Rocket-borne in situ measurements of meteor smoke: Charging properties and implications for seasonal variation

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Received 24 June 2009; revised 9 November 2009; accepted 11 December 2009; published 7 May 2010.

[1] Rocket-borne observations of meteoric smoke particles (MSPs) are presented from three campaigns at polar latitudes (69°N) in September 2006, and in the summers of 2007 and 2008. MSPs are detected using a novel technique based on photoelectron emission from the particles after stimulation by UV photons emitted by a xenon flashlamp. Resulting photoelectron currents are shown to be proportional to particle volume density. September results match model predictions qualitatively at altitudes from 65 to 85 km while measurements at higher altitudes are contaminated by photoelectrons from NO and O2(1Δg). Contamination below this altitude can be excluded based on concurrent satellite observations. The observations show a large variability from flight to flight. Part of this variability can be attributed to differences in the charging of MSPs during day and night. Finally we find that MSP volume density in summer can exceed that during September. Analyzing model simulations of the global transport and microphysics of these particles, we show that our observations are in agreement with the model predictions, even though number densities of particles with radii >1 nm, which have long been thought to be suitable condensation nuclei for mesospheric ice particles, show the opposite behavior. It is shown that this discrepancy is caused by the fact that even larger particles (∼3 nm) dominate the volume density and that transport affects these different particle sizes in different ways. These results reinforce previous model findings according to which seasonal MSP variability is mainly driven by the global circulation and corresponding transport.


1. Introduction

[2] Meteor smoke particles (MSPs) are thought to be the product of meteoroid ablation at altitudes between ∼70 and 110 km and subsequent recondensation into tiny particles in the subnanometer to nanometer size range [Rosinski and Snow, 1961; Hunten et al., 1980]. In recent years these particles have received considerable scientific interest because it was realized that they should be involved in a large number of important atmospheric processes. Among these are the nucleation of mesospheric ice particles [e.g., Rapp and Thomas, 2006], mesospheric metal atom chemistry [Plane, 2004], heterogeneous HOx chemistry [Summers et al., 2001], the charge balance of the D-region plasma [Rapp and Lübken, 2001], strong VHF radar echoes from the middle mesosphere (so-called polar mesosphere winter echoes or PMWE [see Stebel et al., 2004; La Hoz and Havnes, 2008]), and even the nucleation of particles in polar stratospheric clouds which are involved in ozone destruction during polar spring [Voigt et al., 2005].

[3] Despite the obvious scientific interest in MSPs, the corresponding experimental database has been surprisingly scarce. This is mainly because of the tiny dimensions of the particles which had led the community to believe that, for example, the detection of MSPs by optical remote sensing is impossible [e.g., Hunten et al., 1980]. In consequence, initial observations of MSPs were made with in situ techniques following the pioneering work of Havnes et al. [1996] on charged mesospheric ice particles which take advantage of the fact that the particles acquire a net charge in the
environment of the D region. Among these few successful detections are the Faraday cup measurements by Gelinas et al. [1998, 2005], Lynch et al. [2005], Rapp et al. [2005], and Strelnikova et al. [2009], the observations with magnetically shielded probes by Horányi et al. [2000] and Amey et al. [2008], and the Gerdien condensor measurements by Croskey et al. [2001]. Recently, Rapp et al. [2007] and Strelnikova et al. [2007] demonstrated that charged MSPs can be observed from the ground making use of the spectral information contained in Doppler spectra from incoherent scatter radar observations of the D-region plasma. Besides these measurements of charged MSPs, Rapp and Strelnikova [2009] and Strelnikova et al. [2009] recently further presented results from a new in situ technique which relies on the active photoionization of MSPs and subsequent detection of photoelectrons emitted by these particles. On the basis of corresponding measurements during a rocket flight in September 2006, these authors were able to demonstrate that MSPs indeed exist throughout the entire (winter) polar mesosphere as suggested by all available model predictions.

Additional interest in MSPs and their global distribution was recently initiated by the study of Gabrielli et al. [2004], who analyzed concentrations of iodium and platinum of meteoric origin in Greenland ice cores. They suggested that this meteoric material likely originated from MSPs which were transported into the winter polar vortices by the mesospheric meridional circulation by which they were preferentially deposited in the polar ice caps. This idea was further pursued in the model study by Megner et al. [2008b], who combined the two-dimensional circulation model NRL Chem2D with the Community Aerosol and Radiation Model for Atmospheres (CARMA) with which the microphysical processes of MSP growth and sedimentation were treated (see also Megner et al. [2006] for a detailed description and discussion of these microphysical processes). Making the assumptions that MSPs are formed at molecular sizes of about 0.2 nm and then grow by Brownian coagulation only, this study found that the residual circulation from the summer to the winter pole with corresponding mean upward and downward vertical winds in the summer and winter hemisphere leads to a depletion of MSPs with radii ≥1 nm in the polar summer mesopause region and a corresponding enrichment in the winter polar vortex. Importantly, these results were fully confirmed by independent model simulations with the three-dimensional Whole Atmosphere Community Climate Model (WACCM), which was also coupled with the CARMA model for the treatment of microphysical MSP processes [Bardeen et al., 2008].

As a consequence of this depletion of MSPs in the polar summer mesopause region, it was realized that the conventional picture of mesospheric ice nucleation on MSPs must be questioned [Megner et al., 2008a]. That is, if the corresponding model calculations were correct this would imply that there were too few ice nuclei to account for the observed number densities of mesospheric ice particles in NLC and PMSE. Hence, several alternative nucleation pathways such as the nucleation on charged (but significantly smaller) MSPs [Megner et al., 2008a; Gumbel and Megner, 2009; Megner and Gumbel, 2009] and also homogeneous nucleation in extreme temperature variations due to gravity waves [Lübken et al., 2009] have been suggested as alternative nucleation pathways.

[6] Hence, it is obvious that experimental information on the seasonal variation of MSPs is of great scientific interest. A first step in that direction was taken by Fentzke et al. [2009], who applied the method first described by Strelnikova et al. [2007] to dual beam observations with the Areceibo incoherent scatter radar (18.3°N, 66.8°E) from April, August, and September 2006. These authors did not detect any significant variation over these three months. This is in agreement with the expected annual variation of MSPs at low latitudes which is rather small compared to the very pronounced seasonal cycle at polar latitudes [Megner et al., 2008b; Bardeen et al., 2008].

[7] A major breakthrough in this respect was recently made by Hervig et al. [2009a], who presented solar occultation measurements of MSPs by the Solar Occultation For Ice Experiment (SOFIE) on board the Aeronomy of Ice in the Mesosphere (AIM) satellite. Thanks to the impressive sensitivity of the SOFIE extinction measurements, these authors were able to present polar (at a mean latitude of ~75°S) MSP observations from roughly 35 to 85 km altitude during winter, and from 35 to about 70 km during summer. Their observations were largely able to confirm the qualitative picture predicted by the model studies of Megner et al. [2008b] and Bardeen et al. [2008]. As a minor drawback, however, these measurements can by design only provide limited information on the abundance of MSPs in the polar summer mesopause region because observations there are dominated by mesospheric ice particles most of the time. Mesospheric ice particles mask potential MSP observations down to altitudes of about 70 km because of the limb viewing geometry of SOFIE. That is, mesospheric ice clouds at real altitudes between 80 and 90 km which are in the near or far field of the instrument appear at tangent altitudes well below this height range. In consequence, SOFIE registers ice clouds down to tangent altitudes as low as about 70 km [see Hervig et al., 2009b, Figure 11c]. Hence, additional measurements which do not suffer from such effects are necessary in order to quantify the seasonal variation of MSPs as close as possible to the summer mesopause region in order to understand their role in ice microphysics.

In this study, we will present rocket-borne in situ observations of MSPs and mesospheric ice particles from the Northern Norwegian Andøya Rocket Range at a latitude of 69°N in order to address the issue of MSP abundance in the polar summer mesopause region. In section 2, we describe the various observational techniques used in this study. Our observations are then presented in section 3. In section 4.1, these data will be discussed with respect to the role of background ionization and the charging properties of MSPs on the measured altitude profiles. In Appendix A it is further shown that our measurements are not contaminated by photoemission from molecular species like NO and O2(1Δe). Based on these analyses we will then finally turn to a discussion of evidence for a seasonal variation of MSPs in the mesopause region and we will close with a summary and suggestions for future work in section 5.

2. Experimental Details

[9] The observations presented here were obtained during three sounding rocket campaigns from the Northern Norwegian Andøya Rocket Range (69°N, 16°E) in the frame
of the German–Norwegian ECOMA project (ECOMA is an acronym for “Existence and Charge State Of Meteoric smoke particle in the middle Atmosphere”). The rocket campaigns were conducted in September 2006, i.e., outside the polar summer period, and in August 2007 together with the NASA-funded MASS campaign (MASS denotes Mesospheric Aerosol Sampling Spectrometer), and in June–July 2008. While the focus of the first campaign was on the experimental verification that MSPs do exist in the entire mesosphere, the latter two campaigns were dedicated to studying the distribution of MSPs in the presence of mesospheric ice layers and the potential role of MSPs for the nucleation of these ice particles. Some initial results from the campaign in 2006 are published by Rapp and Strelnikova [2009] and Strelnikova et al. [2009]. In addition, initial results of the ECOMA/MASS campaign in August 2007 with a focus on observations of mesospheric ice layers and their environment are presented in a recent special issue of Annales Geophysicae (M. Rapp and S. Robertson, Preface: ECOMA/MASS: Aerosol particles near the polar summer mesopause, 2009, available at http://www.ann-geophys.net/ prefaces/preface219.pdf). In this study, we will use data from all successful flights during the ECOMA project to determine the characteristics of MSPs during polar summer and compare these to our measurements from outside the polar summer season, i.e., from September 2006. A list with details about all these flights is provided in Table 1.

Table 1. Dates, Times, Solar Zenith Angles, and Launch Conditions of the ECOMA Sounding Rocket Flights

<table>
<thead>
<tr>
<th>Label</th>
<th>Date</th>
<th>Time (UT)</th>
<th>Solar Zenith Angle (deg)</th>
<th>Launch Condition</th>
</tr>
</thead>
<tbody>
<tr>
<td>ECOMA01</td>
<td>08 Sep 2006</td>
<td>2217</td>
<td>114.5</td>
<td>moderate ionosphere</td>
</tr>
<tr>
<td>ECOMA03</td>
<td>03 Aug 2007</td>
<td>2322</td>
<td>93.2</td>
<td>NLC and PMSE</td>
</tr>
<tr>
<td>ECOMA04</td>
<td>30 Jun 2008</td>
<td>1322</td>
<td>50.8</td>
<td>NLC and PMSE</td>
</tr>
<tr>
<td>ECOMA05</td>
<td>07 Jul 2008</td>
<td>2124</td>
<td>86.6</td>
<td>NLC, but no PMSE</td>
</tr>
<tr>
<td>ECOMA06</td>
<td>12 Jul 2008</td>
<td>1046</td>
<td>47.5</td>
<td>NLC and PMSE</td>
</tr>
</tbody>
</table>

2.1. Rocket-Borne Instruments

[11] The prime in situ instrument for this article is the ECOMA particle detector which is mounted on the front deck of the ECOMA payload [Rapp and Strelnikova, 2009]. In short, the ECOMA particle detector is a Faraday cup with two biased grids to shield the cup against ambient electrons and positive ions, similar to the one originally developed and successfully used by Havnes et al. [1996]. Heavy particles pass through these biased grids because of their large kinetic energy and, if they carry some charge, produce a current which can be measured by a sensitive electrometer. Note that neutral particles cannot be detected by this method, and neither can small aerosol particles with radii less than ~2 nm reach the detector electrode because of aerodynamical effects [Horányi et al., 1999; Rapp et al., 2005; Hedin et al., 2007]. To detect particles of a broad size range and independent of their charge state, however, the Faraday cup is combined with a xenon flash lamp for the active photoionization (photodetachment) of neutral (negatively charged) particles and the subsequent detection of photoelectrons. Hence, the detector has two data channels, one for the direct Faraday cup measurement and one for the detection of photoelectrons excited by the UV photons of the xenon flash lamp. The detailed detection scheme of the detector is shown in Figure 1. This schematic shows that the xenon flash is initiated every 64 ms, i.e., at a rate of 15.6 Hz (Figure 1, top). The current recorded at the electrode is continuously sampled at a rate of 100 kHz and 16 bit resolution. The first 64 samples after each flash are transmitted to the ground at full time resolution in order to capture the short pulses of photoelectrons emitted by particles which have been actively ionized/photodetached by flash photons (Figure 1, middle, indicated in red). The corresponding data channel is denoted the “photoelectron channel” and corresponding sample data from flight ECOMA05 during the passage of a noctilucent cloud are shown in Figure 2. When discussing photoelectron measurements from our rocket flights, we will usually show profiles of the maxima of the photoelectron peaks shown in Figure 2 (marked with red diamonds) which will be denoted as the “photoelectron current” for brevity. In addition, throughout the entire flight time, 100 data points taken at 100 kHz are averaged to yield “slow” data values at a rate of 1 kHz (Figure 1, bottom). These data are continuously transmitted to the ground and yield information on the slowly varying background particle charge density. These data correspond to classical Faraday cup measurements with the one difference that the first data point after each flash is contaminated by a contribution from artificially created photoelectrons and must be removed. The corresponding data channel is denoted the “Faraday cup channel.” Samples of actual measurements can be found in our earlier papers [see, e.g., Rapp and Strelnikova, 2009, Figures 4 and 5].

[12] In this study we will mainly focus on ECOMA observations of photoelectron currents, so, here we reiterate the most important properties of this novel technique. Accord-
ing to Rapp and Strelnikova [2009], the ECOMA flash current can be written as

\[
I = \left( \int_{c}^{\infty} F_{c} \cdot dF_{c} \cdot \sigma(r_{p}, m, \lambda) \cdot P \cdot dr_{p} \cdot dl \cdot d\lambda \right)
\]

where \( e \) is the electron charge, \( r_{\text{min}} \) is the minimum assumed size of the particles in a particle size distribution, \( v_{e} \) is the velocity of a photoelectron, \( \Delta t = 10 \mu \text{s} \) is the sampling interval during which photoelectrons are recorded, \( h \) is Planck’s constant, \( c \) is the speed of light, and \( W_{p} \) is the threshold energy for photoionization/photodetachment of a particle, i.e., the work function or electron affinity of the corresponding material. The \( dF/d\lambda \) is the number of photons per wavelength interval emitted in one flash [see Rapp and Strelnikova, 2009, Figure 2], and \( l \) is the distance from the particle detector. Note that the relative intensities of the flash lamps used in this study were quantified using the experimental setup described in the context of Rapp and Strelnikova [2009, Figure 9] yielding an overall small variation from lamp to lamp by less than \( \pm 15\% \). \( P = S(4\pi F) \) is the probability that the photoelectron is emitted toward the detector electrode with area \( S \). \( dN_{p}/dr_{p} \) is the number density of particles per size interval \( dr_{p} \), and \( dl \) and \( d\lambda \) are the length and wavelength elements over which the integrations above are carried out. Note that the integration over the wavelength \( \lambda \) starts at 110 nm because of the transmission properties of the MgF\(_{2}\) window of the Xe flashlamp. Finally,

\[
\sigma(r_{p}, m, \lambda) = \pi r_{p}^{2} \cdot Q_{\text{abs}}(r_{p}, m, \lambda) \cdot Y
\]

is the photoionization/photodetachment cross section of particles with radius \( r_{p} \) and complex index of refraction \( m = n + i k \) at photon wavelength \( \lambda \). For any calculations shown in this manuscript, cross sections were calculated using the publicly available Mie code from the textbook by

![Figure 1. Detection scheme of the ECOMA particle detector. See text for details.](image)

![Figure 2. Samples of raw currents measured during flight ECOMA05 in the photoelectron channel at a flight time of \(~269.3 \text{ s}\) corresponding to an altitude of about 84 km (downleg part of the rocket trajectory) when the rocket passed through a noctilucent cloud (see also Figure 5 and corresponding discussion). The blue dotted lines indicate the times when the xenon flashlamp was fired. Black stars indicate the current recorded as a consequence of photoelectrons hitting the electrode. Note that the number of samples shown on the \( x \) axis is not directly proportional to flight time since the flash is fired at a rate of 15.6 Hz only. That is, each of the 64 samples after their corresponding flashes correspond to 0.64 ms of flight time, but the next flash is fired after 64 ms only. Red diamonds mark the maxima of the photoelectron pulses. In subsequent plots, only these maxima will be shown and denoted as the “ECOMA photoelectron current.”](image)
Calculated photoelectron currents expected by the ECOMA particle detector as a function of particle volume density assuming different particle materials (Fe₂O₃ or SiO) and threshold energies (2.0 eV for photodetachment and 5.5 eV for photoionization.) The dashed black line indicates a slope of 1, i.e., a direct proportionality between the expected current and particle volume density.

In equation (2), \( Q_{abs} \) is the Mie absorption efficiency, and \( Y \) is the quantum yield for photodetachment and 5.5 eV for photoionization.) The dashed black line indicates a slope of 1, i.e., a direct proportionality between the expected current and particle volume density.

\[ Q_{abs} = -\frac{4\pi}{3} \cdot \text{Im} \left( \frac{m^2 - 1}{m^2 + 2} \right), \]

where \( \text{Im}(a) \) denotes the imaginary part of the complex number \( a \). For the cross section \( \sigma(r_p, \lambda) \) this finally yields

\[ \sigma(r_p, \lambda) = -\frac{8\pi^2 r_p^3}{3\lambda} \cdot \text{Im} \left( \frac{m^2 - 1}{m^2 + 2} \right) \cdot Y = -\frac{2\pi \text{Im} \left( \frac{m^2 - 1}{m^2 + 2} \right)}{\lambda} \cdot V_p \cdot Y, \]

where \( V_p \) is the particle volume. That this proportionality holds for all conceivable parameters of MSPs is demonstrated in Figure 3 which shows theoretical photoelectron currents as a function of MSP volume density based on equation (1) and assuming different MSP materials. These calculations were carried out for a large number of MSP size distributions of the type predicted by currently available MSP models [Hunten et al., 1980; Saunders et al., 2007; Megner et al., 2008a; Bardeen et al., 2008]. Indeed, Figure 3 demonstrates that the photoelectron current is strictly proportional to the volume density of MSPs hence validating the Rayleigh approximation. We note that the general proportionality between the absorption cross section and particle volume is also valid for nonspherical particles which would, however, lead to a moderate increase of the cross section by up to \( \sim 40\% \) (if for example very extreme randomly oriented spheroids with axial ratios of 0.1 or 10 were assumed) [Bohren and Huffman, 1983; Min et al., 2006].

Importantly, we further note that this general proportionality of photoionization and photodetachment cross sections to the particle volume is generally confirmed by laboratory measurements of atomic and molecular clusters [Brechignac et al., 1991; Zajfman et al., 1992; Maeyama et al., 1997], even though we need to appreciate that the exact properties of MSPs are not known due to the lack of precise knowledge regarding their composition as well as corresponding laboratory investigations.

Each ECOMA payload was further equipped with the CONE (COmbined measurement of Neutrals and Electrons) instrument (see Giebeler et al., 1993 for more details). CONE is a combination of an ionization gauge for the measurement of neutral density and a fixed biased Langmuir probe to measure electron number density. CONE yields measurements with very high spatial resolution and allows the detection of small-scale fluctuations in both species (neutrals and electrons) that arise due to processes such as neutral air turbulence (e.g., Lübken et al., 2002) or plasma instabilities (e.g., Strelnikov et al., 2009). In addition, the height profile of neutral number densities can be integrated assuming hydrostatic equilibrium to yield a temperature profile at \( \sim 200\) m altitude resolution and an accuracy of \( \sim 3\) K [Rapp et al., 2001, 2002].

In addition, high-precision absolute electron density measurements were made using a radio wave propagation experiment [Friedrich et al., 2006]. Basically, this is realized by transmitting a radio signal from the ground and receiving it by a pair of antennas on the rocket. The theoretical basis and practical application of this technique are described for example by Bennett et al. (1972) and Smith (1986). The transmitted linearly polarized electromagnetic wave may be considered to be the resultant of two circularly polarized waves with equal electric field vectors rotating in opposite directions. When propagating through the ionospheric plasma these two waves differ in absorption and phase velocity. The difference in absorption causes the resultant wave to become elliptically polarized (differential absorption). In addition, the difference in phase velocity causes the major axis of the polarization ellipse to rotate as the wave propagates (Faraday rotation). Since both differential absorption and Faraday rotation directly depend on the electron number density along the path of the radio signal through the ionosphere, the height-resolved measurements of both quantities can be used to precisely derive the electron number density profile at a typical vertical resolution of \( \sim 1\) km (defined by the rocket velocity and the rocket spin rate). Importantly, these measurements are not influenced by undesirable effects due to the interaction of the payload with the ambient plasma such as payload charging.

In 2007 and 2008, each ECOMA payload also carried a NLC photometer from the University of Stockholm for the optical in situ detection of mesospheric ice particles [e.g.,
Figure 4. Altitude profile of the photoelectron current (in red) measured during sounding rocket flight ECOMA01 in September 2006 (see Table 1 for details). The upper abscissa converts the measured photoelectron current to volume density assuming that the particles consisted of Fe₂O₃. The thick black line with thick diamonds is a model profile for the same latitude and month taken from the model study by Megner et al. [2008b]. Corresponding thin black lines indicate model uncertainties caused by uncertainties of the meteoroid influx as well as the altitude of maximum ablation. The thin orange line shows the measured photoelectron currents divided by a factor of 3.

Gumbel et al., 2001]. This type of instrument looks at the scattered intensity of sunlight from the particles, as well as the angular distribution of scattering from which the phase function can be derived. The design of the particular photometer on board the ECOMA payloads is described in detail in the paper by Megner et al. [2009].

Finally, independent particle measurements were provided from the mesospheric aerosol sampling spectrometer (MASS) instrument which was launched on board a NASA-funded sounding rocket on 3 August 2007 at 2251:20 UTC, i.e., about 30 min before the launch of ECOMA03 (see Table 1). The MASS instrument has been described in detail by Knappmiller et al. [2008], and results from the rocket flight mentioned above were reported by Robertson et al. [2009]. In short, the MASS instrument is a mass spectrometer which uses electrostatic deflection to separate particles by their mass-to-charge ratio. The instrument possesses 8 channels and is able to discriminate positive and negative particles in the size ranges <0.5 nm, 0.5–1 nm, 1–2 nm, and >3 nm (radii of mesospheric ice particles with a density of 0.93 g/cm³; calculations for an altitude of ~85 km).

2.2. Ground-Based Instruments

Both ECOMA sounding rocket campaigns in August 2007 and June–July 2008 were supported by ground-based measurements with the ALWIN radar, the Andenes MF radar, and the ALOMAR RMR lidar.

The ALOMAR wind (ALWIN) radar and the ALOMAR RMR lidar are two of the prime instruments of the ALOMAR observatory located in close vicinity to Andøya Rocket Range at 69.3°N and 16°E. The ALWIN radar is a phased array consisting of 144 Yagi antennas operating at a frequency of 53.5 MHz and has been extensively used for the study of polar mesosphere summer echoes (PMSE) over the past ~10 years [Latteck et al., 1999; Bremer et al., 2006; Latteck et al., 2007].

The ALOMAR MF radar operates at 1.98 MHz with a peak power of 40 kW applying the spaced antenna technique. A wide-beam antenna transmits vertically radio wave pulses of 4 km length. Their atmospheric returns are received by three crossed horizontal dipoles arranged in an equilateral triangle. The radar continuously provides horizontal winds and tides at altitudes between 70 and 94 km since 1998 using the full correlation analysis method [e.g., Hoffmann et al., 2007].

The ALOMAR Rayleigh/Mie/Raman (RMR) lidar measures relative density profiles and particle (aerosol) properties in the stratosphere and mesosphere and has been described in detail by von Zahn et al. [2000]. Throughout the noctilucent cloud (NLC) season (1 June to 15 August) the lidar is operational 24 h a day to take advantage of even short measurements permitted by the weather conditions. A recent review of corresponding results is presented by Baumgarten et al. [2008]. Furthermore, an overview of the mesospheric ice cloud morphology during the ECOMA/MASS sounding rocket campaign in August 2007 is presented by Baumgarten et al. [2009].

3. Observations

Before we turn to the results of the ECOMA observations in August 2007 and June/July 2008, we recapitulate our findings from flight ECOMA01 from September 2006, i.e., from outside the polar summer season. These data were discussed in detail by Strelnikova et al. [2009] who also attempted to convert the measured photoelectron currents to MSP number densities. However, this required strong assumptions regarding the altitude-dependent size distribution of the MSPs which had to be taken from a model. A significantly more direct way of representing these data and comparing it to model simulations is by recalling that the photoelectron current is directly proportional to the total particle volume density. Hence, making only one assumption about the particle composition, measured and simulated profiles of total particle volume density can be compared in a much more direct way.

The result of this exercise is presented in Figure 4 where we have assumed that the particles consist of hematite, i.e., Fe₂O₃ for which corresponding refractive indices were taken from the Database of Optical Constants for Cosmic Dust made available by the Astrophysical Laboratory of the University of Jena, Germany (see http://www.astro.uni-jena.de/Laboratory/OCDB/oxsul.html). While the actual composition of MSPs is still not known we note that hematite has been named as a plausible candidate based on laboratory experiments [Saunders and Plane, 2006] and actually appears to have been directly observed at least once from the middle atmosphere [Bohren and Olivero, 1984]. We further note that the choice of a different material would simply shift the volume density profile derived from our photoelectron currents by a constant factor since the cross section for photoemission or photodetachment is directly
proportional to volume density and the imaginary part of the particle refractive index (see equations (1) and (4)).

In addition, it must be realized that also the model has large uncertainties as summarized recently in the model study by Megner et al. [2006] (see their Figure 4 and corresponding discussion). For the current model-data comparison, the largest unknowns are caused by uncertainties of the total meteoroid influx as well as by the altitude of maximum ablation. Megner et al. [2008b] assume these two crucial quantities to be 44 t/d and ∼85 km. However, estimates in the literature range from 5 t/d to 400 t/d for the total meteoroid influx [Gabrielli et al., 2004; Love and Brownlee, 1993; Mathews et al., 2001; Ceplecha et al., 1998] and maximum ablation altitudes as high as 100 km have been suggested [e.g., Sparks and Janches, 2009]. In order to demonstrate the large uncertainties of the model results shown here, Figure 4 hence also shows corresponding model error bars which were derived by considering factors of 0.5 and 2 for the meteoroid influx as well as allowing for a moderate 5 km higher ablation altitude.

Having these caveats in mind, we see that measured photoelectron currents (red line) correspond to volume densities between $4 \times 10^{-17}$ cm$^{-3}$cm$^{-2}$ and $3 \times 10^{-16}$ cm$^{-3}$cm$^{-2}$. Comparing this to the model results from Megner et al. [2008b] (thick black line) for the relevant latitude and time and taking into account the corresponding model error bars, we see that the relative shapes of the profiles are consistent with each other between 65 and ∼85 km but that the model results are lower by about a factor of 3. Again, we stress that this relative shift can easily be accounted for if the particles consist of a different material with larger imaginary part of the refractive index (note that we already assumed that the quantum yield $Y$ attained a maximum conceivable value of 1; see section 2.1 above).

Furthermore, observational and theoretical evidence now strongly suggests that there is indeed a large geographical and seasonal variation of this flux [Janches et al., 2004, 2006; Singer et al., 2004; Pentke et al., 2009] which is particularly pronounced at polar latitudes and peaks in September, i.e., at the time of the ECOMA01 launch. This could have resulted in a corresponding underestimate of the MSP volume density in the model. However, above ∼85 km, the model and observations start deviating more and more and it was shown by Rapp and Strelnikova [2009] and Strelnikova et al. [2009] that this difference above ∼85 km likely originates from the contribution of other ionizable species like NO and O$_2$(1D$_g$) to the measured currents. Nevertheless, we emphasize that below this altitude region, the match between measured and modeled MSP profiles (taking into account the above described uncertainties) is
Temperature profiles measured during the rocket flights ECOMA01 in September 2006 (grey line) and ECOMA03, ECOMA04, and ECOMA06 from August 2007 and June and July 2008, respectively. The black dotted and dashed lines show the frost point temperature for water ice using the water vapor saturation pressure according to Murphy and Koop [2005] and assuming water vapor mixing ratios of 1 ppmv and 4 ppmv, respectively, over the whole altitude range.

Figure 6. Temperature profiles measured during the rocket flights ECOMA01 in September 2006 (grey line) and ECOMA03, ECOMA04, and ECOMA06 from August 2007 and June and July 2008, respectively. The black dotted and dashed lines show the frost point temperature for water ice using the water vapor saturation pressure according to Murphy and Koop [2005] and assuming water vapor mixing ratios of 1 ppmv and 4 ppmv, respectively, over the whole altitude range.

satisfactory which underlines the feasibility of this new in situ technique.

We now turn to our corresponding Summer results. The results from all four sounding rocket flights under polar summer conditions are presented in Figure 5. Except for flight ECOMA05, all summer flights were conducted under conditions of radar and lidar observations of PMSE and NLC, hence confirming the presence of mesospheric ice particles at the time of the rocket launches. In addition, the presence of mesospheric ice particles at the time and place of the actual passage of the ECOMA payloads through the altitude ranges from ~80–90 km was directly proven by onboard photometer measurements during all flights (see dashed blue profiles and upper abscissae for these data), including flight ECOMA05. ECOMA05 was launched under the confirmed absence of PMSE, while ground-based information about NLC was not available owing to poor tropospheric viewing conditions for the optical measurements. Nevertheless, the onboard photometer proved the presence of a mesospheric ice layer also during ECOMA05. In all four plots of Figure 5, the red lines show the photoelectron currents measured by the ECOMA particle detector. For comparison, the corresponding currents from the September flight ECOMA01 are overplotted in each plot (black lines). As additional information, we have marked the altitude ranges in which the Faraday cup channel of the ECOMA particle detector registered charged ice aerosol particles which penetrated into the detector and created a direct current on the detector electrode. We further note that the simultaneous temperature measurements with the CONE instrument (for flights ECOMA03, ECOMA04 and ECOMA06, see Figure 6) all yielded temperatures well below the frost point of water ice, hence reinforcing our interpretation that these particles were made of water ice. A temperature profile for flight ECOMA05 is unfortunately not available because of a malfunction of the CONE instrument during that flight. For the purpose of the present study we use the Faraday cup observations in combination with the CONE temperature measurements only to discern altitude ranges with and without the presence of water ice particles. A more thorough interpretation of these data, for example, with respect to the local charge balance is beyond the scope of the present paper and will be presented in a future study (T. Blix et al., manuscript in preparation, 2009).

Turning back to the photoelectron measurements, we see that during all four Summer flights the NLC layers detected by the optical photometer measurements were also identified as pronounced peaks in the photoelectron data (note that there are certainly no NLC and hence no corresponding photoelectron peak during September). For the case of flight ECOMA03, this was analyzed in detail by Rapp et al. [2009] who were able to demonstrate that these currents give direct information on the volume density of the water ice particles (even though the relation between volume and current is not as straight forward as for the MSP particles since ice particles are much bigger, i.e., several tens of nanometers, such that the absorption is no longer strictly in the Rayleigh regime). We note that the NLC peak during flight ECOMA05 actually has a curious shape with two peaks, one at the bottom and the other at the top of the NLC layer recorded by the photometer. A detailed analysis of these NLC observations will be presented in a future study.

The most striking difference between the summer flights and the September flight is the prominent current down to about 60 km for the Fall flight, while the summer flights recorded no current below 70 km. In addition, the four summer flights show a large variability in the current above their corresponding lower cutoff altitudes. For example, during flight ECOMA03 no photoelectron current was actually recorded in the entire altitude range from 60 to 90 km except for the above mentioned NLC peak. During flights ECOMA04 and ECOMA05, a small photoelectron current was recorded down to altitudes of 78 km and ~82–85 km, respectively, which was almost everywhere smaller than the corresponding current from flight ECOMA01 during September 2006. During flight ECOMA06, however, the recorded photoelectron current exceeded that of flight ECOMA01 everywhere above 75 km by as much as a factor of 2, while it shows a sharp bottom scale height below that and vanishes into the detector noise (~200 pA) at about 70 km.

4. Discussion

The natural question that arises from these observations is what is causing the observed variability of the photoelectron currents during polar summer. In this discussion section, we will consider different potential physical mechanisms which could have caused this variability, where we will, however, focus on the variation of the smoke layer outside the ice region. These are particle charging (section 4.1) and a potential underlying seasonal variation of the MSP layer owing to the residual circulation (section 4.2). In addition, possible contaminations from the
4.1. Role of Background Ionization and Particle Charging

[32] The issue of particle charging under conditions of the polar summer mesopause region was recently reconsidered in a study by Rapp [2009]. Motivated by the repeated observation of the coexistence of small positively and larger negatively charged particles in an NLC/PMSE environment [Croskey et al., 2001; Smiley et al., 2006; Robertson et al., 2009; Brattli et al., 2009], Rapp [2009] considered time constants for photodetachment and photoemission from hematite MSPs and ice particles and compared these to time constants for electron capture by neutral and positively charged particles. Photodetachment and photoionization time constants were calculated according to

\[ \tau_p = \left( \int \frac{F(\lambda, \chi) \cdot \sigma(r, m, \lambda) \cdot d\lambda}{(\lambda^* - \lambda)} \right)^{-1}, \]  

where \( F(\lambda, \chi) \) is the flux of solar photons at wavelength \( \lambda \) and solar zenith angle \( \chi \), \( \sigma(r, m, \lambda) \) is the absorption cross section of the particle depending on particle radius, complex refractive index \( m = n + ik \), and wavelength as defined in equation (2). The \( \lambda^* = hc/E^* \) is the critical wavelength up to which the integration is carried out where \( h \) is Planck’s constant, \( c \) is the speed of light, and \( E^* \) is the threshold energy for photodetachment or photoionization.

[33] Values of \( \tau_p \) were compared to time constants for electron attachment which were calculated according to

\[ \tau_e = \frac{1}{\alpha_e \cdot N_e}, \]  

where

\[ \alpha_e^0 = \pi r^2 \cdot v_i \cdot \left(1 + \frac{e^2}{8\epsilon_0 \cdot k \cdot T \cdot r}\right) \]  

and

\[ \alpha_e^+ = \pi r^2 \cdot v_i \cdot \left(1 + \frac{e^2}{4\pi\epsilon_0 \cdot k \cdot T \cdot r}\right) \]  

are the electron capture rate for a neutral and singly positively charged particle, respectively. The rates in equations (7) and (8) were calculated based on the theory developed by Natanson [1960], where \( r \) is the particle radius, \( v_i \) is the mean thermal velocity of the electrons, \( e \) is the electron charge, \( \epsilon_0 \) is the permittivity of space, \( k \) is Boltzmann’s constant, and \( T \) is temperature which is equal for all charged and neutral constituents.

[34] Because of the very different absorption properties of hematite MSPs on the one hand and ice particles on the other hand, Rapp [2009] showed that photodetachment of electrons by solar photons (in the visible and UV spectral range) from charged hematite MSPs is at least 4 orders of magnitudes faster than electron attachment. Hence, these particles cannot become negatively charged but rather become positively charged as long as the electron number density does not increase above a certain limit of about 1000 cm\(^{-3}\). Ice particles, on the other hand, hardly absorb any of these solar photons such that under normal conditions photodetachment and photoionization are irrelevant and, hence, ice particles acquire a negative charge. In summary, the observed coexistence of positive and negative particles in the polar summer mesopause region can be readily explained by the coexistence of positively charged MSPs and negatively charged ice particles. We note that ice particles in the polar summer mesopause region likely contain a meteor smoke core such that in principle this core could also change the charging properties of the ice particles, at least as long as the particles are small. The detailed properties of such ice-coated particles are, however, beyond the scope of the current paper and will be considered in a future publication (S. Knappmiller et al., manuscript in preparation, 2010). Finally, we note that Havnes and Kassa [2009] arrived independently at similar results regarding the importance of photodetachment of electrons from negatively charged MSPs when considering observed temporal characteristics of active HF heating experiments under conditions of PMWE.

[35] For the present purpose, we have repeated a similar analysis as that of Rapp [2009] for the particular conditions of the ECOMA sounding rocket flights in August 2007 and June–July 2008. Referring to Table 1 we see that the solar zenith angle (SZA) during these flights varied between 47.5° and 93°. In order to quantify the importance of photodetachment and photoemission from MSPs for our observations, we have first calculated the solar irradiance for altitudes of 85 km and 60 km (i.e., maximum and minimum altitudes where we can reasonably expect to detect MSPs with our detector) and for solar zenith angles at 47° and 93°. These radiative transfer calculations were carried out using an O2 profile (dominating absorption below wavelengths of \(~200\) nm) from the MSIS reference atmosphere and an ozone profile (dominating the absorption between \(~200–300\) nm) from Microwave Limb Sounder (MLS) observations averaged zonally over the latitude interval 64°N–74°N on 1 July 2008 (C. Randall, personal communication, 2009). For more details about this radiative transfer model we refer readers to the study by Hedin et al. [2008].

[36] Corresponding results are presented in Figure 7 (top) where we have also overplotted the imaginary part of the refractive index of hematite for comparison (recall the expression for the photoionization cross section in equation (4)). In the two bottom plots we show corresponding time constants for photodetachment, photoionization, and electron capture for an ambient electron density of 1000 cm\(^{-3}\). Figure 7 shows that photodetachment is indeed by far the fastest process for all altitudes and zenith angles relevant to the ECOMA rocket flights during summer.

[37] Comparing further the time constants for photoionization and electron attachment to a positively charged particle we see that photoionization is also faster than electron attachment (for an electron density of 1000 cm\(^{-3}\)) for radii larger than about 1.5 nm except for SZA = 93° and an altitude of 60 km. For radii smaller than \(~1.5\) nm, however, electron attachment is fast enough to neutralize positively charged particles. Importantly, we note that the radius below which particles can be neutralized by electron attachment to a positively charged particle depends on the electron density (see equation (6)). This implies that in the presence of more (less) free electrons a larger (smaller) part of the MSPs can be neutral instead of positively charged. This is clearly seen
in Figure 8 in which we show charge distributions of hematite MSPs with a radius of 1 nm for nighttime and daytime conditions (the latter for different electron densities) which were calculated using the discrete charging model first described by Draine and Sutin [1987].

[38] In contrast, we note that the conditions for flight ECOMA01 were much simpler. During the ECOMA01 flight the SZA was 114.5° for which the solar irradiance is zero at the altitudes discussed here. This means that neither photodetachment nor photoionization play any role. Hence, for the case of ECOMA01 the particles had to be negatively charged as confirmed by the direct Faraday cup observations during that flight [Strelnikova et al., 2009].

[39] What do these considerations now imply for the observations during the ECOMA flights? The point that we want to make here is that the photoelectron current measured by the ECOMA particle detector does indeed depend on the particle charge. It is evident from equation (1) that the photoelectron current depends on the threshold energy for photoionization/photodetachment, i.e., $W_p$. The larger this threshold energy is, the smaller the wavelength interval over which the product of photon flux from the flashlamp and the corresponding cross section is integrated. For negatively charged hematite MSPs, the binding energy of the excess electron is given by the electron affinity which is 2 eV [Wang et al., 1996]. In contrast, the work function of a neutral hematite MSP is on the order of 5.5 eV but also depends on the particle radius (i.e., 5.5 eV is the work function for an extended macroscopic surface). Furthermore, if a particle is positively charged, a photoelectron to be emitted from this particle sees a larger retarding electric field than from a neutral particle such that the work function is further increased. According to Wood [1981] and Burtscher et al. [1982] the work function of a nanoparticle depends in the following way on particle radius $r_p$ and the number of positive elementary charges $p$:

$$W_p = W_{p0} + \frac{e^2(p + 1)}{4\pi\varepsilon_0 r_p} - \frac{5}{8} \frac{e^2}{4\pi\varepsilon_0 r_p},$$

(9)

where $W_{p0}$ is the work function of an extended macroscopic object, $e$ is the elementary charge, and $\varepsilon_0$ is the permittivity.
of space. We have used this expression in calculating the photoelectron current as a function of MSP radius and have repeated this calculation for negative, neutral, and singly and doubly positively charged hematite MSPs. Results of these calculations normalized to the current for negatively charged particles are presented in Figure 9. Figure 9 demonstrates that the dependence of the photoelectron current on the particle charge can be quite significant: for example, for a 1 nm particle, the current is reduced by factors of 2.5/17/50 in going from negative particles to neutral/singly positive/doubly positive particles. This effect is even larger for smaller particles (e.g., factors of $\sim 5/100/1000$ for $r_p = 0.5$ nm). Note, however, that the validity of the used equations becomes questionable at such small radii as the particle size approaches molecular dimensions and quantum effects need to be taken into account (see Rapp [2009] for a corresponding discussion). For large particles the reduction factor reaches a constant value of 2.5 for all charging states different than negative at about 10 nm.

Taking into account that the noise level of the photoelectron channel of the ECOMA particle detector is about 200 pA, we see that the maximum signal reduction, for example, from flight ECOMA01 to flight ECOMA03 was on the order of a factor of 50. Given the arguments above, we note that this can be explained if particles during ECOMA01 were negatively charged (as observed) and particles during ECOMA03 were positively charged.

So is there evidence in our data confirming that MSPs during Summer are either positively charged or neutral? In order to investigate this question, we recall from Figure 7 that an electron density of about $1000 \text{ cm}^{-3}$ would result in particles smaller than 1.5–2 nm being neutralized by electron attachment while larger particles would still be positively charged. Likewise, for smaller electron densities, a larger fraction of MSPs (i.e., down to smaller radii) would become positively charged. So, do we see a systematic variation of photoelectron currents measured during summer and simultaneously measured electron densities? A comparison of these two data sets is presented in Figure 10. Comparing the lower altitudes where photoelectron currents were observed below the ice existence region of about 80–90 km, i.e., during flights ECOMA04 and ECOMA06, we see that this altitude corresponds to an electron density of
about 1000 cm\(^{-3}\) in both cases. This is consistent with our idea that photoelectron currents were observed when at least a fraction of the particles became neutral as determined by a minimum electron density of about 1000 cm\(^{-3}\). Below that altitude, we argue that a dominant positive charge prevented significant photoelectron currents to be emitted from the particles (see Figure 9).

In particular, if the arguments above were correct, there should have been positively charged particles during flight ECOMA03. While the Faraday cup channel of this instrument can only provide information on charged particles above an altitude of about 80 km because of aerodynamical effects, and hence from the altitudes where ice particles occur, the aerosol mass spectrometer MASS launched about 30 min before the launch of ECOMA03 is indeed capable of detecting both negatively and positively charged aerosols at altitudes below the ice layer. In order to confirm this quantitatively, a set of aerodynamical calculations as described by Knappmiller et al. [2008] was carried out for an altitude of 75 km, in order to determine the mass sensitivity of the different channels of the MASS instrument for this altitude range. The result of these calculations is shown in Figure 11 (bottom). Figure 11 shows that channel 2 is sensitive to particles with peak sensitivity at about 300 amu. Note that water cluster ions could potentially fall into the same mass range. However, calculations of the sensitivity of the different channels of the MASS instrument show that water clusters should be simultaneously detected in channel 2 and 3 (up and downward orange triangles in Figure 11 (bottom)). This has been shown using measured values of the collision cross section of these cluster ions with air molecules. This allows to distinguish water cluster ions and positively charged MSPs by looking at differences in channels 2 and 3 (see Knappmiller et al. [2008] for more details).

Turning now to the observations made during the MASS flight as shown in Figure 11 (top), we see that at altitudes below 80 km, there is a clear signature of positive particles in channel 2 but no such signature in channel 3. This confirms that the measured signal originates from positively charged aerosol particles, i.e., likely positively charged MSPs, and not from water cluster ions. We also note that channel 2 also recorded a small signal owing to negatively charged particles with an amplitude which is about 20% of the signal measured for positive particles in the same mass range. These results imply that our general considerations about the charging of MSPs in the presence of sunlight are correct, but that still a minor fraction of particles can indeed acquire a negative charge. This is probably the case because our idealized charging calculations do not take into account the detailed electronic properties of the MSPs but rather use refractive indices derived from measurements of much larger macroscopic objects. In addition, we repeat that we have no information about the actual composition of the particles but had to assume that they consisted of hematite. Hence, we here reiterate one of the conclusions of Rapp [2009], namely that laboratory measurements of the charging properties of MSP-type par-

\[\text{Figure 10. (left) Comparison of altitude profiles of photoelectron currents measured during the ECOMA sounding rocket flights in summer and (right) corresponding electron densities measured with the Faraday rotation method on board the same rockets. The dotted horizontal lines indicate altitudes with an electron density of about 1000 cm}^{-3}\ \text{(10}^9 \text{m}^{-3}\text{) for the flights ECOMA04 (light blue) and ECOMA06 (red). The vertical black line marks an electron density of 1000 cm}^{-3}.\]
ticles in the transitions size range from molecular clusters to nanoparticles are needed to derive more quantitative results. We further note that, unfortunately, our measurements can only be compared to this one MASS flight while more observations with this instrument would certainly have been very useful. However, we would also like to point out that previous observations and/or theoretical results under similar ionospheric background conditions have all yielded evidence for a population of small positively charged particles which coexisted with negatively charged particles in NLC/PMSE and partly extended beyond the ice altitude range. These are the Gerdien condensor observations of Croskey et al. [2001] and Mitchell and Croskey [2001], the observations with a simple magnetically shielded probe by Smiley et al. [2006], and the combined experimental/theoretical study by Brattli et al. [2009].

Finally, we address the question to which extent our results actually depend on the specific choice of hematite as the major component of MSPs. We stress that it is not the particular choice of hematite, but that any material with strong absorption (and hence strong photoemission) in the UV and visible wavelength range would lead to the same qualitative results as presented above. As an example, SiO also absorbs strongly in the relevant wavelength range and hence also leads to equivalently strong photoemission (see, e.g., Figure 3). There is, in addition, one important point which we would like to stress here: that is, both the functional principle of our detector and the above charging mechanism by photodetachment and photoemission rely on exactly the same physical processes. This means that if meteor smoke particles consisted of significantly weaker absorbing materials than discussed above, this would imply that there would also be no means to explain the photoelectron currents observed by the ECOMA detector quantitatively. Since we can exclude contamination of our measurements by photoelectrons from other species than MSPs (see Appendix A) as a possible source for our observations, we are hence confident that the interpretation presented above is qualitatively correct.

Summarizing the discussion above we conclude that we have presented both theoretical and observational evidence that at least part of the variability of the photoelectron currents observed during the ECOMA flights is due to different charging states of the MSPs. While MSPs during nighttime are negatively charged and can hence give rise to large photoelectron currents, these currents are reduced in the presence of sunlight owing to photodetachment and photoionization processes. This reduction, in turn can be modulated by the free electron number density; that is, a larger electron number density allows a larger photoelectron current to be detected. In section 4.2, we finally discuss the implications of our observations for the seasonal variation of MSPs in the polar mesopause region.

4.2. Seasonal Variation of MSPs From Photoelectron Measurements

As shown above, part of the variation seen in different photoelectron measurements with our instrument can be explained as being due to different charging states of the MSPs. For possible conclusions on the seasonal variation of MSPs, we hence now focus our discussion on a comparison of flight ECOMA01 and flight ECOMA06 as the two extreme cases. ECOMA01 was conducted during darkness such that photodetachment and photoionization did not play any role for the charging of these particles. Instead, these particles were negatively charged by electron attachment as proven by the Faraday cup channel observations with the same instrument [Sirelnikova et al., 2009]. ECOMA06 on the other hand was conducted during daylight conditions such that photodetachment and photoionization, but also a quite large electron number density need to be taken into account. While direct information about the charging state of these particles is not available (because the Faraday cup channel data are due to ice particles), we argue based on the above considerations that the particles should have been neutral above ~70 km with potentially a fraction of nega-
tively charged particles (in analogy to the MASS observations). However, in any case, owing to the photodetachment and photoionization processes, we argue that there should not have been a larger fraction of negatively charged particles than during the September flight in darkness.

Nevertheless, a direct comparison of the measurements during both flights shows that the ECOMA06 flash currents are actually larger than the ECOMA01 flash currents by about a factor of 2 above an altitude of ~75 km (and we have argued that the cutoff seen in ECOMA06 below that altitude is due to a decreasing free electron density). At first sight, this observation appears to be inconsistent with the model prediction of a severe reduction in MSP abundance during summer as predicted by Megner et al. [2008b] and Bardeen et al. [2008]. However, we will see below that this conclusion would be premature. The point we want to make here is that the above cited model predictions are for number densities of particles with radii in excess of 1 nm (which are thought to be the fraction of MSPs available for nucleation of ice particles) and not for the particle volume density (which scales as $r_p^3$) as observed by the ECOMA photoelectron measurements. Secondly, we note that the above models did not consider the time constant of transition from a summer state with a severe MSP depletion toward a winter state with significantly enhanced MSP number densities (again larger than 1 nm). In order to scrutinize this further we have used the model simulations of Megner et al. [2008b] to look into the predicted seasonal variation of MSP volume density at a latitude of 70°N, i.e., for conditions appropriate for our rocket observations. The result of this analysis is presented in Figure 12. Figure 12 reveals that MSP volume is basically constant above an altitude of 85 km while below that altitude the volume density is in fact larger in the middle of summer than it is in the beginning/middle of September (Figure 12a). Looking at corresponding number densities of particles with sizes larger than 0.2 nm, 1 nm, and 3 nm (Figures 12b–12d) we see that the constant volume density above 85 km is consistent with constant number densities of small MSPs (Figures 12b and 12c). Below 85 km, however, number densities of particles with radii larger than 1 nm show a severe depletion in the middle of Summer and larger number densities in September, i.e., the opposite behavior as the volume density. The reason for this apparent discrepancy can be seen by looking at Figure 12d which shows number densities of particles larger than 3 nm. These show in fact the same qualitative seasonal variation as the volume density below ~85 km which implies that the latter is mainly determined by the few large particles due to its $r_p^3$ dependence on particle radius.

In order to identify the reason for this behavior we need to consider the residual wind fields in the model. Figure 13 shows the seasonal variation of the zonal, meridional, and vertical wind component from the NRL Chem2D model simulations used by Megner et al. [2008b]. This shows that the winter circulation with downward vertical and meridional transport from the winter to the summer pole has already stopped around day 100, i.e., in the middle of April. This implies that MSPs sedimenting down from above (and growing by coagulation on their way in the meantime) are no longer transported downward (since transport by the mean vertical wind dominates gravitational

![Figure 12](image-url)

**Figure 12.** (a) MSP volume density for a latitude of 70°N as a function of season and altitude from the model by Megner et al. [2008b]. Corresponding particle number densities of (b) all MSPs ($r \geq 0.2$ nm) and those with radii larger than (c) 1 nm and (d) 3 nm. In all plots the vertical red lines indicate times of the summer launches (here 15 July) and of the September launch.
settling by far) such that contours of particles larger than \( \sim 3 \) nm start sloping upward until the wind system changes to a well-established summer regime (\( \sim \) day 150) with strong transport away from the pole and corresponding upwelling, hence preventing any more large particles from reaching low altitudes from above. In the subsequent summer weeks, the number density of large particles drops further until a winter circulation regime with mean downwelling is reestablished around day number 270. This implies that our September ECOMA observations (day 251) were made just a few days/weeks before the winter circulation reestablished. This means they were made during a time when we may actually expect that the MSP volume density (caused by the number density of particles with radii larger than \( \sim 5 \) nm) is smaller than during the middle of summer. This hypothesis can actually be tested in comparison to actual wind observations at the location of our rocket launches for the year 2006, i.e., when ECOMA01 was launched. Figure 14 shows the seasonal variation of the zonal and meridional wind at the location of the Andøya Rocket Range obtained with the ALOMAR MF radar [e.g., Hoffmann et al., 2007]. Figure 14 indeed confirms our reasoning above about the timing of circulation changes. Hence, we conclude that the observed difference between ECOMA MSP volume density estimates (from the ECOMA flash currents; see section 2) in summer and at the beginning of September is in fact in agreement with the model predictions of Megner et al. [2008b]. This reinforces the interpretation put forward by Megner et al. [2008b] and Bardeen et al. [2008], namely that MSP variability over season is mainly driven by the prevailing circulation and its effect on MSP transport.

5. Conclusions

[50] In the current paper we have presented rocket-borne observations of MSPs from three different sounding rocket campaigns. These campaigns were conducted in September 2006, i.e., outside the polar summer season, and in August 2007 and June–July 2008, i.e., during the polar summer season. The core instrument considered here is a Faraday cup-type particle detector which is combined with a xenon flashlamp for the active photoionization of aerosol particles (or photodetachment of electrons from negatively charged particles) and subsequent detection of corresponding photoelectrons. It is shown that these photoelectron currents are strictly proportional to the total particle volume density of MSPs and that different MSP materials change the relation between volume density and current by a constant factor only. The measurements from September 2006 were compared to model simulations and show reasonable agreement below an altitude of \( \sim 85 \) km, while above that altitude contamination of the measured photoelectron current by NO and \( \text{O}_2(\text{I}^1\text{D}_g) \) appears to dominate. Measurements during polar summer show clear signatures of mesospheric ice layers as proven by simultaneous photometer observations on all rockets. Outside these mesospheric ice layers, however, the summer observations show a large degree of variability ranging from no particle signatures at all to currents that are even larger than during September 2006. We have then investigated whether this variability could be due to a similar variability in NO and/or \( \text{O}_2(\text{I}^1\text{D}_g) \), and were able to

Figure 13. Seasonal variation of the (top) zonal, (middle) meridional, and (bottom) vertical wind at a latitude of 70°N from the model study by Megner et al. [2008b].
exclude this possibility below an altitude of \( \sim 85 \text{ km} \) based on satellite observations with the ACE-FTS and SABER. We then considered the charging properties of MSPs and find supporting evidence both from modeling as well as from simultaneous electron density measurements that the observed variability is at least in part due to a similar variability in the charging state of the particles with corresponding effects on measured photoelectron currents. While MSPs need to be negatively charged in the absence of solar illumination, we showed that both photodetachment and photoionization from solar photons in the visible and UV are important for characterizing the charging state during polar summer. Assuming that MSPs consist of hematite or similarly strong absorbing species (which are required to explain the observed ECOMA photoelectron currents), our calculations show that MSPs should be positively charged in polar summer in the presence of low electron densities. This is positively confirmed on the basis of independent observations with an aerosol mass spectrometer which was launched 30 min before one of our rocket launches. In addition, cutoff altitudes of photoelectron currents recorded during summer all coincide with a lower electron density limit of about \( 1000 \text{ cm}^{-3} \). This is also the theoretically predicted lower electron density needed to allow the presence of a fraction of neutral MSPs which should then result in larger photoelectron currents. Finally, comparing the September measurements on the one hand, and the summer measurements with maximum photoelectron current (in the presence of largest electron densities) on the other hand, we find that MSPs signatures at altitudes of \( 80 \text{ km} \) are about a factor of 2 larger in Summer than during September.

[51] While this finding appears to contradict model predictions about a Summer depletion of MSPs larger than \( 1 \text{ nm} \) at first sight, we were then able to show that the models actually do predict a pattern of volume density variation as observed (recall that the ECOMA photoelectron current is proportional to volume density, see Figure 3). The underlying reason is that the volume density is dominated by even larger particles with radii larger than say \( 3 \text{ nm} \) whose number density is small, which, however, dominate the particle volume due to the \( r^2 \) dependence. The corresponding variation was explained as being the direct consequences of the seasonal variation of the mesospheric circulation in the model and we found that real wind observations obtained at the location of our rocket launches agree nicely with the qualitative pattern of the model winds. Hence, we conclude that our observations are indeed consistent with the model prediction that the seasonal variation of MSPs at polar latitudes is dominated by transport effects owing to the mean circulation. We note, however, that these results also imply that the volume densities of MSPs which are sensitive to particle volume such as our own measurements (and also the SOFIE observations) are obviously not the best experimental means to quantify the number density of condensation nuclei for mesospheric ice clouds in the polar summer mesopause region. While the SOFIE results of Hervig et al. [2009a] and our own observations do largely confirm the general model predictions by Megner et al. [2008b] and Bardeen et al. [2008], we note that additional measurements using different techniques are needed in order to yield more quantitative results on the availability of condensation nuclei.

[52] In addition, we see the following promising areas for future research: Time constants of MSP microphysics should be studied using the recently developed radar technique described in the paper by Strelnikova et al. [2007]. Corresponding measurements should be carried out over a diurnal cycle in order to see whether the diurnal variation of the meteoroid flux imposes a similar cycle of MSP properties or whether MSP properties are rather controlled by variations on much larger time scales (~10 days and longer).
as suggested by current microphysical models and supported by our current observations. While the initial attempts in that direction presented by Fentzke et al. [2009] for measurements at Arecibo (18.3°N, 66.8°E) are promising, we note that such observations should be carried out at polar latitudes using for example the new Poker Flat Incoherent Scatter Radar (PFISR) since variations of both the meteoroid flux as well as the residual circulation at those latitudes are expected to be largest. Concerning in situ measurements, new measurements with the ECOMA particle detector before, during, and after a meteor shower like the Geminids under otherwise identical geophysical conditions (local time and auroral activity) have the potential of characterizing MSP time constants. In addition, new measurements with a mass- and charge-resolving instrument like MASS should be repeated outside the polar Summer under conditions of daytime and nighttime and also for different ionospheric states. This will yield quantitative insight into the charging properties of MSPs. Of course, such measurements in the field should be accompanied by corresponding laboratory studies and theoretical considerations. Success or failure of any such attempt, however, critically depends on more knowledge about the composition of MSPs. Here every possible attempt should be made in order to solve this decade-old question of aeronomy.

Appendix A: Possible Contamination by NO and \( \text{O}_2(1\Delta_g) \)

[53] This study makes use of satellite-borne observations of NO with the Atmospheric Chemistry Experiment (ACE) on the Canadian SCISAT-1 satellite and the Sounding of the Atmosphere using Broadband Emission Radiometry (SABER) instrument on the NASA Thermosphere Ionosphere Mesosphere Energetics and Dynamics (TIMED) satellite.

[54] ACE is a Canadian satellite mission for remote sensing of the Earth’s atmosphere that was launched in August 2003. The main goal of ACE is to study the atmospheric chemistry and dynamics that affect stratospheric ozone depletion, but ACE measurements are also being used for many other areas of atmospheric science [Bernath et al., 2005]. The instrument which is of interest for this study is the ACE-FTS, a high spectral resolution infrared Fourier Transform Spectrometer (FTS). ACE-FTS operates at wave numbers from 750 to 4400 cm\(^{-1}\) in order to measure the vertical distribution of trace gases and temperature. During sunrise and sunset, the ACE-FTS measures sequences of atmospheric absorption spectra in the limb viewing geometry with different slant paths and tangent heights; when these spectra are analyzed, the results are inverted into vertical profiles of atmospheric constituents. For the purpose of this study, we have considered corresponding altitude profiles of nitric oxide. For details about ACE-FTS NO measurements see Kerzenmacher et al. [2008].

[55] The SABER instrument [Russell et al., 1999] was launched on the NASA TIMED satellite in December 2001. A fundamental goal of SABER is to observe comprehensively the ozone abundance and energy balance of the terrestrial mesosphere [Mlynczak, 1997]. One of the key measurements toward achieving this goal is the global observations of the \( \text{O}_2(1\Delta_g) \) airglow emission at 1.27 \( \mu \)m. 

The purpose of this study, \( \text{O}_2(1\Delta_g) \) number densities (data version 1.07) were directly derived from corresponding measurements of the volume emission rate using an Einstein coefficient for spontaneous emission of \( 2.23 \times 10^{-14} \) s\(^{-1}\). For further details we refer readers to the article by Mlynczak et al. [2007].

[56] We here consider whether the variability seen in the different ECOMA flights could be due to some unexpectedly strong contamination of our measurements due to photoionization of NO and/or \( \text{O}_2(1\Delta_g) \). Rapp and Strelnikova [2009] had shown that NO and \( \text{O}_2(1\Delta_g) \) are the only possible contaminants since all other conceivable molecules or ions either have a too large ionization potential or a too small photoemission cross section. These authors had excluded NO or \( \text{O}_2(1\Delta_g) \) as major contributors to the observed currents below altitudes of about 85 km using a climatological profile for NO from HALOE satellite measurements presented by Siskind et al. [1998] and a rocket-borne \( \text{O}_2(1\Delta_g) \) measurement from the study by Gumbel et al. [1998]. However, Rapp and Strelnikova [2009] did not take into account the variability of these species and did not attempt to use direct observations relevant for the time and location of their rocket launch. In this section, we will do exactly this using NO observations from the ACE-FTS on SCISAT-1 and \( \text{O}_2(1\Delta_g) \) observations from SABER on TIMED.

[57] Before we discuss actual data from these two satellite instruments, however, we clarify how large the NO or \( \text{O}_2(1\Delta_g) \) number densities would need to be in order to explain the observed photoelectron currents. For that purpose, measured photoelectron currents were converted to apparent NO or \( \text{O}_2(1\Delta_g) \) number densities on the basis of equation (1) but replacing the cross section for photoionization of NO and/or \( \text{O}_2(1\Delta_g) \) in equation (1) with the wave-length-dependent absorption cross sections of these two species from laboratory measurements, i.e., from Watanabe et al. [1953] for the case of NO and from Clark and Wayne...
[1970] for the case of $O_2(1\Delta_g)$. The result of this conversion is shown in Figure A1. This shows that the observed photoelectron currents below 90 km (i.e., at those altitudes where we expect to observe MSPs) would correspond to NO number densities between $10^8$ cm$^{-3}$ and $3 \times 10^9$ cm$^{-3}$ and to $O_2(1\Delta_g)$ number densities between $10^{10}$ cm$^{-3}$ and $2 \times 10^{12}$ cm$^{-3}$.

We next show NO number densities inferred from solar occultation measurements with the ACE-FTS in Figure A2 in the latitude band from 60°N–80°N in September 2006 (Figure A2, left) and July 2008 (Figure A2, right). Figure A2 reveals that the NO number density could indeed reach maximum values of up to $10^8$ cm$^{-3}$ at an altitude of 90 km. Between 60 and 90 km altitude, however, even maximum values do not exceed $2 \times 10^7$ cm$^{-3}$. These values are 1 to 2 orders of magnitude too small to account for the observed photoelectron currents at these altitudes.

Turning next to corresponding $O_2(1\Delta_g)$ number density profiles obtained with SABER shown in Figure A3, we see that corresponding number densities vary between $10^9$ cm$^{-3}$ and $2 \times 10^{10}$ cm$^{-3}$. Again, these values are between 1 to 2 orders of magnitude too small to account for the observed photoelectron currents exceeding the noise level of the instrument (corresponding to an $O_2(1\Delta_g)$ number density of about $10^{10}$ cm$^{-3}$) at altitudes between 60 and 90 km.

Based on these comparisons, we conclude that we may safely exclude a possible influence of NO and $O_2(1\Delta_g)$ on the observed currents below ~85 km. Hence, the observed currents cannot be explained by any other molecular species such that photoemission/photodetachment from MSPs (or ice particles above 80 km) is the only feasible explanation.

Acknowledgments. This work was supported by the German Space Agency (DLR) under grants 50 OE 0301 and 50 OE 0801 (Project ECOMA). The Norwegian part of the project was supported by the Norwegian Space Centre and the Research Council of Norway (grant 177295). The Austrian contribution was funded under grant 18560 of the Austrian Science Foundation (FWF). Excellent support of the rocket campaigns by the Mobile Rocket Base (Moraba) of DLR and the staff of the Andøya Rocket Range is gratefully appreciated. The Atmospheric Chemistry Experiment (ACE), also known as SCISAT, is a Canadian-led mission mainly supported by the Canadian Space Agency and the Natural
 Sciences and Engineering Research Council of Canada. The National Center for Atmospheric Research is operated by the University Corporation for Atmospheric Research under sponsorship of the National Science Foundation. Microwave Limb Sounder (MLS) data have been made available by the NASA Goddard Earth Science Data and Information Services Center. Finally, we greatly acknowledge the efforts of Heiner Asmus (University of Rostock) in carrying out important laboratory tests of the ECOMA instruments.

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