Problems with ionization rate in the research of atmospheric aerosols

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Cluster ions are ubiquitous in the Earth's atmosphere. They are produced by galactic cosmic rays and by the radiation of radioactive bodies. The cluster ions continually interact with aerosol particles transferring them electric charges. The general evolution of cluster ions can be expressed by the following equation

$$\frac{dc}{dt} = I - \alpha c^2(t) - S_{ion} c(t), \qquad (1)$$

where c is the average concentration of negative and positive cluster ions, t is time, I is the ionization rate, α is the coefficient of mutual recombination, and S_{ion} is the term, which characterizes the ion sink. Besides the effect of aerosols, the term of ion sink may also contain a sink caused by ion-induced nucleation.

In steady state conditions, the equation (1) can be presented in a simpler manner (as a balance equation):

$$I = \alpha c_\infty^2 + S_{ion} c_\infty, \qquad (2)$$

where c_∞ is the concentration of cluster ions in the steady state.

The ionization rate is a factor of ion-induced aerosol nucleation and is included in the models of atmospheric aerosol formation (Tammet and Kulmala, 2005; 2007; Luts *et al*., 2015). The existing models usually consider the ionization rate as a homogeneous scalar, characterized by a single number or by few numbers. In reality, the ionizing radiation near the ground appears to be largely dependent on location and environmental conditions, especially when the ground is covered with tall vegetation. Our special study of literature showed that researchers have measured the ionization rate values from 0.8 to 117 ion pair $cm^{-3}s^{-1}$ near the ground. Disagreements between the results of different studies have been considerable. Thus, the progress in the study of atmospheric aerosol formation requires an adjustment of the air ionization characteristics and development of new measurement methods.

The authors propose a new experimental method, which relies on the theory of balance and evolution of cluster ions and incorporates both the time-dependent and steady state equations (1) and (2) (Hõrrak *et al.*, 2014). It does make no attempt to specify the sources of either ionization rate I or ion sink S_{ion}.

We accept the assumption of symmetric charging of aerosol particles by cluster ions and weak polarity symmetry, which states that the attachment rates (sinks) of small ions to neutral aerosol particles are equal: $\beta_o^+ c^+ = \beta_o^- c^-$ and use symbols $\beta_o = (\beta_o^+ + \beta_o^-)/2$ and $c = (c^+ + c^-)/2$, where the terms with upper indexes " $^+$ " and " $^-$ " mean that the corresponding terms are measured or calculated for a particular polarity.

The analytical solution of Equation (1) is known, but the numerical integrating is usually more convenient. The solution of Equation (1) can be interpreted as a special function $c = f(c_0, I, S_{ion}, t)$, where c_0 is the initial value of c at $t = 0$.

We employ a special measurement method using a long metal tube with a controlled electrostatic gate of air ions at the inlet and an air ion spectrometer at the outlet of the tube (Tammet, 1977; Semenov *et al.*, 1988; Hõrrak *et al.*, 2014). The electrostatic gate periodically blocks the passage of cluster ions into the tube; then they are produced only within the tube. When the gate is open, the ambient air reaches the spectrometer in normal conditions. The measured c values during both the periods enable to calculate both I and S_{ion} values.

For preliminary testing of the experimental system, it was installed in indoor conditions at the attic room of the former building of the Institute of Physics at 4 Tähe St, Tartu. The results demonstrate substantial variations in the measured ambient concentrations of cluster ions, ranging for about 400 cm^{-3} up to 1700 cm^{-3}. The ionization rates varied between about 9 cm^{-3} s^{-1} and 21 cm^{-3} s^{-1}. Such a large variation within few days was not a surprise, if to consider the effect of radon. The calculated sinks varied between about 0.005 s^{-1} and 0.025 s^{-1}; such values are adequate as well. Thus the new method can be recommended for further testing.

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