DIURNAL VARIATION IN THE CONCENTRATION OF AIR IONS OF DIFFERENT MOBILITY CLASSES

U. Hõrrak, J. Salm, and H. Tammet

Institute of Environmental Physics, University of Tartu, Estonia

ABSTRACT: Analyzed data consists of 8900 hourly average mobility distributions measured in the mobility range of 0.00041–3.2 $cm^2V^{-1}s^{-1}$ (diameter range 0.36–79 nm) at Tahkuse Observatory, Estonia, in 1993–1994. The average diurnal variation in the concentration of cluster ions is typical for continental stations: the maximum in the early morning hours and the minimum in the afternoon; this is explained by variations in radon concentration. The variation for big cluster ions (0.5–1.3 $cm^2V^{-1}s^{-1}$) differs from that for small cluster ions (1.3–3.14 $cm^2V^{-1}s^{-1}$). The size distribution of intermediate and light large ions in the range of 1.6–22 nm is strongly affected by the nucleation bursts of nanometer particles. On the burst days, the maximum concentration of intermediate ions (1.6–7.4 nm) is at about the noontime, and that of light large ions (7.4–22 nm) at about 2 hours later. The concentration of heavy large ions (charged Aitken particles of diameters of 22–79 nm) is enhanced in the afternoon, which is explained by the bursts of nanometer particles and the subsequent growth of particles by condensation and coagulation. If the burst days are excluded, then in the warm season the concentration of Aitken particles increases during night. In the cold season, the diurnal variation is different, and all the classes of aerosol ions (2.1–79 nm) show a similar variation with the minimum at 6 LST and the maximum in the afternoon; exceptions are the rare nucleation burst days.

INTRODUCTION

The diurnal variations in air ion concentration and in air conductivity have been discussed since the 1930s. Overviews of that period can be found in the monographs [*Israël*, 1970]. Later the diurnal variation in the concentration of air ions was considered in several papers [e.g. *Misaki and Kanazawa*, 1969; *Prüller*, 1970; *Retalis et al.*, 1991; *Dhanorkar and Kamra*, 1993; *Israelsson and Tammet*, 2001]. The integral Gerdien condensers were commonly applied in those measurements. The sensitivity and resolution of devices, as well as the techniques of mobility distribution measurements were unsatisfactory in many cases [see *Israël*, 1970; *Tammet*, 1970]. Often the mobility ranges of air ion classes were very wide and with quite uncertain boundaries. Since the conductivity is mainly determined by the concentration of small air ions, the diurnal variations of these two quantities are similar. The diurnal variation in the concentration of large air ions, or charged aerosol particles, is to a greater extent affected by meteorological conditions and by local air pollution.

Due to the complexity and large-scale variability of atmospheric processes, the long-term measurements of air ions in a wide range of mobility are necessary to draw statistically founded conclusions about the air ion characteristics. This was the main argument for the installation of wide-range automated multichannel mobility spectrometers at Tahkuse Observatory in 1988 [*Hõrrak et al.*, 1994]. So far the problems of the bursts of intermediate ions (the generation of charged nanometer aerosol particles driven by photochemistry and boundary layer meteorology) and of the classification of air ions have been studied especially on the basis of regular measurements at Tahkuse Observatory [*Hõrrak et al.*, 1998, 2000]. The present paper deals with the average diurnal variations of different air ion classes.

A statistical analysis revealed that the entire mobility spectrum of air ions can be divided into two main classes: *cluster ions* with mobilities above 0.5 $cm^2\,V^{-1}\,s^{-1}$ and *aerosol ions* (charged aerosol particles) with mobilities below 0.5 $cm^2\,V^{-1}\,s^{-1}$ [*Hõrrak et al.*, 2000]. These two classes can in turn be divided into 5 almost independent air ion classes: small cluster ions (1.3–3.2); big cluster ions (0.5–1.3); intermediate ions (0.034–0.5); light large ions (0.0042–0.034); heavy large ions (0.00087–0.0042 $cm^2\,V^{-1}\,s^{-1}$).

MEASUREMENTS

The measurements of air ion mobility distributions were carried out at Tahkuse Observatory (58°31'N 24°56'E), which is located in a sparsely populated rural region, 27 km northeast of the city of Pärnu, and 100 km south of Tallinn, the capital of Estonia. Pärnu, with 52000 inhabitants, is on the coast of the Gulf of Riga, at the east coast of the Baltic Sea.

A complex of air ion spectrometers covering a mobility range of 0.00041–3.14 $cm^2\,V^{-1}\,s^{-1}$ was installed at Tahkuse Observatory in 1988 [*Hõrrak et al.*, 1994, 2000; *Hõrrak*, 2001]. The complex consists of three original multichannel aspiration spectrometers designed according to the principle of the second-order differential mobility analyzer [*Tammet*, 1970]. The whole mobility range was logarithmically divided into 20 intervals: 9 intervals in the subrange of 0.00041–0.29 $cm^2\,V^{-1}\,s^{-1}$ and 11 intervals in the subrange of 0.25–3.14 $cm^2\,V^{-1}\,s^{-1}$. The analysis is based on the data from September 1, 1993 to October 27, 1994.

RESULTS AND DISCUSSION

The average diurnal variation in the concentration of cluster ions had the ordinary shape for continental stations: the maximum in the early morning hours and the minimum in the afternoon. The diurnal variation was considerable in the warm season, and it was very weak in the cold season when the soil was wet or it was frozen and covered by snow. These results are in accordance with the hypothesis that radon is the main ionizing agent, which causes the variation in the ionization rate, and therefore in the concentration of cluster ions near the ground. Subdividing cluster ions (0.5–3.14 $cm^2V^{-1}s^{-1}$) into two classes of small and big cluster ions with the boundary at 1.3 $cm^2V^{-1}s^{-1}$ for negative ions and 1.0 $cm^2V^{-1}s^{-1}$ for positive ions leads to more distinct shape of the diurnal variation in the concentration of these ion classes (Figure 1). The average diurnal variation of the entire group of cluster ions is mainly caused by small cluster ions. The average diurnal variation in the concentration of big cluster ions was weak in general, and sometimes it was slightly in an opposite phase compared to that of small cluster ions. Considering the days with the nucleation bursts of nanometer particles (intermediate ions), the average diurnal variation in the concentration of big cluster ions showed the maximum at noon; in the warm season the second maximum was recorded near midnight. The different behavior of small and big cluster ions causes diurnal variation in the mean mobility of small ions. The median mobility of positive ions had the maximum of 1.39 $cm^2V^{-1}s^{-1}$ (negative ions 1.59 $cm^2V^{-1}s^{-1}$) in the nighttime and the minimum of 1.29 $cm^2V^{-1}s^{-1}$ (negative ions 1.42 $cm^2V^{-1}s^{-1}$) in the afternoon.

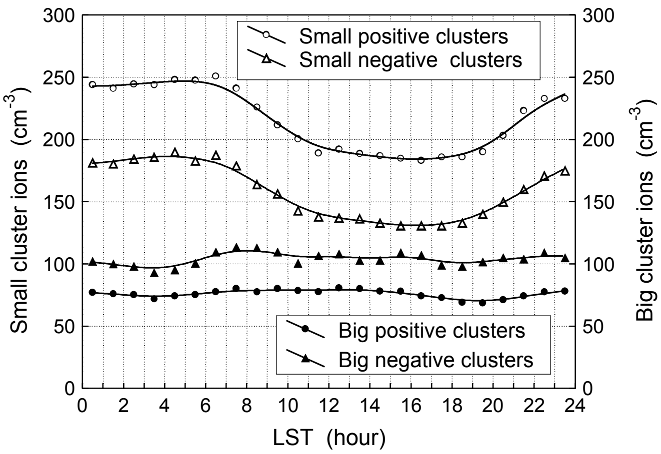

Figure 1. Diurnal variation in the median concentration of positive and negative small cluster ions and big cluster ions in the warm season (Sept. 1993, May – Sept. 1994).

The diurnal variation in the concentration of aerosol ions (intermediate and light large ions) of the diameters of 1.6–22 nm is strongly affected by the photochemical nucleation bursts of nanometer aerosol particles in favorable meteorological conditions, e.g. after the inflow of clean and cool Arctic air masses in spring and autumn [*Hõrrak et al.*, 1998]. In the burst days, the maximum in the concentration of intermediate ions (1.6–7.4 nm) was found at about the noontime (Figure 2).

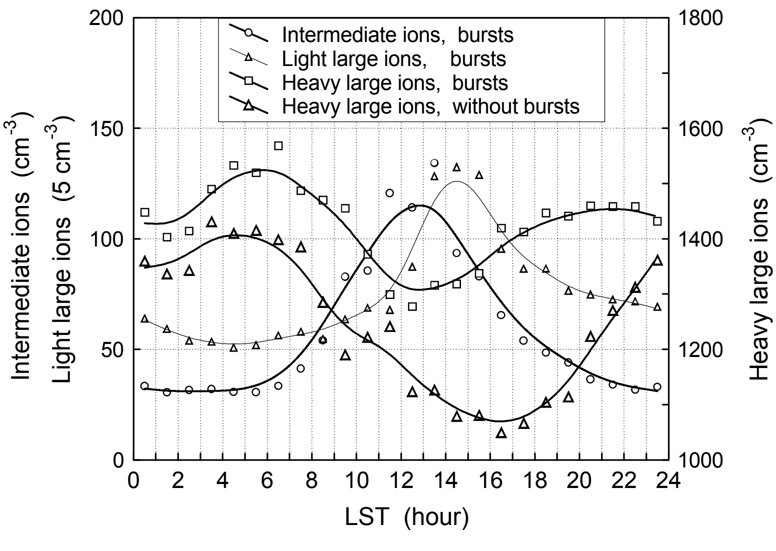

Figure 2. Diurnal variation in the median concentration of intermediate ions (diameter 2.1–7.4 nm), light large ions (7.4–22 nm) and heavy large ions (22–79 nm) in the warm season separately for days with and without burst events of intermediate ions. Sept. 1993, May – Sept. 1994.

Light large ions (7.4–22 nm) varied differently from intermediate ions; their maximum concentrations were shifted to somewhat later hours. In the case of nucleation bursts an evolution process was observed in the range of intermediate and light large ions: the spectral peak drifted slowly from smaller diameters to larger ones.

The average diurnal variation in the concentration of heavy large ions (charged Aitken particles) with the diameters of 22–79 nm was usually very weak. The average diurnal variations displayed different behavior during the cold season and during the warm season. When the rare nucleation events were excluded then in the cold season all the classes of aerosol ions (2.1–79 nm) showed a similar average diurnal variation with the minimum at 6 LST (Figure 3). The concentration of intermediate ions exhibits the maximum in the afternoon. The heavy large ions reach the maximum in the evening around 20 LST. A decrease during the nighttime is probably due to the deposition, coagulation and growth of particles by condensation.

In the warm season, the concentration of heavy large ions (charged Aitken particles of 22–79 nm) increases during the nighttime, it reaches the maximum at 6–7 LST and later decreases gradually up to about 17 LST. In the days with nucleation bursts the concentration of heavy large ions was enhanced in the afternoon compared with non-burst days (see Figure 2). This is explained as a consequence of the particle growth towards large sizes.

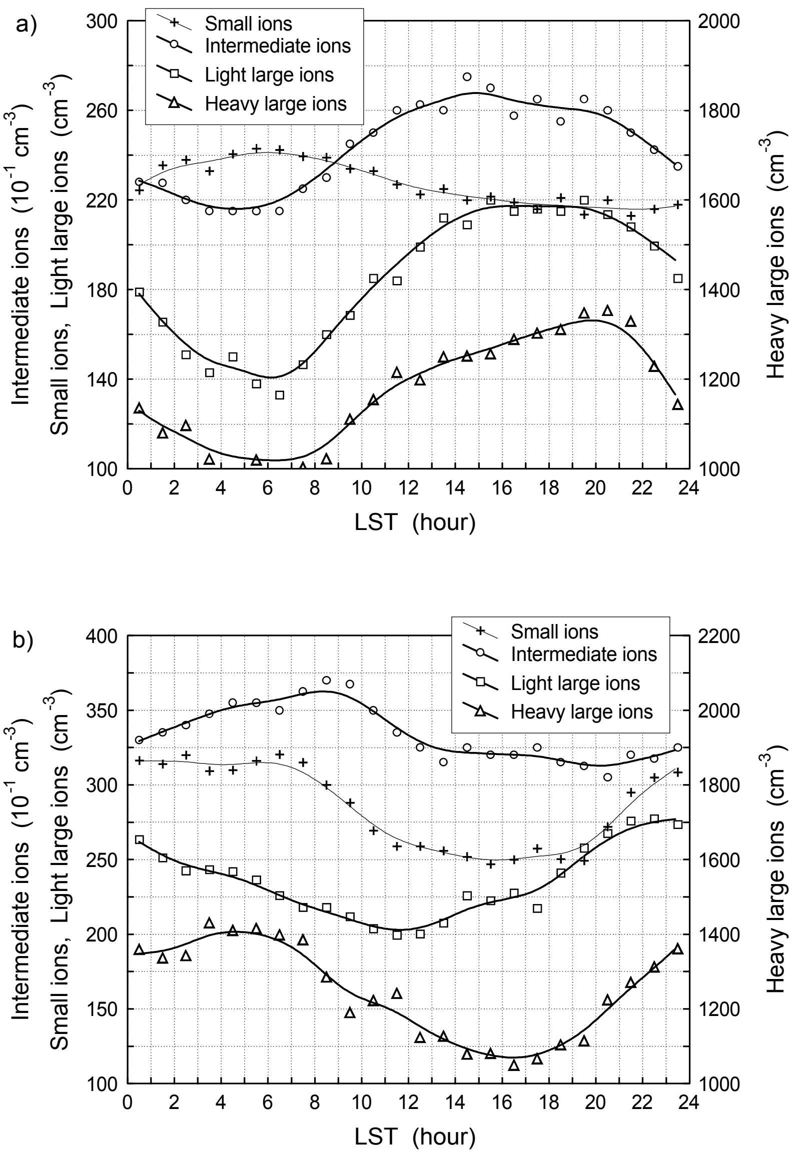

Figure 3. Diurnal variation in the median concentration of positive cluster ions (0.5–3.14 $cm^2V^{-1}s^{-1}$), intermediate ions (2.1–7.4 nm), light large ions (7.4–22 nm) and heavy large ions (22–79 nm): a) in the cold season (November – March), b) in the warm season (May – September). The days with the bursts of intermediate ions are excluded.

In the cold season the average diurnal variation in the concentration of heavy large ions is nearly in the opposite phase to that in the warm season. This contrast can be explained by the different processes of aerosol particle generation (combustion versus radiolytic processes). In winter the concentrations of charged Aitken particles and NO_2 were positively correlated (the correlation coefficients were 41–77%).

The variation in different classes of air ions provides information about different processes in the atmosphere: e.g. the accumulation of radon near the ground dependent on stability of boundary layer, the generation and growth of nanometer particles during nucleation bursts and changes in Aitken particle concentration.

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