

TITLE: Vertical profiles of cloud condensation nuclei, condensation nuclei, optical aerosol, aerosol optical properties, and aerosol volatility measured from balloons.
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Under the support of this grant a balloon-borne gondola containing a variety of aerosol instruments was developed and flown from Laramie, Wyoming, (41°N, 105°W) and from Lauder, New Zealand (45°S, 170°E). The gondola includes instruments to measure the concentrations of condensation nuclei (CN), cloud condensation nuclei (CCN), optically detectable aerosol (OA) ($r \geq 0.15 - 2.0 \mu\text{m}$), and optical scattering properties using a nephelometer ($\lambda = 530 \text{ nm}$). All instruments sampled from a common inlet which was heated to 40°C on ascent and to 160°C on descent. Flights with the CN counter, OA counter, and nephelometer began in July 1994. The CCN counter was added in November 1994, and the engineering problems were solved by June 1995. Since then the flights have included all four instruments, and were completed in January 1998. Altogether there were 20 flights from Laramie, approximately 5 per year, and 2 from Lauder. Of these there were one or more engineering problems on 6 of the flights from Laramie, hence the data are somewhat limited on those 6 flights, while a complete data set was obtained from the other 14 flights. Good CCN data are available from 12 of the Laramie flights. The two flights from Lauder in January 1998 were successful for all measurements.

Laramie is a mid-continental site, without major local pollution sources so that measurements are expected to be representative of free-tropospheric conditions from just short distances above the surface. Lauder, New Zealand, offers a nice contrast to Laramie since measurements there are representative of the remote free troposphere in the mid-latitude southern hemisphere.

The results from these flights, and the development of the balloon-borne CCN counter have formed the basis for five conference presentations (Snider and Deshler, 1995; Deshler et al., 1995; Wechsler et al., 1996; Delene and Deshler, 1996; Snider et al., 1997), two manuscripts (Delene et al., 1998; Hofmann et al., 1998), one M. S. thesis (Lei, 1998) and one Ph. D. dissertation, in progress, (Delene, 1998).

The heated and unheated CN and OA measurements have been used to estimate the mass fraction of the aerosol volatile at 160°C, $(V_{40} - V_{160})/V_{40}$, while comparisons of the nephelometer measurements were used to estimate the light scattering (b_{sp}, m^{-1}) associated with the volatile aerosol. These estimates were calculated for 0.5 km averages of the ascent and descent data between 2.5 km and the tropopause, near 11.5 km. Since the selected temperatures should result in the removal of chemically bound water (at 40°C) and both water and sulfuric acid (at 160°C) we assume that the relative differences, $(V_{40} - V_{160})/V_{40}$ and $(b_{sp}(40) - b_{sp}(160))/b_{sp}(40)$, result from the volatilization of sulfuric acid. Values of both $(V_{40} - V_{160})/V_{40}$ and $(b_{sp}(40) - b_{sp}(160))/b_{sp}(40)$ were approximately 0.3 at 2.5 km and increased to 0.8 at the tropopause. These results indicate that a

significant fraction of the aerosol mass behaved in a manner consistent with that expected for sulfuric acid, that this fraction increased through the upper troposphere, and that the optical properties of the aerosol was strongly influenced by a sulfuric acid component. Values of the sulfate scattering efficiency corresponding to the volatilized aerosol mass were also derived. Results from these measurements have been presented at the International Union of Geodesy and Geophysics meeting, Boulder (Snider and Deshler, 1995), and at the Aerosol Interdisciplinary Program Workshop, Columbia, Maryland (Deshler et al., 1995).

For the CCN counter a significant effort was devoted to developing an effective calibration system. The calibration system, associating the measured intensity of the scattered light with the droplet concentration in the chamber, now provides a reproducible correlation between these two quantities. The successful CCN flights have provided vertical profiles of CCN concentration between the surface and 200 mb, where the instrument is turned off to avoid difficulties in controlling the plate temperatures in the stratosphere. Comparing the CCN profiles with CN and aerosol with radius $\geq 0.15 \mu\text{m}$ indicates that the rate of concentration change with height for all three of these aerosol types are similar and that layers in the vertical profiles are usually reflected in all three aerosol types. On some flights comparison of the heated and unheated profiles show distinct layers where sulfuric acid appears to provide a major component to the CCN population, while on other flights there is less of a difference between the unheated and heated measurements. These CCN measurements may aid in characterizing patterns of CCN source distributions, and in narrowing, to the extent possible with volatility measurements, the chemical composition of the mid and upper tropospheric CCN. Correlative measurement of CCN with OA and CN will allow a firmer understanding of the variability and sources for CCN.

Presentations on the CCN measurements were given at the 14th International Conference on Nucleation and Atmospheric Aerosols, Helsinki, (Wechsler et al., 1996) and at the American Geophysical Union fall meeting, San Francisco (Delene and Deshler, 1996). A manuscript describing the instrument, the calibration system, and some of the vertical CCN profiles has been accepted for publication in *J. Geophys. Res.* (Delene et al., 1998). This effort has also been the focus of D. Delene's Ph. D. work (Delene, 1998).

Comparing the optical scattering measured with the nephelometer with that calculated from the CN and OA measurements of aerosol size distribution has been the subject of additional work. The measured size distributions from the CN and OA counter are used, along with Mie theory, to estimate the extinction expected from the tropospheric aerosol. These estimates are compared with the nephelometer measurements of b_{sp} , which were corrected to account for two known systematic errors: 1) angular truncation of the scattered light, and 2) the non ideal illumination source. This comparison has been done for field measurements, where an index of refraction is assumed, and for measurements in the laboratory where particle index of refraction is known. In the laboratory, using polystyrene latex beads, the calculated and observed scattering coefficients agree to within 10% for particles with radii $< 0.30 \mu\text{m}$ and $> 0.40 \mu\text{m}$. For measurements between $0.3 - 0.4 \mu\text{m}$ the calculated scattering is 20 - 40% below that measured. In the lowest layer of the atmosphere above Laramie (2.5 - 5.0 km) the comparison is not so good. The optical depth calculated from the CN and OA

measurements is approximately a factor of two below the optical depth measured by the nephelometer. The source of the discrepancy between the measured and calculated optical depths from the field measurements is still under investigation. This work has been presented at the American Association for Aerosol Research's annual meeting, Denver (Snider, et al., 1997) and is nearly complete as an M. S. thesis (Lei, 1998).

Presentations/Publications resulting from this grant

Conference Presentations:

- Snider, J. R., and T. Deshler, Optical and physical properties of upper tropospheric aerosol sampled through a heated inlet over Laramie, Wyoming, *International Union of Geodesy and Geophysics XXI General Assembly*, Boulder, Colorado, July 1995.
- Deshler, T., J. R. Snider, G. Vali, Vertical profiles of cloud condensation nuclei, optical aerosol, aerosol optical properties, and aerosol volatility measured from balloons, Aerosol interdisciplinary program workshop, Columbia, Maryland, October 1995.
- Wechsler, P., G. Vali, J. R. Snider, and T. Deshler, Vertical profiles of cloud condensation nuclei from a balloon-borne instrument, *Fourteenth international conference on nucleation and atmospheric aerosols*, Helsinki, (*Nucleation and atmospheric aerosols 1996*, 911-914) August 1996.
- Delene, D. J., and T. Deshler, Balloon-based measurements of cloud condensation nuclei at Laramie, Wyoming, *American Geophysical Union*, San Francisco, December, 1996.
- Snider J. R., W. Lei, and T. Deshler, Comparison of calculated and measured aerosol scattering coefficients obtained from balloon-borne measurements, *American Association for Aerosol Research*, Denver, Colorado, October, 1997.

Manuscripts:

- Delene, D., T. Deshler, G. Vali, and P. Wechsler, A balloon-borne cloud condensation nuclei counter, *J. Geophys. Res.*, in press, 1998.
- Abstract: A balloon-borne instrument was constructed for observations of vertical profiles of cloud condensation nucleus (CCN) concentrations, active at 1% supersaturation. Droplet concentration in the static thermal-gradient diffusion chamber is deduced from the amount of scattered laser light detected by a photodetector. The photodetector is calibrated using a video camera and computer system to count the number of droplets produced from NaCl aerosol. Preliminary data are available from nine early morning profiles obtained at Laramie, Wyoming, between June 1995 and January 1997. To complement the CCN measurements, instruments that measure condensation nuclei (CN) and aerosols with diameter greater than 0.30 μm ($D_{0.3}$) were also included on the balloon package. CCN concentrations exhibited a general decrease from the surface to the top of the boundary layers, were generally uniform through well-mixed layers, and show variability above well mixed layers. In general, the structure of

the CCN profile appears to be closely related to the structure in the CN and $D_{0.3}$ profiles. Summer profiles generally have CCN concentration greater than 200 cm^{-3} up to 500 mb, whereas winter profiles are less than 200 cm^{-3} at all levels.

Hofmann, D. J., R. Stone, M. Wood, T. Deshler and J. Harris, An analysis of 25 years of balloonborne aerosol data in search of a signature of the subsonic commercial aircraft fleet. *Geophys. Res. Lett.*, in press, 1998.

Abstract: The University of Wyoming balloonborne condensation nuclei (CN) record of 25 years (1983-1997) was analyzed to determine possible effects of commercial aircraft on the 8.6 - 12.7 km altitude range of the atmosphere, which includes the primary commercial air lanes between 29 and 41 kft. Thin layers of highly concentrated small particles are often observed in this region of the atmosphere. However, this region is also one of maximum production of natural particles related to photochemical processes. Generally, aircraft flight information is not available for past balloon soundings making it impossible to ascribe a source to any specific observed CN layers. However, a CN layer observed in March 1997 was traced to a particular aircraft, with the help of the Federal Aviation Administration, thus supporting the hypothesis that at least some of the observed layers are contrail remnants. Using the Laramie data set, a method was developed to quantify the enhancement in CN concentration induced by aircraft in comparison with natural background levels. We estimate conservatively that the contribution of the commercial aircraft fleet in the vicinity of Laramie, Wyoming, amounts to about 5-13% of the total measured concentration, depending on season.

Theses:

Lei, W., Comparison of calculated and measured aerosol optical scattering coefficients from vertical profile measurements, M. S. Thesis, University of Wyoming, May 1998.

Delene, D., Vertical profiles of cloud condensation nuclei above a clean mid-continental site. Ph. D. thesis, University of Wyoming, December 1998.