Experimental evidence for ice particle interaction with metal atoms at the high latitude summer mesopause region

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[1] Potassium number densities were measured in 2001–2003 with a resonance lidar in Spitzbergen (78°N). Typical number densities in the 85–100 km height range are 5–70 atoms/cm^3 with a maximum around 92 km in summer. Comparison of the seasonal and height variation of K number densities with similar measurements at mid latitudes (54°N) shows a remarkable reduction in the lower part of the layer during the summer months. This reduction strongly correlates with the appearance of ice particles detected as noctilucent clouds (NLC) and polar mesosphere summer echoes (PMSE) by the same lidar and by a VHF radar, respectively. In a total of 226 hours of simultaneous K and NLC observations, the upper edge of the NLC layer was always detected below the lower edge of the K layer (i.e., no overlap), even when both edges vary substantially with height and time. The ice particle effect on potassium seems to correlate with the ice particle size: 'large' particles (>10–20 nm, detectable as NLC) completely remove all available K atoms, whereas smaller particles (but still large enough to create PMSE) gradually reduce the number of K atoms. Our observations suggest that the loss of K atoms on ice is just in the right order of magnitude to compete with the major production and loss terms. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0340 Atmospheric Composition and Structure: Middle atmosphere—composition and chemistry. **Citation:** Lübken, F.-J., and J. Höffner (2004), Experimental evidence for ice particle interaction with metal atoms at the high latitude summer mesopause region, *Geophys. Res. Lett.*, *31*, L08103, doi:10.1029/2004GL019586.

1. Introduction

[2] The very low temperatures at the summer mesopause region lead to water ice particles that can be seen from the ground by the naked eye or can be detected by ground based lidars or by satellite borne instruments. In this paper we refer to these 'visible' ice particles as 'noctilucent clouds' (NLC) which require a minimum radius of approximately 10–20 nm to be detectable for lidars and which occur at ~82–85 km [*Gadsden and Schröder*, 1989; *von Cossart et al.*, 1999]. At roughly the same latitudes and season but to somewhat higher altitudes very strong radar backscatter signals are detected which are called 'polar mesosphere summer echoes' (PMSE). It has been speculated early after the first detection of PMSE that these echoes are directly linked to charged ice particles which affect the electron gas such that the diffusive destruction of small scale structures is reduced [*Cho et al.*, 1992]. A more quantitative analysis of turbulence generation of PMSE and plasma diffusion in the presence of charged ice particles has shown that a minimum ice particle radius of $r_A \sim 3$ nm is required to significantly affect the plasma and to create PMSE [*Rapp and Lübken*, 2003].

[3] Lidar measurements at the south pole have shown strong indications of a depletion of iron atoms at NLC altitudes (J. M. Plane et al., Removal of meteoric iron on polar mesospheric clouds, submitted to *Science*, 2004). Since the detection of NLC by a lidar is most sensitive to 'large' particles (see above) the effect of small ice particles cannot be detected by this technique.

[4] In this paper we present experimental evidence that mesospheric ice particles can significantly reduce the metal atom concentration, in our case potassium. We make use of simultaneous and common-volume measurements of K number densities and NLC by our K lidar, and PMSE by a VHF radar. These measurements took place in the years 2001–2003 in the polar cap on the archipelago Svalbard close to Longyearbyen (78°N).

2. Observations of Potassium, NLC, and PMSE

[5] Potassium number densities, noctilucent clouds, and atmospheric temperatures are observed by the mobile potassium lidar of the Leibniz-Institute of Atmospheric Physics in Kühlungsborn [*von Zahn and Höffner*, 1996]. The lidar container was transported to Svalbard in May 2001 and observations were performed from time to time until autumn 2003. In this paper we concentrate on measurements of potassium number densities and noctilucent clouds. Raw potassium densities were multiplied by a factor of 3 to take into account saturation effects (only relevant for daylight measurements). The uncertainty in this factor is approximately 50%. It is important to note, that the same correction factor was applied for all profiles, i.e., the correction affects the absolute number densities only, not the relative changes in height and season. NLC are detected as an enhanced signal (relative to the background noise and the air molecule signal) that does not vary when the laser frequency is tuned over the potassium resonance line. More details and first results on the statistics of NLC observations are published in *Höffner et al.* [2003]. For the summer season, measurements are available from 12 June until 5 October 2001, and 31 March until 21 August 2003. It should be noted that the ability to separate a NLC from the K layer depends on the strength of the NLC relative to the potassium signal. Generally, a strong K signal reduces the sensitivity to

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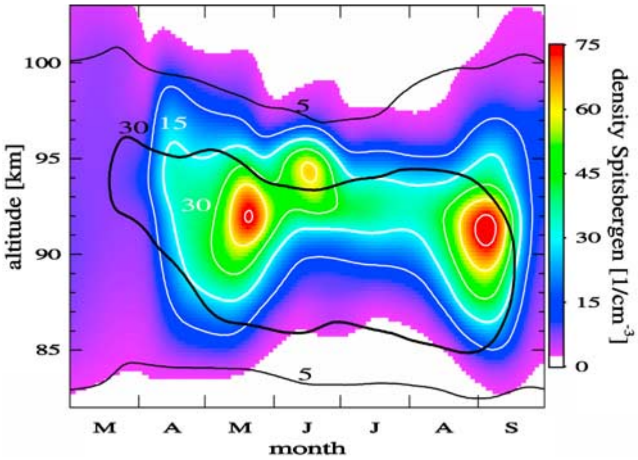

Figure 1. Potassium number densities at 78°N as a function of altitude and season (color contour and white isolines). For comparison the K densities from Kühlungsborn (54°N) are shown as black isolines.

detect a NLC, and vice versa. For example, a conservative estimate shows that within an integration time of 2 minutes and an altitude bin of 200 m a NLC with a volume backscatter coefficient of $1 \cdot 10^{-10}$/m/sr can be detected in a K layer with a density of up to $\sim$7/cm^3. Such a NLC corresponds to ice particles with a radius of larger then $\sim$12 nm and number densities of $\sim$1000/cm^3.

[6] Potassium number densities as a function of season and height are shown in Figure 1. The instrumental sensitivity is appr. 0.5 atoms/cm^3 during daylight conditions (much better in darkness). Maximum number densities of $\sim$90 atoms/cm^3 are observed in the $\sim$90–95 km height range from April to September. We note that this seasonal variation of [K] with a maximum in summer is different from sodium and is not yet understood.

[7] The K lidar observed NLC from approximately mid June until mid August at altitudes of $\sim$81–87 km (the first and last dates of NLC in the combined data set from 2001 and 2003 are 11 June and 21 August). The combined occurrence rate (2001 and 2003) is 74% at $\sim$82–85 km with very little inter-annual variability. In the first year of NLC observations (2001) the mean NLC peak altitude was 83.6 km (variability: 1.1 km) which did not vary significantly with season. The average top and bottom altitude of the NLC layer was 85.1 and 82.5 km, respectively (see *Höffner et al.* [2003] for more details). A first analysis of the 2003 season shows very similar mean values.

[8] The SOUSY Svalbard Radar is an MST VHF radar installed very close to Longyearbyen by the Max-Planck-Institut für Aeronomie in 1998. It has been operated routinely on a campaign basis since summer 1999. First results regarding the seasonal and height variation of PMSE and comparison with in-situ measurements of temperatures were published recently [*Rüster et al.*, 2001; *Lübken et al.*, 2004]. PMSE are present practically permanently at this latitude in the main summer season and occur between $\sim$81–92 km with decreasing occurrence frequencies at the edges (see Figure 2 in *Lübken et al.* [2004]). The first and last occurrence of PMSE in 2001 was on 22 May and 27 August, respectively.

3. Simultaneous Observations

[9] In Figure 2 we show simultaneous observations of K number densities, NLC, and PMSE on July 31, 2001. It is important to note that the NLC layer is located below the potassium layer which is a very typical result. In a total of 226 hours of K and NLC observations (2001 + 2003) we have always observed the upper edge of the NLC layer below the lower edge of the K layer (i.e., no overlap), even when both edges vary substantially with height and time as for example in Figure 2. We note that the lidar NLC signal strength varies drastically with the ice particle radius. As mentioned before our lower detection limit is approximately $r_A \sim 12$ nm. The fact that there are indeed smaller ice particles present above the NLC layer is demonstrated in Figure 2 by the presence of PMSE up to 90–92 km. The characteristics of the NLC and PMSE layers in this figure are typical: the PMSE extends to higher altitudes compared to NLC whereas the lower edges of both layers coincide nearly perfectly.

[10] The upper edge of the PMSE layer occasionally shows strong correlation with a decrease in K number density (see e.g., around 2–5 UT in Figure 2). This could indicate an interaction of ice particles with K atoms. This interaction is obviously not always dominating compared to other sources and sinks, since there are still K atoms present in the vicinity of PMSE and, even more so, there are times of no correlation between PMSE and a substantial reduction in [K] (see, for example, around 10–11 UT in Figure 2).

[11] In Figure 3 individual altitude profiles of potassium, NLC, and PMSE at 5:09 UT on July 31 are shown. The profiles are integrated over a time period of 2 minutes and have been slightly smoothed. In this case there is a clear ledge in the K profile at the top of the PMSE profile

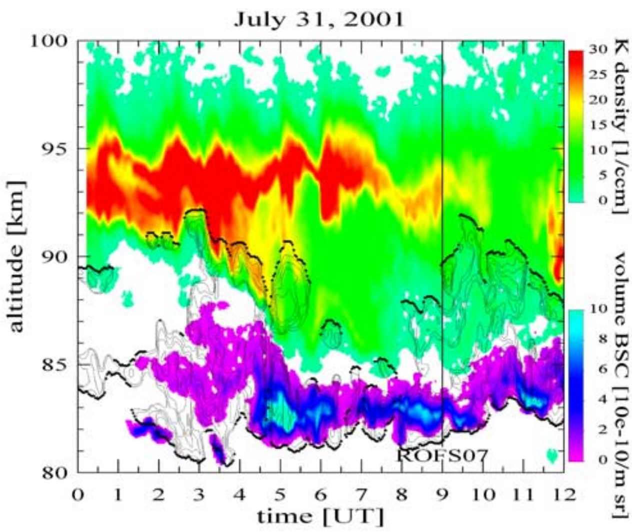

Figure 2. Potassium number densites on July 31, 2001 (color scale from green to red), and simultaneous observations of NLC (color scale from violet to blue) and PMSE (black isolines). The upper and lower edge of the PMSE have been highlighted to facilitate intercomparison. The vertical line labeled ROFS07 indicates the launch of a meteorological rocket (see *Lübken et al.* [2004]).

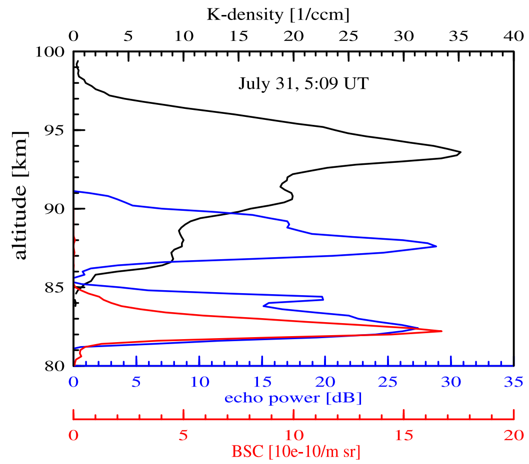

Figure 3. Altitude profiles of potassium (black), NLC (red), and PMSE (blue) measured at Spitsbergen on 31 July 2001 at 5:09 ± 1 min.

(~91 km) which is presumably caused by the PMSE particles. We note that such a ledge is not always observed, i.e. the influence of 'small' ice particles (PMSE type) may not always be large enough to be detectable in individual profiles. As mentioned earlier, no K atoms are present at NLC altitudes, and the lower edges of NLC and PMSE agree nearly perfectly.

4. Discussion and Conclusion

[12] The depletion of potassium atoms by interaction with ice particles may be difficult to detect in single cases, in particular if the ice particles are very small (typical for PMSE). However, their effect on the mean K density distribution is apparent when comparing the seasonal and height variation of K at 78° with mid latitudes (54°N) where NLC and (P)MSE are very seldom (see Figure 1). Whereas the lower edge of the K layer is nearly constant in time at 54° during summer, it rises by several kilometers at 78°N and leaves a region of depletion from approximately 85–93 km from mid May until end of August. This depletion is also very evident in the seasonal variation at, for example, 91 km: at mid and low latitudes (Kühlungsborn and Arecibo) the K density is nearly constant in time from beginning of May until end of August whereas at Spitzbergen there is a substantial decrease in June and July (see Figure 1) [*Eska et al.*, 1999; *Friedman et al.*, 2002].

[13] From the comparison of K densities at Spitzbergen and Kühlungsborn alone we cannot exclude that the variation of the K layer in summer is caused by natural variation other than by the influence of ice particles. This interaction is evident, however, when comparing the K density distribution with the seasonal and height variation of NLC and PMSE (Figure 4). There is a significant reduction of the Spitsbergen K number densities at times and altitudes where PSME is present and potassium atoms are completely removed at NLC heights. The PMSE and potassium density isolines in Figure 4 nearly perfectly overlap from beginning of June until the end of August. The lower edge of the K layer deviates from a constant height exactly in the period of PMSE appearance.

[14] It is interesting to compare the appearance of NLC and PMSE in Figure 4 which can largely be explained by the different particle size dependence (NLC: $r_A > 12$ nm; PMSE: $r_A > 3$ nm). The particles fall and grow until they are large enough to be seen as NLC, but they influence the plasma already in their early stage. We explain the early occurrence of PMSE (in mid May) without NLC by the seasonal variation of water vapor: assuming that the maximum of H_2O concentration at 78°N appears in mid August (similar to 69°N [*Seele and Hartogh*, 1999]) implies that the amount of water vapor available in mid May is comparatively small, i.e. the ice particles cannot grow large enough to be detectable as NLC.

[15] We note that the seasonal and height variation of PMSE strongly correlates with low temperatures, more precisely with super-saturation conditions (water vapor concentration plays a minor role) as has been shown by a comparison of in-situ temperatures measurements and PMSE [*Lübken et al.*, 2004].

[16] Our measurements strongly indicate an interaction of ice particles with metal atoms and a dependence of this interaction on the ice particle size. K atoms seem to disappear completely when particles are 'large' (i.e., large enough to be detectable as NLC), whereas their number density is reduced when ice particles are 'small' (but still large enough to create PMSE). According to measurements and microphysical modeling other relevant parameters, such as atmospheric temperature and ice particle number density, don't vary substantially in the height range of interest.

[17] We note that measurements at other latitudes and seasons suggest that the K layer is fading below 85 km which makes it difficult to estimate the undisturbed distribution and to quantify the removal of potassium atoms by ice particles. Furthermore, the lidar NLC signal is roughly proportional to the volume of the ice particles whereas the uptake of metal atoms is presumably proportional to the surface which is very difficult to determine (see below).

[18] The observed potassium number density is a consequence of a balance between production and loss. Our results suggest that this balance is disturbed but not completely dominated by the presence of ice particles. This implies that the loss of K atoms on ice is just in the right order of magnitude to compete with the major production and loss terms, for example with meteoric influx which is on the order of $10^{-4}/(\mathrm{cm}^3 \cdot \mathrm{s})$ (see *Eska et al.* [1999] for measurements and modeling of the K layer).

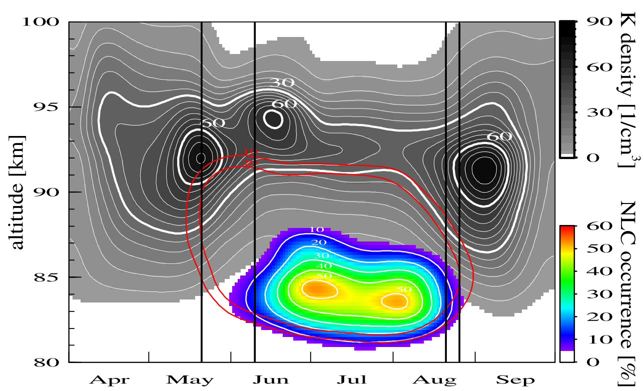

Figure 4. Seasonal and height variation of potassium (grey contours; same as Figure 1) compared with the occurrence of PMSE (red isolines) and NLC (color contours). The vertical lines indicate the first and last appearance of PMSE and NLC, respectively (see text for exact dates).

[19] The number of K atoms which disappear per unit volume and time due to uptake on ice is given by

$$\frac{1}{4} \cdot \bar{c} \cdot \gamma \cdot ([ice] \cdot F) \cdot [K], \qquad (1)$$

where γ is the uptake coefficient, $\bar{c}$ is the mean velocity of K atoms (~400 m/s), A = ([ice] · F) is the volumetric surface area of ice ([ice] is the number density and F the mean surface area of ice), and [K] is the potassium number density. As a rough estimate we take $\gamma = 0.1$ (B. Murray, personal communication, 2003), A = 10^{-8} cm^2/cm^3 [*von Zahn and Berger*, 2003], and [K] = $10/cm^3$. This leads to an uptake rate of $10^{-4}/(cm^3 \cdot s)$ which is comparable with the meteoric production rate. We note, however, that this estimate is very uncertain, since some of the parameters in equation (1) are only poorly known. For example, the ice particles are most likely not cubic, i.e. their surface is probably much larger than that of the volume equivalent cube. Furthermore, the value for γ stated above is a sensible lower limit but, if similar to iron, could be as large as $\gamma = 1$ [*Murray and Plane*, 2003]. A complete understanding of the effect of ice particles on potassium atoms (and metals in general) can only be achieved if these uncertainties are solved and if the mean and undisturbed background distribution is understood. This certainly requires more laboratory and field measurements and comprehensive modeling. An important consequence of our observation is that the reduction of metal atoms by ice particles limits the capability of resonance lidars to measure temperatures within PMSE and NLC.

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