Charge balance in the mesosphere with meteoric dust particles

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Abstract

An aerosol particle charging model (Reid, 1997) developed initially for noctilucent clouds has been extended in several steps in order to better explain data for charged meteoric smoke particles (MSPs) returned by the nighttime and daytime CHAMPS rockets (Robertson et al., 2013) launched from the Andøya rocket Range, Norway, in October 2011. Addition of photodetachment to the model shows that this process reduces the number density of positively charged MSPs as well as the number density of negatively charged MSPs as a consequence of the photodetached electrons neutralizing the positively charged MSPs. In addition, the model shows that the ionization rate can be deduced from the electron number density and the electron-ion recombination rate only at the highest altitudes as a consequence of recombination of electrons on the MSPs at lower altitudes. The differences between the daytime and nighttime data place constraints on the photodetachment rate. A further extension of the model to include the formation of negative ions and their destruction by atomic oxygen helps explain the ledge seen in the number density of the lightest negatively charged particles. MSP particle densities from the CARMA/CHEM2D model are in better agreement with rocket data for assumed values of the meteor input flux that are at the low end of the generally accepted range.

Reid, G.C., Geophys. Res. Lett. 24, 1095–1098 (1997).
Robertson, S., et al., Detection of Meteoric Smoke Particles in the Mesosphere by a Rocket-borne Mass Spectrometer, to appear in JASTP (2013).

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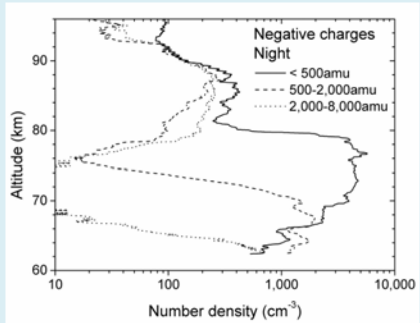
Summary the CHAMPS rocket data

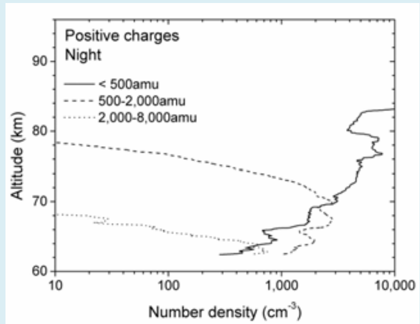
The CHAMPS rocket carried a multichannel mass spectrometer (Knappmiller et al., 2008) with channels sensitive to MSPs with mass in the ranges <500 amu, 500 – 2,000 amu, 2,000 – 8,000 amu, 8,000 – 20,000 amu and >50,000 amu. The spectrometer recorded the charge number density as a function of altitude for both positively and negatively charged particles. The signal was negligible in both the daytime and the nighttime data for positive and negatively charged particles in the 8,000 – 20,000 amu and >50,000 amu mass ranges (not shown in the figures) indicating few MSPs (<10 cm^{-3}) in this range and suggesting an upper limit in the radius of ~1.2 nm if a mass density of 2 g/cm^3 is assumed. Particles with masses greater than 500 amu were identified as MSPs and the particles with masses <500 amu were assumed to be a mixture of "ordinary" light molecular ions, cluster ions and MSPs. The spectrometer was not sensitive to electrons as a consequence of the payload potential being negative.

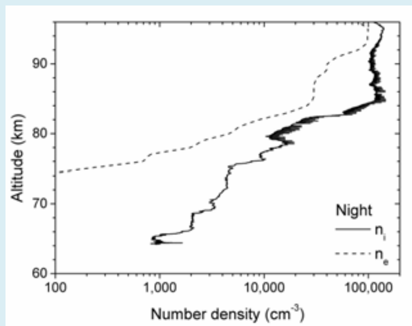
Knappmiller, S., Robertson, S., Sternovsky, Z., Friedrich, M., 2008. Rev. Sci. Instrum. 79, 104502.

Nighttime data

11 October 21:50 UT, SZA 117 degrees

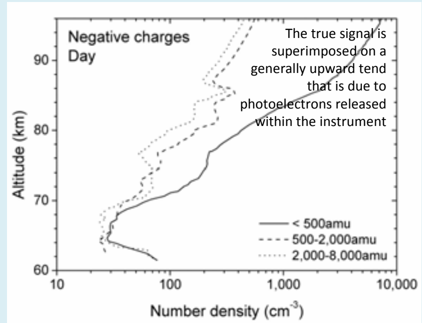


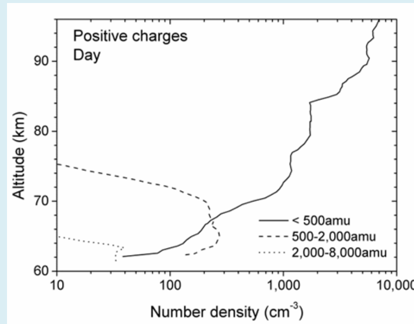


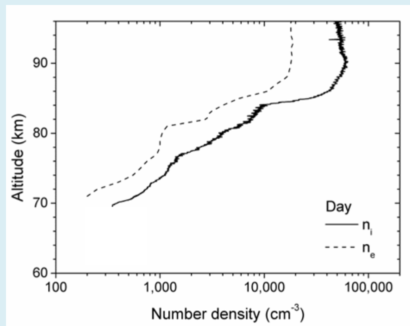


Daytime data

13 October 13:50 UT, SZA 83.5 degrees







The Motivation

The motivation is to explain unexpected features of the data

For Model 1:

The daytime data show both the positive and negative MSPs reduced in density. Photodetachment directly affects only the negatively charged MSPs.

For Model 2:

The 0 – 500 amu data for negative MSPs (and ions) show a ledge at about 80 km in the nighttime data. There are MORE negative <500 amu particles where the number of electrons is LESS, suggesting that these negative particles are getting charge by transfer of electrons from negative ions (which probably have a greater density below 80 km).

Model 1

We have extended Reid's model by adding photodetachment of electrons from the negatively charged MSPs. The independent parameters are an ionization rate and a photodetachment rate that assumed independent of altitude. The model is dependent upon altitude only through the ionization rate. The model is simplified by assuming that the MSPs have a single radius of 0.8 nm and a single (homogeneous) number density. This model reveals the effect of photodetachment on the number densities of electrons, ions, and both positively and negatively charged MSPs.

Model 1 equations

The adjustable parameters in the model are the ionization rate Q, the total number density of MSPs N_{tot}, and the photodetachment rate β. The altitude dependence enters only through the altitude dependence of Q. The outputs of the model are the number densities of the electrons, of a single species of positive ions, of positively charged MSPs, and of negatively charged MSPs. The number of uncharged MSPs is the total number N_{tot} reduced by the number that are charged. The data from the nighttime flight, for which photodetachment is inactive, are used to find the likely value for the N_{tot} given the measured numbers of charged MSPs.

Electrons
$$\frac{d}{dt}n_e = Q - \alpha_{ie} n_e n_i - n_e \sum_Z \alpha_e(Z) N_{MSP}(Z) + \beta N_{MSP}(-1),$$

Ions
$$\frac{d}{dt}n_i = Q - \alpha_{ie} n_e n_i - n_i \sum_Z \alpha_i(Z) N_{MSP}(Z),$$

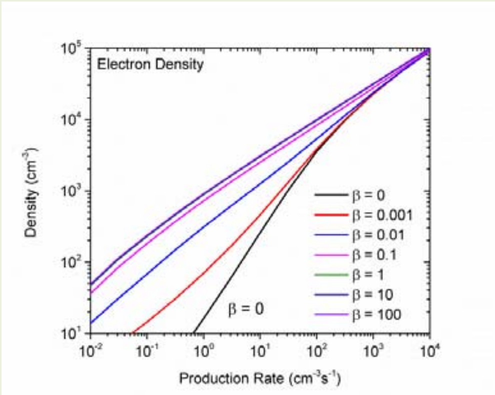
MSPs, Z = 1
$$\frac{d}{dt}N_{MSP}(1) = \alpha_i(0) N_{MSP}(0) n_i - [\alpha_e(1) n_e + \alpha_i(1) n_i] N_{MSP}(1),$$

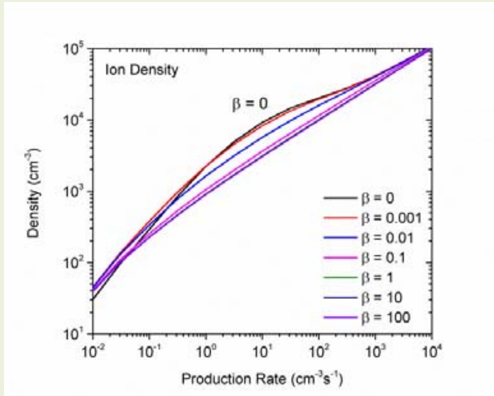
MSPs, Z = 0
$$\frac{d}{dt}N_{MSP}(0) = \alpha_i(-1) N_{MSP}(-1) n_i - [\alpha_e(0) n_e + \alpha_i(0) n_i] N_{MSP}(Z) + \alpha_e(1) N_{MSP}(1) n_e + \beta N_{MSP}(-1),$$

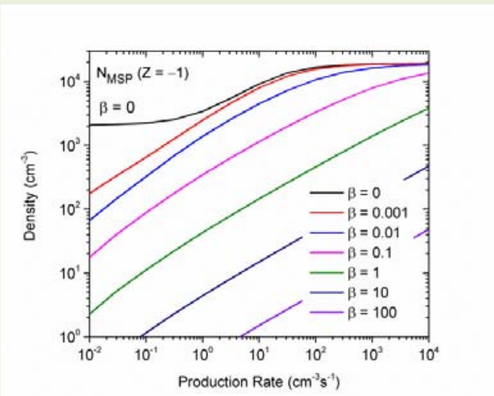
MSPs, z = -1
$$\frac{d}{dt}N_{MSP}(-1) = -[\alpha_e(-1) n_e + \alpha_i(-1) n_i] N_{MSP}(-1) + \alpha_e(0) N_{MSP}(0) n_e - \beta N_{MSP}(-1).$$

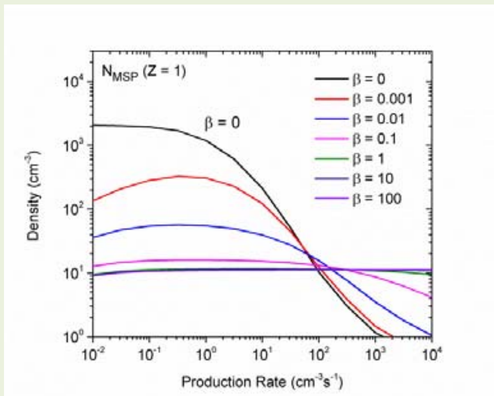
where n_e is the electron number density, n_i is the ion density, α_{ie} is the electron-ion recombination rate coefficient, Z is the charge number which can have values 1, 0, and –1, $N_{MSP}(Z)$ is the number of MSPs with charge number Z, $\alpha_e(Z)$ is the rate coefficient for attachment of electrons to MSPs with charge number Z, $\alpha_i(Z)$ is the coefficient for ion attachment and t is time.

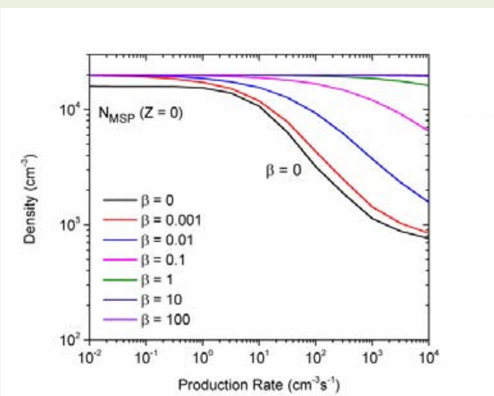
Model 1 Results Plotted











The most interesting result is the graph above which shows that <u>positive</u> MSPs are reduced by photodetachment. The electrons created by photodetachment from negative MSPs recombine with the positive MSPs reducing their number.

Discussion – Model 1

Model 1 results are plotted at left with a range of β values. Rapp (2009), using Mie scattering theory, has shown that β can have values as large as 100 s^{-1} for a radius of 1 nm for hematite. The <u>electron number density</u> is increased by increasing photodetachment for all values of Q. In the absence of MSPs and negative ions, the production is balanced by recombination of electrons with ions and the expected electron density is $n_e=\sqrt{(Q/\alpha_{ie})}$. For the highest photodetachment rates the electrons attach to MSPs and then are immediately removed; hence the electron density approaches the value without MSPs at altitudes where there are no negative ions.

The <u>ion number density</u> is decreased by increasing photodetachment for Q values greater than about 1 s^{-1}. This is likely a result of their being more electrons available for recombination. For the lower values of Q, the electrons from photodetachment are likely to attach to MSPs rather than recombine with ions and the effect of photodetachment on ions is less easily interpreted.

The <u>number density of negatively charged MSPs</u> is reduced by photodetachment for all Q values. At 65 km, the numbers of negatively charges MSPs in the 500 – 2,000 amu mass range is reduced from ~2,000 cm^{-3} at night to less than 30 cm^{-3} in the daytime, indicating β greater than about 0.3 s^{-1}. For sufficiently high photodetachment rates, attachment of an electron to a neutral MSP occurs at the same rate as photodetachment from negative MSPs, if loss of charge by attachment of ions to negative MSPs can be ignored which should be the case for sufficiently low Q values. The simplified expression for negative MSP density is $\alpha_e(0)N_{MSP}(0) = \beta\, N_{MSP}(-1)$ or $N_{MSP}(-1) = \alpha_e(0)N_{MSP}(0)/\beta$. This implies that the number of negatively charged MSPs should vary inversely with β, which is seen in the plot of the density of $N_{MSP}(-1)$. The <u>number density of positively charged MSPs</u> is reduced by increasing photodetachment for all values of Q as a result of recombination with the electrons from photodetachment. The <u>number density of uncharged MSPs</u> is increased by photodetachment for all Q values and approaches N_{tot} for the highest values of Q and β.

Rapp, M., Ann. Geophys. 27, 2417-2422 (2009)

Model 2 ideas

Since the smaller MSPs are in the sub nanometer range we assumed that they act more or less like heavy ions. Hence we divided the MSPs in two different groups determined by size. First we have the bigger ones above 500 amu which are represented by particles with a radius of 0.8 nm. This group is treated as in Model 1. MSPs below 500 amu represented by 0.4 nm particles were now treated differently. We assumed that these particles have the following properties:

- Positive charging of MSPs by ions is assumed not to happen
- Negative charging of MSPs is done by electron transfer from negative ions
- Negative charging of MSPs by electrons in two-body collisions is assumed not to happen because of the energy defect
- Negatively charged small MSPs can recombine with positive ions
- Negative small MSPs can photodetach at the highest altitudes

Atomic O and negative ions

Negative ions are included in our model by adding a rate equation for their formation and destruction that begins with attachment of electrons to diatomic oxygen $e + O_2 + M \rightarrow O_2^- + M$, where e is an electron and M is a third body (N_2, O_2). Other negative species have not been included here. There are two different rates for the creation of an O_2^-. For N_2 as the third body M the rate coefficient k_{N2} is 10^{-31} cm^6s^{-1} and for O_2 as M the rate coefficient k_{O2} is $4 \cdot 10^{-31}\exp(-193/T_e)$ cm^6s^{-1}. The density profiles of N_2, O_2 are taken from the MSIS90 model for the CHAMPS conditions. The negative ions are destroyed by the reaction $O_2^- + O \rightarrow O_3 + e$. This reaction is thought to be the origin of ledge in electron density as a consequence of the ledge in the density of O, which is an input parameter to the model.

Model 2 additional equations

Negative ions: $$\frac{d}{dt}n_{i-} = \left[k_{N_2}N_{N_2} + k_{O_2}N_{O_2}\right]N_{O_2}n_e - k_O N_O n_{i-} - \sum_Z \left[\alpha_{i-}(Z)N_{MSP}(Z) + \alpha_{i-,small}(Z)N_{MSPsmall}(Z)\right]n_{i-} - \beta_{O_2^-}n_{i-}$$

Neutral MSPs: $$\frac{d}{dt}N_{MSP_{small}}(0) = \alpha_{i-}(-1, r_{small})N_{MSP_{small}}(-1)n_{i-} - \alpha_{i-}(0, r_{small})N_{MSP_{small}}(0)n_{i-} + \beta N_{MSP_{small}}(-1)$$

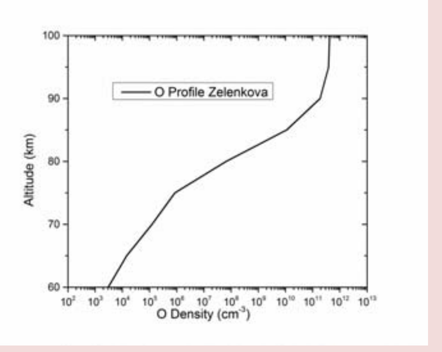
Negative MSPs: $$\frac{d}{dt}N_{MSP_{small}}(-1) = \alpha_{i-}(0, r_{small})N_{MSP_{small}}(0)n_{i-} - \alpha_{i-}(-1, r_{small})N_{MSP_{small}}(-1)n_{i-} - \beta N_{MSP_{small}}(-1)$$

where n_{i-} is the density of negative ions, N_{O2} is the density of diatomic oxygen, N_O is the density of atomic oxygen, and $N_{MSPsmall}(Z)$ is the density of MSPs in the 0 – 500 amu mass range with charge number Z. The new rate coefficients are for the attachment of electrons and ions to the small MSPs. In addition to these added equations, there are added terms in the Model 1 equations (not shown).

Atomic O input function

Model 2 requires atomic O as an input variable shown as the black line. This profile from Zelenkova et al. (1993) provides the needed ledge in the density of negative ions. MSIS is used above 90 km.

Rates for reactions have been taken from Dieminger et al., 1996.



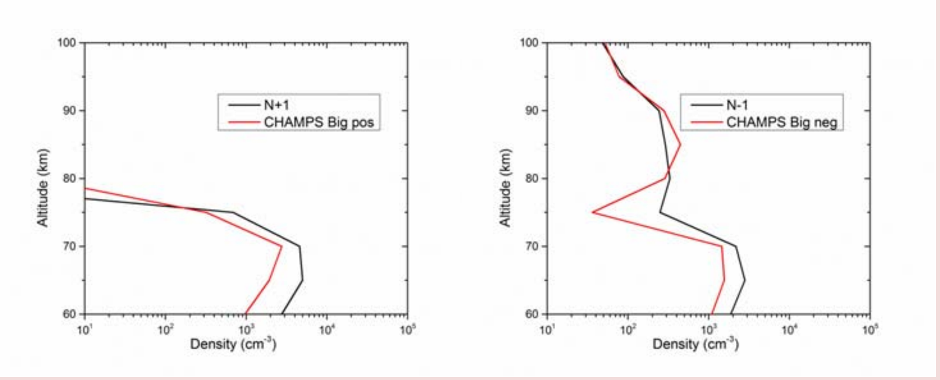
Zelenkova, L., Soldatov, V., and Broznets, M., 1993. Ozone and atomic oxygen distribution variations on heights of ionospheric D-region during disturbances PCA and AA, SPIE Vol. 2047 Atmospheric Ozone pp. 237-241.

Gumbel, J. at al, 2003. Influences of ice particles on the ion chemisty of the polar summer mesosphere, J. Geophys. Res. 108(D8) 8436, doi:10.1029/2002JD002413.

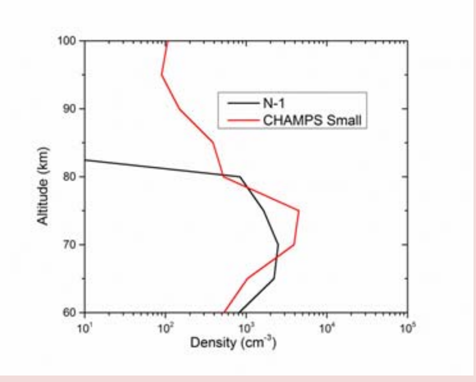
Dieminger, Walter; Hartmann, G. K.; Leitinger, R. (1996): The upper atmosphere. Data analysis and interpretation. Berlin, New York: Springer-Verlag.

Model 2 preliminary results

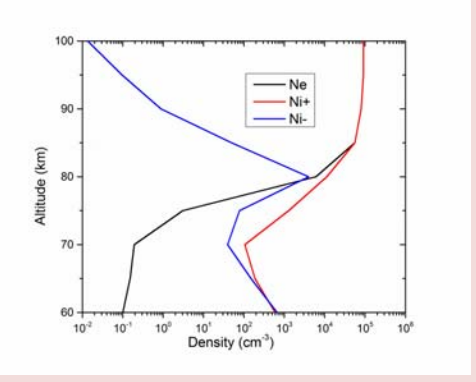
The model with atomic O reproduces features in the density of MSPs (>500 amu). The altitude atomic O profile selected is the one that best fits the data.



The model reproduces the ledge in the density of small negative MSPs (<500 amu), but the ledge is not as pronounced.



The model reproduces the ledge in electron density but does not give electron and ion densities smoothly increasing with altitude.



In progress: We are finding additional solutions to the model with different assumption about the input parameters.

Conclusions

Model 1:

1.Photodetachment decreases both the number density of negative MSPs and positive MSPs because the photodetached electrons neutralize the positively charged MSPs. Positive and negative MSPs should be about equally abundant at low *Q* values, in the absence of significant numbers of negative ions.

2.The ledge in density of negatively charged MSPs at 80 km suggests that negative MSPs below 80 km are charged by charge transfer from negative ions and not by attachment of electrons.

Model 2:

1.Addition of the negative ion O_2^- helps explain the disappearance of electrons below about 80 km.

2.Charge transfer from negative ions to small MSPs helps explain negative MSPs being abundant where there are negative ions and few electrons.

3.The cutoff at 80 km for positive MSPs is partially explained by their being neutralized by electrons above the ledge.