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Bursts of intermediate ions in atmospheric air

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Abstract. The mobility spectrum of air ions has been measured at Tahkuse Observatory in Estonia for several years. The average concentration of intermediate ions with mobilities of 0.05–0.5 $cm^2 V^{-1} s^{-1}$ in atmospheric air is about 50 cm^{-3}. On the level of this low background, high concentration bursts of intermediate air ions occur occasionally. A burst can be followed by subsequent evolution of intermediate ions into larger ones. To explain the bursts of intermediate air ions, two hypotheses can be advanced: (1) A burst of neutral particles occurs due to homogeneous nucleation, and the particles are charged by the attachment of cluster ions. (2) The cluster ions grow by ion-induced nucleation in proper environmental conditions.

1. Introduction

The concept of intermediate ions has been gradually established during its history. *Pollock* [1915] was the first, who distinctly wrote about intermediate ions. *Weiss and Steinmaurer* [1937] proposed the mobility interval of 0.02–1.0 $cm^2 V^{-1} s^{-1}$ for intermediate ions. *Misaki et al.* [1972] measured the mobility spectra of air ions in a continuous interval from 0.0001 to 3.2 $cm^2 V^{-1} s^{-1}$. The spectra have a deep depression between small and large ions. The range of depression could be considered as the range of intermediate ions. The upper boundary of the depressed mobility range lies at about 0.5–0.6 $cm^2 V^{-1} s^{-1}$ and the lower boundary at about 0.03–0.1 $cm^2 V^{-1} s^{-1}$, depending on the measurement site and time. A deep depression in the average mobility spectrum between 0.05 and 0.5 $cm^2 V^{-1} s^{-1}$ was found by measurements in urban air [*Salm and Reinart,* 1983, 1992; *Dolezalek et al.*, 1985]. *Kojima* [1984] measured the mobility spectrum of intermediate ions in the interval of 0.0085–0.24 $cm^2 V^{-1} s^{-1}$ as 10 logarithmically distributed fractions. *Dhanorkar and Kamra* [1991] designed and built a spectrometer with three aspiration condensers for measuring in a wide mobility range. The condenser for intermediate ions has a range of limiting mobilities of 0.0243–0.248 $cm^2 V^{-1} s^{-1}$. The above measurements have been carried out episodically, and the duration of measurement periods has been limited. Some atmospheric processes have a long correlation time, and long-term measurements are required to study statistical behavior of the air ion mobility spectrum in the atmosphere.

In 1985–1986 the mobility spectrum of air ions in an interval of 0.32–3.2 $cm^2 V^{-1} s^{-1}$ was measured at Tahkuse Observatory during a 1-year period. The mobility of 0.5 $cm^2 V^{-1} s^{-1}$ appeared as a quite distinct boundary between small and intermediate ions [*Tammet et al.*, 1987, 1988, 1992; *Hõrrak et al.*, 1994]. According to the physical nature, the air ions of mobility above 0.5 $cm^2 V^{-1} s^{-1}$ are charged molecular clusters or cluster ions. The boundary 0.5 $cm^2 V^{-1} s^{-1}$ is considered as the lower limit of small ion range as well as the upper limit of intermediate ion range. The lower limit of intermediate ion range cannot be so distinctly determined; the statistical analysis of measured mobility spectra in a wide range could yield a basis for a conventional agreement. The ions with mobilities less than 0.5 $cm^2 V^{-1} s^{-1}$ can be ordinary aerosol particles charged by diffusion mechanism; enhanced concentrations of such particles can arise in the process of homogenous nucleation. On the other hand, intermediate ions can arise through ion-induced nucleation, like a growth of cluster ions.

An extended system for regular measuring in a wide range of mobilities was set into operation at Tahkuse Observatory in 1988. The average mobility spectrum for 6 months (July 1988 to January 1989) [*Hõrrak et al.*, 1992, 1994] showed a similar depression in the range of 0.05–0.5 $cm^2 V^{-1} s^{-1}$ also mentioned by *Misaki et al.* [1972] and by *Salm and Reinart* [1983, 1992].

In the measurements at Tahkuse Observatory in 1985–1986, the narrow fraction of intermediate air ions in the range of 0.32–0.5 $cm^2 V^{-1} s^{-1}$ showed occasional bursts (enhancements of concentration) [*Tammet et al.*, 1988, 1992; *Hõrrak et al.*, 1994]. The yearly average concentration of this fraction was about 25 cm^{-3}. Enhanced concentrations over 60 cm^{-3} were observed during 4% of the hours. In a few occasions some very high concentrations of intermediate ions were observed, and in some cases the enhancement of the concentration was observed only for one polarity. The average diurnal variation had a maximum at noon of local time. A hypothesis was advanced stating that a burst of the intermediate air ion concentration could be explained by ion-induced nucleation in atmospheric air [*Tammet et al.*, 1988; *Hõrrak et al.*, 1994, 1995]. The ingredients of the air responsible for nucleation remained unknown. A possibility that the ingredients were of local anthropogenic origin was not excluded. An analysis of the regular measurements about intermediate ions in a wide mobility range is presented in this paper.

2. Measurements

The measurement site is located in a sparsely populated rural region, 27 km northeast of the city of Pärnu, Estonia. The geographical coordinates of the observatory are 58°31'N, 24°56'E [*Hõrrak et al.*, 1994].

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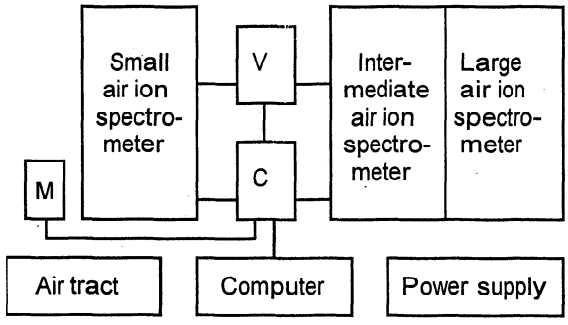

Figure 1. Schematic diagram of the instrumentation for air ion measurements. V is voltage supply, C is controller, and M stands for meteorological sensors.

The first intermediate ion measurements at Tahkuse were carried out in 1985–1986. An original mobility spectrometer was designed and built for these measurements [*Tammet et al.*, 1987]. The mobility range of 0.32–3.2 $cm^2 V^{-1} s^{-1}$ was logarithmically divided into 10 fractions. The air is sucked into the instrument through an opening in the vertical fronton of the attic of a building at a height of about 5 m from the ground. The air intake is shielded from the atmospheric electric field by tall trees surrounding the building. The lack of considerable electrode effect under the trees was verified by the small ion measurements in 1985 resulting in average polar conductivities of $\lambda_+ = 9.07$ fS m^{-1} and $\lambda_- = 8.96$ fS m^{-1} [*Tammet et al.*, 1992].

The same spectrometer was applied in a special measurement campaign near Zvenigorod, Russia, in June 1986 [*Kikas et al.*, 1990] simultaneously with the electric aerosol spectrometer (EAS) of Tartu University [*Mirme et al.*, 1984].

A complex air ion spectrometer covering the mobility range of 0.00032–3.2 $cm^2 V^{-1} s^{-1}$ has been operating at Tahkuse Observatory since 1988 [*Hõrrak et al.*, 1990; *Tammet*, 1990]. A diagram of the instrumentation is presented in Figure 1. The complex air ion spectrometer consists of three original multichannel aspiration spectrometers designed according to the principle of second-order aspiration measuring condenser. All three aspiration measuring condensers have a coaxial shape. At the entrance of each measuring condenser, the incoming airflow is divided into two parts by a preliminary condenser. The outer coaxial layer is deionized, while the central flow passes through, retaining its natural condition. Behind the preliminary condenser the measuring condenser has one inner electrode and multiple outer electrodes. Each outer electrode is connected with the input of individual electrometric amplifier. The measuring condenser of the small ion spectrometer is the same as during earlier measurement campaigns. The air intake is the same as described above for 1985–1986. The range of mobility is logarithmically divided into 20 intervals: 9 intervals in the subrange of 0.00032–0.25 $cm^2 V^{-1} s^{-1}$ and 11 in the subrange of 0.25–3.2 $cm^2 V^{-1} s^{-1}$. The positive and negative air ion mobility spectra are measured every 5 min. The hourly averages and standard deviations of spectral components during hourly periods are recorded and saved on the hard disk of a PC.

Besides the atmospheric electrical parameters the concentration of NO_2, wind direction, wind speed, atmospheric pressure, temperature, and relative humidity were measured. A detailed description of the instrumentation is given by [*Hõrrak et al.*, 1990].

A statistical analysis is presented below for data collected during 14 months from September 1993 to October 1994. The

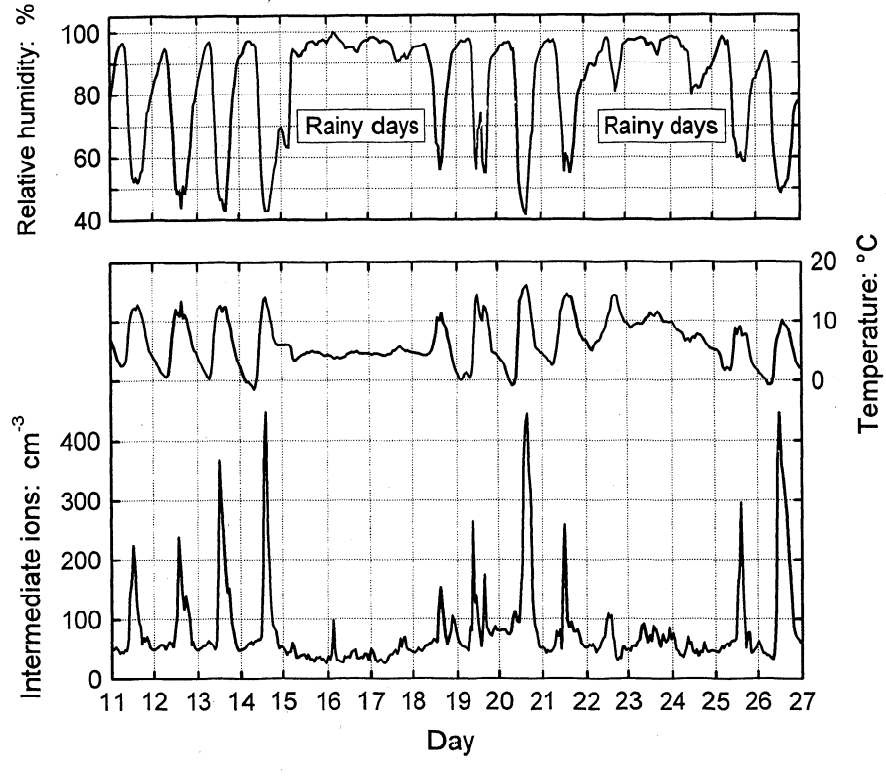

Figure 2. Variation of the concentration of positive ions of a mobility of 0.034–0.5 $cm^2 V^{-1} s^{-1}$, air temperature, and relative humidity at Tahkuse Observatory, September 11–27, 1993.

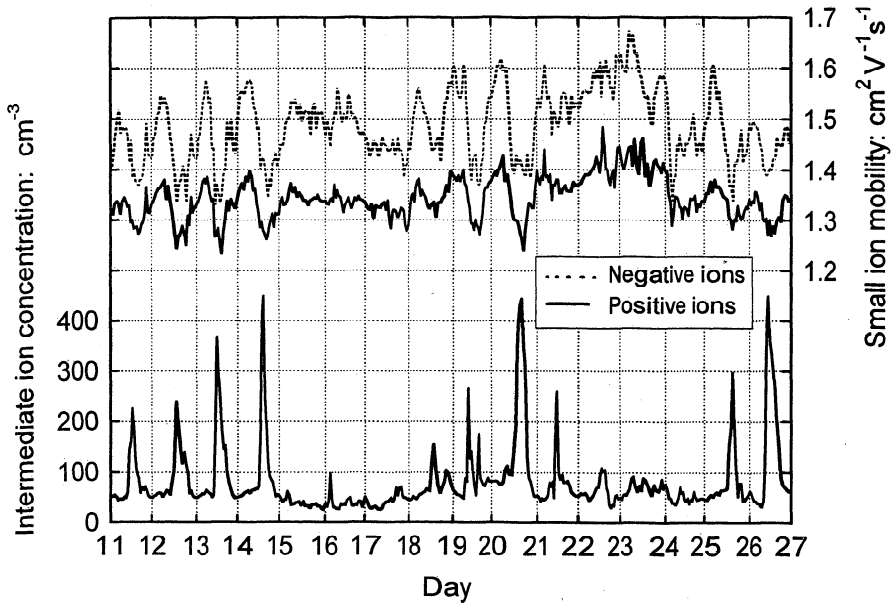

Figure 3. Variation of the concentration of positive ions of a mobility of 0.034–0.5 cm^2 V^{-1} s^{-1} and the mean mobility of negative and positive small ions of 0.5–3.2 cm^2 V^{-1} s^{-1} at Tahkuse Observatory, September 11–27, 1993.

statistical distribution of intermediate air ion concentrations is not Gaussian. Thus the complete set of hourly averages of air ion concentrations in the mobility interval of 0.034–0.5 cm^2 V^{-1} s^{-1} (corresponding diameter interval is 1.6–7.4 nm) is characterized by the average concentration and by the quantiles of the statistical distribution. The average polar concentration of positive and negative ions is 57 cm^{-3}, the median is 40 cm^{-3}, and the upper quartile is 58 cm^{-3}. There is no significant difference between average concentrations and the quartiles of the positive and negative air ions. The 90% quantile is 110 cm^{-3} for positive ions and 125 cm^{-3} for negative ions. The maximum hourly average concentration in the above interval was 960 cm^{-3} for positive ions and 1010 cm^{-3} for negative ions.

3. Results

Sometimes the enhanced concentrations of intermediate air ions, called bursts, occur. Polar concentration of intermediate air ions over 100 cm^{-3} was observed during 14% of the measurement period. A typical time variation is shown in Figure 2. On the background of concentration of about 50 cm^{-3}, bursts up to 10 times over the background are noticeable. As a rule, a burst occurs around noontime. The burst of intermediate air ions coincides with a temperature maximum at daytime.

The concentration of intermediate ions is strongly correlated with temperature during the diurnal cycle. The solar radiation is considered as an essential factor of a burst. The hypothesis about the solar radiation factor is confirmed by the

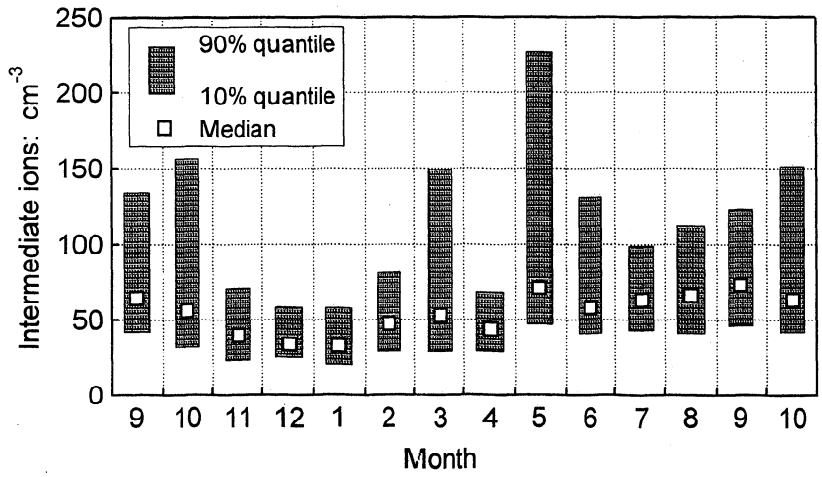

Figure 4. Annual variation of intermediate ions of a mobility of 0.034–0.5 cm^2 V^{-1} s^{-1} at Tahkuse Observatory, September 1, 1993 – October 27, 1994.

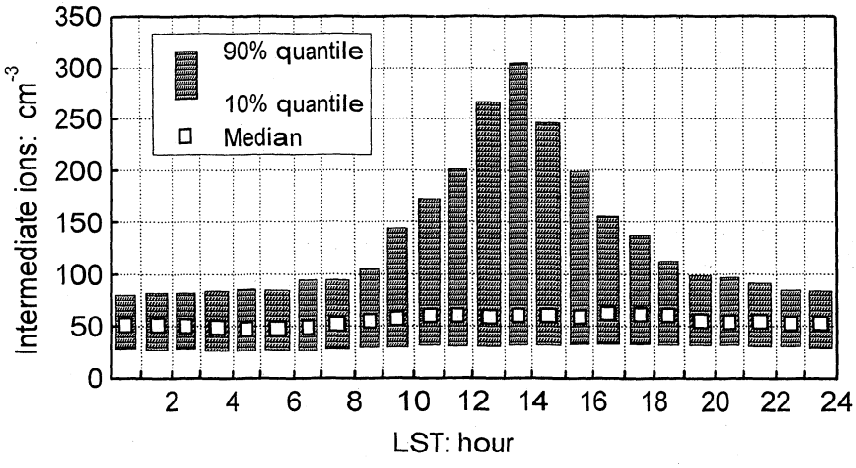

Figure 5. Diurnal variation of the concentration of positive intermediate ions of 0.034–0.5 $cm^2 V^{-1} s^{-1}$ at Tahkuse Observatory, September 1, 1993 – October 27, 1994.

time variation diagram of intermediate ion concentration and of relative air humidity (Figure 2). The extensive variations of temperature and relative humidity are present at the time of bursts, as a rule. There are no bursts in cloudy and rainy days. Bursts occur when relative humidity is less than 70%. The concentration of intermediate ions is negatively correlated with relative humidity (e.g. linear correlation coefficient about –65% in September). Consequently, the humidity (as a condensable vapor) is not a factor of generating intermediate air ions.

The mean mobility of small air ions of both polarities is calculated by averaging over the mobility interval from 0.5 to 3.2 $cm^2 V^{-1} s^{-1}$. The hourly mean mobilities averaged over the whole measurement period of 14 months, and their standard deviations are $k_{-ave} = 1.54 \pm 0.10\ cm^2 V^{-1} s^{-1}$, and $k_{+ave} = 1.37 \pm 0.06\ cm^2 V^{-1} s^{-1}$. The mean mobility of small air ions in winter is higher, as in summer; the ratio of annual maximum and minimum is about 1.1. The bursts of intermediate ions are related to the changes in the mobility spectrum of small ions; the changes take place mainly in the low-mobility wing of the spectrum (below 1 $cm^2 V^{-1} s^{-1}$). The bursts coincide with the minima of the mean mobility of small ions in the diurnal cycle (Figure 3). We suppose that the changes in the mobility spectrum of small ions are due to the changes in the composition and concentration of some trace gases in the air. Considering large ions with mobilities of 0.00041–0.0042 $cm^2 V^{-1} s^{-1}$ (charged Aitken particles with diameters of 23–79 nm), it is possible to conclude that a burst occurs when the concentration of large ions has been low for a few hours (probably due to atmospheric exchange).

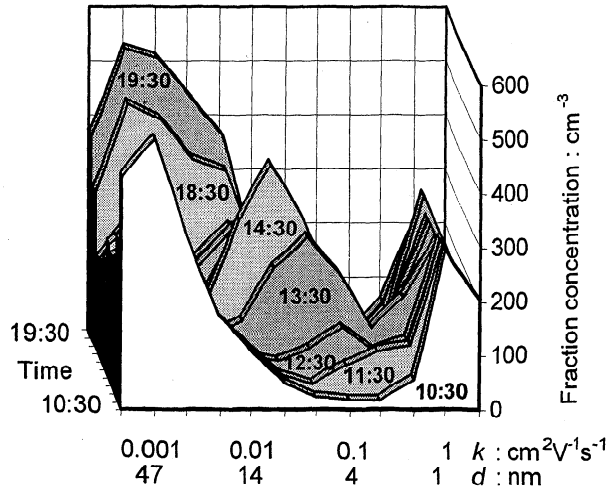

Figure 6. Evolution of air ion mobility spectrum at Tahkuse Observatory, October 20, 1994.

The annual variation of the intermediate ion concentration is weakly expressed with a minimum in December to January (Figure 4). The annual variation of the 90% quantile of concentration is more expressive with a maximum in May. The main peak in May can probably, in part, be explained by the beginning of the early vegetation period and agricultural activity [*Hõrrak et al.*, 1996]. The weather in July was extremely stable, hot and sunny with the average temperature of 20.2°C, often (more than 15 days) over 30°C at noon. The atmospheric pressure with its standard deviation was 1013 ± 4 mbar (three durable anticyclones). The average relative humidity was 66.7%, it was below 95% in 95% of all hours. Nevertheless, the 90% quantile in Figure 4 is lower than that in June and in August; the bursts were relatively low, only four of them reached a value of about 200 cm^{-3}. The stability of weather conditions can be a factor in this connection.

The examination of the time series of atmospheric pressure and intermediate ion concentration enables to conclude that intensive bursts (up to 900 cm^{-3}) occur in fine weather conditions when pressure is considerably changing. Sometimes bursts followed drastic changes in air temperature and relative humidity; the drastic changes of meteorological parameters in their turn are related to the exchange of air masses.

The diurnal variation of the intermediate ion concentration quantiles calculated for the whole measurement period is shown in Figure 5. It has one peak value around noontime by local time. The ratio of 90% quantile and median indicates that the distribution of intermediate air ion concentration is essentially different from the normal distribution at noon. The median concentrations of different mobility fractions reach peak values at different times in the afternoon. The time of the peak value is the later the lower the mobility of the fraction is.

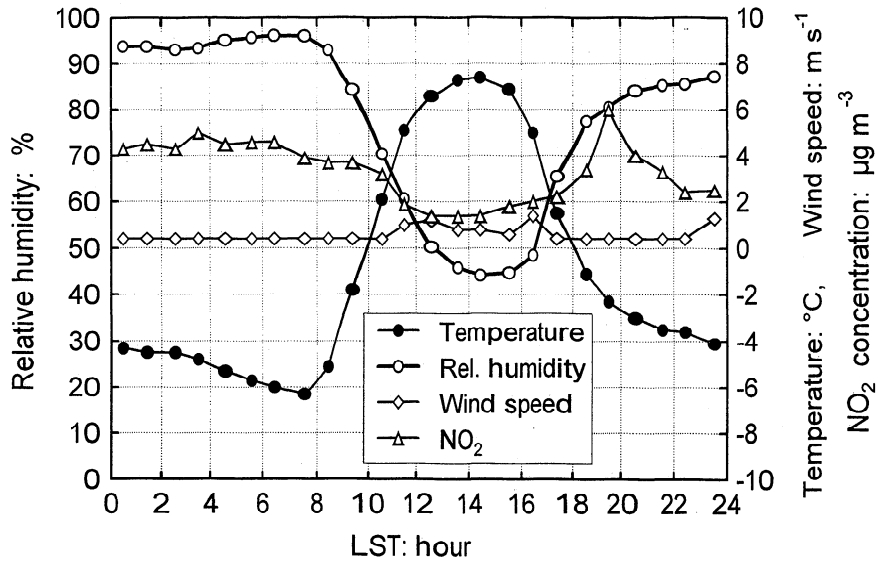

Figure 7. Variation of air temperature, relative humidity, wind speed, and NO_2 concentration at Tahkuse Observatory, October 20, 1994.

The examination of the time series collected during 1988–1994 enables to conclude that often a process of the evolution of the mobility spectrum in a wide mobility range followed a burst of intermediate air ions. An example is demonstrated in Figure 6. The first hourly average spectrum of positive air ions at 1030 LST is similar to the long time average and can be considered as an undisturbed spectrum of air ions. During the next hour a considerable amount of intermediate air ions of diameter 2–4 nm has arisen. Henceforth, the maximum in the spectrum of intermediate air ions gradually shifted toward lower mobility and larger size up to a size range of 9.7–15 nm (corresponding mobility range of single-charged particles is 0.009–0.021 $cm^2 V^{-1} s^{-1}$). The process of evolution from intermediate ion size range to nucleation mode size range took about 4 hours. During next 3 hours, intermediate ion concentration decreased simultaneously with a decrease in nucleation mode size range. The nucleation mode disappeared after a sudden increase in Aitken mode size range of 23–79 nm (corresponding mobility range is 0.00041–0.0042 $cm^2 V^{-1} s^{-1}$). The evolution of the spectrum of negative ions was similar. The weather was sunny and almost calm, with weak south wind. The variations of air temperature, relative humidity, wind speed, and NO_2 concentration are presented in Figure 7. Sunrise was at about 0730 LST. The variation of the concentration of intermediate, small, and large ions is shown in Figure 8. An enhancement of the concentration of intermediate ions began at about 1100 LST, at the time of quick rise of air temperature. In general, a burst of intermediate ions occurred

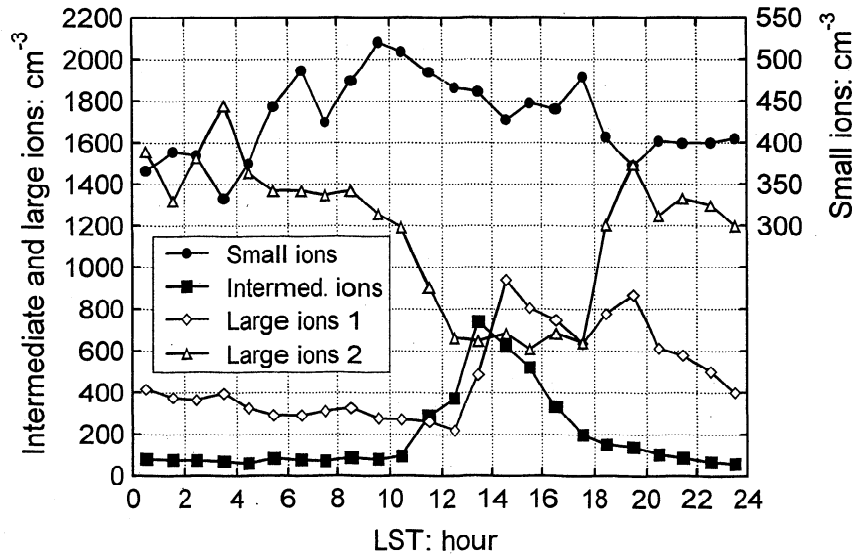

Figure 8. Variation of the concentration of positive intermediate ions of a mobility of 0.034–0.5 $cm^2 V^{-1} s^{-1}$, small ions of 0.5–3.2 $cm^2 V^{-1} s^{-1}$, large ions 1 of 0.0042–0.034 $cm^2 V^{-1} s^{-1}$, and large ions 2 of 0.00041–0.0042 $cm^2 V^{-1} s^{-1}$ at Tahkuse Observatory, October 20, 1994.

Table 1. Maximum Correlation Lag Between Mobility Fractions of Air Ions and Fraction of Intermediate Ions of Mobility 0.034–0.5 $cm^2 V^{-1} s^{-1}$ at Tahkuse Observatory, May 1994

Mobility, $cm^2 V^{-1} s^{-1}$	Lag, Hours	Correlation Coefficient
0.034–0.5	0.0	1.0
0.016–0.034	0.0	0.9
0.009–0.02	0.5	0.8
0.004–0.009	1.0	0.7
0.0019–0.004	2.0	0.5
0.0009–0.0019	3.0	0.2
0.0004–0.0009	4.5	<0.2

not always during a rapid change in temperature and relative humidity, but if it occurred, then a synchronism of these processes was noted.

The above example was selected as the most expressive from seven bursts during the period of October 16–23. This period began with the inflow of a cool high-pressure air mass; the daily average temperature suddenly decreased from 8°C to 0.3°C, remained roughly on this level during the whole period, and raised to 7.2°C on October 24.

Besides the process of the evolution described above, the process of another character was also observed: a spectral mode suddenly appeared in the nucleation size range of 9.7–23 nm (mobility range of 0.0042–0.021 $cm^2 V^{-1} s^{-1}$) and remained there for 4–6 hours, changing in height. The meteorological conditions for this process were the same as for the evolution process.

4. Discussion

The process of the evolution of mobility spectrum is similar, as observed by *Misaki* [1964] in the New Mexico semidesert for large air ions in a mobility range of 0.00018–0.01 $cm^2 V^{-1} s^{-1}$. He observed a gradual shift of a spectral peak toward lower mobilities (larger sizes) for large ions of both polarities during several hours in daytime; the weather was fine at that time.

The evolution of a mobility spectrum after a burst is characterized by a correlation coefficient between the concentrations of intermediate and large air ions provided that the large ions are measured later than the intermediate ions. The correlation coefficient depends on the delay of large ion measurements and has a maximum at a delay called the maximum correlation lag. The maximum correlation lag calculated for various mobility fractions of large ions characterizes the time of the transformation of air ions from higher to lower mobility. Some values of the maximum correlation lag for the transformation of intermediate ions into different fractions of large ions are shown in Table 1. These values are consistent with the estimates of correlation lags for size fractions of atmospheric submicrometric aerosols [*Mirme et al.*, 1988].

Some indications on the phenomenon of the same physical nature as the bursts of intermediate ions are known and described in the literature.

The short measurement campaign near Zvenigorod in June 1986 showed several events when at first the concentration of air ions in the range of 0.32–0.5 $cm^2 V^{-1} s^{-1}$ arose, and then after 1–2 hours, an enhancement of the concentration of aerosol particles in the diameter range of 10–18 nm (measured by EAS) followed [*Kikas et al.*, 1990].

Covert et al. [1992] measured the total number concentration of aerosol particles with diameters above 3 nm and aerosol number size distribution in the range from 20 to 600 nm in the marine boundary layer (MBL) along the coast of Washington State in 1991. They have found local mesoscale bursts of new particle production in a size range less than 15 nm. The generation of particles was favored in the conditions of low particulate surface area, intensive solar radiation, and high concentration of SO_2.

Occasional enhanced generations of nanometer particles in natural conditions have been described by *Aalto et al.* [1995] at the Värriö environmental measurement station in Finland. They measured aerosol size spectrum in the range of 10–200 nm by means of a TSI diffusion battery. In some situations, when air masses came from the Arctic Sea, they observed high concentrations of particles in the fraction of 10–30 nm.

McMurry et al. [1995] experimentally studied nucleation and particle growth in the free troposphere on Mauna Loa and in the Colorado Rockies. The observed rates of particle formation in these measurements exceeded values predicted by the theory by 5 or 6 orders of magnitude when neglecting the possibility of ion-induced nucleation. However, the measured neutral particle nucleation rate was not high enough to explain the bursts of intermediate air ions as a result of diffusion charging of initially neutral nanometer particles.

McGovern et al. [1996] measured the concentration of Aitken nuclei at the Mace Head Atmospheric Research Station in 1991, and occasionally very high Aitken nuclei concentrations were observed around noontime. Clear diurnal periodicity and dependence on solar radiation were revealed.

Covert et al. [1996] made aerosol measurements onboard the Swedish icebreaker *Oden* during the International Arctic Ocean Expedition in 1991 (IAOE-91). They measured aerosol number size spectra in a range from 3 nm to 500 nm and recorded three simultaneously existing distinct number modes with mean geometric diameters of around 170 nm, 45 nm, and 14 nm referred to as accumulation, Aitken, and ultrafine modes, respectively. They concluded that the generation of ultrafine particles is not associated with the MBL but occurs near the top of the MBL or in the free troposphere. The air parcels containing a high concentration of ultrafine particles are mixed into the MBL with Aitken and accumulation mode particles.

For the explanation of the bursts of intermediate air ions, several hypotheses can be advanced. The two alternative mechanisms are as follows:

1. The nanometer particles are generated in neutral state (by homogeneous nucleation) and then charged by the attachment of small air ions. This is the same process of charging that is responsible for generating large air ions. In this case, the concentration of nanometer particles during a burst is estimated tens of thousands per cubic centimeter which was never observed in atmospheric aerosol studies. Such high concentrations of neutral particles should suppress the concentration of small ions, but no considerable suppression was observed during the bursts. An additional argument against the diffusion-charging hypothesis is the charge asymmetry of the concentrations of intermediate air ions which was sometimes observed [*Tammet et al.*, 1988, 1992].

2. A burst of intermediate ions is a result of an enhanced ion-induced gas-to-particle conversion following the bursts of some (still unknown) trace gases in atmospheric air. These

gases are probably not emitted by a local anthropogenic source, because the long duration of the evolution process (Figure 6) shows evidence that it is running in an air mass of considerable horizontal size. Up to now we can state that H_2O and NO_2 are not the factors for the bursts. According to the second hypothesis, some amount of intermediate air ions (majority during a burst) are former small ions grown up to the size of nanometer aerosol particles. They are inherently charged and could be called the primary aerosol ions.

The phenomenon of the spectral peak evolution toward larger ions (after a burst) is still without quantitative explanation. *Noppel* [1995] tried to simulate the evolution by a theoretical model supposing the generation of a condensable vapor with a constant rate, ion-induced nucleation, and condensation of the vapor on aerosol particles. The simplified model was in a sufficient accordance with measurements. However, an improvement of the model, taking into account coagulation with preexisting aerosol particles and recombination with small air ions, forfeited the accordance; the shift of the spectral peak toward larger ions in the model disappeared. Obviously, the factors not considered in simplified theoretical models (nonstationary concentration of nucleating trace gases, binary nucleation etc.) are responsible for the evolution of atmospheric ion spectrum in the occasion of a burst of intermediate ions.

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