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Stratospheric aerosol measurements in the Arctic winter of 1996/1997 with the M-55 Geophysika high-altitude research aircraft

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ABSTRACT

In-situ aerosol measurements were performed in the northern hemispheric stratosphere up to altitudes of 21 km between 13 November 1996 and 14 January 1997, inside and outside of the polar vortex during the Airborne Polar Experiment (APE) field campaign. These are measurements of particle size distributions with a laser optical particle counter of the FSSP-300 type operated during 9 flights on the Russian M-55 high-altitude research aircraft Geophysika. For specific flights, the FSSP-300 measurements are compared with balloon-borne data (launched from Kiruna, Sweden). It was found that the stratospheric aerosol content reached levels well below the background concentrations measured by the NASA operated ER-2 in 1988/89 in the northern hemisphere. During the APE campaign, no PSC particle formation was observed at flight altitudes although the temperatures were below the NAT condensation point during one flight. The measured correlations between ozone and aerosol give an indication of the subsidence inside the 1996/97 polar vortex. Despite the lower aerosol content in the winter 1996/97 compared to the 1989 background, the heterogeneous reactivity of the aerosol (as calculated from the measured data with additional model input) is comparable. This is due to the dependency of the reactive uptake coefficients on the atmospheric water vapor content. Under the described assumptions the reaction rates on the background aerosol are significantly smaller than for competing gas phase chlorine activation, as can be expected for stratospheric background conditions especially inside the polar vortex.

1. Introduction

High-altitude in-situ aircraft-borne aerosol measurements of particles with sizes larger than 0.1 μm were conducted in the recent years inside and near the northern hemispheric polar vortex on the NASA operated ER-2 (Jonsson et al., 1996; Wilson et al., 1993; Brock et al., 1993; Dye et al., 1992; Borrmann, 1996). These cover periods of relative volcanic quiescence, as well as the time from shortly after the Mount Pinatubo eruption (in June 1991) until 3 years later. The ER-2 measurements showed the dependency of the observed total aerosol volume on temperature and gave experimental evidence of PSC type-I clouds consisting of liquid solution droplets (Dye et al.,

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1992). The temporal evolution of the stratospheric aerosol and its relation to microphysics after the Mount Pinatubo eruption was described (Wilson et al., 1993; Jonsson et al., 1996; Borrmann et al., 1993) in detail. Fahey et al. (1993) provided evidence of the heterogeneous reaction of N_2O_5 with water proceeding on the binary liquid solution droplets of the volcanic aerosol and pointed out that possible saturation effects of this reaction can occur at low levels of atmospheric aerosol surface area. The availability of the Russian M-55 Geophysika high-altitude aircraft (Stefanutti et al., 1999a; Borrmann et al., 1995a) for stratospheric research provided the opportunity to conduct aerosol measurements in the northern hemispheric winter 1996/97 inside and outside of the polar vortex in a very clean background atmosphere 5.5 years after the Mount Pinatubo eruption. The aircraft characteristics, its instrumentation and performance are described in detail by Stefanutti et al. (1999a; 1999b), together with a summary of the encountered meteorological conditions and with the geographical locations covered by the research flights during the APE campaign. In this paper examples of the measured aerosol properties are shown and these 1996/97 measurements are related to the pre- and post-Pinatubo data sets obtained from the ER-2 during the Airborne Arctic Stratospheric Expeditions and the Stratospheric Photochemistry, Aerosol and Dynamics Experiment (AASE in 1988/89, AASE II in 1991/92, and SPADE in 1992/93). Instrument intercomparisons between the Geophysika instrument and the balloon-borne particle counter of the University of Wyoming are also described. Borrmann et al. (1995b) have shown the use of stratospheric aerosol properties as dynamical tracers after the Pinatubo eruption in the mid-latitude outside vortex air by considering correlations with ozone, where the ozone is used as reference tracer. In this paper the measured ozone versus aerosol correlations could demonstrate the subsidence inside the vortex in the time between mid-December 1996 and mid-January 1997. The dependency of the measured aerosol volume on ambient temperature is shown for temperatures close to the NAT condensation point under conditions of very low atmospheric aerosol loading, and calculated estimations of the heterogeneous reactivity of this aerosol are provided.

2. Methodology

The aerosol particle size distributions were measured on Geophysika (as on the ER-2 during AASE, AASE II, and SPADE) with a FSSP-300 (Forward Scattering Spectrometer Probe) covering a particle diameter size range from 0.41 μm to 23 μm in 31 size bins (Baumgardner et al., 1992). The instrument implementation on Geophysika is similar to that on the ER-2, although special arrangements needed to be made to accomodate for the heating requirements due to the instrument's unprotected exposure in the free flow under the wing of Geophysika. Fig. 1 shows the airplane shortly before takeoff in Rovaniemi, Finland, where the cylindrical instrument container can be seen under the wing. The instrument's response was monitored during the campaign through a series of size calibrations before and after each flight. Experiments on the stability of the scattered light signals as function of decreasing temperatures have been performed in a cold chamber facility utilizing a pulsed laser and a fiber optic system to excite the FSSP's detection optics and electronics. This laser fiber optic setup was adopted because it was not possible to generate and inject Polystyrene Latex calibration beads into the cold chamber. Down to the lowest temperatures near $-40^{\circ}C$ that could be obtained inside the cold chamber the FSSP's signal processing system exhibited remarkable stability with variations of the pulse heights generated by the individual laser flashes of less than 10%. During the Geophysika flights such low temperatures of the optical system were not reached due to the modified heating control system. Since no PSC particles were encountered during the APE campaign the conversions of detected scattered light signals to particle sizes were performed for all measurements by means of Mie theory. For these calculations an optical refractive index of 1.44 was assumed for the stratospheric sulfuric acid aerosol droplets. Inside thin cirrus clouds consisting of presumably aspherical particles the Mie theory calculations were replaced by T-Matrix results for the refractive index of 1.31 according to Borrmann et al. (2000). Due to the very low atmospheric particle concentrations data sampling periods larger than 100 s (compared to 10 s periods on the ER-2 during the Pinatubo period) were necessary for the particle size distribution measurements in order to

Fig. 1. Photograph of the Russian high-altitude research aircraft Geophysika on the taxiway in Rovaniemi, Finland. The sonde of the FSSP-300 system is contained in the cylinder underneath the wing, while the data acquisition computer is located inside the left tail boom behind the position of the retracted wheel. The probe containing cylinder is covered by a sheet of aircraft metal to provide space for additional heating circuitry and insulation.

accomodate the requirements imposed by counting statistics.

The ozone mixing ratio was measured by an electrochemical cell sensor (ECOC) from the Central Aerological Observatory (CAO) in Moscow, and water vapor by the FLASH (FLuorescent Airborne Stratospheric Hygrometer) instrument, also from CAO (see Stefanutti et al., 1999a, and references therein).

3. Stratospheric aerosol loading in the 1996/97 winter

Since a FSSP-300 type aerosol measurement system was installed on the ER-2 during the Pinatubo period (Baumgardner et al., 1992) and on Geophysika during the testflights and the APE campaign, direct comparisons of the measurements are readily available. Fig. 2 shows the measured particle number density for size diameters between 0.4 and 23 μm as function of flight time for flights conducted at various northern hemispheric latitudes. The time scale on the abscissa has been normalized with respect to the duration of the flight between takeoff and landing (usually between 4 and 8 h). This implies that each flight has its individual scaling of the abscissa slightly differing from the others. On the left and right sides of the figure the concentration increases because these are the measurements from the takeoff and landing phases of the flights. The center region of the graph represents values measured by both aircraft at altitudes between 18 and 21 km pressure altitude. The uppermost line indicates the aerosol levels encountered by the ER-2 during flights between January and March 1992 (Borrmann et al., 1993). The solid single thick line in the middle of the figure shows the levels measured by the ER-2 during a flight from Wallops Island, USA, to Stavanger, Norway, on 31 December 1988, mostly outside of the polar vortex. This flight took place in a period of relative volcanic quiescence and represents what were thought to be background conditions at that time. The group of 3 dotted lines underneath the 31 December 1988, ER-2 flight are the measurements by Geophysika during testflights from Pratica di Mare (near Rome, Italy) in November and early December 1996. These represent outside vortex, mid-latitude data from similar latitudes as the 31 December 1988, ER-2 flight. The lowermost 3 dashed lines are the Geophysika flights from January 1997 from Rovaniemi, Finland, mostly inside the polar vortex. Although these data are not binned with respect to latitude, and inside

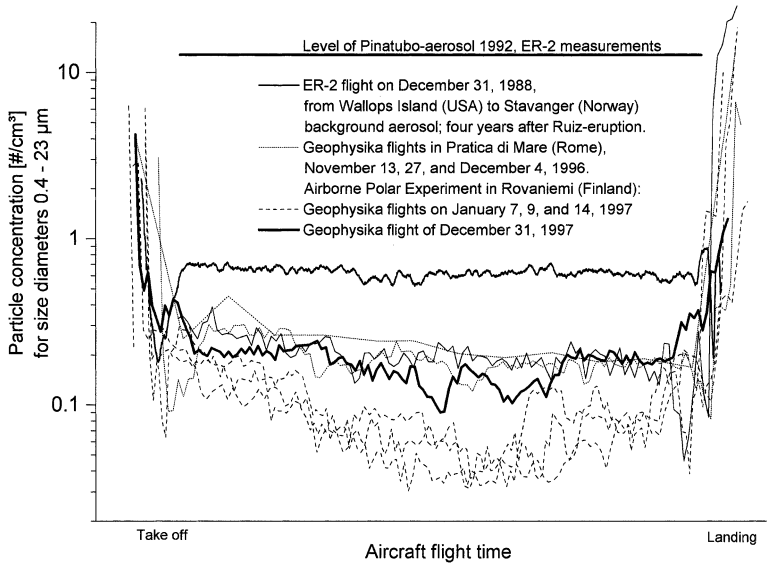

Fig. 2. Particle number densities from FSSP-300 measurements on the ER-2 and Geophysika from northern hemispheric locations for the pre-Pinatubo background period, the time after the Pinatubo eruption, and the 1996/97 winter (see text for details).

vortex air parcels are represented, as well as air from outside, this figure graphically demonstrates the atmospheric changes of the aerosol number densities from before until long after the Mount Pinatubo eruption. Also, the decrease of the atmospheric particle concentrations in the winter 1996/97 beyond the previous background case of 31 December 1988, is highlighted this way. These data are put into the context of the periods before, during, and after the Mount Pinatubo eruption in Fig. 3 for the 450 K potential temperature level in the northern hemisphere. The data are 10 s flight

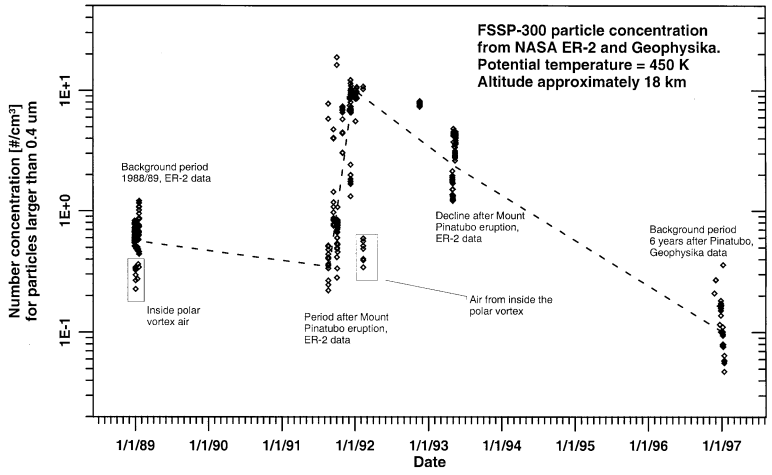

Fig. 3. The particle number density measured in-situ by FSSP-300 instruments on the NASA operated ER-2 and on the Russian M-55 Geophysika for the periods before, during, and up to 6 years after the Mount Pinatubo eruption on the 450 K potential temperature level in the northern hemisphere. The data points represent 10 s flight time sampling periods.

time sampling periods and the scatter for the individual flights results from counting statistics as well as differences in the geographic locations covered during the flights. Air from inside the polar vortex has been identified for the ER-2 data sets by the concurrently measured values of ozone and N_2O. The background periods of January 1989 and January 1997 are clearly evident, as is the enhancement by a factor of approximately 20 during the time directly after the eruption in June 1991.

In Fig. 4 examples of particle size distributions from the 1996/97 winter are shown from measurements inside (middle and lower curve) and outside (upper curve) of the polar vortex on similar levels of potential temperature. It is interesting to note that the middle curve was obtained closer to the vortex boundary and the lower curve towards the vortex center. Apparently the concentrations decreased in the middle of the 1996/97 polar vortex as was the case for the volcanic conditions after the Mount Pinatubo eruption in the 1991/92 winter (Borrmann et al., 1993). A second set of vertical profile data of the measured densities of aerosol particle number, surface areas and volumes is shown together with ambient temperature and ozone in Fig. 5, where Geophysika executed a "dive" on 29 December 1997, over Novaja Zemlya, which means the plane descended from cruising altitude (19 km) down to 9 km subsequently re-ascending up to 21 km altitude. At the bottom of this dive the vicinity of the tropopause was reached as indicated by the temperature and ozone data, which were found below 300 ppbv for altitudes lower than 11 km. In this altitude band near the bottom of the dive an increase in measured particulate surface area and volume is visible in Fig. 5 indicating the presence of larger particles while the particle number density remains fairly constant between 9 and 15 km.

It has to be emphasized that the use of a FSSP-300 system in general, and especially under the given low background aerosol levels in the stratosphere is insufficient, because much of the aerosol surface area is supplied by particles with sizes below this instrument's lower detection threshold. Therefore the implementation of other aerosol instrumentation on Geophysika extending towards smaller sizes as, e.g., in Jonsson et al. (1996) is needed, as well as a condensation nuclei

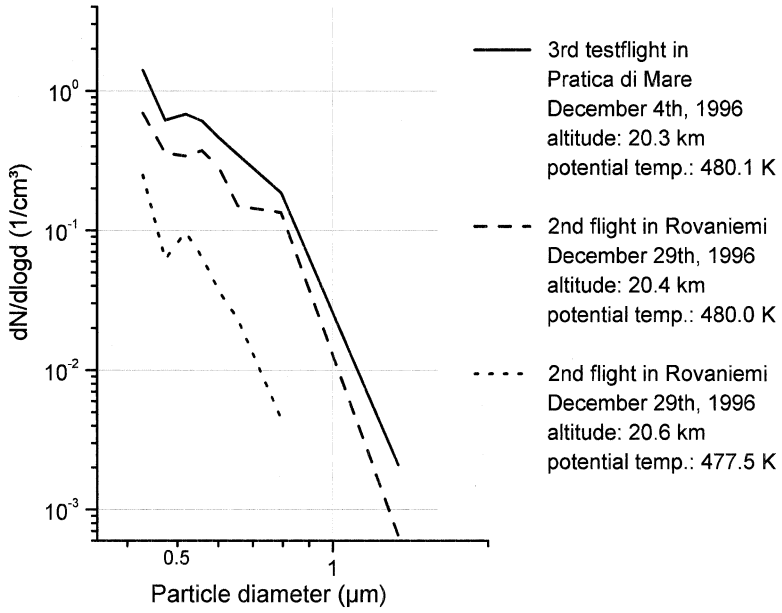

Fig. 4. Size distribution measurements of the FSSP-300 system on Geophysika on similar potential temperature surfaces outside of the vortex (4 December 1996, flight) and inside the vortex (29 December 1996). The averaging periods for these size distributions are between 300 and 1000 s of flight time depending on the counting statistics.

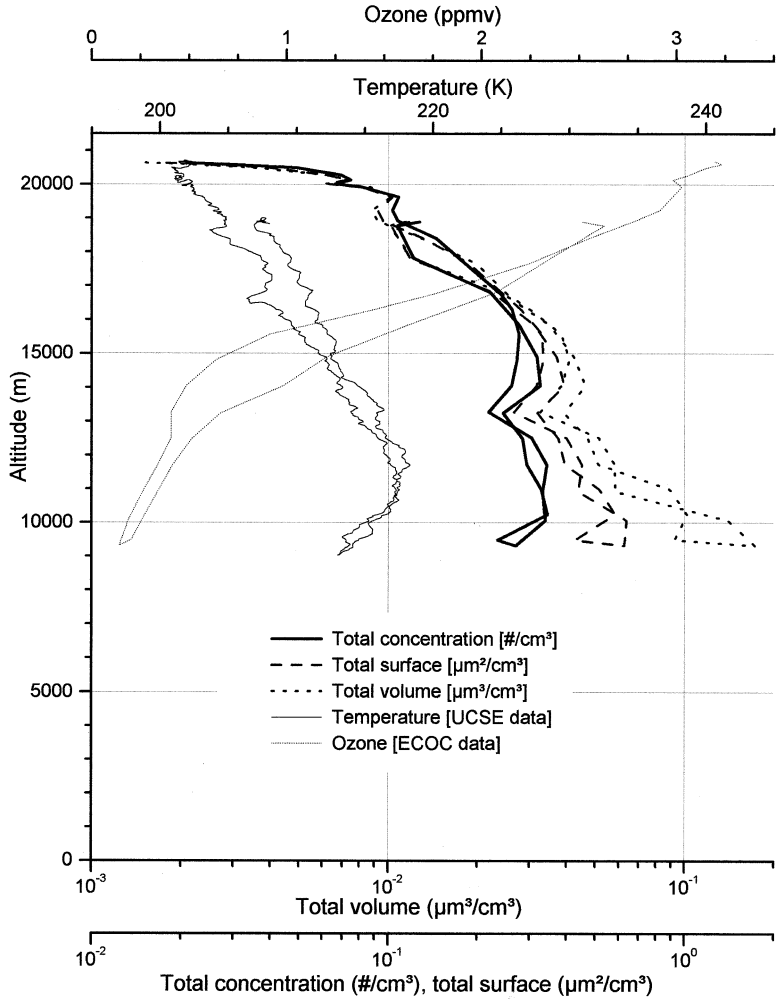

Fig. 5. Vertical profile of the measured aerosol number, surface area, and volume densities (3 curves at right side) from the descent and re-ascent of the mid-flight "dive" of Geophysika from 29 December 1996, over Novaja Zemlya. Also indicated are the ambient temperature (middle curve) and the ozone mixing ratio measured by ECOC (left curve).

counter for the smallest particles (Wilson et al., 1983; Brock et al., 1995). Thus, in order to cover the stratospheric particle sizes from freshly nucleated particles to polar stratospheric particles of type II at least 3 different instruments are required as implemented for example on the American ER-2.

4. Intercomparison with balloon-borne data

In Figs. 6 and 8, the measured particle number density from 2 vertical profiles of the Geophysika ascent and descent on 14 January 1997 (Rovaniemi, Finland), are intercompared with a balloon-borne particle instrument (Deshler and Oltmans, 1998), which was launched on 19 January 1997 (T. Deshler, private communication, 1999) from Kiruna, Sweden (i.e., approximately 275 km from Rovaniemi). The particle concentrations indicated by the lines in Fig. 6 for particles with diameters larger than 0.3 and 0.5 μm for altitudes between 13 and 17 km are broadly comparable with the total concentrations measured by the FSSP-300 (i.e., number densities for

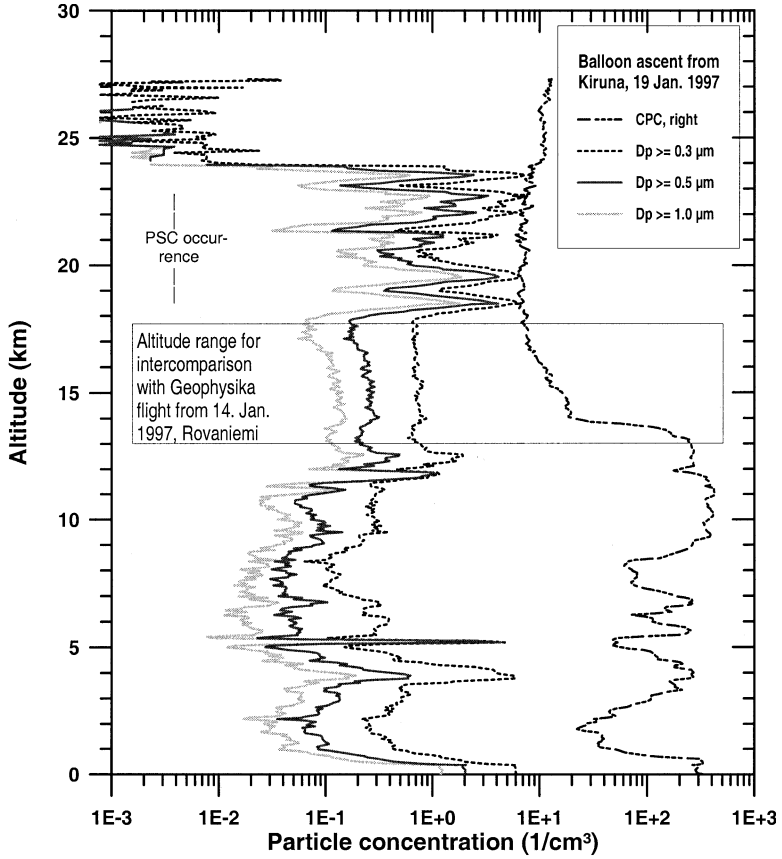

Fig. 6. Vertical profile of aerosol particle and condensation nuclei measurements (CN), and of temperature from the ballon launch of 19 January 1997, from Kiruna, Sweden. The aerosol particles with size diameters $\geqslant$0.3–1.0 µm are binned into 3 classes. (The data also measured for larger particle sizes are not shown for clarity.) A cirrus cloud is present between 11 and 12 km, and a polar stratospheric cloud between 18 and 24 km. The tropopause is at 12 km.

particles with diameters larger than 0.41 µm) in Fig. 8. Only this altitude band can be intercompared because at higher altitudes PSCs were encountered during the balloon flight and at lower altitudes differences in tropospheric clouds cover prevent direct comparison. Based on ECMWF analyses the measurements of both data sets took place in the vortex boundary region inside a potential vorticity (PV) band with values between 36 and 42 PV-units. Due to the differences in geographical location (Kiruna in Sweden at 67.82°N, 20.32°E for Fig. 6 and Rovaniemi, Finland, at 66.57°N and 25.83°E for Fig. 8) as well as the different times and physical conditions of the measurements, and considering the differences between ascent and descent of Geophysika inherent in one single day (Fig. 8), the measured concentrations cannot be expected to coincide. Also errors due to counting statistics and the uncertainty in the optical volume of the FSSP-300 (Baumgardner et al., 1992) play a role. Thus this intercomparison is only of limited value, however, the results are close enough to establish some confidence in the Geophysika measurements.

Note also the polar stratospheric cloud layers encountered at altitudes above 17 km as indicated by the measurements in Fig. 6 and the profiles in Fig. 7 with concentrations near 10 particles per cm^3 of air. In some of these layers the number of the measured condensation nuclei (CN), i.e., particles with size diameters above 10 nm, is identical to the number of measured cloud particles per cm^3 of air.

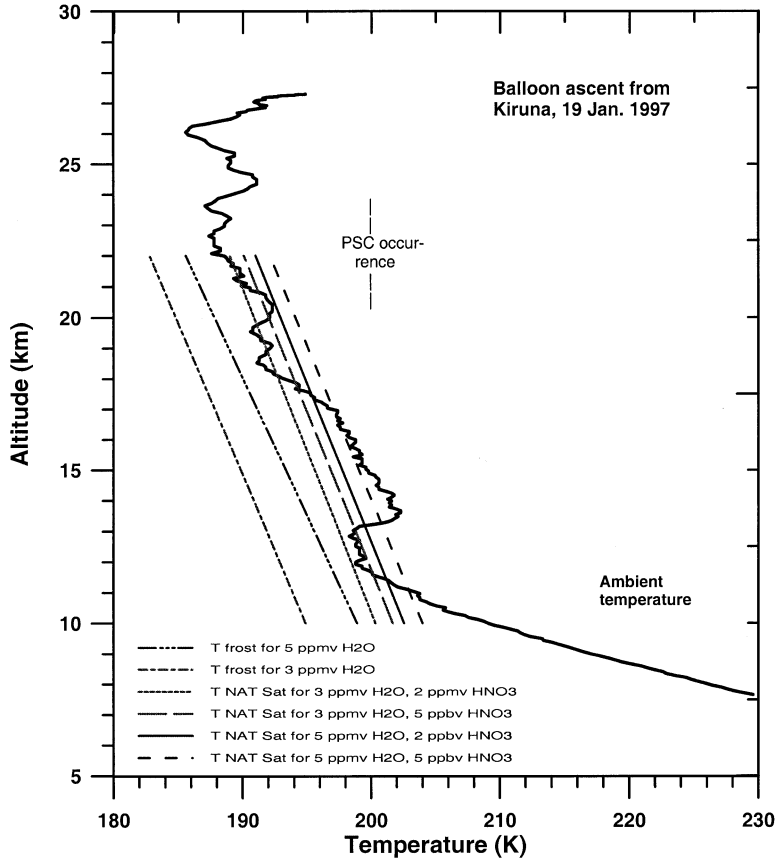

Fig. 7. Vertical profile of the measured temperature and calculated saturation temperatures from the balloon launch of 19 January 1997, from Kiruna, Sweden. "T frost" denotes the temperature of saturation over ice for the indicated water vapor mixing ratios. "T NAT Sat" designates the saturation temperature over Nitric Acid Trihydrate (NAT) under the given assumptions for the gas phase abundances of water vapor and nitric acid.

This indicates near complete activation of the available background aerosol particles. The NAT formation threshold in these layers was only reached assuming water vapor abundances near 5 ppmv.

5. The dependency of aerosol volume on temperature

For the coldest temperature sections encountered by Geophysika during the APE campaign the measured aerosol volume is plotted versus temperature in Figs. 9, 10 in a way similar to the Dye et al. (1992) study. Here the data are binned with respect to ozone because there were no other in-situ tracer measurements available on Geophysika. Fig. 9 shows a section of the flight from 7 January 1997, where the data are from a narrow ozone bin indicating the sampling of one single air mass. The scatter in the data points is largely due to counting statistics and its propagation into the aerosol volumes (which are precise to ± 30–50% depending on the shape of the size distribution and the integration time period). Although temperatures lower than the NAT formation temperature was reached (assuming the — not measured — ambient gas phase abundances indicated in the figure) no significant increase in particle volume occurred as for the 24 January 1989 case in Dye et al. (1992). Also the absolute aerosol volumes in Fig. 9 are by up to 2 orders of magnitude lower than those

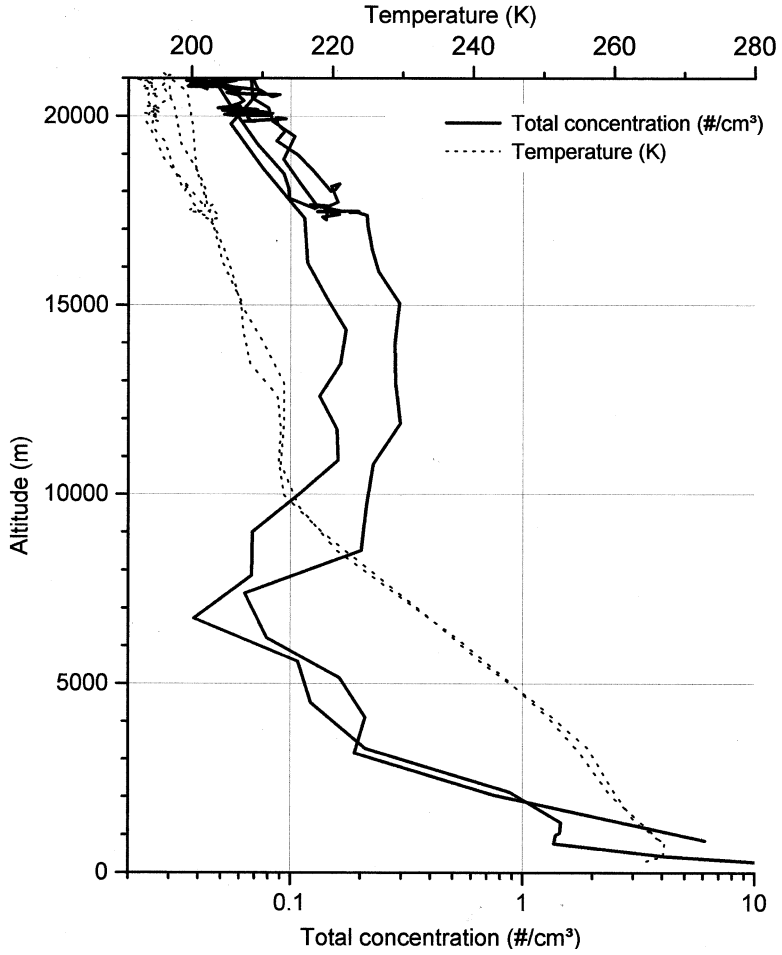

Fig. 8. Vertical profile of the FSSP-300 aerosol number densities from the ascent (right solid line) and descent (left solid line) of Geophysika from 14 January 1997, in Rovaniemi, Finland, together with the measured temperature profiles (dashed lines).

observed for similar temperatures in Dye et al. (1992), which indicates again the clean state of the arctic, inner polar vortex stratospheric air. The same applies to the flight of 14 January 1997, in Fig. 10. Here the ozone values varied considerably showing that a selection of different air masses was sampled. The observed low concentrations of background aerosol near the PSC formation temperature threshold raise the question inasmuch the PSC formation process itself is influenced by the absence of higher amounts of nuclei. If the temperature drops below the NAT formation threshold (or the threshold where growing ternary solution droplets start to form by significant uptake of HNO_3 into the present H_2SO_4–H_2O solution droplets) the number of available nuclei also plays a role for PSC formation because the speed at which particles increase their size depends on the rate of temperature change as well as the number of particles competing for condensable vapors (Meilinger et al., 1995).

6. The ozone versus aerosol correlation inside the 1996/97 polar vortex

Graphs showing correlations of in-situ measured stratospheric aerosol properties with ozone

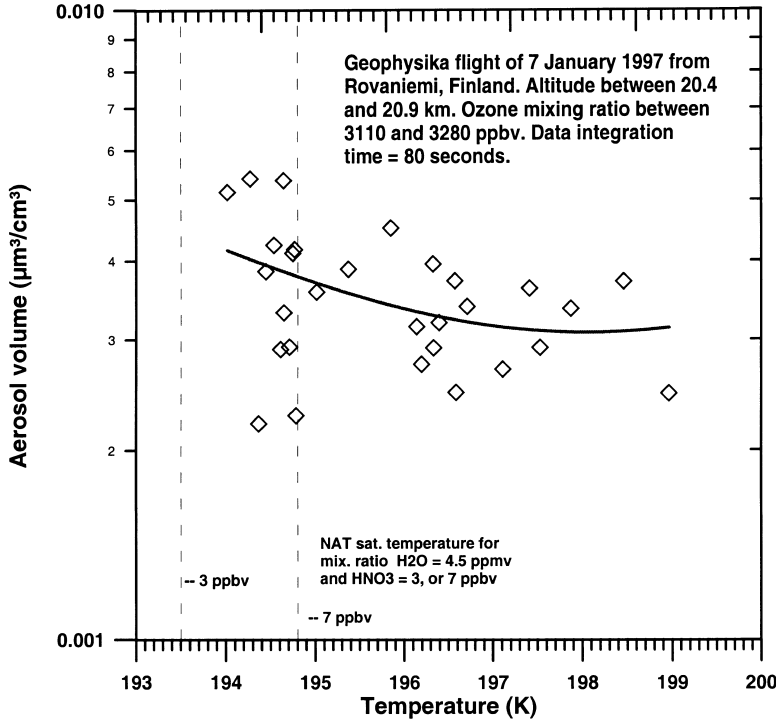

Fig. 9. The measured particulate volume as function of ambient temperature for parts of the flight of 7 January 1997. The dashed vertical lines show the temperatures at which saturation with respect to NAT occurs under the indicated assumed mixing rations for water vapor and nitric acid.

have been used to document the aerosol's suitability as dynamical tracer under certain circumstances by Borrmann et al. (1995b) for mid-latitude locations. Since N_2O, or other species suitable as dynamical tracers were not measured during APE on Geophysika the available ozone measurements need be adopted as a reference tracer instead. This is possible on timescales of the weeks between November 1996 and January 1997, because chemical ozone loss occurred later in 1997 inside the northern hemispheric polar vortex. Manney et al. (1997) showed by means of MLS data that on the 465 K level (i.e., near typical Geophysika flight altitudes) ozone did not decrease between 1 December 1996 and 1 March 1997. Figs. 11, 12 display the correlations between the mixing ratios of measured ozone and aerosol number density for 6 flights of Geophysika from Rovaniemi, Finland, in December 1996 and January 1997. Although graphs for aerosol surface area instead of number density show similar behavior, the surface area property of the aerosol is not a useful quantity in this context for the low temperatures encountered during the polar region flights. The particles grow or shrink by water vapor uptake in dependency of ambient temperature which results in changes of the measured surface area and prevents its use as "conserved quantity". The aerosol number density is not subject to such temperature induced changes for the considered particle sizes. Also processes with influence on number density, like coagulation, do not play a role for the considered short timescale and for particles larger than 0.4 μm at the low number concentrations encountered during the 1996/97 winter.

With altitude the ozone increases monotonically above the tropopause, while the aerosol also increases but only towards the (latitude dependent) altitude of the Junge layer maximum. Thus below the Junge layer maximum increasing ozone is associated with increasing aerosol number mixing ratios. Above the Junge layer maximum there is an aerosol decline with ozone still increas-

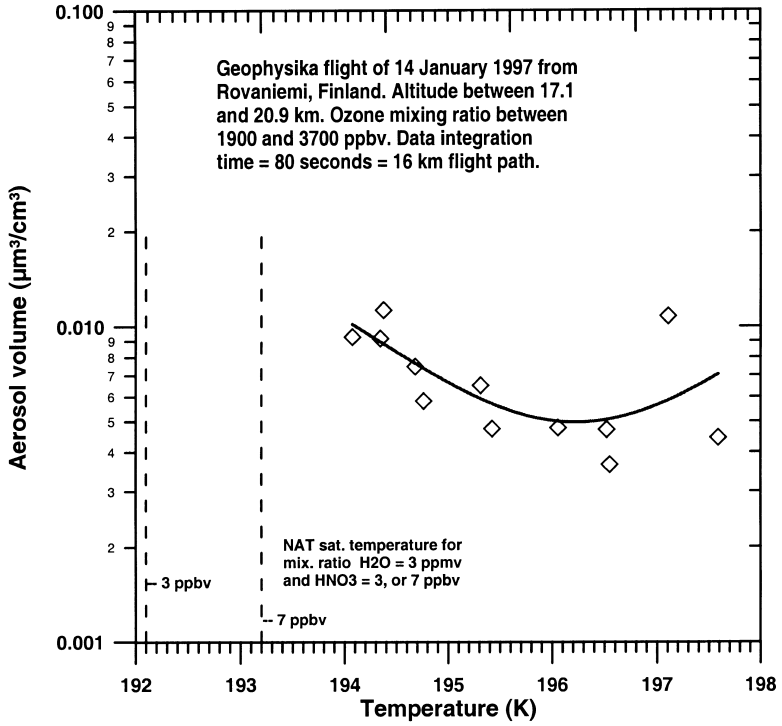

Fig. 10. The measured particulate volume in dependency of temperature for parts of the Geophysika flight of 14 January 1997, as in Fig. 9.

ing, which explains the shape of the observed correlation curve between ozone and aerosol. The shapes of the correlograms in Figs. 11, 12 closely resemble those of the correlation graphs between N_2O and aerosol reported by Borrmann et al. (1993) for the ER-2 flight of 10 February 1989 under background conditions. Despite the considerable scatter in the data of Figs. 11, 12 it is evident that for most of the given values of ozone the corresponding particle number densities are lower in the January 1997 flights than in the December flights, i.e., the correlation curve between the 2 variables has shifted towards lower values on the abscissa from December to January. We tentatively interpret this as a consequence of the subsidence proceeding with time inside the polar vortex. Apparently during December 1996 and January 1997 different air masses were sampled. If there was no mixing of aerosol from mid-latitudes into the vortex, then lower aerosol number density mixing ratios associated with the same or higher values for ozone most likely represent air masses differing in altitude.

7. Comparison of the heterogeneous reactivity of the 1988/89 and 1996/97 background aerosol

Based on assumptions concerning the abundances of reactive gas phase species the measured aerosol surface areas and the corresponding in-situ data of other variables (pressure, temperature, etc.) can be used to calculate the heterogeneous reaction rates for the reactions of $ClONO_2$ with water and HCl, of HOCl with HCl, and of N_2O_5 with H_2O (Hanson et al., 1994) as in Borrmann et al. (1997). This delivers estimates of the aerosol's potential for heterogeneous chlorine activation. Since the gas phase reactand species were not in-situ measured by the ER-2 or Geophysika, assumed values as given from a 2-D model (here:

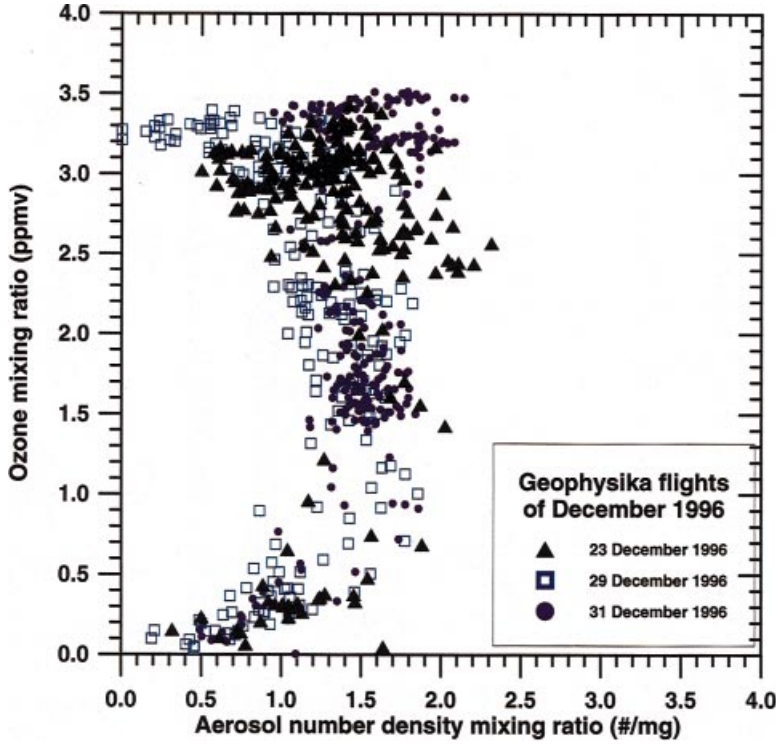

Fig. 11. Correlation between measured ozone and aerosol surface area mixing ratios for altitudes above 7 km inside the polar vortex for the ascents and descents of the Geophysika flights of December 1996 from Rovaniemi, Finland. The averaging periods for each data point are between 60 and 120 s.

Garcia and Solomon, 1994) and listed in Table 1 have been utilized for these rate calculations. Fig. 13 shows the resulting heterogeneous reaction rates as function of temperature for the Geophysika flight of 7 January 1997 (open and solid symbols) under the same assumptions (from Table 1) as adopted for the ER-2 flight of 31 December 1988 (solid and dashed, numbered lines) in Borrmann et al. (1997). For example the lowest, solid straight line labelled with "(5)" refers to the ER-2 measurements of the HOCl plus HCl reaction from 31 December 1988, and is to be compared with the open diamonds of the Geophysika data from 7 January 1997, of the same reaction. The dashed straight line labelled "(4)" indicates the results for the $ClONO_2$ plus HCl reaction of 31 December 1988, to be compared with the small open triangles of 7 January 1997. The single solid curved line with number "(6)" below the asterisks indicates the rates of the homogeneous gas phase reaction for the Geophysika flight (which competes with the heterogeneous chlorine activating reactions) of OH with HCl forming H_2O and Cl. These rates are compared with the corresponding values of the ER-2 data, i.e., the solid line with the mark "(2)". The details of these rate calculations and inherent limitations and caveats, as well as remarks concerning the non chlorine related heterogeneous N_2O_5 reaction, are described in Borrmann et al. (1997).

Although the aerosol surface areas in 1997 are by factors of 2 to 4 lower than in the 1998 flight, the reaction rates are significantly higher. This is due to the fact that the water vapor mixing ratio used for the Geophysika flight is 5 ppmv (in accordance with the water vapor measurements from a few days later (T. Deshler, private communication, 1998), while the measured values for the ER-2 flight on 31 December 1988, are between 3.5 and 4 ppmv. The difference in available aerosol surface area plays a smaller role for the heterogen-

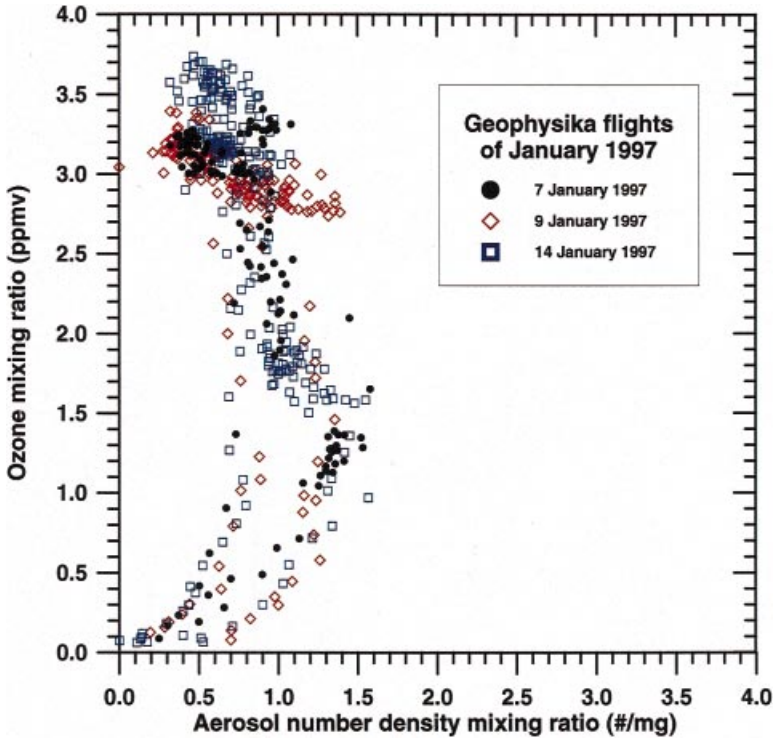

Fig. 12. Correlation between measured ozone and aerosol surface area mixing ratios as in Fig. 11 for January 1997.

Table 1. *Input data for the calculation of the heterogeneous reaction rates from Fig. 13; the assumed values for the gas phase abundances are from the 2-D model by Garcia and Solomon (1994) except for water vapor*

Variable	Dimension	Assumed value
OH number density	#/cm^3	$8 \cdot 10^5$
HCl mixing ratio	ppbv	0.3
H_2O mixing ratio	ppmv	5.0 on Geophysika
H_2O mixing ratio	ppmv	measured on the ER-2
$ClONO_2$ mixing ratio	pptv	30
HOCl mixing ratio	pptv	1.0
N_2O_5 mixing ratio	pptv	90

eous reactivity of the aerosol than the available water vapor because of the strong dependence of the reactive uptake coefficients γ (Hanson et al., 1994) characterizing the heterogeneous reactions on the atmospheric water vapor content. This point emphasizes the need for very accurate and precise water vapor measurements where heterogeneous reactions are of concern. Under the given measured conditions and the assumed gas phase abundances of Table 1 the reaction rates on the background aerosol are by far smaller compared with the competing gas phase reaction of OH with HCl leading to chlorine activation.

8. Conclusions

The Russian M-55 Geophysika has demonstrated to be a suitable aircraft for atmospheric research during the testflights and the mission flights of the Airborne Polar Experiment (APE). The current instrumentation payload, however, needs to be extended to include dynamical tracers, other aerosol instruments, instruments for gas phase species, radicals as ClO and BrO, radiation, and other variables. The in-situ measurements of the stratospheric aerosol by means of a FSSP-300 system shown in this paper demonstrate the low

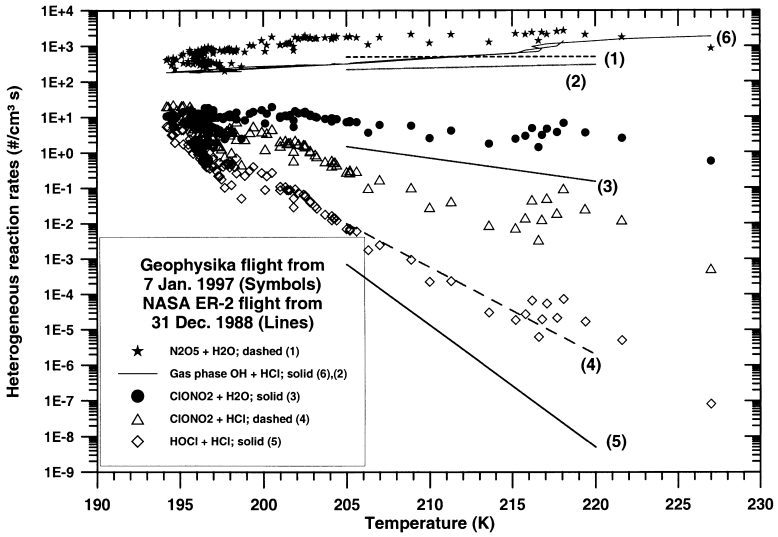

Fig. 13. Comparison between the heterogeneous reaction rates (as function of temperature) from the ER-2 flight of 31 December 1988 (solid and dashed straight lines) and the Geophysika flight of 7 January 1997 (open and solid symbols) for different background aerosol conditions for altitudes above 12 km. The solid and dashed lines with the numbers in parentheses refer to the heterogeneous reaction with the number indicated in the figure. The unnumbered solid curve below the asterisks refers to the homogeneous gas phase reaction of OH with HCl. (See text for details.)

background aerosol levels encountered by Geophysika in the northern hemispheric stratosphere during the winter 1996/97 season in comparison with pre- and post-Pinatubo data from the ER-2. The aerosol number, surface and volume densities in the lower stratosphere encountered by Geophysika were up to 4 times lower compared with the ER-2 measurements from December 1988. Intercomparison between the Geophysika aerosol measurements from Rovaniemi, Finland, with the data of a balloon ascent from Kiruna, Sweden, show reasonable agreement under the given experimental circumstances. Polar stratospheric clouds were not encountered on the Geophysika flight altitudes during APE. Consideration of the correlation between aerosol surface area and ozone mixing ratios could give an indication for the inner vortex subsidence. Despite the lower aerosol surface areas compared with the 31 December 1988 ER-2 flight, the estimated reaction rates are significantly higher in the 7 January 1997, background aerosol encountered by Geophysika. This can be attributed to the different water vapor mixing ratios and the nonlinear dependence of the heterogeneous reactive uptake coefficients on water vapor and temperature. As typical for stratospheric background conditions, the heterogeneous reaction rates on the background aerosol are significantly below those of the gas phase reaction leading to chlorine activation.

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