties are determined by the use of relationships found from Mie scattering calculations. Extinction and backscattering are related to surface-area concentration A and volume concentration V of the particles in the following way. α and β can be expressed by

$$\alpha = \int Q_\alpha(r)\, N(r)\, \pi r^2\, dr = \tilde{Q}_\alpha \int N(r)\, \pi r^2\, dr\,, \tag{1}$$

$$\beta = \int Q_\beta(r)\, N(r)\, \pi r^2\, dr = \tilde{Q}_\beta \int N(r)\, \pi r^2\, dr\,. \tag{2}$$

$N(r)$ is the particle number density in the radius interval $r\,dr$. $Q_\alpha(r)$ is the extinction efficiency and $Q_\beta(r)$ the backscattering efficiency for a single sulfuric-acid droplet with radius r. $\tilde{Q}_\alpha$ and $\tilde{Q}_\beta$ are introduced as the appropriate effective efficiencies of the particle size distribution. A and V can then be expressed by

$$A = 4\pi \int N(r)\, r^2\, dr = \frac{4\alpha}{\tilde{Q}_\alpha} = \frac{4\beta}{\tilde{Q}_\beta}\,, \tag{3}$$

$$V = \frac{4\pi}{3} \int N(r)\, r^3\, dr = \frac{4\tilde{r}\alpha}{3\tilde{Q}_\alpha} = \frac{4\tilde{r}\beta}{3\tilde{Q}_\beta}\,, \tag{4}$$

with the effective radius

$$\tilde{r} = \frac{\int N(r)\, r^3\, dr}{\int N(r)\, r^2\, dr} = \frac{V}{3A}\,. \tag{5}$$

By the use of Fig. 1, in which Mie scattering calculations for realistic bimodal stratospheric aerosol size distributions measured between April 1991 and December 1992 (Deshler et al. 1993) are shown, first $\tilde{r}$, $\tilde{Q}_\alpha$, and $\tilde{Q}_\beta$ and finally A and V are determined from the profiles of α and β. The lidar ratio

$$S = \frac{\alpha}{\beta} = \frac{\tilde{Q}_\alpha}{\tilde{Q}_\beta} \tag{6}$$

contains the information on the effective radius of the particle size distribution. The fitted

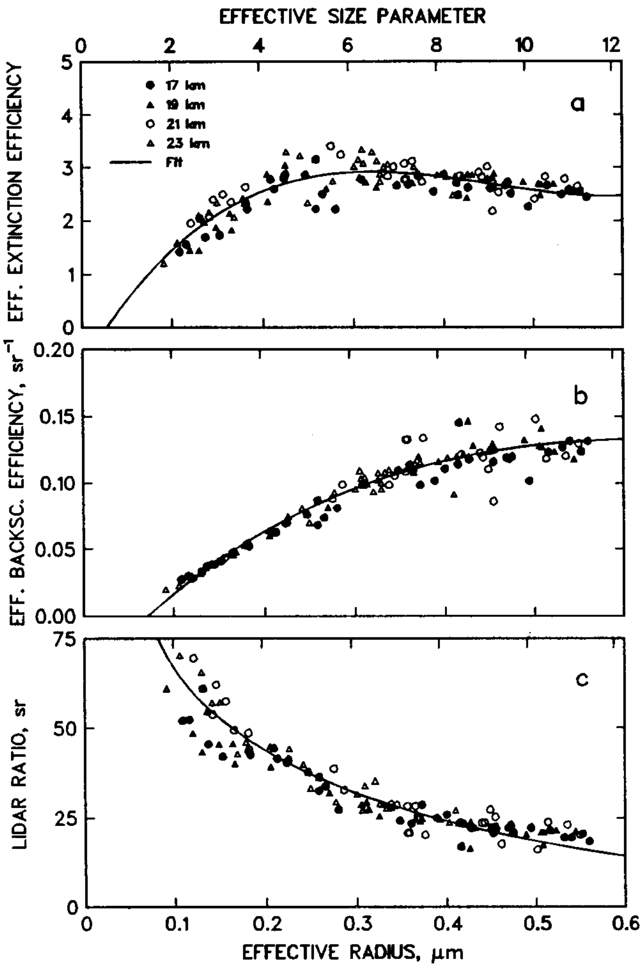

Fig. 1: Effective extinction efficiency (a), effective backscattering efficieny (b), and lidar ratio (c) related to the effective radius of aerosol size distributions measured at different heights in 1991 and 1992 (Deshler et al. 1993). The values are calculated after the Mie scattering theory for the laser wavelength of 308 nm of the GKSS Raman lidar and a refractive index of $1.45 + i10^{-8}$.

curve in Fig. 1c gives

$$\tilde{r} = 0.534\, S^{\frac{1}{2}} - 1.75\, S^{\frac{1}{3}} + 2.833. \quad (7)$$

As long as the effective size parameter $\tilde{x} = 2\pi\tilde{r}/\lambda$ is less than 8, i.e. $\tilde{r} < 0.4\,\mu$m for the laser wavelength $0.308\,\mu$m, the effective radius can be determined with a relative error of less than 25 %. A lidar ratio < 25 sr indicates an effective radius $> 0.4\,\mu$m. The uncertainty in this region is large. In the next step $\tilde{Q}_\alpha$ and $\tilde{Q}_\beta$ are found from Fig. 1a and 1b (fitted curves) by using the formulas

$$\tilde{Q}_\alpha(\tilde{r}) = 49.74\,\tilde{r}^3 - 67.34\,\tilde{r}^2 + 27.79\,\tilde{r} - 0.70, \quad (8)$$

$$\tilde{Q}_\beta(\tilde{r}) = 0.40\,\tilde{r}^3 - 0.94\,\tilde{r}^2 + 0.72\,\tilde{r} - 0.05. \quad (9)$$

Due to the uncertainty in the estimate of $\tilde{r}$, the relative errors for the range $\tilde{r} < 0.4\,\mu$m ($x < 8$) are 40 % and 30 % for $\tilde{Q}_\alpha$ and $\tilde{Q}_\beta$, respectively. For $\tilde{r} > 0.4\,\mu$m ($x > 8$) the relative errors of $\tilde{Q}_\alpha$ and $\tilde{Q}_\beta$ are about 20 % and 30 %, respectively. Thus, assuming statistical errors of α and β of the order of 20 % and 10 %, respectively, the surface-area concentration can be determined with an error of about 35 % from the β profile for $\tilde{r} < 0.4\,\mu$m and with an error of about 30 % from the α profile for $\tilde{r} > 0.4\,\mu$m.

Finally, the volume or mass concentration can be determined after Eq. (4) with a relative error of 40 % for $\tilde{r} < 0.4\,\mu$m. Only a rough estimate is possible for lidar ratios < 25 sr and corresponding $\tilde{r}$ values $> 0.4\,\mu$m at a laser wavelength of 308 nm. Since $\tilde{Q}_\beta$ is linearly related to $\tilde{r}$ for $x < 6$, so that $\tilde{r} = b\,\tilde{Q}_\beta$ could be used in Eq. (4), a good determination of the volume concentration directly from the β profile is possible for wavelengths > 500 nm if b is known from Mie scattering calculations.

Fig. 2 presents examples of evaluated lidar data. Two cases measured with the Raman lidar of GKSS in the winters 1992 and 1993 are shown. The error bars include statistical errors of α and β and errors due to the uncertainties in the determination of $\tilde{r}$, $\tilde{Q}_\alpha$, and $\tilde{Q}_\beta$ from Fig. 1. An additional error of roughly 25 % due to a necessary correction of ozone absorption at 308 nm is not shown here. For stratospheric aerosol studies laser wavelengths $\lambda > 350$ nm should be used to avoid the ozone interference. More details of the Pinatubo-aerosol measurements performed at Geesthacht (53.5° N) since August 1991 will be presented at the conference.

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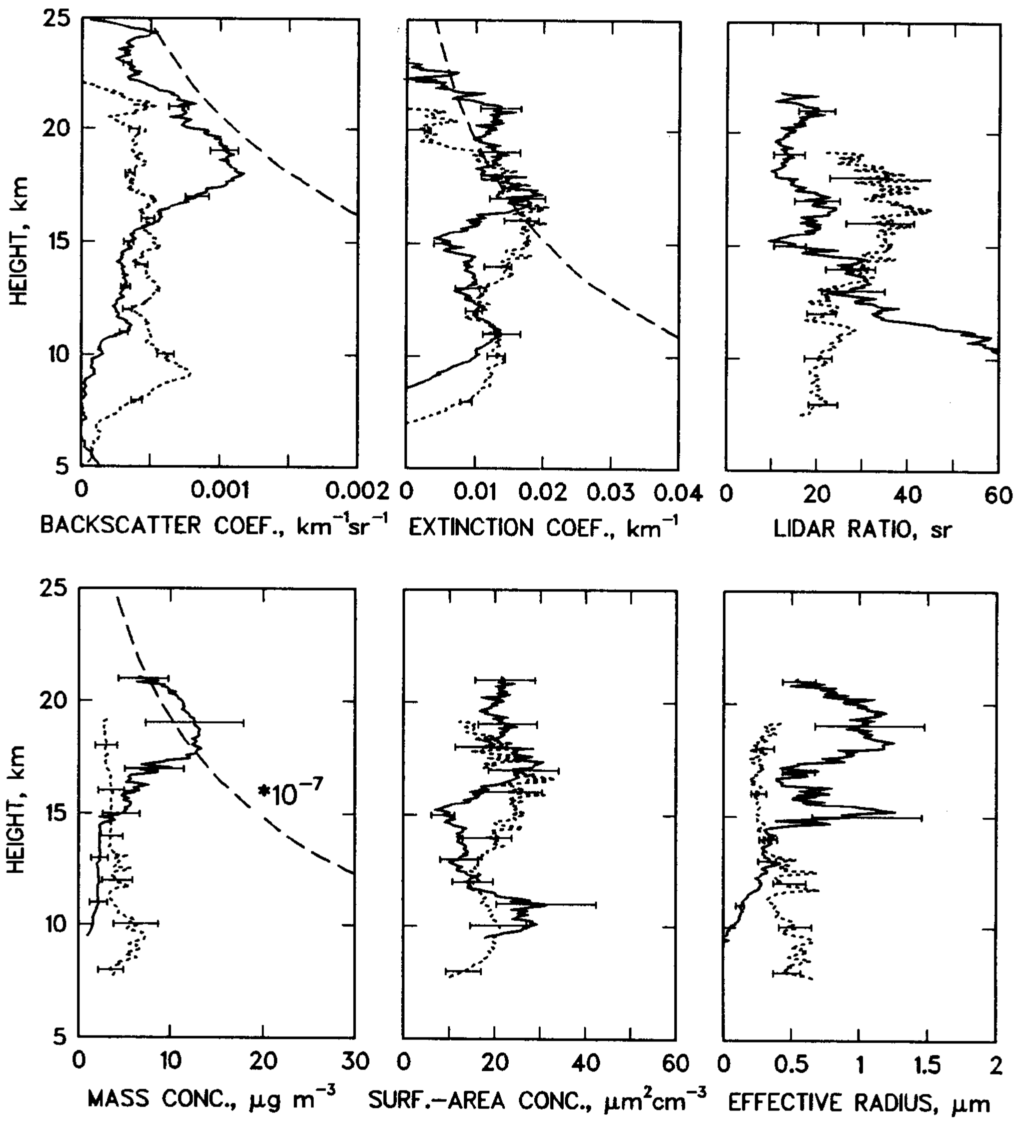

Fig. 2: Backscatter coefficient, extinction coefficient, lidar ratio, mass concentration, surface-area concentration, and effective radius of the stratospheric aerosol measured with the Raman lidar at 308 nm over Northern Germany on 22 January 1992 (solid) and on 18 January 1993 (dotted). Dashed lines show Rayleigh backscatter and extinction coefficients and air density for comparison.