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AIR IONS AND ELECTRICAL AEROSOL ANALYSIS

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SPECTRUM OF ATMOSPHERIC IONS IN THE MOBILITY RANGE 0.32-3.2 $cm^2/(V\cdot s)$

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Introduction

The authors have carried out continuous measurements of ion spectra in the mobility range from 0.32 to 3.2 $cm^2/(V\cdot s)$, whereas the whole range was logarithmically divided into 10 intervals. The spectrum was represented by a corresponding set of fraction concentrations. Instruments and methods used are described in [1]. The present paper presents preliminary conclusions from the results of measurements carried out from 04.06.85 to 10.06.85 in the city of Tartu, Estonia, and from 10.06.85 to 15.09.85 at Tahkuse village, Estonia. In 1984 systematic measurements of mobility spectra of small ions of one-second age were carried out at the same village [2]. The measurement point at Tahkuse was situated in a typical sparsely populated rural region 27 km to the north-east of the city of Pärnu.

In the measurement point at Tahkuse the spectrometers with the sensors were located in the attic of a one-storeyed country house. The air was sucked in through an opening in the vertical frontone of the attic at a height of about 5 m from the ground. The house was surrounded by tall trees and there were fields nearby. The computer, controller, and the sensor of pressure were situated in a room with a microclimate of normal living premises.

The measurements were carried out round the clock, interruptions were due to technical failures and power cuts. All the measurements were controlled by the computer program described in [1].

Processing of results

The results were first recorded on a compact cassette and subsequently rewritten on a hard disk of *Iskra-226* computer with the help of the system described in [3]. Further processing was carried out by *Iskra-226* computer using Basic-02 programming system.

The first stage of processing included technical recoding of information and elimination of all the data where at least one overloading of at least one electrometer had occurred. At the second stage fraction concentrations of ion spectra were computed and the times when measurement precision had failed to meet certain criteria, were eliminated. The method of the computation of spectra is presented in [1]. The elimination of results is described in Table 1. The eliminated times were not uniformly distributed over the whole day; a heightened probability of insulation failures was observed in high-humidity morning hours. Similarily, the probability of failures and noise in power input was increased in the morning hours.

Table 1

The extent of measurements

Place	Time (Moscow, Summer)		No. of hrs.			
	Beginning	End	possible	actual	passed stage 1	passed stage 2
Tartu	04.08.85. 18:00	10.08.85. 7:00	133	102	91	90
Tahkuse	10.08.85. 19:00	15.09.85. 19:00	2328	2025	1763	1700

The results obtained at stage 2 were the basis of all the statistical conclusions. *No further elimination of* results on the basis of quality was carried out.

It can be hypothesized that the apparatus matrix proposed in [1] is designed on the basis of an overly modest estimate of apparatus smoothing of the spectrum and leads to systematic "undercorrection" of spectra, especially in the conditions of strong turbulence. The apparatus matrix can be improved after further experiments in the laboratory. As it is, we have considered it best to stick to the lower limit of the estimate of apparatus smoothing, as "overcorrection" would lead to even more considerable distortions of results, than the retention of residual apparatus smoothing in the case of "undercorrection".

At the second stage of processing, an additional correction was used for the adsorption of ions on the input grid of the measurement capacitor. According to a semi-empirical estimate [4] the adsorption is about 4% for a mobility of 2 $cm^2/(V \cdot s)$.

The form of the spectrum and the classification of ions according to their mobilities.

To determine the average spectra of small air ions on the basis of 1700 hours of measurements at Tahkuse, only the hours where none of the four main parameters exceeded the interval $x \pm \sigma_x$ (neither for positive, nor for negative ions) were chosen. There were 443 such hours and they were more or less uniformly distributed over the whole period of measurement. The respective spectra were averaged arithmetically. The results are presented in Fig.1.

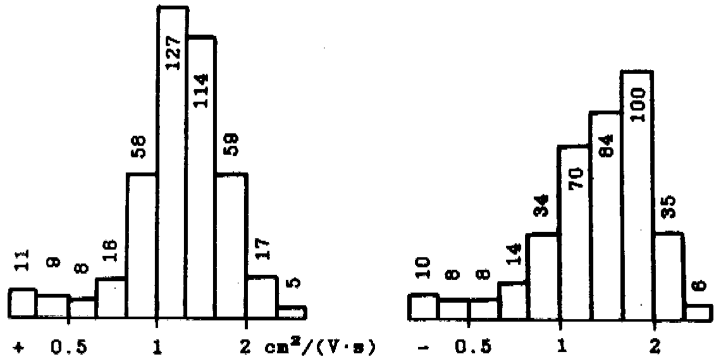

Fig. 1. Average spectra of small atmospheric ions. Henceforth the fractions will be indicated in the descending order of numbers and ascending order of the mobilities. The number of ions/cm^3 is indicated at every column. The limits of mobility intervals are: 0.32; 0.40; 0.50; 0.63; 0.79; 1.00; 1.26; 1.58; 2.00; 2.51; 3.16.

The standard deviation of random error of the measurement of fraction concentrations in an one-hour spectrum was about several ions in cm^3. Random errors in the average spectra in Fig. 1 were much lower. However, considerable systematic errors which do not diminish in averaging are not excluded. One of the possible sources of errors is residual apparatus smoothing of the spectrum. It is very likely that positive results in the mobility interval over 2.5 $cm^2/(V \cdot s)$ are only a result of smoothing, and that actually there are no such ions. Results pointing to the existence of high-mobility ions in the atmosphere have been published for several times, e.g. [5, 6]; however, in all these cases it cannot be excluded that the apparent presence of such ions has been caused by low resolution of the instruments and resulting smoothing of spectrum.

The presence of low-mobility ions in the results cannot be explained by apparatus distortions. Such ions that are not always present, are of special interest.

In the spectra with a clear-cut "heavy tail" a minimum is observed in the mobility range 0.5-0.6 cm^2/(V·s). The minimum is observed also in the average spectra (Fig. 1). An analysis of a set of spectra creates an impression that the physical nature of ions changes at the limit 0.5-0.6 cm^2/(V·s). It seems that in the case of high mobilities the ions could be considered as molecular clusters, whereas in the case of low mobilities the properties of macroscopic particles are dominant.

The above regularity confirms the correctness of using the boundary 0.5-0.6 cm^2/(V·s) for the classification of ions. Henceforth, in accordance with numerous other studies, we will use 0.5 cm^2/(V·s) as the standard value of the border, whereas only ions with higher mobilities will be called small ions. Ions with lower mobilities will be classified as intermediate ions. In the following discussion of the intermediate ions we will consider only the ions with mobilities 0.32--0.5 cm^2/(V·s), recorded during the measurements.

Intermediate ions are mostly distributed symmetrically by polarities. However, there are cases where intense formation of intermediate ions takes place only for one of the polarities. Respective examples are presented in Fig. 2, the examples are extreme also with a view to relative proportions of intermediate and small ions.

These examples permit to hypotesize that at the division of ions into small and intermediate according to their physical nature it is necessary to consider the respective mobility intervals as partially overlapping. For instance, positive ions with the mobilities 0.63-0.8 cm^2/(V·s) in the spectrum (Fig. 2a) are likely to be intermediate ions physically.

In the spectrum of small positive ions at the achieved resolution always only one maximum is discovered, whereas its location is highly variable. In the spectrum of negative ions often two peaks were observed (see Fig. 2b). In ordinary conditions the more mobile peak in the spectrum is strongly dominant and there is no intermediate minimum. Fig. 3 presents an extremely narrow spectrum. This sample also demonstrates an extremely high average mobility and a rather low concentration of small ions.

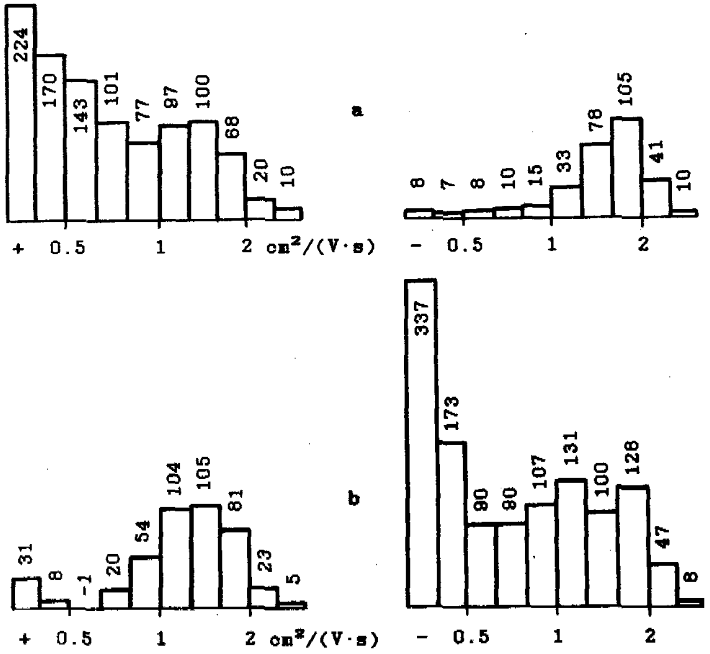

Fig. 2. Examples of extreme spectra.
a - at 3 a.m. 13.06.85; b - at 5 p.m. 05.08.85.
Estimates of standard deviation of random errors are less than 3 ions/cm^3 for any fraction.

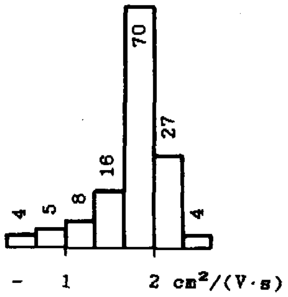

Fig. 3. Extremely narrow spectrum of small negative ions. Midnight 30.08.85.

Our results concerning the form of spectrum agree well with an example by Yunker [7], with the observations of Misaki et al. [8], and also with the results of [9].

Parameters of the spectrum

For statistical analysis it is necessary to describe the basic properties of the spectrum by means of a small amount of numeric parameters. The following set of parameters is based on the above conclusion about the expedience of dividing the whole spectral range into a subrange of small ions with mobilities over 0.5 cm^2/(V·s) (8 first fractions) and a subrange of intermediate ions with mobilities below this limit (2 last fractions). The main parameters are determined and denoted as follows:

1. Concentration of small ions $n = \sum_{j=1}^{8} \varphi_j$.

2. Average mobility of small ions for standard conditions

   $\bar{k} = \sum_{j=1}^{8} k_j \varphi_j / n$, where k_j is geometric mean of boundary mobilities of the fraction.

3. Relative width of the spectrum of small ions

$$s = \sqrt{\left(\sum_{j=1}^{8} k_j^2 \varphi_j / n - \bar{k}^2 \right)} / \bar{k}.$$

4. Fraction concentration of intermediate ions

$$m = \varphi_9 + \varphi_{10}$$

Natural average mobility of small ions is viewed as a supplementary parameter.

$$\bar{k}' = \left(\frac{101325 \text{ Pa}}{p} \frac{T}{273.15 \text{ K}} \right) \bar{k}.$$

Statistical averages and distributions

Average values and standard deviations of the parameters of all 1700 pairs of spectra recorded at Tahkuse are presented in Table 2.

Table 2

Average parameters of spectra at Tahkuse

Parameter	Polarity	Average	Standard deviation	Unit of measurement
n	+	408	145	cm^{-3}
n	-	359	136	"
m	+	24	16	"
m	-	24	20	"
$\bar{k}$	+	1.31	0.10	cm^2/(V·s)
$\bar{k}$	-	1.47	0.13	"
$\bar{k}'$	+	1.39	0.10	"
$\bar{k}'$	-	1.56	0.13	"
s	+	0.305	0.022	1
s	-	0.317	0.033	"

By way of comparison it can be pointed out that in summer 1984 on the island of Vilsandi it was found that $k_+ = 1.35 \pm 0.49$ cm^2/(V·s) and $k_- = 1.59 \pm 0.44$ cm^2/(V·s) [10]. These values were obtained using an integral aspiration counter and respectively higher random errors of measurement are reflected in the high estimates of standard deviations.

Average conductivities $\lambda_+ = 9.20$ fS/m and $\lambda_- \approx 9.12$ fS/m are nearly equal; this is evidence of the suppression of the electrode effect under tall trees. The presented values do not include the conductivity caused by ions with mobilities below 0.32 cm^2/(V·s).

The statistical distribution of hourly averages of mobilities is described by Table 3.

Table 3

Distribution of hourly averages of small ion mobilities at Tahkuse

	1.0	1.1	1.2	1.3	1.4	1.5	1.6	1.7	1.8	1.9
$\bar{k}_+$	12	235	584	577	234	53	5	0	0	
$\bar{k}_-$	2	41	135	321	463	475	196	65	2	
$\bar{k}'_+$	0	28	294	652	520	179	25	2	0	
$\bar{k}'_-$	0	5	47	150	324	487	457	168	41	

Mobilities below 1.1 cm^2/(V·s) occurred only when ions which were intermediate according to their physical nature, formally fell into the mobility range of small ions. The Sherman distributions [11] differ from those in Table 3 first of all in their greater width. A possible explanation is the effect of random measurement errors.

The statistical distribution of the average mobility is rather close to normal distribution. The distribution of small ion concentration has a noticeable positive asymmetry and a large excess. The distribution of intermediate ion concentration is significantly different from normal distribution: the coefficient of asymmetry exceeds 9 and the coefficient of excess is 180 (see also Table 9).

The distribution of the coefficient of unipolarity is shown in Table 4.

Table 4

Distribution of the unipolarity coefficient n_+/n_- at Tahkuse. The first line contains the values and the second line the frequencies.

0.8	0.9	1.0	1.1	1.2	1.3	1.4	1.5	1.6
4	66	440	747	353	72	12	3	

Daily variation

The daily variation of main spectral parameters is weakly expressed. The data are presented in Table 5.

The strong daily variation of the concentration of small ions in inhabited locations is caused by the daily rhythm of anthropogenic pollution of the air. The concentration of small air ions is determined by both the intensity of ion formation and aerosol density. In a relatively sparsely inhabited rural location variations of both factors evidently cancel each other.

The nightly maximum of the concentration of small ions was caused by rare cases of anomalously heightened concentration exceeding $n + 3\sigma_n$. None of these anomalies was accompanied by anomalies of unipolarity. A likely reason of high concentrations is an anomalously low concentration of aerosols which occurred only at night. If to eliminate 2% of the data on the basis of the criterion of high ion concentration, then the average daily variation is strongly suppressed - peaks of n_+ at 23:00 and 3:00 fall to 418 cm^{-3}.

The daily variation of the average mobility is closely connected with the daily variation of temperature which will be dealt with separately.

Table 5

Daily variation of spectral parameters at Tahkuse. First column - beginning of the interval by Moscow Summer Time. υ - number of days covered, t - temperature in °C, η - correlation ratio.

	υ	t	n_+	m_+	$\bar{k}_+$	$\bar{k}'_+$	n_-	m_-	$\bar{k}_-$	$\bar{k}'_-$
0	78	14.3	441	23	1.332	1.406	386	23	1.481	1.562
1	74	13.6	443	23	1.338	1.409	381	22	1.500	1.579
2	75	13,0	437	22	1.343	1.410	384	20	1.510	1.588
3	73	12.5	452	28	1.343	1.409	392	21	1.514	1.588
4	72	12.3	440	23	1.355	1.419	380	22	1.516	1.588
5	71	11.6	437	23	1.356	1.417	376	21	1.526	1.595
6	72	11.5	406	24	1.354	1.414	350	22	1.528	1.597
7	67	11.9	399	23	1.347	1.410	344	20	1.526	1.598
8	57	12.5	307	23	1.336	1.401	341	22	1.504	1.578
9	52	14.5	401	27	1.298	1.371	348	24	1.462	1.546
10	58	15.8	391	26	1.299	1.380	338	21	1.463	1.554
11	61	16.9	385	25	1.293	1.377	334	25	1.467	1.562
12	66	18.0	396	28	1.267	1.354	347	31	1.431	1.544
13	70	18.9	399	29	1.262	1.352	348	30	1.441	1.544
14	72	19.3	392	26	1.269	1.361	343	28	1.460	1.566
15	74	19,7	369	27	1.265	1.360	341	30	1.449	1.557
16	76	19.8	379	27	1.266	1.361	334	30	1.435	1.543
17	77	20.0	381	26	1.263	1.358	338	35	1.424	1.532
18	75	19.7	373	23	1.278	1.373	331	24	1.441	1.548
19	75	18.9	381	21	1.281	1.373	340	21	1.443	1.547
20	78	18.7	387	21	1.285	1.376	344	20	1.438	1.540
21	78	17.7	400	20	1.297	1.384	364	19	1.436	1.532
22	75	16.5	420	21	1.307	1.389	381	19	1.443	1.534
23	74	15.4	444	23	1.309	1.387	405	23	1.444	1.530
η	-	64%	17%	17%	32%	23%	17%	19%	26%	19%

Dependence of mobility on temperature

The dependence of mobility on temperature is described in Table 6.

In the case of ions with stable structure the increase of temperature leads to the growth of natural mobility which is inversely proportional to the density of the air. Actually,

Table 6

Dependence of average mobility
on temperature at Tahkuse

°C	$\bar{k}'_+$	$\bar{k}_+$	$\bar{k}'_-$	$\bar{k}_-$
0-5	1.52	1.48	1.71	1.63
5-10	1.45	1.40	1.62	1.55
10-15	1.40	1.33	1.57	1.49
15-20	1.38	1.29	1.54	1.46
20-25	1.34	1.24	1.51	1.41
25-30	1.32	1.21	1.52	1.39
η	33%	48%	26%	35%

natural mobility decreases with the growth of temperature. This gives evidence of the dependence of the structure of ions on temperature. This dependence may partially be caused by the growth of the concentration of air trace-gases together with the temperature.

The data in Table 6 can be taken to pose a problem of the reduction of mobilities proportionally to the density of the air. To obtain more information, the dependence of the reduced mobility on air pressure was computed. The results shown in Table 7 demonstrate a lack of a linear tendency, at the same time a non-linear tendency is evident.

Table 7

Dependence of average reduced
mobility on air pressure

Pressure	995-1005	1005-1015	1015-1025	1025-1035	hPa
$\bar{k}_+$	1.35	1.30	1.30	1.36	cm²/(V·s)
Frequency	66	795	669	158	

The investigation of the mobilities of one-second aged ions did not reveal a dependence of the reduced mobility on temperature [2]. It seems that the dependence on temperature is caused by admixtures of such low concentrations that the relaxation time of their reactions with ions is substantially over one second.

Linear correlations

The estimate of linear statistical correlations in the set of basic parameters is presented in Table 8. Disregarding the problem discussed above, the data in Table 8 do not contradict the accepted understanding of the regularities of the behaviour of air ions.

Table 8

Estimates of the coefficients of linear correlation
r for the whole period of measurement. Number of
realizations is 1790, and the 99% confidence level
is r_{99} = 6%.

r: %	t	n_+	m_+	$\bar{k}_+$	n_-	m_-	$\bar{k}_-$
t	+100	-12	-7	-49	-10	+2	-39
n_+	-12	+100	+20	-23	+97	+15	-28
m_+	-7	+20	+100	-24	+17	+63	-23
$\bar{k}_+$	-49	-23	-24	+100	-24	-28	+86
n_-	-10	+97	+17	-24	+100	+20	-33
m_-	+2	+15	+63	-28	+20	+100	-37
$\bar{k}_-$	-39	-28	-23	+86	-33	-37	+100

Ionization and aerosol formation

The formation of intermediate air ions is one of the most frequently discussed mechanisms of atmospheric aerosol generation. Especially interesting is the condensation on ions of substances other than water which can form stable particles. Some mechanisms of this process are known [12]. The results of the present measurements clearly demonstrate and quantitatively describe the formation of intermediate ions in the atmosphere which makes it possible to draw several conclusions about aerosol formation.

In earlier measurements of the spectra of one-second aged ions at Tahkuse intermediate ions were not discovered [2]. In a special experiment (22:00. 09.06.85) with the air of the city of Tartu a weak radioactive substance was used, this increased the concentration of small ions to 1500 cm^{-3} but did not lead to the growth of the concentration of intermediate ions which remained at a level of 20 cm^{-3}. This indicates that the concentration of intermediate ions does not grow together with increased ionization rate, but is kept at a certain low level by some limiting factor. This factor can be the concentration of certain trace-gases in the air which is necessary for the formation of intermediate ions. As the effect of intermediate ion formation is almost always saturated at the level of natural ionization rate, it is understandable that artificial ion formation in laboratory conditions does not lead to intense aerosol formation.

The air component controlling the formation of intermediate ions cannot be water, as the amount of water in the air cannot be responsible for the effect of saturation. According to measurement data the concentration of intermediate ions is *not any function of humidity. Spectra with and without* intermediate ions can be accompanied by both foggy and fair weathers. Examples of strong charge asymmetry presented in Fig. 2 point towards the existence of different trace-gases capable of forming intermediate ions of either one or the other polarity. The chemical nature of such trace-gases is *presently unknown. The task of identification of these trace-gases* would be easier, if it were possible to find conditions where intermediate ions are generated with stability. According to [18] it can be supposed that such conditions were in the caves of Carlsbad, USA where average mobilities +0.35 cm^2/(V·s) and -0.50 cm^2/(V·s) were recorded for a sum of small and intermediate ions, whereas the concentrations were above 600 000 cm^{-3}.

In Tartu the average $m_+ = m_- = 57$ cm^{-3} was observed from 04.06.85 to 10.06.85 (for Tahkuse the respective figure was 24 cm^{-3}). The limited measurement period in the city (90 hrs) does not make it possible to draw any conclusions about the increased concentration of intermediate ions in the conditions of higher anthropogenic air pollution.

A quantitative description of statistical distribution of concentration of intermediate ions and charge symmetry are presented in Table 9. On the background of low concentrations detailed in Table 9 there are some conspicuous samples. Some of those are the samples depicted in Fig. 2, where m_+ achieves the value 394 cm^{-3} (a), and m_- 510 cm^{-3} (b).

Above it was assumed that intermediate ions are formed as a result of condensation of substances on small ions. However, there is another hypotesis which claims that particles are formed in a neutral state and only subsequently charged,

Two-dimensional distribution of concentrations of intermediate ions of different polarities with mobilities from 0.32 to 0.5 cm^2/(V·s) at Tahkuse

m_+ \ m_-	<10	10-20	20-30	30-40	40-50	50-60	>60	Σ
<10	22	39	7	3	0	0	0	71
10-20	109	375	106	27	4	1	0	622
20-30	28	232	291	70	16	2	6	645
30-40	2	27	66	70	31	10	4	210
40-50	1	2	6	21	21	15	11	77
50-60	0	0	0	1	7	13	17	38
>60	1	1	1	0	2	5	27	37
Σ	163	676	477	192	81	46	65	1700

as a result of diffusion of small ions to particles similarly to large ions. The mobility range 0.32-0.5 cm^2/(V·s) corresponds to the diameter range 2-2.7 nm. According to the most reliable of the available results [14] the mean probability of the presence of charge of one polarity for this range is about 0.007. If m = 500 (see Fig. 2), then in one cm^3, there should be about 70000 neutral particles of the size 2-2.7 nm. Data in [15] show that the average concentration of such particles in air does not exceed several hundreds in cm^3 which leads to the formation of several intermediate ions in cm^3. The hypothesis of intermediate ion formation from neutral particles also runs into difficulties if it has to be used to explain the observed cases of charge asymmetry.

Be that as it may, the above-said cannot be taken to refute conclusively the hypothesis that a certain part of intermediate ions is formed from neutral particles. It is possible that both mechanisms have their own role in the real atmosphere. The sample in Fig. 2b could have the following hypothetical explanation. Particles arise due to rapid growth of negative small ions. These particles are neutralized in recombination with positive ions and this causes the emergence of an anomalously high quantity of small neutral particles. Due to the diffusion of positive ions to the neutral particles positive ions with the mobility 0.32-0.5 cm^2/(V·s) emerge (see Fig. 2b).

On the basis of the above measurement results and speculative argumentations a double classification of atmospheric ions can be proposed. The traditional classification distinguishes *small*, *intermediate* and *large ions* on the basis of mobility. The conditional limit 0.5 cm^2/(V·s) between small and intermediate ions found empirical proof in this investigation. To increase the exactness of the formal limit further measurements are necessary.

The other classification is based on the physical nature of ions:
- *cluster ions* - particles with the properties of clusters,
- *condensation ions* - particles with the properties of macroscopic bodies, emerging from cluster ions by a growth of their size.
- *aerosol ions* - particles emerging as a result of adsorption of cluster or condensation ions to aerosol particles.

The difference between the physical properties of molecular clusters and those of microscopic bodies has been dealt with e.g. in [16].

The mobility ranges of different groups of ions overlap. If to accept the above explanation of the spectra in Fig. 2b. it can be said that negative ions with mobilities 0.32-0.5 cm^2/(V·s) are condensation ions in this particular case but positive ions in this same mobility range are aerosol ions.

To check the proposed hypothesis and to find out about the role of atmospheric ions in the balance of atmospheric aerosol it is necessary to carry out systematic measurements of atmospheric ion spectra in a wider mobility range.

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