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Sporadic Ca and Ca^+ layers at mid-latitudes: Simultaneous observations and implications for their formation

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Abstract. We report on the observations of 188 sporadic layers of either Ca atoms and/or Ca ions that we have observed during 112 nights of lidar soundings of Ca, and 58 nights of Ca^+ soundings, at Kühlungsborn, Germany (54°N, 12°E). The Ca^+ soundings have been performed simultaneously and in a common volume with the Ca soundings by two separate lidars. Correlations between sporadic neutral and ionized metal layers are demonstrated through four case studies. A systematic study of the variations of occurrence of sporadic Ca and Ca^+ layers reveals that neutral and ionized Ca layers are not as closely correlated as expected earlier: (a) The altitude distribution shows the simultaneous occurrence of both sporadic Ca and Ca^+ layers to be most likely only in the narrow altitude range between 90 and 95 km. Above that region, in the lower thermosphere, the sporadic ion layers are much more frequent than atom layers. Below 90 km only very few sporadic layers have been observed; (b) The seasonal variation of sporadic Ca layers exhibits a minimum of occurrence in summer, while sporadic Ca^+ layers do not show a significant seasonal variation (only the dense Ca^+ layers appear to have a maximum in summer). At mid-latitudes sporadic Ca layers are more frequent than sporadic layers of other atmospheric metals like Na or K. For the explanation of our observations new formation mechanisms are discussed.

Key words. Ionosphere (ion chemistry and composition; ionosphere-atmosphere interactions; mid-latitude ionosphere)

1 Introduction

Metal atoms reside in the upper atmosphere in two types of layers: (1) In permanent, broad “regular” layers covering the altitude range 80 to 110 km and (2) in transient, narrow “irregular” layers occuring almost exclusively above 90 km altitude. Even the very first lidar observation of an atmospheric metal layer by Bowman et al. (1969) exhibited an irregular Na profile. Thereafter, the lidar experiments of Gibson and Sandford (1971) yielded an irregular second maximum in their Na profile. Seven years later, Clemesha et al. (1978) reported their observation of a strong irregular Na layer which had a FWHM of 2.5 km. But the irregular layers remained of minor interest for almost two decades until the observations of von Zahn et al. (1987, 1988) and von Zahn and Hansen (1988) concerning irregular layers observed at high latitudes which those authors called “sudden” sodium layers. Thereafter, many publications similar to Kwon et al. (1988) or Beatty et al. (1988) appeared in quick succession which dealt with observations of irregular layers at high, middle, and low latitudes. It became common use to call them “sporadic” metal layers, a nomenclature which we will use here too. In his review paper, Clemesha (1995) summarized already more than 40 publications dealing with sporadic metal layers.

Clemesha (1995) concluded in his review that a detailed explanation for the formation of sporadic metal layers, their occurrence, their geographical distribution, and their high variability cannot be given yet. Still, a strong link between the neutral and the ionized sporadic metal layers is generally assumed (e.g. Alpers et al., 1993; Qian et al., 1998). For the case of Ca atoms and Ca^+ ions, this link can be studied time-resolved by ground-based lidar observations. Initial simultaneous and common-volume Ca and Ca^+ observations have been performed by Alpers et al. (1996). They found various examples of coupled and independent sporadic ion and neutral layers during summer.

In 1997/1998 we performed during 58 nights simultaneous soundings of the Ca and Ca^+ layers with two ground-based lidars at the Leibniz-Institute of Atmospheric Physics (IAP), Kühlungsborn, Germany (54°N, 12°E). By using the same telescope the observed atmospheric volumes are exactly the same for Ca and Ca^+ soundings. In addition, 54 nights of Ca-

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only observations improve the statistics on the regular and sporadic Ca layers. Gerding et al. (2000) have presented a description of the lidars in use and of their results concerning the regular Ca and Ca^+ layers. Here we report on the almost 200 sporadic layers observed during the same observation period. We give examples for sporadic neutral and ionized Ca layers and demonstrate their high variability in altitude, density, and shape. Based on the complete data set we show variations of sporadic layer occurrence with altitude and season for both Ca and Ca^+. Finally, the implications of our observations for the formation of sporadic neutral and ionized metal layers are discussed.

2 Observations of neutral and ionized sporadic metal layers: Four case studies

Sporadic layers are thin layers of metal atoms and/or ions in the altitude range 90 to 130 km. Their altitude extension (FWHM) is typically between 2 and 3 km and always less than 5 km. Their occurrence is transient ("sporadic") and independent of the regular metal layers. The peak density and altitude of sporadic layers may change with a time scale of minutes. Some authors have used the observed rate of number density change ("growth rate") as a criterion for sporadic metal layers. Stationary lidars like ours cannot tell us, however, whether the observed temporal density changes are caused by local chemical processes or the transport of an inhomogeneous layer through the lidar field-of-view. Thus, here we define sporadic layers only by their altitude extension, having a FWHM of less than 5 km. Sporadic layers are marked with a subscript "s" (Ca_s, Ca_s^+), while the regular layers have no subscript. The following four case studies give examples for the degree of coupling between sporadic Ca and Ca^+ layers, and their temporal variations in altitude and density.

2.1 The night April 7, 1997: A case of slightly coupled Ca_s^+ and Ca_s layers

Figure 1 shows the temporal variations of the Ca^+ (upper panel) and Ca (lower panel) layers during the night of April 7, 1997. Each individual profile is the integrated backscatter signal of 2000 laser pulses (about 2:10 min of observations). The profiles are smoothed by a 1.8 km wide running Hanning filter and averaged by a running mean of three profiles. Gaps in our lidar soundings are caused either by clouds or by changes of the dyes used in the lasers. Here, as in following figures, the absolute density scales are given within the panels. We point out that the four case studies are representative by the fact that the most sporadic Ca layers show much higher ion abundance than atom abundance.

Strong sporadic Ca^+ and Ca layers existed already when we started the record of density profiles at 19:30 and about 20:00 UT, respectively. Both the Ca_s and Ca_s^+ layers were observed near 95 km altitude and both increased in density. A second, much weaker ionized Ca_s^+ layer formed later that night between about 100 and 105 km altitude. Below the sporadic Ca_s layer, the regular Ca layer above 85 km is visible. This Ca layer is very weak and reaches a maximum number density of only 8 cm^{-3}.

The strong sporadic layers in the 90 to 95 km altitude range totally dominate the layer picture during this night. The altitudes of the peaks of the Ca_s^+ and Ca layers track each other roughly for more than 3 hours, but not perfectly, indicating a lack of strong coupling between the two layers. Figure 2 gives details of the altitude changes of the sporadic layer maxima. At the beginning of the night, both Ca_s and Ca_s^+ are in fact double-peaked layers with the upper peak of the ion layer being stronger than the lower peak, and vice versa for the neutral maxima. (Between 19:45 and 20:00 UT only qualitative information on Ca^+ layer shape has been obtained, so these profiles are neglected in Fig. 1.) The minor maxima decreases in density and disappears around 20 UT. From then on the main Ca_s^+ layer is found 1–2 km above the peak of the Ca_s layer until about 20:30 UT when the Ca_s^+ peak falls on top of the Ca_s peak. Thirty minutes later, when clouds interrupted our observations, the ionized layer has crossed the neutral one and was observed about one km lower than the Ca_s. After 21:45 UT, the temporal variations of layer altitude match each other quite well, but now with a persistent gap of about 1.5 km.

2.2 The night August 1/2, 1997: A case of a high altitude Ca layer without a Ca^+ companion

During the night of August 1/2, 1997 (Fig. 3), we observed between 84 and 91 km altitude a typical narrow regular summer Ca layer, similar to the ones reported by Granier et al. (1989), Alpers et al. (1996), and Gerding et al. (2000). Throughout the observation, its lower ledge ascended wave-like from 83 to 86 km.

The outstanding feature of this example is a second Ca layer between 108 and 120 km altitude which is as dense as the regular layer at the beginning of the night. Its FWHM of at least 6 km disqualifies this layer to be called a "sporadic" layer. Even though the peak number density of this upper layer decreased during the night, its column density changes rather little. The layer remained significantly above the noise level of the lidar until the end of the night (compare the signal level at 104 km with background signal above 120 km). We found such dense high layers in 7 out of 112 nights of Ca observations and weaker high altitude layers on 12 additional nights. In the example of Fig. 3, the high altitude Ca layer persists without any accompanying Ca^+ layer. For the entire night, the Ca^+ concentration was found to be below 1 cm^{-3} at the altitude of the high Ca layer. For further discussion of these high neutral layers, see below.

The altitude development of the neutral and ionized sporadic layers is shown in Fig. 4. At 21:55 UT, a narrow and hence sporadic Ca_s was formed at about 107 km altitude and just below the high Ca layer. Subsequently, this Ca_s descended with a rate of about 2.5 km/h, reaching 97 km altitude at 1:55 UT. With regards to Ca_s^+ layers, a strong one

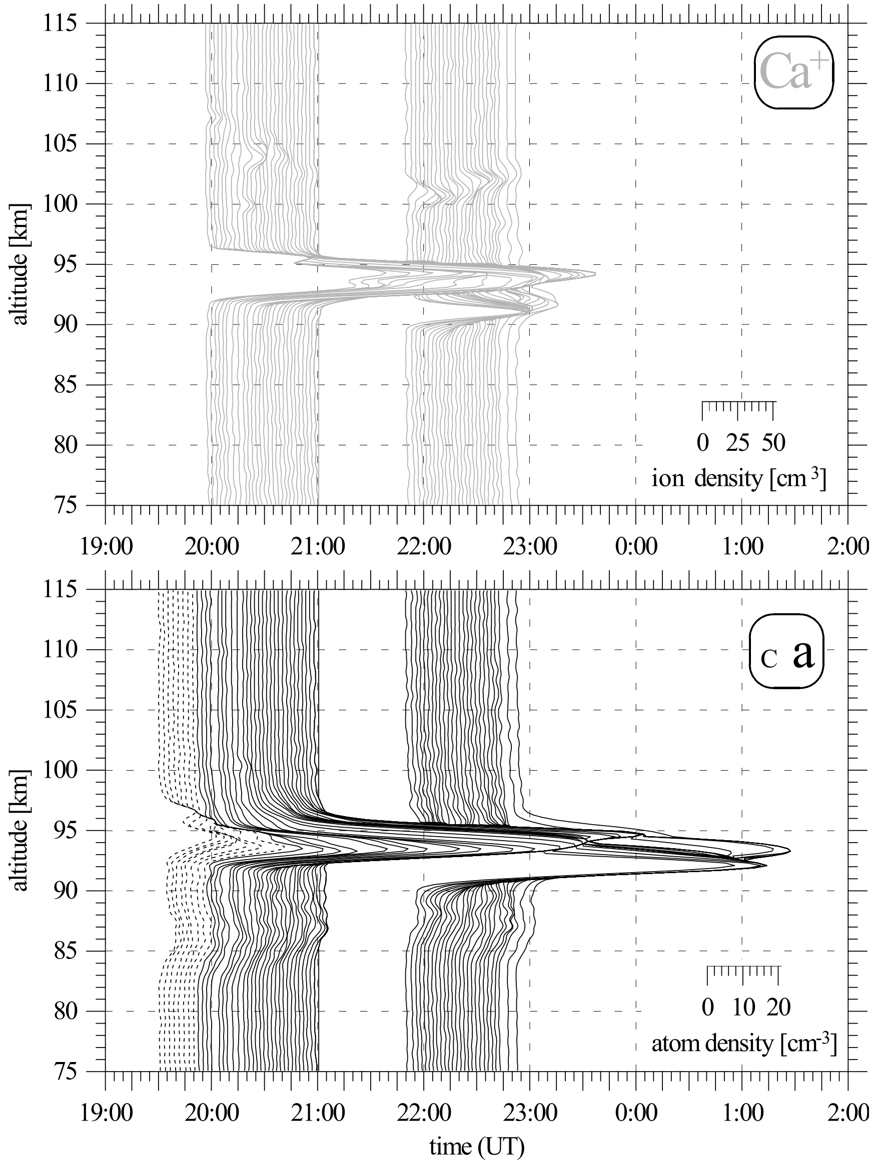

Fig. 1. Series of density profiles obtained in the evening of April 7, 1997 for Ca^+ (upper panel) and Ca (lower panel). The Ca^+ profiles show a strong layer around 94 km and weak layers above 100 km altitude. The Ca profiles show a strong sporadic layer around 94 km only. Note the different density scales in the two panels.

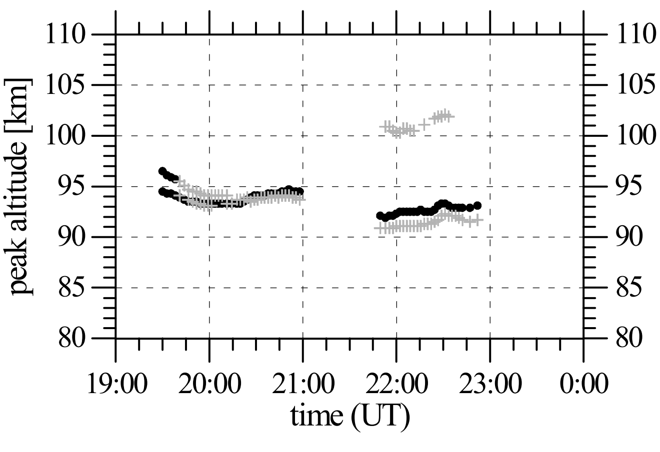

Fig. 2. Altitude variation of the sporadic Ca_s (dots) and Ca_s^+ (crosses) around 95 km in the evening of April 7, 1997.

was present already at the beginning of the night in the 100–105 km altitude range with concentrations of up to 500 cm^{-3}. At about 22:20 UT this Ca_s^+ split into a double layer with maxima separated by up to 3 km. At 23:30 UT the lower maximum had disappeared and the upper maximum descended in altitude at about 2.5 km/h.

2.3 The night March 4/5, 1997: A case of near-simultaneous occurrences of decoupled and of coupled Ca_s^+ and Ca_s layers

During the night March 4/5, 1997, a number of Ca_s^+ and Ca_s layers were observed over a wide range of altitudes (Fig. 5). During the first two hours of the soundings, a weak Ca_s^+ layer was observed between 90 and 95 km. In addition, at about 19:00 UT, a Ca_s^+ developed at about 110 km altitude, exhibiting rapid changes in both altitude and density. This layer had no corresponding Ca_s layer and disappeared at about 20:30

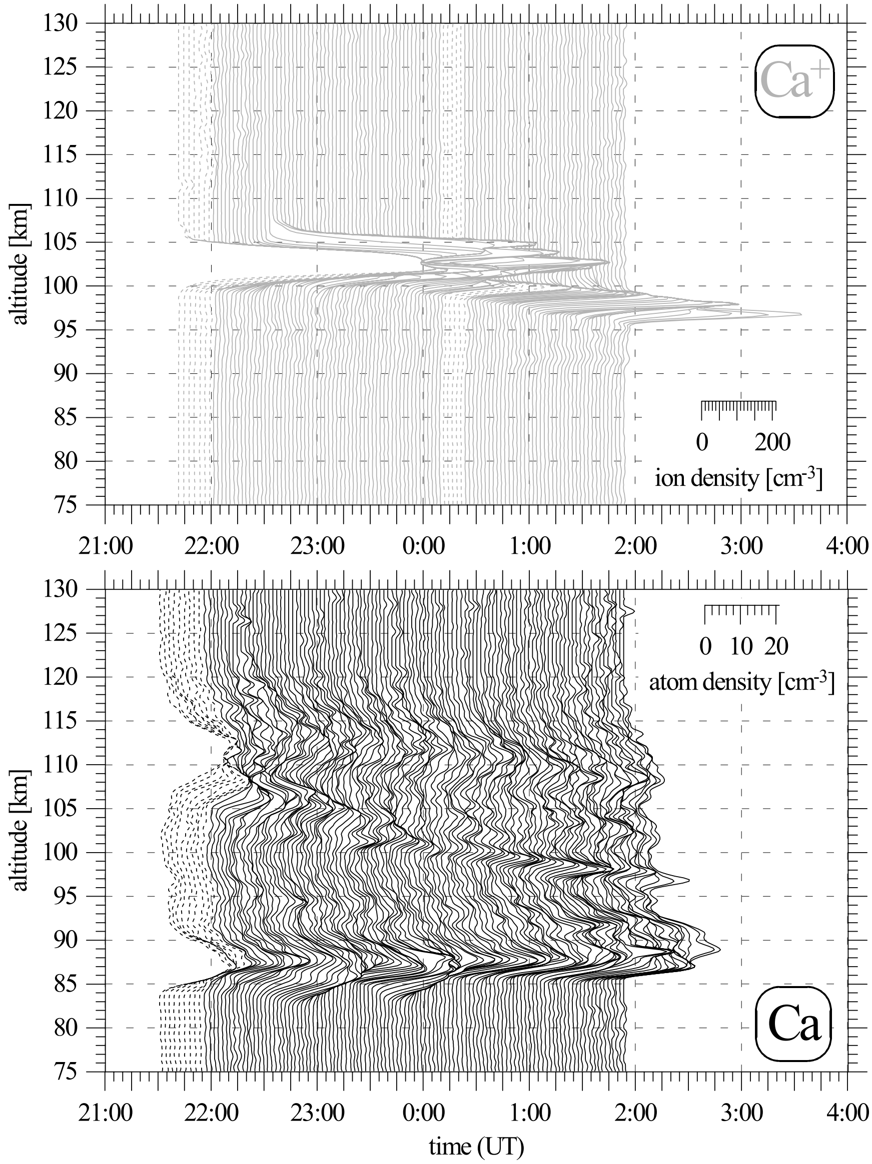

Fig. 3. Series of density profiles obtained during the night August 1/2, 1997 for Ca^+ (upper panel) and Ca (lower panel). Note the very different density scales for the ionized and neutral Ca profiles.

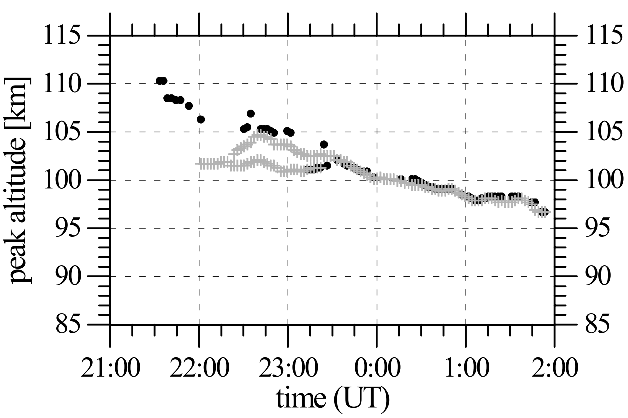

Fig. 4. Altitude variation of the Ca_s (dots) and Ca_s^+ (crosses) during the night August 1/2, 1997.

UT. After 21:00 UT, another Ca_s^+ appeared at 102 km altitude. It started to descend about ten minutes after appearing, reached 92 km at 00:30 UT and ascended thereafter, to 95 km, where it hovered for about an hour at the end of the night.

The regular Ca layer was observed at the beginning of the night as a broad layer with a maximum between 90 and 95 km. There were several small, short-time density peaks indicating a highly variable layer profile. With 10 to 15 cm^{-3}, the Ca atom density is about a factor of 2–3 lower than the ion density at the same altitude. About 23:00 UT, a Ca_s peak was observed for a few minutes at about 99 km, located a few kilometers above the strengthening Ca_s^+. Starting at about 23:10 UT, a high neutral Ca_s developed at 106 km, which descended to 103 km. During the whole 3 h presence of this Ca_s layer, no Ca^+ ions were detected by our lidar within this atmospheric volume.

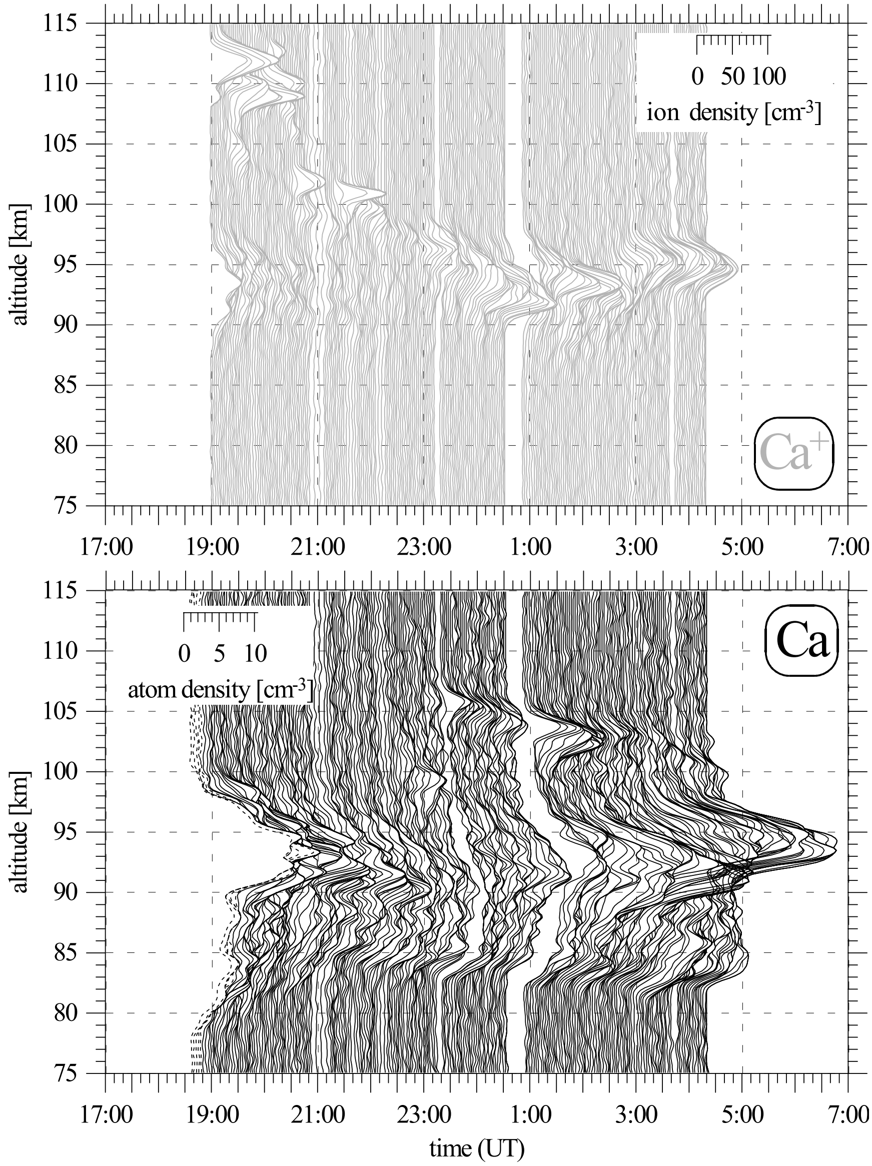

Fig. 5. Series of density profiles obtained during the night March 4/5, 1997 for Ca^+ (upper panel) and Ca (lower panel). Various simultaneous and single Ca^+ or Ca sporadic layers are visible and will be identified in the text.

As outlined above, extended Ca_s layers occured without accompanying Ca_s^+ layers and vice versa during this night. Nevertheless, highly correlated layers also developed. Strong similarities in layer heights persisted past 1:30 UT: The Ca_s^+ between 93 and 96 coincided with an increase in neutral Ca density in the same altitude with larger layer width between 3 and 7 km.

It should be noted here that two nights later on March 6/7, 1997 a strong meteor shower was observed by the IAP Ca and Ca^+ lidars, with sporadic ion layers occuring additionally (Gerding et al., 1999). But for the night of March 4/5, the number of lidar meteor trails does not exceed the typical background level, as reported by the same publication. Due to this normal meteor rate and the small amount of metal deposited by a single meteoroid, a connection between sporadic layers in these nights and ablating meteoroids can be negated.

2.4 The night June 6/7, 1997: A case of decoupled Ca^+ and Ca layers

The night of June 6/7, 1997 may serve as a dramatic example for the possibility of a simultaneous existence of totally decoupled Ca atom and Ca ion layers. The Ca^+ ion profiles showed strong sporadic layer activity above 105 km altitude from the beginning of the observations until the end (Fig. 6). One of the Ca_s^+ layers reached the unusually high altitude of 130 km, while another one reached a maximum density of 800 cm^{-3} at 110 km altitude. The fast changes in sporadic layer heights and densities give reason to assume the presence of strong electric fields shifting and collecting the ions from a bigger atmospheric volume (see e.g. Kirkwood and von Zahn (1991)). In addition to the strong Ca_s^+ layers above 105 km, a weak and broadly distributed Ca^+ layer was observed between 87 and 100 km, reaching maximum densities

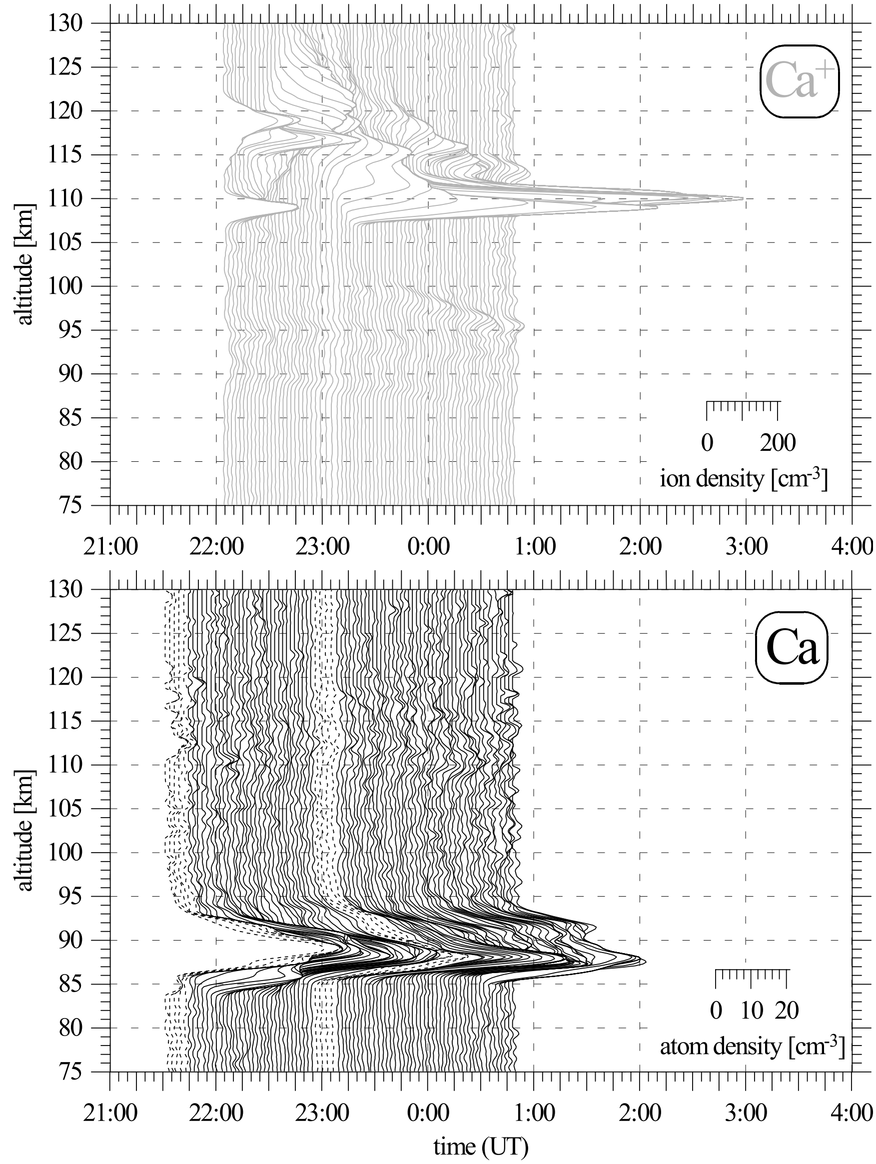

Fig. 6. Series of density profiles obtained during the night June 6/7, 1997 for Ca^+ (upper panel) and Ca (lower panel).

of about 25 cm^{-3} below 95 km and 40 cm^{-3} above 95 km.

Within the neutral Ca profiles, no signs of sporadic layer activity were found. We observed only the regular layer between 85 and 94 km with a density of up to 40 cm^{-3}. During the time of the strongest Ca_s^+ layer (around 1 UT), a slightly increased Ca count rate was found above 109 km, indicating a Ca density of about 3 cm^{-3}.

3 Altitude distribution of neutral and ionized sporadic Ca layers

The previous case studies show that simultaneous sporadic layers in Ca^+ and Ca can be observed both at the same altitude and well separated by many kilometers. Hence, it is desirable to examine quantitatively the distribution of Ca_s^+ and Ca_s with altitude and season. In the following, we will present first the altitude distribution of simultaneous and single-species sporadic layers and then their seasonal distribution.

For the examination of the altitude distribution, all sporadic layers are sorted into 5-km-bins. Descending and ascending sporadic layers are assigned to the bin where they have been observed for the longest period. The full data set contains 89 sporadic Ca layers (from 112 nights) and 99 sporadic Ca^+ layers (from 58 nights). Table 1 gives the altitude-bins in the first column and the number of Ca_s^+ and Ca_s in columns 2 and 3, respectively. The distribution is plotted in Fig. 7. There is a prominent maximum of Ca_s rate between 90 and 95 km and a rapidly decreasing rate higher up. Furthermore, only about 10% of all Ca_s have been observed below 90 km. For the ionized sporadic layers we found a nearly constant occurrence rate between 90 and 105 km with about 25% of all Ca_s^+ in each 5-km-bin. Only 1 out of 99 Ca_s^+ layers has been observed below 90 km.

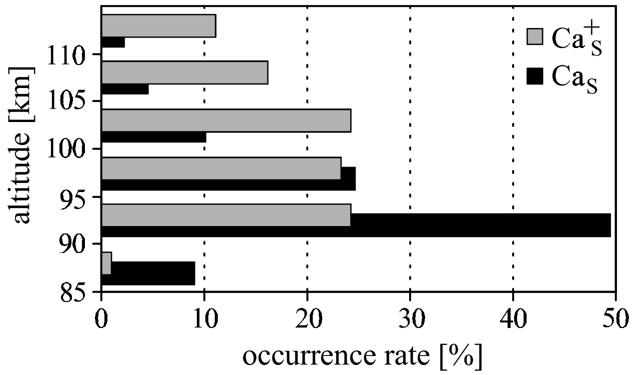

Fig. 7. Altitude distribution of sporadic Ca and Ca$^+$ layers as listed in Table 1 (columns 2 and 3).

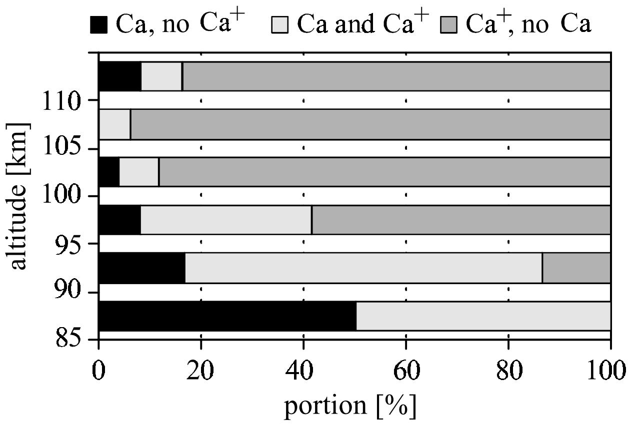

Fig. 8. Altitude distribution of joint and single-species sporadic Ca and Ca$^+$ layers as listed in Table 1 (columns 4 to 6).

The 58 nights of simultaneous, common-volume Ca and Ca$^+$ observations give an overview of the distribution of simultaneous Ca$_s$/Ca$_s^+$ and of single-species sporadic layers (Table 1, columns 4 to 6). The result is plotted in Fig. 8. The proportion of Ca-only sporadic layers without Ca$_s^+$ decreases with altitude. Joint Ca$_s$/Ca$_s^+$ dominate between 90 and 95 km altitude (70%), but become less frequent with increasing altitude. Even above 95 km the Ca$^+$-only sporadic layers become most probable, with a proportion of 90% or more above 100 km. Neutral Ca$_s$ without Ca$_s^+$ have been observed only twice above 100 km. This reflects the low concentration of Ca at these altitudes (Gerding et al., 2000).

4 Seasonal variation of sporadic layer occurrence at Kühlungsborn

For the calculation of the seasonal variation of sporadic layer occurrence, the periods of lidar observations and those of sporadic layer occurrences were integrated separately for each month. If two or more sporadic layers have been observed in a single profile, the occurrence times for all sporadic layers

Table 1. Altitude distribution of Ca$_s$ and Ca$_s^+$. The total numbers N per bin (columns 2 and 3) are summarized for all observations of this species, the numbers in columns 4 to 6 only for nights with simultaneous observations

Altitude [km]	N(Ca$_s$)	N(Ca$_s^+$)	N(Ca$_s$ and Ca$_s^+$)	N(Ca$_s$ without Ca$_s^+$)	N(Ca$_s^+$ without Ca$_s$)
> 110	2	11	1	1	10
105–110	4	16	1	0	15
100–105	9	24	2	1	22
95–100	22	23	8	2	14
90–95	44	24	21	5	4
85–90	8	1	1	1	0
Total	89	99	34	10	65

have been added. Thus, the "occurrence index", defined as the quotient of the occurrence period and observation time, can be bigger than 100%. The calculation is not weighted with the strength of a layer.

For the Ca$_s$ layers, the occurrence index is lowest in summer (about 10%) and increases until early winter (see Fig. 9 and Table 2). For October and November, indexes of more than 60% have been observed, while in December, the occurrence index reaches a maximum of more than 95%. Even though the December value is very high, the January value becomes one of the lowest monthly indexes. The low values in January and summer indicate a semi-annual variation, although somewhat perturbed by the high December occurrence index.

The low numbers of Ca$_s$ layers in summer coincide with the observation of so-called "high layers" above 100 km, which are only observable between May and September. A high Ca layer is defined here by either (a) a night mean number density of more than 1 cm^{-3} at 105 km altitude with a density minimum between the high layer and the permanent layer or (b) a night mean number density of more than 1 cm^{-3} at 110 km altitude or above. An example of a high layer is shown in Fig. 3. The percentage of nights with high layers is also indicated in Fig. 9. The most prominent feature of the occurrence of such layers is the annual variation with a maximum in summer. In addition, high layers have been observed with number densities of more than 5 cm^{-3} above 105 km. These intense high layers also show a distinct maximum in summer.

Due to the anti-correlation of the occurrence of Ca$_s$ and high Ca layers in summer, the seasonal variation of the Ca$_s$ occurrence has been examined for different altitudes. Figure 9 shows the portions of Ca$_s$ observed below and above 100 km altitude. Like the high Ca layers, the sporadic Ca above 100 km maximize in summer with proportions of more than 30% of all Ca$_s$ between May and August. During the rest of the year the Ca$_s$ are observed above 100 km in less

Table 2. Occurrence index of sporadic neutral and ionized Ca layers for each month of the year, based on 89 Ca_s and 99 Ca_s^+, and of high Ca layers (for definition of the latter, see text)

Month	total Ca observations [h]	Ca_s [%]	Ca_s (<100 km) [%]	Ca_s (>100 km) [%]	High Ca layers	Total Ca^+ observations [h]	Ca_s^+ [%]	Ca_s^+ (<100 km) [%]	Ca_s^+ (>100 km) [%]
Jan.	58.0	12	8	3	0	24.4	43	16	27
Febr.	16.8	59	56	3	0	14.9	97	50	47
March	68.6	31	28	4	0	43.1	66	32	33
April	69.8	60	57	3	0	36.6	123	96	28
May	46.6	22	15	6	1	15.3	131	38	93
June	31.0	9	5	3	8	17.5	98	20	78
July	18.6	13	5	8	1	13.1	130	50	80
Aug.	65.4	22	15	8	7	10.1	102	0	102
Sept.	39.8	54	54	0	2	16.7	126	94	31
Oct.	28.6	68	61	6	0	20.2	185	118	67
Nov.	22.5	64	64	0	0	5.9	187	109	78
Dec.	21.7	98	93	5	0	12.1	132	112	21

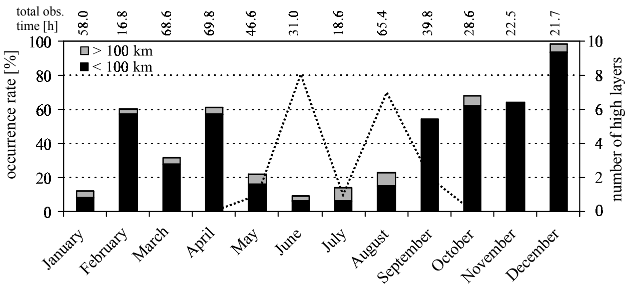

Fig. 9. Seasonal variation of the occurrence of sporadic neutral Ca layers (columns, based on exact observation time) and of so-called high Ca layers (dotted line, based on night mean profiles). The portions of Ca_s above and below 100 km altitude are marked by different colors.

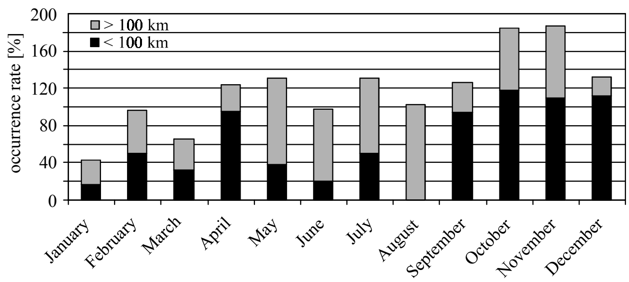

Fig. 10. Seasonal variation of the occurrence of sporadic ionized Ca_s^+ layers (based on exact observation time). The portions of Ca_s^+ above and below 100 km altitude are marked by different colors.

than 30% of all Ca_s events.

Is this seasonal variation of sporadic layers also valid for ionized sporadic layers? As Table 2 shows, there is no distinct seasonal variation of total Ca_s^+ occurrence. Ca_s^+ layers are observed in about 40 to 180% of total Ca^+ observation time. In general there is a higher index of Ca_s^+ occurrence in the second half of the year than in the first. In Fig. 10 the sporadic Ca^+ layers above and below 100 km altitude are separated: Below 100 km a semi-annual variation of Ca_s^+ occurrence is observed. The occurrence index of Ca_s^+ minimizes in summer months and in January, and increases like the occurrence of neutral Ca_s in spring and autumn. In addition, the higher Ca_s^+ (above 100 km) are dominated by annual variation, with maximum in summer.

5 Discussion of the observations

On average, we find the occurrence index of sporadic Ca (Fig. 7) to maximize in altitude about 3–4 km higher (with 5 km bin-width) than the density of the regular Ca layer (Gerding et al., 2000). This is similar to the behavior of sodium: Hansen and von Zahn (1990) reported the mean altitude of 75 Na_s observed above Andenes, Norway (69°N, 16°E) to be 95 km, while the density peak of the regular Na layer was found to be at 90 km altitude. For the location of Sao José dos Campos, Brasil (23°S, 46°W) Clemesha (1995) found the mean Na_s height to be 2 km higher than the regular layer maximum.

Case studies of direct comparisons of sporadic Ca and Ca^+ layers have been reported by two groups (Granier et al., 1989; Alpers et al., 1996). Both studies gave examples for simultaneous Ca_s+Ca_s^+ layers as well as for Ca_s^+ without Ca_s. For the case of sodium, Friedman et al. (2000) report on Na enrichments below 97 km without the occurrence of Na^+ layers in the same volume. Height differences between ionized and neutral sporadic metal layers have been reported by Alpers et al. (1993) and Gardner et al. (1993). Alpers et al. (1993) measured the altitude of a Fe_s layer by lidar and that of a simultaneous Fe_s^+ layer by a rocket-borne mass spectrometer. They found the neutral layer 1 km higher than the ionized layer. They suggested that this separation in altitude could have been due to a horizontal distance of 26 km between the two sampled volumes. Gardner et al. (1993) recorded three single Ca^+ profiles during Na and Fe soundings. He assumed that the observed height difference of 1 km between the neutral sporadic layers and the Ca_s^+ was caused by different ion masses of Fe^+, Na^+, and Ca^+. But this explanation fails for the observed identical altitude of Na_s and Fe_s (Gardner et al., 1993) as well as for the observed height differences between Ca_s and Ca_s^+, as reported here.

Seasonal variations of sporadic layer occurrences are still poorly known. The 288 hours of sodium observation of Hansen and von Zahn (1990) were carried out between July and August and between October and March, without observations in April, May, June and September. They yielded a rate of 0.4–0.5 sporadic Na layers per hour in July and August, while the rate varied between 0.03 in their first November-March period 1985/86 and 0.2–0.3 during the other autumn/winter campaigns (January to March 1987 and October/November 1987) (Hansen and von Zahn, 1990). So there are hints for a slightly increased rate of sporadic layers in summer at 69°N. Batista et al. (1989) reported on 65 sporadic sodium layers observed during 3500 hours of observation between 1975 and 1987 at Sao José dos Campos (23°S, 46°W). They identified the highest number of sporadic layers in August, while the total duration of Na_s maximized in May, June, and August, with a pronounced minimum in July. But their observational time increased during the summer months, also. They extracted a higher rate of Na_s in the second half of the year than in the first half, and compared this observation with the increased rate of visual meteors between July and December (Batista et al., 1989). So both Batista et al. (1989) and Hansen and von Zahn (1990) reported on only weak seasonal variation of sporadic Na layer occurrence.

Nagasawa and Abo (1995) gave the occurrence a total of 37 Na_s above Tokyo, Japan (36°N, 140°E) on a monthly basis and compared this with the seasonal variation in occurrence of local sporadic E layers. Their observations are shown in Fig. 11 (upper panel). The data from Nagasawa and Abo (1995) show a strong positive correlation between seasonal variations of Na_s and E_s. We compare their results with ours obtained at Juliusruh, Germany (54°N, 13°E) (Fig. 11, lower panel). The lower panel shows the Ca_s occurrence index from Fig. 9 and the occurrence of sporadic E layers above Juliusruh (J. Bremer, private communication). For these species, we find, unexpectedly, for their seasonal variation a clear anti-correlation. The situation for the ionized sporadic Ca layers is different from that of the neutral sporadic Ca layers. Also shown in the lower panel of Fig. 11, the occurrence index of the denser Ca_s^+ layers (with more than 300 Ca^+ cm^{-3}) shows a positive correlation with the E_s occurrence above Juliusruh, as expected.

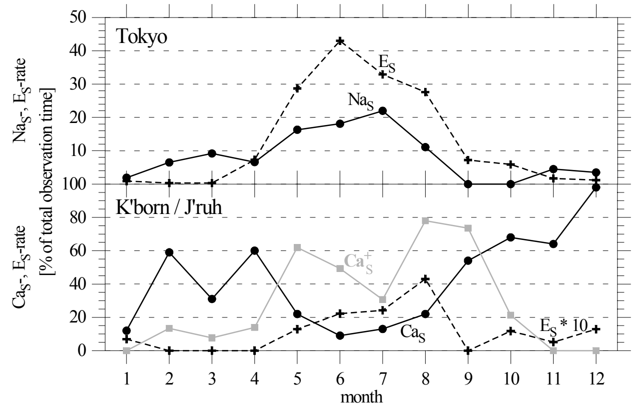

Fig. 11. Seasonal variation of sporadic metal and electron layers: Na_s and E_s as observed above Tokyo (Nagasawa and Abo, 1995) (upper panel), Ca_s and E_s as observed above Kühlungsborn and Juliusruh, respectively (lower panel). The occurrence of strong sporadic Ca_s^+ with number densities higher than 300 cm^{-3} is shown for comparison (lower panel).

The higher value of Ca_s occurrence is obvious from Fig. 11 when compared with the Na_s numbers as observed by Nagasawa and Abo (1995). These authors found an average of 6.2% occurrence of strong Na_s, although cited as much higher than observed at other mid-latitude locations. Kane and Gardner (1993) report on a total Fe_s occurrence rate of 27% for the site of Urbana, Illinois (40°N, 88°W). In spite of this, the Ca_s rate at Kühlungsborn, Germany was found to be 43% in annual mean. Also for the site of Kühlungsborn, Eska et al. (1998) report on 16 nights with strong K_s out of 110, thus roughly a rate of 15% on a nightly basis. Even this limited set of mid-latitude sporadic layer statistics give a Ca_s rate that is at least by a factor of 1.5 higher than observed for other metals.

The phenomenon of high neutral metal layers that are well above the regular metal layers has not been reported in literature. Treating these layers as distinct phenomena beside the regular and sporadic layers may be justified as follows: (a) With more than 5 km, the FWHM of high layers was found to be larger than the generally accepted upper limit for sporadic layers; (b) The fact that Ca atoms can form layers with significant abundances above 100 km without Ca^+ ions at the same altitude makes this an interesting phenomenon; (c) Neither a large variabilitiy of altitude nor concentration – typical for sporadic layers – has been observed in these broad layers. There are several examples with additional sporadic Ca layers between the high and the permanent layer. Sporadic ion layers (Ca_s^+) have been observed below the high neutral Ca layer, but at no time at the altitude of the high layer itself. In the following chapter we suggest mechanisms for neutral Ca_s formation that may explain the observed height differences between Ca_s and Ca_s^+, the seasonal variation and the observations of the high, broad layers.

6 Interpretation of variations and implications for the formation of sporadic layers

Currently, the most favored process for formation of neutral layers is the recombination of metal ions in sporadic ion layers, the latter correlating well with sporadic E layers (see e.g. von Zahn and Hansen (1988), Alpers et al. (1993), Kirkwood and von Zahn (1993), Clemesha (1995)). For the case of fast conversion of Ca ions to Ca atoms, the seasonal variations of both species are expected to correlate positively. The same positive correlation can be expected for the seasonal variation of Ca atoms and sporadic electron layers because electrons and Ca ions are correlating well, as pointed out above and proofed in Fig. 11. But this formation mechanism of direct and fast recombination seems to fail, however, in explaining the seasonal structure of Ca_s. This is because our observations show considerable differences between the seasonal variations of sporadic Ca_s and E_s layers. In addition, none of the other formation processes discussed in the literature can explain the observed seasonal variations and the altitude differences between Ca_s and Ca_s^+ layers (cf. review by Clemesha (1995)). Nevertheless, there is a general consensus on the importance of the combination of wind shears and Lorentz forces as triggers for the formation of sporadic ion layers (Chimonas and Axford, 1968; Kirkwood and von Zahn, 1991; Höffner and von Zahn, 1994). Lacking quantitative information on the ambient winds and electric fields in our experiments, we have little to add to the discussion of the latter scenario.

6.1 The height difference between Ca_s and Ca_s^+

Forty-four Ca_s layers and 99 Ca_s^+ layers monitored during our simultaneous lidar observations show either (a) various events with either Ca_s or Ca_s^+, (b) events with both Ca_s and Ca_s^+ at the same altitude, or (c) events with both Ca_s and Ca_s^+, but the two layers being separated by at least several hundred meters in altitude. Again we emphasize that the examined atmospheric volumes are exactly the same for Ca and Ca^+ observations. Thus there is no horizontal distance in sounded areas that is responsible for the observed variations. In particular, the high number of events of type (c) seems to indicate that there is no fast chemical process forming Ca directly out of Ca^+ or vice versa. In the case that such a direct and fast conversion process between Ca ions and atoms exists, one would also expect that both types of sporadic layers would match and follow each other much closer in vertical variation.

Recently, Gerding et al. (2000) have presented a detailed ion chemistry scheme for Ca and Ca^+. These authors have Ca^+ only reacting with N_2, O_2, or O_3 forming CaX^+ (with X = N_2, O_2, O). The conversion takes about 10 s or 1 h for the three body reaction $Ca^+ + Y + M \rightarrow CaY^+ + M$ (with Y = N_2, O_2 and M = collision partner) and about 15 s or 6 min for the reaction $Ca^+ + O_3 \rightarrow CaO^+ + O_2$ at 90 or 105 km altitude, respectively. Subsequently, the CaO^+ ion can react with O, giving back Ca^+ (and O_2), while the CaY^+ reacts quickly with atomic O forming CaO^+ (Plane and Helmer, 1994; Gerding et al., 2000). CaX^+ can dissociate and recombine with e^-, giving back the Ca atom. This is the only reaction leading from ion molecules to the metal atoms because direct recombination $Ca^+ + e^- \rightarrow Ca + h\nu$ is extremely slow at upper atmospheric conditions (Bates and Dalgarno, 1962).

Both reactions, CaY^+ going to CaO^+ and CaO^+ going to Ca^+, convert the reactants within less than 1 s at all heights. Therefore, the dissociative recombination of CaX^+ with e^- forming neutral Ca becomes very slow, taking several hours under typical atmospheric conditions and electron density (Gerding et al., 2000). But the reaction rates are strongly dependent on the densities of atomic O and electrons. Decreasing the O density shifts the chemical equilibrium from Ca^+ towards CaX^+, while increasing the e^- density accelerates the recombination. The ionization of Ca is most easily accomplished by charge transfer from NO^+ or O_2^+. But this is, nevertheless, a slow chemical process for ion formation (Rutherford et al., 1972; Gerding et al., 2000). Under typical ionospheric conditions, it takes several days to convert 63% of the neutral Ca into Ca^+ by these reactions, with the velocity of Ca^+-forming reactions increasing only by a factor of 4, from 90 to 105 km (Gerding et al., 2000). Due to this rather loose chemical coupling between Ca^+ and Ca, the sporadic Ca^+ and Ca layers cannot generally be expected to occur at the same altitude. As suggested here, an existing Ca_s^+ layer will first be converted into CaX^+ before recombining into Ca. This CaX^+ layer is invisible for lidars tuned to the resonance wavelengths of Ca or Ca^+. The abundance ratio of CaX^+ over all calcium ions depends strongly on the local O density and changes quickly with changing O (see above). The various Ca species (Ca^+, CaX^+, and Ca) are transported by the atmospheric wind field and the ions, in addition, are acted upon by Lorentz forces and the species can become separated horizontally as well as vertically. This can result in different heights of ionized and neutral sporadic Ca layers, as observed by our lidars.

If indeed Ca^+ ions can be converted into Ca atoms only via an initial formation of ion molecules, it would help to explain why we observe an increasing correlation of Ca_s^+ and Ca_s with decreasing height down to 90 km. Because the reactions forming molecular ions from Ca^+ are three body reactions (Plane and Helmer, 1994; Gerding et al., 2000), the reaction rate increases with the square of the air density and hence, with decreasing altitude. Furthermore, the recombination rate of molecular ions increases with decreasing altitude and the chemical equilibrium between ions and atoms tends towards a larger share of atoms with decreasing altitude. Similarly, the chemistry-based model studies of Cox and Plane (1998) for the formation of sporadic Na layers confirm the role of dissociative recombination of molecular ions for forming Na atoms. The delayed conversion of Ca^+ ions into Ca atoms leads us to expect that the lidar will be able to detect simultaneous neutral Ca_s layers at the altitudes of only those Ca_s^+ layers which were either strong or long-lasting (see Fig. 6).

The slow and indirect conversion from Ca^+ ions to Ca atoms may also explain qualitatively the phenomenon of high Ca layers above 100 km. The high neutral Ca layers have often been observed together with strong Ca_s^+ layers several kilometers below them. The neutral layers could have been formed earlier from very strong Ca_s^+ layers provided the ion layers stayed near the same altitude for an extended period. The latter condition has only a small, but finite chance to be fulfilled above 100 km. Because the charge transfer reaction $Ca + Z^+ \rightarrow Ca^+ + Z$ (with $Z = NO^+, O_2^+$) is slow, the high Ca layer may remain at these high altitudes during an entire night with only a small loss of Ca density.

6.2 The differences of seasonal variations of Ca_s, E_s, and Ca_s^+

As pointed out above, the different seasonal variations of Ca_s and E_s argue against the importance of a process of rapid neutralization of Ca^+ ions in forming neutral Ca_s layers. This mechanism just cannot explain the majority of our observed Ca_s. This situation may be, however, specific to Ca. The mean profiles of the regular metal layers differ considerably between the various metals (Helmer et al., 1998; Plane et al., 1999; Eska et al., 1999; Gerding et al., 2000). Observational and modeling results show, that for Ca, the ion concentration outnumbers the atom concentration above 91 km altitude (Gerding et al., 2000), for Fe above 93 km (Helmer et al., 1998), for K above 99 km (Eska et al., 1999), and for Na only above 101 km (Plane et al., 1999). Hence, above 90 km altitude, the density ratio of ions over atoms is highest for Ca. Furthermore, we recall that in our observations, weak sporadic Ca^+ ion layers may not correlate with sporadic E_s layers due to the worse detection limit of ionosondes. It can be shown that these weak layers can be easily formed by propagating waves and their interactions with electric and magnetic fields (Höffner and von Zahn, 1994). Therefore, local Ca^+ enrichments can be expected throughout the whole year, possibly correlating with electron layers invisible to ionosondes.

Gerding et al. (2000) have shown that the Ca^+ concentration is positively correlated with the ambient O density because of the reactions $CaY^+ + O \rightarrow CaO^+ + Y$ (with $Y = O_2, N_2$) and $CaO^+ + O \rightarrow Ca^+ + O_2$. Therefore, an increased Ca^+ total abundance can be expected during the O-rich summer season (Garcia and Solomon, 1983, 1994). This also gives reason for a decreased summer rate of conversion from Ca^+ to Ca (and therefore, a low number of sporadic neutral layers built from Ca_s^+), because there is no direct recombination, only the recombination path $Ca^+ \rightarrow CaO^+ \rightarrow Ca$.

Can the increased number of spring and autumn Ca_s also be explained by metal chemistry? The recombination rate from metal ions into the neutral atomic state can be enhanced by an increase in ozone density. In other words, if a Ca_s^+ layer coincides with an enhanced ozone abundance at this altitude, the production of neutral Ca in a small layer would result. Gerding et al. (2000) obtained a summer minimum of ozone concentration above 100 km, calculated from O concentration using the temperature dependent rate coefficients of DeMore et al. (1994). But in spring and autumn there is an increased O_3 concentration in the 100 km altitude area, enabling the Ca_s formation process quoted above.

7 Summary and conclusions

Our comprehensive lidar observations of the regular and sporadic neutral and ionized Ca layers demonstrate the high variability of sporadic Ca and Ca^+ layers as well as differences in the occurrences and movements of neutral and ionized layers. The altitude distribution of sporadic layers shows an increasing fraction of Ca^+-only sporadic layers with altitude. Layers containing both Ca and Ca^+ dominate only between 90 and 95 km. Above 100 km, Ca-only sporadic layers are rare, but have been observed in two cases. The seasonal distribution shows a minimum of Ca_s occurrence in summer, which is unusual when compared with other metals. The total number of sporadic ion layers is more or less equally distributed throughout the year, but the number of strong Ca_s^+ layers (density higher than 300 cm^{-3}) peaks in summer similar to the number of E_s observed at other locations.

The observations reported cannot be explained with the published mechanisms for sporadic layer formation. Therefore, we propose a new mechanism in which Ca^+ can recombine to neutral Ca via first forming CaX^+ (with $X = O, O_2, N_2$). This mechanism may explain both the frequently observed height difference between Ca_s^+ and Ca_s and the unusual seasonal variation of Ca_s layer occurrence. In order to verify the quantitative viability of the proposed mechanism, both new experiments and 3-dimensional time dependent chemistry-transportation modeling are needed.

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