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Condensation nuclei at the German Antarctic Station "Georg von Neumayer"

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ABSTRACT

Evaluations of measurements of the tropospheric surface condensation nuclei at the German Antarctic station for the last 8 years are presented. They show clearly the annual variation of the concentration with a maximum in austral summer, as previously seen by other investigators. In addition, a pronounced increase of the concentration of 9.63 % per year has been found. The size distributions of the condensation nuclei of measurements over 2 years are evaluated and discussed. On average, they are of mono-modal shape, but individually most of them show a bimodal shape.

1. Introduction

The Antarctic atmosphere certainly belongs to the clean air regime and our understanding of the dynamics and properties still is poor. In 1981, the German Antarctic Station "Georg von Neumayer" (GvN) was opened and equipped with a condensation nuclei (CN) counter. Measurements have been carried out continuously since then. The concentration of the CN is one parameter of the atmosphere closely related to the pollution of the atmosphere, the cloud and rain forming process, and the climate of the earth. In addition, the concentration of CN is a very sensitive indicator for human activities. If a change in concentration is observed, our attention is needed to explain it and further observations are recommended.

2. Location and instrumentation

The German Antarctic station is located at 70°37′S and 8°22′W at the so called Atka Bay on the Ekström Shelf Ice. The station itself is dug under the surface about 7 km on the shelf, west of the Atka Bay. The station is manned year round with about 11 scientists and technicians in winter and an increased number of visiting scientists during late December to mid February.

From the main station a power line supplies a geophysical observatory and the air chemistry container 1.5 km to the South. This container is equipped with various instruments for measuring atmospheric trace constituents (Münnich and Jaenicke, 1989). The CN counter is a commercially available instrument (Model 3020 of Thermo Systems Inc), based on alcohol supersaturation. The measurements serve two purposes: The investigation of the CN and the protection of other air chemistry measurements from local pollution. It is well known that human activities produce large concentrations of CN. Therefore the counter shuts off the power supply to other air chemistry measurements (mostly collecting and concentrating devices) if the concentration exceeds a certain threshold, usually 2000 cm^{-3}. Only the CN counter continues to measure and returns power to the other devices, if for half an hour the concentration remains lower than the threshold. Besides CN observations, the air chemistry program at the Georg von Neumayer-station now includes: ozone, non-methane hydrocarbons, carbon isotopes in carbon dioxide, major ions (sea salt, sulphate, nitrate) in aerosols, trace elements (Mn, Pb, radioactive) of the aerosol mass, ^{85}Kr, and ^{222}Rn.

The CN counter runs year round. The instrument is replaced once every year with a second instrument. The instruments are shipped to the

laboratory in Mainz, maintained, calibrated and eventually returned to Antarctica in the following year. The setup is completely self contained and needs maintenance usually once a week to adjust the alcohol supply, change printouts and magnetic data tapes. The instrument is equipped with temperature sensors, protecting it from running at temperatures exceeding the operating range. Such cases might happen after the air chemistry container is disconnected from the pwer supply in case of an emergency exercise.

The recorder registers the concentration and additional parameters of the instrument (power supply, temperature, flow rate, etc.) continuously on paper as backup. Once every hour the data are recorded additionally on tape (or disks) for easy data transfer. In some of the years (1985, 1988) the tape recording failed. For those years the data were later transfered manually from the backup.

The CN counter grows the particles to about 10 μm droplets and runs in two modes: scatter and count mode. In scatter mode, the light scattered from the particles in the optical sensitive volume is taken as a measure for the total particle concentration. The count mode registers individual droplets. Our laboratory did additional calibration (Weber and Jaenicke, 1988). The counting mode is sensitive to concentrations below 1 cm^{-3}. This permits the use of diffusion screens for estimating the size distribution. As the concentration increases in count mode corrections because of coincidence loss are needed. With further increase the automatic switch to scatter mode for higher concentrations is reached.

The use and comparison with other condensation nuclei counters make sense only, if the counting efficiency and the smallest particle size detectable are known. As in our case, the instrument is adjusted in comparing it with a water based Aitken particle counter (Jaenicke, 1980). This counter is tuned to see all aerosol particles larger than the size of small ions. In other experiments (Haaf and Jaenicke, 1977) a comparable Aitken nuclei counter has been shown to detect particles as small as 1.6 $(+0.2/-0.4)$ nm. This device does very well in international comparisons (Liu et al., 1980). The comparison between the counter used in Antarctica and the Aitken nuclei counter mentioned above is carried out with urban atmospheric aerosols containing steep concentrations of gas-to-particle conversion (GPC) aerosols (There is a simple and qualitative test for small particles being detected. If, in urban aerosols with steep particle concentrations, a 60 cm long hose (8 mm diameter) is attached to the inlet and the recorded concentration drops, particles much smaller than 5 nm are detected.).

During 1982/83 and 1986/87 the CN counter was supplemented with a diffusion battery (Model 3042 of Thermo Systems Inc) of the screen type in order to obtain information about the size distribution of the CN. To serve the twofold purpose, measurements and protection, the measurements were arranged as follows. Every 24 h all other instruments in the air chemistry container were disconnected from power for about one hour. This time was needed for measuring the size distribution because of the low concentrations in Antarctica and sufficient statistical accuracy. Each channel of the diffusion battery was connected to the instrument for about 5 min with 1 min allowing for adjustment. Thus a complete set of channel measurements was obtained within one hour. The size distributions were evaluated only if the total concentration at the beginning and end of the measuring period have been constant (within 1% difference). The time shift of 24 h plus 1 h for measurement permitted obtaining size distributions not only at a specific hour but through the whole day.

3. Data processing and evaluation

For 1983 to 1989, the concentration data are available on tape once every hour. The data of 1982 could only be evaluated roughly as monthly averages. Negative data were removed indicating failures of the recording. Additionally data exceeding 10^5 cm^{-3} were removed. This is a rather arbitrary limit, but it was selected, because anthropogenic pollution cannot be excluded at this location, especially in summer time. With the prevailing easterly winds and the anchor location of the supply vessels again to the East, commuter vehicle exhaust can be carried to the air chemistry container. However, the supply vessels visits only for a few days and in winter time the station crew controls the local pollution. Northerly winds are rather infrequent, so pollution from the main station usually can be excluded. The 10^5 cm^{-3}

seems justified compared to Bigg's et al. (1984) selection.

The data were screened further and some months excluded from evaluation. These are March 1984, October 1984, and May 1985. During these months the recording changed frequently on an hourly basis between concentrations up to 10^5 cm^{-3} and below 10 cm^{-3}. The average of these months were larger than 1000 cm^{-3}. The reason for such obvious failures remains unexplained. Further, the data were corrected for coincidence loss in the counting mode according to Jaenicke (1972).

In 1986/87, 167 aerosol size distributions could be evaluated. Often concern is expressed that bimodal diffusion battery size distributions are suspect. Indeed, the algorithm of retrieving the size distribution needs attention. The inversion procedure used in this paper follows Tichonov's regularisation (Bashurova et al., 1992) which results in numerically stable size distributions. Careful examinations (Koutsenoguii, 1992) of the evaluation procedure showed in addition the channels of the diffusion battery being not completely independent. The complete information is contained in fewer channels than used. As a consequence, at the most two modes can be evaluated from those measurements. To obtain size distributions, the set of 10 penetrations for every run of diffusion battery data was calculated. The penetrations were determined as the ratios of concentrations measured to the total number concentration. All the penetration sets for one period discussed below were averaged and an average set obtained. This set than was multiplied with the average concentration for that period and numerically inverted.

4. Results and discussion

The presentation and discussion of the total particle concentration and the size distributions will be done separately.

4.1. Concentration of condensation nuclei

Fig. 1 shows the monthly averages of the 8 years of observation at the Georg von Neumayer-station. They confirm the annual variation with the maximum in summer and the minimum in winter as reported by Bigg et al. (1984) and Samson et al. (1990).

The average over the years 1982 to 1989 is 349 cm^{-3}. That has to be compared with the South Pole Amundsen–Scott station's 130 cm^{-3} (Standard Temperature and Pressure, STP) for the years 1974 to 1985 and 105 cm^{-3} (STP) for 1984 to 1988.

A regression for the concentration as function of time was calculated and added to Fig. 1 together with the confidence interval (95%). This regression shows a significant constant increase over the years totaling in 28.9 cm^{-3} yr^{-1} or 9.63%/yr (if the base is the beginning of each year, 8.7%/yr if the base is the average concentration of the observation period and the period is considered as one unit). This is a rather dramatic increase never reported before and certainly should be monitored over the following years. For the Amundsen–Scott station (Samson et al., 1990) a slight decrease has been calculated.

From the data, the monthly averages during the year were calculated, modified with the above calculated increase of 28.9 cm^{-3} yr^{-1} and added to Fig. 1. Thus it reflects the change of the average months with the overlaying increase. Because of the fluctuations superimposed on the yearly variations, the average year is not smooth, rather it shows two maxima, in December and March. A similar two peaked curve can be calculated for the South Pole. Early in 1984 the concentrations are exceptionally low, even lower than at the South Pole.

The correlation coefficient between the data at GvN and Amundsen–Scott is +0.44 (significant at the 95% level), indicating some common history of the particles. However, deviations occur. Such deviations most probably can be attributed to sudden increases reported by Ito and Iwai (1981). Such sudden increases are observed rather regularly at the Georg von Neumayer-Station. Optical measurements (Leiterer and Sakunow, 1989) suggest a vertical profile with high concentrations aloft for larger particles. Thus the sudden increase might be explained with subsiding air.

The variation in concentration between summer and winter exceeds 1.5 orders of magnitude, more than reported for Mawson and the South Pole (Bigg et al., 1984; Samson et al., 1990). The Georg von Neumayer-station is rather close to open waters in late summer, thus participating directly from the maritime particle production. Other aerosol parameters such as sulphate mass, show

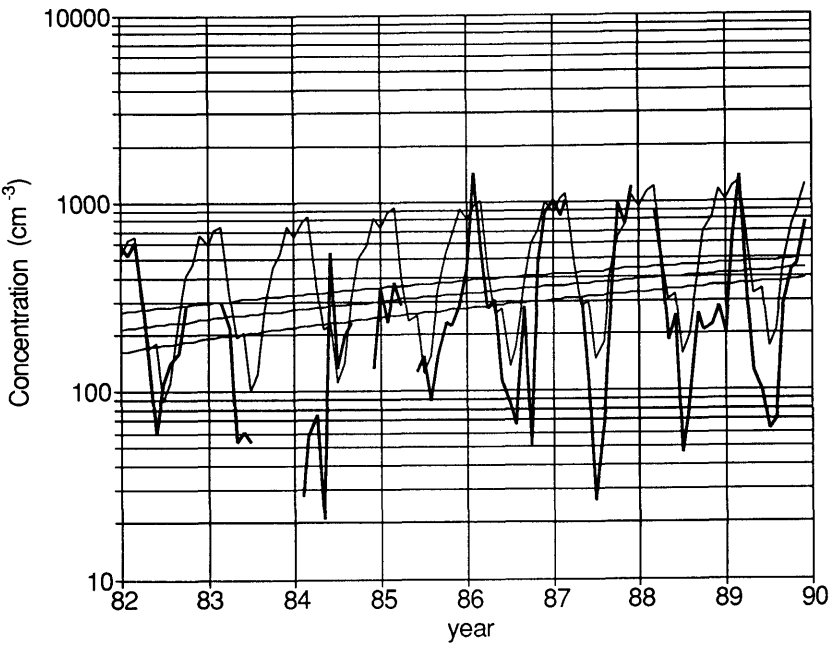

Fig. 1. Monthly averages of CN concentration at the Georg von Neumayer-station (solid bold line, interrupted by missing observations). 1982 is based on monthly estimates only, while the other months have been calculated from hourly observations. The slightly increasing line represents the regression together with the confidence intervals, and the repeating pattern (solid thin line) are the average months plus the increase. The modification reflects the overlaying increase of the concentration of 9.63 %/yr.

a similar variation. However that maximum is rather narrow around January at the time of maximum sun radiation (Münnich and Jaenicke, 1984). The maximum of seasalt aerosol loading occurs in April or even May. This is rather difficult to explain, because at that time the extent of open waters has passed its maximum and the source is farther away from the station than in March. One explanation might be the sea salt particles being of secondary origin. Secondary in that sense means that sea salt particles have been incorporated into clouds, precipitated and later resuspended from the snow surface as lifted ice crystals with increasing wind velocities, a noticeable but unexplored aerosol particle source in Antarctica (Hogan, 1975; Jaenicke and Matthias-Maser, 1992).

4.2. Size distribution of condensation nuclei

The averaged CN size distribution showed a rather uniform picture. In Fig. 2, the average summer (October to April) and winter (May to September) distributions are presented. These distributions are "monodisperse" with the average radius at $R = 33$ nm (winter) for 55 cm^{-3} concentration and $R = 29$ nm (summer) for 269 cm^{-3}, respectively. The distributions have been obtained by a smoothing procedure recommended by Tichonov and Arsenin (1979). The smoothing is

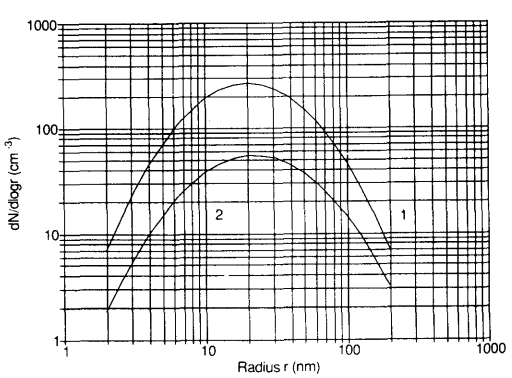

Fig. 2. Average summer (1) and winter (2) aerosol size distribution. The data support the overwhelming occurrence of "monodisperse" distributions.

recommended because the backward calculation of the obtained size distributions to penetration data had a much better accuracy of 1.2% than the deviation of the penetration data during the averaging process.

Both distributions could be written log-normally with the following parameters: summer – $R^* =$ 20 nm, $\log \sigma = 0.37$, $N = 269\ \mathrm{cm}^{-3}$, winter – $R^* =$ 22 nm, $\log \sigma = 0.40$, $N = 55\ \mathrm{cm}^{-3}$ for

$$\frac{\mathrm{d}N}{\mathrm{d}\log r} = N \exp\left(-\frac{\log^2(r/R^*)}{2\log^2 \sigma}\right),$$

with

R^* = maximum (normalizing) radius in same units as radius r,

$\log \sigma$ = a measure of polydispersity,

$\frac{\mathrm{d}N}{\mathrm{d}\log r}$ = size distribution (a density on the ordinate) (cm^{-3}),

N = total concentration (cm^{-3}).

The distributions reflect the monodisperse distributions published by Shaw (1986) for Antarctica and Jaenicke and Schütz (1982) for the Arctic, later summarized in Jaenicke (1988). However those distributions seem to peak around 70 nm. In terms of monodispersity ($\log \sigma$), the distributions of this paper are somewhat broader than suggested earlier.

The evaluation was carried on calculating an average radius as

$$R = \int_{r=0}^{r=\infty} r f(\log r)\, \mathrm{d}\log r.$$

Table 1. *Frequency of observing the average radius of the average size distribution in various size ranges; types and ranges given in the header*

Radius range	Type 2 $15 < R < 25$ nm (%)	Type 3 $25 < R < 35$ nm (%)	Type 4 $35 < R < 45$ nm (%)
winter	17	56	27
summer	39	48	13

The size distributions can be sorted by their maximum radius. Table 1 defines type 2 to 4 in terms of radius range. Type 1 and 5 include distributions outside of that table, namely type 1: $R < 15$ nm and type 5: $R > 45$ nm, respectively. Table 1 indicates in winter, often the size distribution being of type 3 and 4 and peaking in the 25 to 45 nm radius range, while in summer the particles are smaller of type 2 and 3 and peak in the 15 to 35 nm range. From the 167 distribution evaluated so far, only 14 belong to the type 1 and type 5. Fig. 3 shows the size distributions of the three different types. There is only a shift in the maximum, but no obvious change in shape.

Another possibility to classify our distributions is to classify them in groups according to the shape. Three shapes of distributions were identified. Fig. 4 shows two of them. First there are clearly one-modal distributions similar to the average summer or winter distribution. The distributions of the second shape are single-mode modified distributions which seem to have two modes of different peak height close together. The third shape is clearly bi-modal. Both last shapes can be considered as multimodal in contrast to the first shape. Of all distribution shapes, multimodal occur 45% in summer (December–February), 69% in fall (March–May), 76% in winter (June–August), and 52% in spring (September–

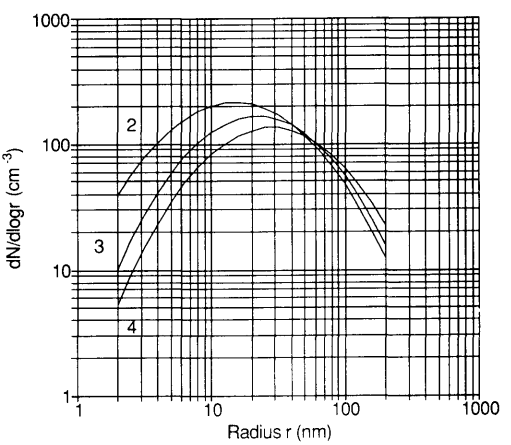

Fig. 3. The size distributions for the various types, defined in the text and Table 1. The types select distributions according to their maximum radius. All these distributions occur in winter as well as in summer.

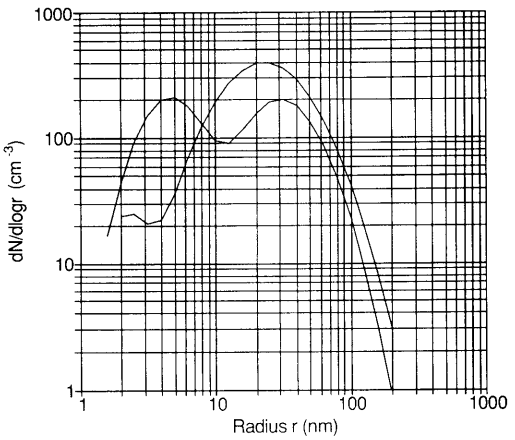

Fig. 4. Two individual size distributions reflecting the different multimodal shapes. Averaging over several multimodal distributions tends to result in a "monodisperse" distribution.

November). It should be emphasized that, while multimodal shapes definitely exist, averaging might transform them into single-mode shapes, because of the varying location of the modes. The percentage observed is rather puzzling and in contrast to expectations (aged "monodisperse" distributions in winter).

Using an observation method (a Pollak counter (like Pollak and O'Connor 1955) and diffusion pipes) rather similar to ours, but a different evaluation procedure (diffusion decay technique) Ito (1982) found exclusively bimodal distributions, even as monthly averages. The mean distribution of all measurements at the Japanese Syowa-Station during 23 August to 31 December 1978 had maxima at 5 nm and 40 nm. While the larger maximum certainly agrees with our observation, our data do not confirm the maximum at 5 nm. The ordinate value ($dN/d \log r$) at the larger maximum agrees very nicely with our observation for summer, and indeed the Japanese measuring period mostly reflects this time span. It remains open, whether both sets of observation would agree, if similar evaluation procedures had been applied. In winter, the size distribution at Georg von Neumayer-station has its maximum ordinate value ($dN/d \log r$) at 30 cm^{-3}.

Considering the particle formation through gas-to-particle conversion (GPC), the situation now is rather unsatisfactorily. It is the present understanding that particles formed by GPC usually are in the range slightly above 1 nm in radius. These particles tend to coagulate rapidly, forming larger ones. That process only continues if a constant production occurs, which should be reflected in the shape of the size distribution. The distributions presented in this paper do not support this idea, whether for summer nor for winter. Ito's (1982) differently shaped distributions are also lacking particles close to 1 nm. It still remains an open question how the particles are formed being observed in the size distributions.

5. Conclusion

The observations of CN concentration during 8 years at the German Antarctic station have shown a remarkable increase of concentration with the rate of 9.63 %/yr. The annual variation clearly is indicated with a minimum in winter and a maximum in summer time. All these observations are overlayed with short sudden increases, propelling the concentrations a factor of ten up for no obvious reason.

The size distributions measured for the CN are often multimodal with particles below 10 nm, but no particles close to 1 nm as one should expect it for GPC. Monodisperse distributions occur with maximum radii in the 15 to 45 nm range. Further studies are needed to explain the production mechanism. As long as this production mechanism is not understood, it is difficult to speculate about the observed increase in total particle concentration.

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REFERENCES

Bashurova, V. S., Dreiling, V., Hodger, T. V., Jaenicke, R., Koutsenogii, K. P., Koutsenogii, P. K., Krämer, M., Makarov, V. I., Obolkin, V. A., Potjomkin, V. L. and Pusep, A. Y. 1992. Measurements of condensation nuclei size distribution in remote continental area, Lake Baikal, Siberia. *J. Aerosol Sci. 23*, in press.

Bigg, E. K., Gras, J. L. and Evans, C. 1984. Origin of Aitken particles in remote regions of the southern hemisphere. *J. Atmosph. Chemistry 1*, 203–214.

Haaf, W. and Jaenicke, R. 1977. Determination of the smallest particle detectable in condensation nucleus counters by observation of the coagulation of SO_2 photo-oxidation products. *J. Aerosol Sci. 8*, 447–456.

Hogan, A. W. 1975. Collection of drifting snow by a track-roughened surface. *Applied Meteorology 14*, 428–429.

Ito, T. 1982. On the size distribution of submicron aerosols in the antarctic atmosphere. *Antarctic Record 76*, 1–19.

Ito, T. and Iwai, K. 1981. On the sudden increase in the concentration of Aitken particles in the antarctic atmosphere. *J. Meteorol. Society Japan 59*, 262–271.

Jaenicke, R. 1972. The optical particle counter: cross-sensitivity and coincidence. *J. Aerosol Sci. 3*, 95–111.

Jaenicke, R. 1980. The Scholz pocket-counter and its modification—a direct condensation nuclei counter. *J. Aerosol Sci. 11*, 263–265.

Jaenicke, R. 1988. Aerosol physics and chemistry. In: *Landolt-Börnstein Numerical data and functional relationships in science and technology. New Series V Geophysics and Space Research, 4 Meteorology, (b): Physical and chemical properties of the air* (ed. G. Fischer). Springer, 391–457.

Jaenicke, R. and Matthias-Maser, S. 1992. Natural sources of atmospheric aerosol particles. In: *Fifth Int. Conf. on Precip. Scav. and Atmos., Surface Exchange Processes*, in press.

Jaenicke, R. and Schütz, L. 1982. Arctic aerosols in surface air. *Időjárás 86*, 235–241.

Koutsenoguii, P. 1992. Reconstruction of aerosol size distribution battery TSI 3040. *J. Aerosol Science 23*, in press.

Leiterer, U. and Sakunow, G. 1989. Messungen von Aerosolpartikeln im Größenbereich 0,2 bis 4,0 μm in der Antarktis. *Z. Meteorologie 39*, 309–316.

Liu, B. Y. H., Pui, D. Y. D., McKenzie, R. L., Agarwal, J. K., Jaenicke, R., Pohl, F. G., Preining, O., Reischl, G., Szymanski, W. and Wagner, P. E. 1980. Recent experiments of the working group on ultrafine aerosols, The WUFA Workshop. *J. Aerosol Sci. 11*, 261–263.

Münnich, K. O. and Jaenicke, R. 1989. Luftchemische Untersuchungen an der Georg-von-Neumayer-Station. *Promet Meteorologische Fortbildung 1/2 '89*, 48–56.

Pollak, L. W. and O'Connor, T. C. 1955. A Photo-electric condensation nucleus counter of high precision. *Geofisica Pura e Applicata 32*, 139–146.

Samson, J. A., Barbard, S. C., Obremski, J. S., Riley, D. C., Black, J. J. and Hogan, A. W. 1990. On the systematic variation in surface aerosol concentration at the south pole. *Atmosph. Res. 25*, 385–396.

Shaw, G. E. 1986. On physical properties of aerosol at Ross Island, Antarctica. *Aerosol Sci. 17*, 937–945.

Tichonov, A. N. and Arsenin, V. Ya. 1979. *Methods of solution of incorrect problems.* Nauka, Moscow (in Russian).

Weber, S. and Jaenicke, R. 1988. The vertical profile of condensation nuclei on cloudy days. In: *Atmospheric aerosols and nucleation* (eds. P. E. Wagner, G. Vali), Lecture Notes in Physics 309, Springer Berlin, 56–60.