RĒZEKNES TEHNOLOĢIJU AKADĒMIJA

Andris Skromulis

IMPACT OF ANTHROPOGENIC POLLUTION ON AIR ION CONCENTRATION

Summary of the Doctoral Thesis

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Faculty of Engineering

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DOCTORAL THESIS PROPOSED TO RIGA TECHNICAL UNIVERISTY FOR THE PROMOTION TO THE SCIENTIFIC DEGREE OF DOCTOR OF ENVIRONMENTAL ENGINEERING

To be granted the scientific degree of Doctor of Environmental Engineering, the present Doctoral Thesis will be publicly defended on 1 November 2018 at 2 pm at the Faculty of Power and Electrical Engineering of Riga Technical University, Āzenes iela 12/1.

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DECLARATION OF ACADEMIC INTEGRITY

I hereby declare that the Doctoral Thesis submitted for the review to Riga Technical University for the promotion to the scientific degree of Doctor of Environmental Engineering is my own and does not contain any unacknowledged material from any source. I confirm that this Thesis has not been submitted to any other university for the promotion to other scientific degree.

Andris Skromulis ……………………………….. (signature)

Date ………………………..

The Doctoral Thesis has been written in Latvian. It consists of an introduction; three chapters; Conclusion; 51 figures; 24 tables; the total number of pages is 108. The bibliography contains 121 titles.

CONTENTS

TOPICALITY OF THE RESEARCH

Chemical and mechanical air pollution is one of the most crucial problems in scientific studies nowadays. Atmospheric pollution is very hard to be localised or prevented. It can spread not only regionally, but also on a global scale.

The notion of air pollution may not be seen only as content of chemical substances or physical components in the air. The air is environment containing various physical fields. The intensity of electromagnetic fields of different frequency (electrostatic and high frequency fields, as well as light, heat, radioactive radiation and other fields) may exceed the level provided in legislation. In this case, the issue of energy pollution appears. Many of the above-mentioned physical fields have an impact on the level of air ionisation providing the air molecules with the energy they need to overcome the ionisation barrier, which makes the air an environment that continuously contains a certain amount of energy (Sinicina et al., 2013). Consequently, it is possible to speak of the energetic air pollution as a negative phenomenon or to see the energy saturation as a positive feature of the air.

For almost a century, scientific studies have been describing the impact of air ions on human beings and other living organisms (Чижевский, 1969; Rim, 1977). Air ions may have favourable and healing impact or can affect harmfully human health. It depends on their concentration level in the air and on mutual proportions of positive and negative ions.

Over the past two decades, open air studies of air ions have become topical again aiming at understanding the interactions between air ions and other natural biotic and abiotic components that affect not only the ecosystem but even the global climate (Hari, Kulmala, 2005). There is still a lack of relevant information on the impact of anthropogenic pollution on air ions both in urban and rural environments. Up to now, there have not been any significant studies on air ion concentrations in Latvia. There is also a lack of information on concentrations of positive and negative air ions in cities, industrial areas, transport nodes, rural areas and resorts. Knowing both the curative and sometimes negative impact of air ions on human health, getting information about the situation in Latvia is very important and topical.

Information on the dynamics of concentration of air ions of different size and charge marks would allow the development of a complete and comprehensive air monitoring system as well as provide additional criteria for planning urban infrastructure and optimisation of transport flows. By obtaining information on the impact of anthropogenic pollution on air ions and by finding existing interconnections, it is possible to introduce pollution abatement measures and to control air ion concentration, which would in turn contribute to the improvement of overall quality of the living environment.

THE AIM AND TASKS OF THE RESEARCH

The aim of the research: to study the impact of human-created atmospheric pollution on air ions and to elaborate a mathematical model for characterisation of this interaction.

To achieve the aim of the thesis, the following tasks were set and undertaken:

1. Analyse scientific literature and existing studies.
2. Study changes in air ion concentrations both in urban and natural environment.

3. Study relations and interaction between air ions and air pollutants.
4. Study changes of air ion types and concentration depending on various external factors.
5. Elaborate a mathematical model for description of changes of air ion concentration depending on chemical and physical parameters of the air.
6. Give recommendations for the improvement of air quality monitoring.

Originality of the research

1. It is the first long-term research on air ion concentration and dynamics of its changes in various areas of Rezekne city.
2. Seasonal fluctuations of light air ion concentration in one of Latvian cities were studied for the first time.
3. It is the first study in Latvia where the light air ion concentration has been studied in flue gases of various fuels.
4. Basing on outdoor measurements, interaction of air physical and chemical pollution, meteorological parameters and concentration ofa wide spectrum of air ions was analysed for the first time.
5. It is the first time when a basis has been developed for a possible mathematical model that could help in assessing and forecasting the concentration of air ions of different classes depending on external environmental factors, basing on non-laboratory studies.

Thesis statements set up for defence

1. Anthropogenic pollution significantly affects air ion concentration.
2. Major impact of anthropogenic pollution is decrease in concentration of negative cluster ions.
3. CO, NO and NO_2 are the main chemical air pollutants affecting air ion concentration in urban environment.
4. In order to improve the air quality monitoring in populated areas, it should also involve measurements of air ion concentration.

APPROBATION OF THE SCIENTIFIC WORK

For approbation of the scientific work, 11 scientific papers have been published in peer-reviewed scientific journals. The results have been reported at international conferences in Latvia, Estonia, Bulgaria, Macedonia, and Turkey.

The results of the work have been reported and discussed at 16 scientific and scientific-practical conferences.

Oral reports

1. Gaisa kvalitātes indikatoru kompleksa pamatojums. The 6^{th} Scientific-practical conference “Nature. Technology. Resources”. Rezekne, Latvia, 26 June 2009.
2. Gaisa enerģētiskā stāvokļa ietekmes analīze uz pilsētu infrastruktūru. Ilgtspējīga telpiskā attīstība. RTU, Riga, 20 November 2009.
3. Gaisa enerģētiskā stāvokļa izvērtējums Rēzeknes pilsētā. The 68th scientific conference of University of Latvia, Riga, Latvia, 4 February 2010.
4. Research of Changes of Ionization and Meteorological Factors of Atmospheric Air in Rezekne City, Latvia. Conference on water observation and information

system for decision support Balwois 2010. Republic of Macedonia, Ohrid, 25–29 May 2010.

5. Research of Atmospheric Light Air Ion Concentration and Meteorological Factor Changes in Rezekne City, Latvia. The 2nd International Geography Symposium GEOMED 2010. Antalya/Kemer, Turkey, 2–5 June 2010.
6. Air Ion Measurements in Rezekne, Latvia. The 14th Finnish-Estonian air ion and aerosol workshop, Pühajärve, Estonia, 14–16 June 2010.
7. Evaluation of Air Ionisation Level in Rezekne City. The 4th international conference "Environmental Science and Education in Latvia and Europe: From Green Projects to Green Society", Jelgava, Latvia, 22 October 2010.
8. Comparison of Air Ionisation Level in Different Ecosystems. Symposium "Ecology & Safety" for a cleaner and safer world, Varna, Bulgaria, 4–8 June 2011.
9. Complex Evaluation of Air Quality in Rezekne. The 8th scientific-practical conference "Nature. Technology. Resources". Rezekne, Latvia, 20–22 June 2011.
10. Air Ion Measurements in Rezekne. Joint MUSCATEN – ABBA workshop on Biogenic VOC: emissions, aerosol formation, modelling. Tartu, Estonia, 9–21 October 2011.
11. Method for Forecasting the Number of Air Cluster Ions. The 17th Finnish-Estonian Air Ion and Aerosol Workshop. Hyytiälä, Finland, 11–12 June 2013.
12. Impact of Microclimate and Indoor Plants on Cluster Ions. The 17th Finnish-Estonian Air Ion and Aerosol Workshop. Hyytiälä, Finland, 11–12 June 2013.
13. Impact of Microclimate and Indoor Plants on Air Ion Concentration. The 9th scientific-practical conference "Nature. Technology. Resources". Rezekne, Latvia, 20–22 June 2013.
14. Amount of Air Ions Depending on Indoor Plant Activity. The 10th scientific-practical conference "Nature. Technology. Resources". Rezekne, Latvia, 18–20 June 2015.
15. Effect of Environmental Factors on Air Ion Concentration. The 10th scientific-practical conference "Nature. Technology. Resources". Rezekne, Latvia, 18–20 June 2015.
16. Effect of Atmospheric Pollution on Air Ion Concentration. "CONECT" Conference of Environmental and Climate Technologies. RTU, Riga, Latvia, 12–14 October 2016.

The results of the research have been reported in 11 publications in Latvian and English.

International publication in peer-reviewed journals

1. Skromulis, A.(2010) Research of Changes of Ionization and Meteorological Factors of Atmospheric Air in Rezekne City, Latvia. In: Conference on Water Observation and Information System for Decision Support: Proceedings of the Conference BALWOIS 2010. Ohrid, Republic of Macedonia, 2010. Published at: http://www.balwois.com/balwois/administration/full_paper/ffp-1646.pdf

2. Skromulis, A., Noviks, G. (2011) Comparison of Air Ionization Levels in Different Ecosystems. In: Journal of International Scientific Publications "Ecology & Safety", Volume 5, Part 1. Info Invest, Bulgaria, 2011, pp. 21–31. Published at: http://www.science-journals.eu

International publications in peer-reviewed journals indexed in the database "Scopus"

1. Noviks, G., Skromulis ,A. (2009) Gaisa vides kvalitātes monitoringa pilnveidošanas iespēju analīze. In: Environment. Technology. Resources: Proceedings of the 7th International Scientific and Practical Conference. Volume 1. Rezekne Higher Education Institution, Rezekne, 2009, pp. 273–283.
2. Skromulis, A., Matisovs, I., Noviks, G. (2011) Complex Evaluation of Air Quality in Rezekne. In: Environment. Technology. Resources: Proceedings of the 8th International Scientific and Practical Conference. (www.ru.lv) Volume 1. Rezekne Higher Education Institution, Rezekne, 2011, pp. 160–168.
3. Manninen, H. E., Franchin, A., Schobesberger, S., Hirsikko, A., Hakala, J., Skromulis, A., Kangasluoma, J., Ehn, M., Junninen, H., Mirme, A., Mrme, S., Sipilä, M., Petäjä, T., Worsnop, D. R., Kulmala, M. (2011) Characterization of Corona-Generated Ions Used in a Neutral cluster and Air Ion Spectrometer (NAIS). In: Atmospheric Measurement Techniques, No. 4, pp. 2767–2776. ISSN 1867–1381, eISSN 1867–8548.
4. Skromulis, A., Noviks G. (2012) Atmospheric Light Air Ion Concentrations and Related Meteorologic Factors in Rezekne City, Latvia. In: Journal of Environmental Biology, No. 4, pp. ISSN: 0254–8704; CODEN: JEBIDP
5. Sinicina N., Skromulis, A., Martinovs, A. (2013) Impact of Microclimate and Indoor Plants on Air Ion Concentration. Proceedings of the 9th International Scientific and Practical Conference "Environment. Technology. Resources". ISSN 1691–5402 (SCOPUS).
6. Sinicina, N., Skromulis, A., Martinovs, A. (2015) Amount of Air Ions Depending on Indoor Plant Activity. Proceedings of the 10th International Scientific and Practical Conference "Environment. Technology. Resources", Volume II, pp. 267–273. ISSN 1691–5402 (SCOPUS).
7. Skromulis, A., Breidaks, J. (2015) Effect of Environmental Factors on Air Ion Concentration. Proceedings of the 10th International Scientific and Practical Conference "Environment. Technology. Resources", Volume II, pp. 274–279. ISSN 1691–5402 (SCOPUS).
8. Skromulis, A., Breidaks, J., Teirumnieks, E. (2017) Effect of Atmospheric Pollution on Air Ion Concentration. In: Energy Procedia, 113, pp. 231–237. ISSN 1876–6102 (SCOPUS).

Other international publications

1. Skromulis, A., Grigorjeva, J. (2010) Gaisa enerģētiskā stāvokļa ietekme uz pilsētu infrastruktūru. In: Scientific Journal of Riga Technical University "Sustainable Spatial Development", Volume 1. Rezekne Higher Education Institution, Rezekne, 2010, pp. 89–93.

1. THEORETICAL BACKGROUND

In atmospheric physics the term "air ion" means any particle with an electric charge located in the air and ensuring electrical conductivity of the air (Dolezalek et al., 1985; Tammet, 1998). In a narrower sense, the term "air ion" means any charged particle in the air whose size is 0.36 to 79 nm and whose mobility is 1.3 to 0.0042 $cm^2 \cdot V^{-1} \cdot s^{-1}$, i.e. from monomolecular ion migrating in the air to aerosol particle with no definite upper limit. It is usually considered that the characteristics of this upper limit is particle size of 79 nm and mobility of 0.00041 $cm^2 \cdot V^{-1} \cdot s^{-1}$. All charged particles within these limits are research subjects in outdoor studies of air ions of natural origin. Specifics of modern devices used for ion measurements also comply with these limits. (Tammet, 2011)

Recently, a classification has been created based on the physical structure of air ions (Horak, 2001). Cluster ions consist exclusively of ionized and polarized gas molecules that form clusters, while the heaviest ions are aerosol ions that are made not only of ionized and polarized gas molecules, but also of atmospheric aerosol particles such as ion condensation kernels.

Table 1.1

Classification of air ions (Horrak, 2001)

Air ion class		Diameter, nm	Mobility, $cm^2 \cdot V^{-1} \cdot s^{-1}$
Cluster ions	Small cluster ions	0.36–0.85	1.33–3.2
	Large cluster ions	0.85–1.6	0.5–1.3
Aerosol ions	Middle-sized ions	1.6–7.4	0.034–0.5
	Light large ions	7.4–22	0.0042–0.034
	Heavy large ions	22–79	0.00041–0.0042

Physical characteristics of the above mentioned air ion classes are as follows.

- Small cluster ions. Their mobility is 1.3 to 3.2 $cm^2 \cdot V^{-1} \cdot s^{-1}$, diameter 0.36 to 0.85 nm, mass 30 to 400 u, typical life expectancy 5 to 60 s. Given the diameters of this ion class, it may be considered that the cluster kernel contains one non-organic molecule surrounded by a one-molecule layer of water molecules. After recombination, small cluster ions are decomposed into initial components, i.e. non-organic molecule and water molecules.
- Large cluster ions. Their mobility is 0.5 to1.3 $cm^2 \cdot V^{-1} \cdot s^{-1}$, diameter 0.85 to 1.6 nm, mass 400 to 2500 u, typical life expectancy 5 to 60 s. The cluster kernel may consist of one organic molecule surrounded by a one-molecule layer of water molecules. Elevated concentration of this ion class in atmosphere can be observed when concentration of large ions is low. It prolongs the life expectancy of this ion class, thus making it possible to achieve bigger ion sizes.
- Middle-sized ions. Their mobility is 0.034 to 0.5 $cm^2 \cdot V^{-1} \cdot s^{-1}$, diameter 1.6 to 7.4 nm. This ion class complies with the aerosol class of small nanometre-size particles (14 to 722 nm (Despiau and Croci, 2007)). Some of middle-sized ions are products of ion-induced nucleation, resulting from condensing of nucleation steam onto cluster ions whose size increases to the size of middle-sized ions. They are called primary aerosol ions. Secondary aerosol ions are formed when cluster ions are connected with neutral aerosol particles.
- Light large ions. Their mobility is 0.0042 to 0.034 $cm^2 \cdot V^{-1} \cdot s^{-1}$, diameter 7.4 to 22 nm. This air ion class complies with the aerosol class of ultrafine particles or

big (coarse) nanometre particles. Ions of this class have one charge and often it is in balance with the charge of stochastic cluster ions.

- Heavy large ions. Their mobility is less than 0.0042 $cm^2 \cdot V^{-1} \cdot s^{-1}$, diameter exceeds 22 nm. Aerosol class corresponding with these ions is called the class of Aitken particles. These ions are almost in balance with the stochastic charge of cluster ions, and some ions of this class may have several charges.

Basically, chemical composition of air ions depends on two factors: chemical composition of the atmosphere and ionisation barrier of its components. The higher the concentration of any specific gas in the atmosphere, the higher the possibility for air ions to contain molecules of this particular gas. The lower the ionisation barrier of a specific gas, the higher the possibility for a molecule of this particular gas to be ionised. This statement can be applied more to small cluster ions than to aerosol ions because, when speaking about aerosol ions, we should take into account possible chemical reactions between air ions and aerosols-condensation kernels. Therefore, aerosol ions have the highest chemical variety and their composition is difficult to identify.

Ionisation potential or ionisation energy is the minimum necessary energy that, when conveyed to an atom or molecule, enables an outer shell electron to overcome the ionisation barrier and to leave the atom that becomes a positive ion. Electron affinity, in turn, is the amount of released energy, which is formed when an atom or molecule gains electron and becomes a negative ion. The higher the electron affinity, the higher the possibility for an atom or molecule to become an electron acceptor. On the other hand, the lower the affinity, the higher the possibility that the atom or molecule shall lose an electron. These and some other physical-chemical parameters of gaseous components of the air are summarised in Table 1.2, assuming that 1 cm^3 of the air contains $2.7 \cdot 10^{19}$ of molecules (Parts, Luts 2006).

Table 1.2

Physical-chemical properties of chemical components of the air

Title	Chem. formula	Molecular weight, amu	Ionisation energy, eV	Electron affinity, eV	Concentration in the atmosphere, atoms in cm^3
Nitrogen	N_2	28	15.58	-	$2.1 \cdot 10^{19}$
Oxygen	O_2	32	12.07	0.45	$5.6 \cdot 10^{18}$
Carbon dioxide	CO_2	44	13.77	-	$9.0 \cdot 10^{15}$
Sulphur dioxide	SO_2	64	12.32	1.1	$1.0 \cdot 10^{9}$
Nitrogen dioxide	NO_2	46	9.75	2.27	$7.0 \cdot 10^{10}$
Water	H_2O	18	12.61	0.9	
OH-radical	OH	17	13.00	1.82	$1.6 \cdot 10^{6}$
NO_3-radical	NO_3	62	11.9 (N_2O_5)	3.94	
Chlorine	Cl_2	71	11.48	2.38	$1.2 \cdot 10^{10}$
Bromine	Br_2	160	10.15	2.55	
Iodine	I_2	254	9.40	2.55	

Air ion properties and related processes in the ground-level atmosphere may be described using mathematical relationships that mainly involve ion mobility and recombination coefficient. Ion mobility*k*is defined as the average speed of ion mobility in one unit of electric field. It is easier to detect air ion mobility in the ground-level atmosphere than its mass and chemical composition (there is no universal correlation between ion mobility and mass). Average air ion mobility in the ground-level atmosphere is 1.15 to 1.25 $cm^2 \cdot V^{-1} \cdot s^{-1}$ (Mohnen, 1977). Air ion mobility is inversely proportional to the air density and does not depend on the electric field intensity. Ideally, if ions are formed in pairs under the influence of ionizing radiation and are anihilated in pairs in the process of ion-ion recombination, the concentration of negative and positive ions in the air is equal and the amount of ions can be expressed by the following formula:

$$n_{\pm} = (q / \alpha)^{1/2} \tag{1.1}$$

where
$n_{\pm}$– the amount of positive and negative ions in 1 cm^3;
q – degree of ionisation;
α – recombination coefficient.

Degree of ionisation is the number of ion pairs formed in 1 cm^3 of air, 1 s. Recombination coefficient is the number of ion pairs lost during recombination of positive and negative ions in 1 cm^3 of air, 1 s. In case there are no aerosols in the atmosphere, life expectancy of ions can be expressed by the following formula:

$$\tau = n_{\pm} / q = (1 / q\alpha)^{1/2} , \tag{1.2}$$

where τ is the life expectancy of ions in the atmosphere, s.

Actually, in the troposphere, ion recombination is a “three particle” process, because recombination involves neutral aerosols.

Ion movement in the electric field ensures atmospheric conductivity described by the following equation:

$$\lambda = e(n_{+}k_{+} + n_{-}k_{-}) , \tag{1.3}$$

where λ is atmospheric conductivity, e is elementary charge, and $k_{\pm}$ is air ion mobility.

Based on the above mathematical equations, air ion concentration near the land surface should be around 3000 ion/cm^3, their life expectancy around 5 min, but the atmospheric electrical conductivity should be approximately $1 \cdot 10^{-13}$ Ω^{-1} if the degree of ionization is 10 ion pairs per $cm^{-3}s^{-1}$. In reality, these values are significantly lower because the ions are destroyed not only during the process of recombination, but also by joining neutral aerosol particles.

Ground-level atmosphere contains many suspended fine particles with a typical radius of 0.01 to 0.05 μm. Concentration of aerosol particles varies from a few hundred per cubic centimetre in remote areas above the oceans to 100000 particles/cm^3 in polluted urban environments. Air ions are diffused onto aerosol particles and, through contact, give them their charge. Thus, aerosol particles act as ion recombination centres. In continental areas, air ion losses are usually caused by the contact with aerosol particles, not by the direct recombination process of ions with opposite charges. Contact of air ions with aerosol particles also ensures the transfer of static charge to aerosols.

To fully describe interaction between air ions and aerosols and the transfer of charges, it would be necessary to have a balanced equation system for ions and aerosol particles with different amount of elementary particles. However, in order to describe only the effect of air ions on the decrease in the number of ions, the following ion balance equation may be applied:

$$\frac{dn}{dt} = q - \alpha n^2 - \beta n Z \qquad (1.4)$$

where Z is the total concentration of charges, regardless of the charge of the specific particle, and β is ion-aerosol effective contact rate.

Under real conditions, if the concentration of aerosol particles exceeds 1000 per cm^3, ion concentration in the ground-level atmosphere is more affected by the exposure to aerosols than by ion recombination.

If aerosol particles are hygroscopic, their average radius increases when the level of air relative humidity is higher. Such increase in the size of particles is particularly visible when relative humidity exceeds 90 %. If particles become bigger, air ions are more exposed to such particles, which lowers air ion concentration thus decreasing electrical conductivity of the air. When relative humidity of the air exceeds 100 %, the radius of some aerosol particles immediately increases even 2 times, reaching more than 1 µm, and creates droplets of mist and clouds. These droplets are the most effective attractors of air ions that cause low atmospheric electrical conductivity in the fog and clouds. The process of particle “growth” greatly enhances the phenomenon in which the atmospheric conductivity decreases, while the intensity of the electric field increases shortly before the formation of the fog.

The main source of ions in the atmosphere is ionising radiation. As a result of radioactive ionisation, the electron is detached from the oxygen or nitrogen molecule. The detached electron immediately joins a neutral molecule and forms a negative ion. Within a few milliseconds, the positive and negative ion undergoes a series of chemical, charge-exchange and cluster-forming reactions with molecules of gas in the air. Cosmic radiation is the main source of ions in areas above the oceans and at a few kilometres high atmospheric layers, while, in the ground-level atmosphere, the impact of cosmic radiation on the formation of air ions is only 1 to 2 ion pairs per cubic centimetre a second. Besides this variable is relatively constant all the time (Hoppel et al., 1986). In the ground-based atmosphere above dryland, the main source of ions is the natural radioactivity of rocks of the earth's crust. This source of ionisation can be divided into 2 groups:

1) radioactive radiation of particles α, β and γ straight from the land cover;
2) radioactive gas emanations from the earth and degradation products of these gases.

Figure 1.1 shows vertical changes of ionisation levels both in ground-level atmosphere and in its higher layers. Ionisation level, due to the cosmic radiation, remains almost constant from the ground surface to the height of several kilometres. Ionisation level resulting from the Earth’s β and γ radiationis variable geographically, depending on the concentration of radioactive elements in the soil. Up to the height of 1 mionisation is determined primarily by the concentration of β particles, but above this level, up to several hundred meters high, by γ particles. The curves Q_{max} and Q_{min} represent the aggregate ionisation caused by cosmic radiation, β and γ particles from the Earth's crust, as well as ^{220}Rn, ^{222}Rn and their decomposition products in the atmosphere. The marked area between the two curves shows the probability of variation of the degree of ionization depending on the atmospheric and geological conditions at the respective geographical point. The high degree of ionization up to 1 m (curve Q_{max}) is possible due to the accumulation of ^{220}Rn, under conditions of windless weather and pronounced inversion of the temperature near the Earth's surface. Under normal conditions, when there is convection in the atmosphere, the degree of ionisation approaches a Q_{min} curve at a height of 10 m. Above the oceans and far away from dryland, the degree of ionisation is determined only by cosmic radiation. Theoretically, the advection of ^{222}Rn above the oceans is possible even at a height of 100 km, but its impact on ionisation is insignificant compared to the background created by cosmic rays.

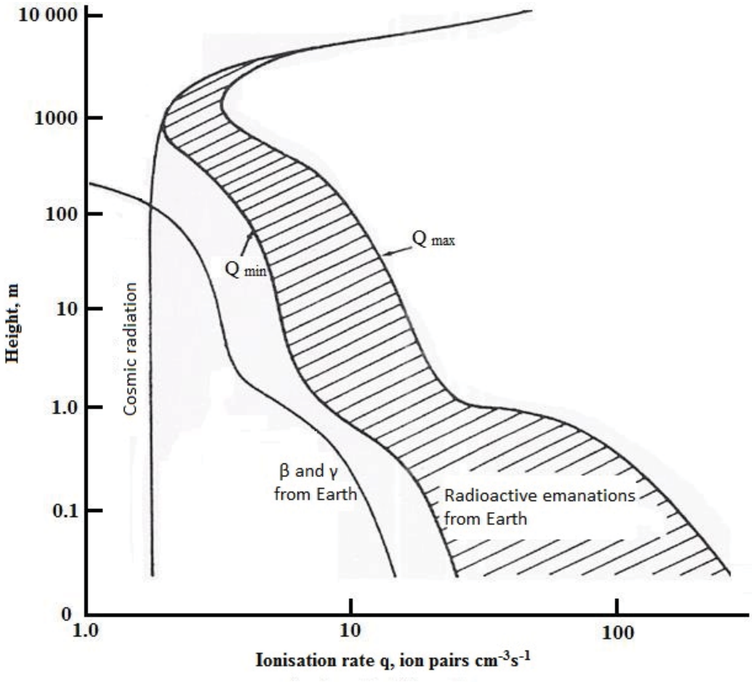


Fig.1.1. Vertical profile of the atmosphere ionisation intensity.

Hydro ionisation or Lenard effect is a process resulting in the formation of two types of ions: atmospheric (air ions) and hydro ions. Instead of ionising radiation energy, hydro ionisation requires mechanical energy that causes water spraying in the form of tiny water droplets. Due to fluctuations, charges may be concentrated even in a very small volume of water (Iribarne and Thomson, 1976).

At least in some of the falling water droplets,accumulation of free charges takes place in different areas of the particular droplet due to auto-ionisation, charge transfer or surface deformation because the droplet always has its specific shape with a sharper "tail".Collision of falling droplets results in their fragmentation, i.e. splitting into even smaller droplets, which often takes place by loosing areas where charges are concentrated. Generation of negative air ions is greatly influenced by the Coulomb explosion, which is a process where water molecules (droplet fragment), moving with a high speed, hit a solid substance; because of the strong collision, electrons of water molecules are thrown away from orbits which turns molecules into clusters of charged atom components that immediately break down because of electrostatic (Coulomb) forces of repulsion. In such case, in order to overcome the ionisation barrier, sufficient mechanical energy is supplied when a water droplet hits rocks, stones or other hard surfaces. Electrons or OH^- ions detached during the Coulomb explosion join small clusters of water molecules, stay for a period of time in the air near waterfalls and give their charge to other atmospheric aerosols thus creating negative air ions of various sizes.Evaporation of negatively and positively charged water molecules is possible both from water droplets and from their fragments, and such molecules join air ions of average size. It should be noted that fragments of water droplets subjected to the possibility of evaporation and to the Coulomb explosion contain slightly more negative ions because the positive charge left over after different droplet collisions is mainly concentrated in the heaviest water droplets that enter the earth or get back into the water.

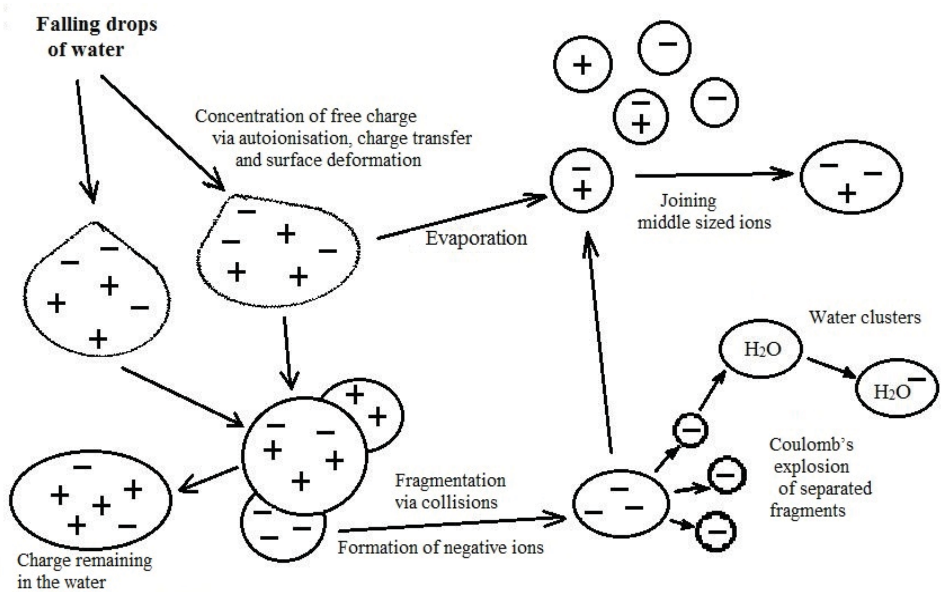


Fig. 1.2. Model of hydro ionisation process.

Table 1.3

Comparison of the impact of positive and negative air ions on human health (Deoux, 1993)

Negative air ions	Positive air ions
- Reduced serotonin levels in the blood; - Increased glucocorticoid levels in adrenal glands; - Better functioning of thyroid gland; - Better ovarian functioning; - Better testicular functioning; - Deep, persistent sleep; - Reduced aggression and anxiety; - Enhanced learning and memory; - Regulated arterial blood pressure; - Increased tolerance to pain; - Improved brain function.	- Increased serotonin levels in the blood; - More pronounced corticoid mineralisation in adrenal glands; - Slower thyroid gland function; - Reduced ovarian function; - Reduced vigilance and concentration capacity; - Light, shallow sleep; - Increased aggression and anxiety; - Impaired memory; - Reduced tolerance to pain.

Air ionsaffect awiderange ofwildlife, such asmicroorganisms, animals, plants, human beings, as well asecosystems andevenglobalclimate.The impact of air ions on the environment and human body depends not only on the level of air ion concentration but also on the ratio of ion polarities, which is characterized by the unipolarity coefficient K that expresses the concentration of positive ions in relation to the concentration of negative ions in 1 cm^3 of air (Sinicina et al., 2015).

$$K = \frac{n^+}{n^-} , \quad (1.5)$$

Where K is unipolarity coefficient,n^+ is concentration of positive cluster ions per 1 cm^3, and n^- is concentration of negative cluster ions per 1 cm^3.

Overall, human health is favourably affected by the negative air ions and harmfully by thepositive ones. Negative oxygen ions absorbed by human body through breathing immediately start taking part in the formation of oxygen partial pressure thus affecting all life processes in the body.

2. MATERIALS AND METHODS

2.1. Outdoor measurements of air ion concentration

Outdoor measurements of air ion concentration are important for better understanding the impact of geophysical, geochemical, meteorological, relief and other aspects on air ion concentration in a particular area. As the effects of all these conditions are unique in each location, it also contributes to the development of a different, site-specific level of air ion concentration.

Table 2.1

Short description of sites chosen for air ion measurements in Rezekne city

No.	Site	Short description
1.	Rezekne Meat Factory	The territory of Rezekne Meat Factory is not considered to be highly polluted, but there is often a specific odour. An important street crossroad and a railway line are nearby.
2.	Pharmacy “Lana”	Parking near the pharmacy “Lana” is located in the city centre. There is also the only air monitoring station of Rezekne located nearby.
3.	Latgales Street	The crossroad of Latgales and Atbrivosanas Streets is the busiest in Rezekne. In the mornings and evenings there are sometimes traffic jams. The central bus station is located nearby.
4.	Miera Street	Residential district opposite the graveyard at Miera Street is quiet, with minimal transport intensity.
5.	Tuberculosis hospital	The area is similar to a park with various tree species. There is almost no traffic.
6.	Railway station “Rezekne-2”	The territory of the railway station “Rezekne-2” is considered contaminated not only in terms of air but also in terms of soil and water. There is an active movement of goods and passenger trains. One of the main streets of Rezekne city is also nearby.
7.	Rezekne Secondary School No. 5	This measurement point represents air pollution in the residential district of the northern part of Rezekne.
8.	“REBIR”	Parking near the factory “REBIR” represents air pollution in the industrial zone of Rezekne.
9.	Bakery “Hanzasmaiznīca”	This measurement point is located near the northern border of the city, near the main street.
10.	Rupnicas Street	The measurement point is located near the crossroads of Rupnicas and Atbrivosanas Street in the territory of former Milk Factory.
11.	Monument “VienotiLatvijai”	The monument is the symbolic centre of the city. It is located on the main street and the traffic intensity around it is average.
12.	State Blood Donor Centre, Latgale branch	Latgale Branch of the State Blood Donor Centre is located in a peaceful and quiet place near the river Rezekne.

In order to assess the overall amount of air ions in Rezekne city and to obtain information on its daily or seasonal fluctuations and on the impact of anthropogenic pollution, measurements of the concentration of light air ions were carried out in different areas of Rezekne city. Measurements were performed from March 2009 until March 2012. They were carried out for a period of five days in each season, i.e. winter, spring, summer and autumn, thrice a day: in the morning (7:00 to 10:00), afternoon (12:00 to 14:00) and evening (17:00 to 19:00) local time. Measurement points in Rezekne city were chosen with the aim of covering the widest possible area with the most diverse effects of anthropogenic pollution on the air quality. To carry out the measurements, the author used the portable bipolar air ion counter "Sapfir-3M". This device provides simultaneous measurements of positive and negative air ions with a minimum resolution of 10 ions per cm^3. It measures the concentration of air ions with a mobility of $k \geq 0{,}4 cm^2 \cdot V^{-1} \cdot s^{-1}$in 1 cm^3 of air. Such interval of mobility is approximately equivalent to the class of cluster ions (Horak, 2001). At each measurement point, average values of 16 s were recorded 10 times consecutively, from which, by mathematical processing, the average concentration of positive and negative air ions at the given measurement point and at the given time was calculated. Each time the measuring device was placedso that the centre of the aspiration chamber was about 1.5 m above the ground.

On 4 September 2011, air ion concentration in exhaust gases of a vehicle was measured. The car chosen for the measurements was "Ford Escort" of 1991 with 1.4 l petrol engine. The air ion counter "Sapfir-3M" was positioned on the ground (mown lawn), on a wooden pallet, 10 cm away from the exhaust pipe, perpendicularly to the flow of flue gases, so as to get as much flue gases as possible into the counter. At the beginning of the experiment, the background concentration was measured for 10 minutes, then the engine was started and it operated at idle speed of 1000 rpm for 10 minutes. For the last 10 minutes of the experiment the engine was switched off and the background concentration of light air ions was measured again. The experiment was carried out in a remote rural area far from roads and industrial structures, therefore, when measuring background concentrations, the effect of anthropogenic factors on air ions could be ignored. At the same time the ambient temperature and relative humidity were recorded as well.

In order to study the concentration of air ions in different locations and to analyse the factors affecting it, outdoor measurements were carried out in October 2010 using the portable air ion counter "Sapfir-3M". Three types of ecosystems were selected for measurements: city ecosystem and two forest ecosystems, where in one case it was a northern coniferous (boreal) forest, and in two other cases it was a suburban forest. In all cases, the concentration of air ions was measured for 30 minutes. The ion counter was always placed at a height of about 0.5 m above the ground.

To analyse the potential dependence of air ion concentrations on various anthropogenic pollutants, the author analysed data from the Estonian Environmental Research Centre's air monitoring station, located to the Southwest from the city centre of Tartu. It also has a "Neutral Cluster and Air Ion Spectrometer NAIS", which provides measurement of concentrations of air ions and neutral aerosols according to their masses. It has 16 positive and 16 negative ion channels, as well as 16 neutral aerosol channels (Mirme et al., 2007). As a result, the monitoring station provides the measurement of the whole spectrum of air ions and neutral aerosols, as well as the measurement of NO, NO_2, NO_x, CO, SO_2, O_3, air temperature, relative humidity φ, wind direction and velocity, atmospheric pressure P, PM 10 and PM 2.5. All devices used for measurements of chemical pollution and meteorological parameters are certified.

Table 2.2

Short description of the places of measurement

Place of measure ment	Type	Short description	Date of measuring
Rezekne	City	Crossroads of Atbrivosanas and Latgales Street (see Table 2.1).	04.10.2010
Ancupani	Suburban forest	Mixed forest, dominated by coniferous trees, located in the northern part of Rezekne, poorly maintained but used for recreation.	04.10.2010
Riga	City	Measurements were carried out at Brivibas Street 71 opposite the air monitoring station of Riga City Council. Traffic intensity – moderate.	03.10.2010
Mezaparks	Suburban forest	Mixed forest, prevalence of coniferous trees, well-maintained, used for recreation.	03.10.2010
Helsinki	City	Crossroads of Mannerheimintie and Arkadiankatu Street, traversed by 39,000 transport units a day on average. There is also a railway station nearby.	02.10.2010
Hyytiälä	Boreal forest	Natural forest with a homogeneous growth of pine species *pinussylvestris*, located 60 km from Tampere in a flat, rocky area. Measurements were performed next to the atmosphere monitoring station "SMEAR II".	01.10.2010

The "Neutral Cluster and Aerosol Spectrometer NAIS" is a large-scale scientific measuring device. By combining its different particle chargers and electro filters, it is possible to use at least six operating modes, including the possibility of using artificial air ionisation, however, at the monitoring station, the spectrometer "NAIS" works only in the mode appropriate for measuring natural air ions. In order to determine mathematical relationships between air ions and other air parameters applying the stepwise regression method, the author used the results of measurements carried out from 11/03/2011 to 27/04/2012.

2.2. Indoor measurements of air ion concentration

From 14 to 16 November 2011 air ion measurements were carried out in both natural and artificial ionisation conditions at the Atmospheric Physics Laboratory of the University of Tartu. Two measuring devices were used: the portable air ion counter "Sapfir-3M" and air ion spectrometer "SIGMA" (*Symmetric Inclined Grid Mobility Analyser*) designed for the studies of spectra of atmospheric aerosol and aerosol masses, appropriate for particle sizes of 0.4–7.5 nm and corresponding mobilities of 0.032 to 3.2 $cm^2 V^{-1} \cdot s^{-1}$. "SIGMA" and "Sapfir-3M" were used simultaneously to measure the air ion concentration.The following experiments were conducted.

Experiment 1. Comparative measurements of "Sapfir-3M" and "SIGMA" in laboratory conditions without additional ionisation. Radon concentration was recorded as well.

Experiment 2. Comparative measurements of "Sapfir-3M" and "SIGMA" in laboratory conditions, using the isotope ^{239}Pu as the ioniser, through which compressed air was released at a different speed, thus achieving various degrees of artificial ionisation.

Experiment 3. Comparative measurements of "Sapfir-3M" and "SIGMA" under conditions of increased radon concentration. The experiment was conducted in the basement of the Institute of Environmental Physics at the University of Tartu, in a closed space where radon gas was being released. The experiment was performed for almost 48 hours with a constant increase of radon concentration and exceeding the maximum allowable concentration ten times.

3. RESULTS AND DISCUSSION

Summarising the results of measurements of light air ions or cluster ions in Rezekne city for three years, the following average concentrations were calculated: 262 negative ions per cm^3 and 233 positive ions per cm^3, which results in unipolarity coefficient $K = 0.89$. These figures were obtained by calculating the average value of all systematic measurements made in Rezekne city at all measuring points and in all seasons of the year. Such degree of ionisation is typical for urbanized areas where the concentration of air ions, regardless of the charge sign, is in the range of 100 to 500 ions per cm^3. It is true that the average concentration of negative ions is slightly higher than the concentration of positive ions, which is good from the point of view of human health, because negative cluster ions have a beneficial impact on human beings. Nevertheless, it cannot be stated that the entire city of Rezekne is a favourable place for health because the concentration of air ions is subject to considerable fluctuations and variability in time and space, as discussed in the following sections.

3.1. Particularities of ionisation level in different areas of Rezekne city

The highest value of the coefficient K was found at the measuring points with the highest level of air pollution. Overall, the following results were obtained: railway station "Rezekne-2": $K = 1.12$; Latgales Street: $K = 1.07$; Rezekne Meat Factory: $K = 0.99$. At both measuring points with the lowest level of air pollution, i.e. Miera Street and Tuberculosis Hospital, the unipolarity coefficient is the lowest ($K = 0.77$). At other measurement points the value of K varies within these limits, respectively, near the pharmacy "Lana" and near the factory "REBIR" $K = 0.82$, while near the Secondary school No. 5 $K = 0.86$. The highest concentration of air ions was found near the Secondary school No. 5 (324 negative and 278 positive ions per cm^3), while the lowest concentration was found on Latgales Street, i.e. 207 negative ions per cm^{-3}, and near the Tuberculosis Hospital, i.e. 207 positive ions per cm^3. On Miera Street and near the Tuberculosis Hospital where air pollution is minimal, relatively high levels of negative ions and relatively low concentrations of positive ions were observed.

Measurement results show the impact of urban anthropogenic air pollution on the reduction of air ion concentration. From the given results it cannot be concluded that the pollution would cause the formation of more positive air ions than the negative ones. The effect of pollution is rather manifested in a greater loss of air ions when they settle on the particles of pollution aerosols and give them their own charge. Particles with opposing charges can then coagulate in larger aggregates whose sedimentation is possible under the influence of gravitational forces. This can explain the role of air ions in purification of contaminated air. The imbalance between positive and negative air ions indicates that negative air ions are more rapidly consumed in the process of air purification than the positive ones. Perhaps this is due to the slightly higher mobility of negative air ions. Another possible mechanism reducing the concentration of cluster ions is the formation of larger ions after the negative air ions have joined the molecules of pollutant gases. Some pollutants have a high degree of electron affinity, i.e. for NO_2 it is 2.27 eV.

3.2. Seasonal variations of ionisation level

The level of ionisation can be considerably various over different seasons. The total ionisation level is the lowest in winter and the highest in autumn, although, at some measurement points, higher concentrations of air ions were observed in spring. Consequently, air temperature is not the decisive factor in the promotion of ionisation in the ground-level atmosphere, because in summer the concentration of negative air ions is only slightly increasing compared to spring, while the concentration of positive ions in summer is even lower than in spring and in autumn.

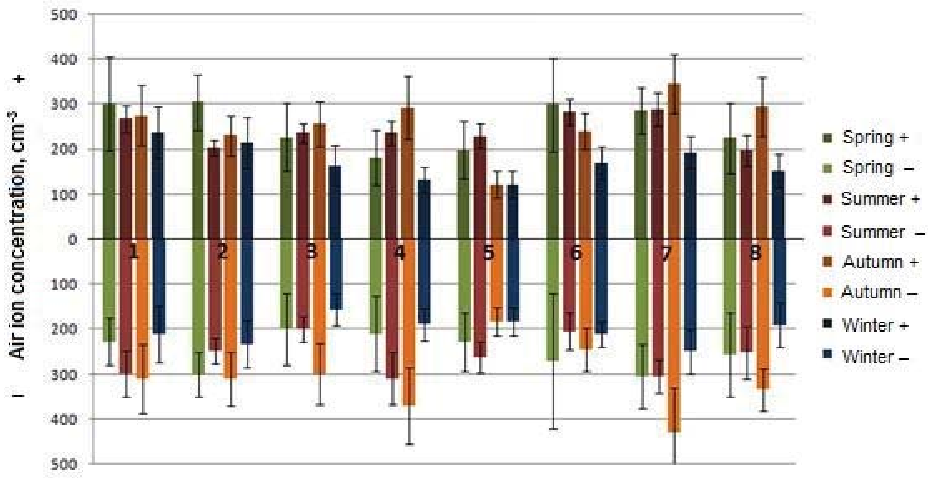


Fig. 3.1. Average concentration of air ions in different areas of Rezekne over different seasons.

Bars from 0 up: concentration of positive air ions; bars from 0 down: concentration of negative ions. The digit is used to designate the measurement point.

Air ion concentrations are much more affected by factors such as air humidity, turbulent air flows that increase the concentration in spring and autumn, and pollution whose influence on the decrease of air ion concentration can be mostly felt in summer. According to meteorological observations made in parallel, higher wind speed and dustiness levelwas detected in spring and autumn. These factors contribute to vertical convection of the ground-level air and provide air exchange even in the air layer near the earth surface, thereby dispersing more evenly the aerosols formed therein over larger volumes of air.

In summer, under windless conditions, when temperature inversion is possible, it becomes much more difficult for air ions formed in the ground-level atmosphere to reach higher layers, and therefore their concentration at the altitude of one meter can be considerably lower than at the ground level. In the autumn and winter period, the proportion of negative ions is higher, while the value of *K* is lower and rarely exceeds 1. In areas contaminated with transport flue gases (e.g. Latgales Street or railway station), the value of *K* often reaches the peak in dry and sunny summer days, mainly due to the minimum concentrations of negative air ions rather than to maximum concentrations of the positive ones. In less polluted areas, the concentration of negative air ions in summer is not the minimal.

3.3. Air ion concentration depending on the time of day

Air ion concentrations are subjected to fluctuations caused not only by the season, but also by the time of day. At almost every measurement point, the concentration in the evening was significantly lower than at midday or in the morning. This phenomenon characterises negative air ions more than the positive ones. Daily variation of unipolarity coefficient was different at each measurement point. Reduction of unipolarity coefficient was observed at four measurement points, but it is not related to the fact that the measurement point was located in an area with relatively clean or contaminated air.

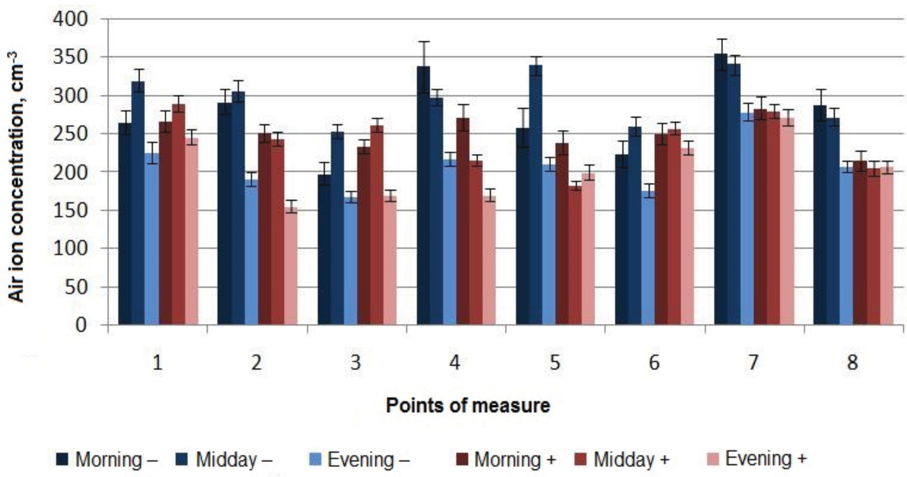


Fig. 3.2. Dynamics of air ion concentration changes at different times of day.

However, the overall results of the entire three year measurement period do not allow precise description of daily changes in air ion concentration typical for each season. In spring, the concentration of negative ions tends to increase at noon, which was observed at all measuring points. In most cases, concentrations of positive air ions increase at noon, too. In summer, concentration of negative ions tends to decrease gradually during the day. This is typical for three measurement points with potentially highest levels of air pollution: Meat Factory, Latgales Street and railway station "Rezekne-2". At other measurement points, this regularity manifests itself irregularly. A pronounced gradual decrease in the concentration of positive ions was observed near the railway station "Rezekne-2". At other measurement points such a trend was not pronounced, however, the concentration of positive ions was almost always lower in the evening than in the morning or in the daytime. Air ion concentration changes observed in summer could also be partly noticed in the autumn period, but overall, the 24-hour concentration changes were rather chaotic. Changes in daily air ion concentrations observed in winter had a relatively small amplitude, e.g. at measurement point No. 1 the amplitude was only 6 ions per cm^3. At other measurement points, the amplitude of changes was wider, but the changes themselves were chaotic.

Many fluctuations in air ion concentration during the day can be explained by 24-hour fluctuations in atmospheric pollution. In urban conditions, air pollution is related with the intensity of road traffic in the streets. Figure 3.6 shows that the level of NO_2 and benzene increases rapidly at the beginning of a working day (around 7:00 to 7:30) as the number of cars increases, creating congestions. The level of pollution remains high throughout the whole working day, with a maximum value at around 17:00 to 18:00. Concentration of NO_2 and benzene is mainly related to mobile emission sources, i.e. road transport, while concentration of SO_2 is related to stationary sources, such as boilers or thermoelectric power plants, whose activity is not so dependent on the population's daily routine. Pollutant aerosols created by road transport, interacting with air ions, cause their disappearance due to recombination and sedimentation on aerosols. Pollution accumulated in urban air during the day is thus the reason for gradual reduction of air ion concentration.

3.4. Air ion concentration in different ecosystems

The highest concentration of air ions of both polarities has been recorded in the monitoring station of Hyytiälä. It is probably due to the proximity of the bedrock to the Earth's surface, which is the source of radioactive element emissions. In the area around the place where measurements were performed the thickness of the soil layer varies from 0 (open cliffs) to 2 m, while in the measurement place itself the soil layer is several centimetres to a few dozen centimetres thick. It is known that the most significant source of ionisation in the atmosphere is the radioactive gases released from the Earth, as well as radioactive radiation from the rocks. At the beginning of measurements, a sudden peak of negative ions was observed until the 5th minute after the measurement had began. Since data from any parallelly working air ion counter are not available, this cannot be interpreted unambiguously. It could have been a relatively strong atomic activity of radioactive elements coming into the apparatus, or a spontaneous ion formation process (nucleation). Later, relatively high concentrations of air ions were observed, but the concentrations of positive and negative air ions were similar in proportions.

Measurements were taken in Helsinki, on the crossroads of Mannerheimintie and Arkadiankatu Streets, on the sunny side of the street. The morning was cool, the air temperature was about 12 °C and relative air humidity was 80 %. During the measurements that lasted for half an hour it was found that the concentration of positive air ions exceeded the proportion of negative air ions. Besides, concentrations of air ions showed wide fluctuations with some expressed peaks of the concentration of positive air ions. On the crossroads, traffic lights functioned in such a way that a moderate traffic flow through the crossroads was always ensured, so it is not possible to relate the peaks of air ion concentrations with the flow of any particular vehicles.

The measurements performed duringa working day in Riga, on Brivibas Street, showed the predominance of positive air ions over the negative ones, K = 1.39. The concentration peaks recurred cyclically, this phenomenon especially characterised positive ions. It is probably due to the organisation of traffic flows on Brivibas Street depending on traffic signals. The duration of the green signal is 35 s; a full signal cycle recurs every 1 min, 20 s. Many concentration peaks can be explained by the traffic flow regulated by traffic lights. Figure 5.3 shows that two full signal algorithm cycles take place within 160 s and the green light is switched on twice to allow the traffic light on Brivibas Street. As can be seen from the Figure, two visible peaks of air ion concentration are often observed in each 160 s measurement. Some of the peaks are related to the traffic of heavy vehicles, for example, a bus passed the ion counter placed near the road in the 480th second. A similar effect on air ion concentration is also described in Chapter 4.

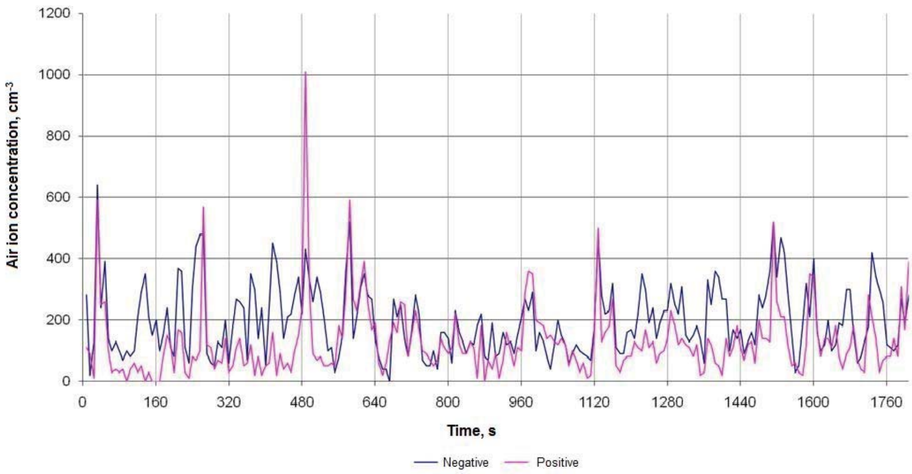


Fig.3.3. Concentration of cluster ions in Riga on 3 October 2010.

During the measurements in Mezaparks significant fluctuations of concentration were detected (0 to 600 positive ions per cm^{-3}). Such fluctuations were not detected at any other measurement point. There was no strong wind in the forest that could cause such fluctuations by mixing atmospheric layers or by bringing dust or sand particles into the measuring device. This phenomenon was probably caused by atmospheric pollution from Riga city located nearby. Potential sources of pollution affecting Mezaparks are transport flows, boiler houses and central heating plant "Riga TEC-1". In general, the level of air ionisation in Mezaparks is higher than in the centre of Riga, which means that the air is less contaminated with atmospheric aerosols than in the city centre where most air ions are recombined taking part in the natural air purification. The value of unipolarity coefficient K = 0.73 shows a stable predominance of negative air ions, which is an important factor for Mezaparks as a recreation area.

On 4 November, in the afternoon, at the time of measurements in Rezekne on Latgales street, there was a small but constant congestion of cars (about 5 vehicles in each direction). Weather conditions: sunny, relative humidity 70 %, temperature 16 ºC. The total level of air ionisation is comparable to that measured in Ancupani, i.e. the average concentration of negative ions is slightly lower than the concentration of positive ions and the value of K is 2.03. It is the highest value of unipolarity coefficient among all measurement points in this experiment. It proves the predominance of positive ions over the negative ones in locations where air pollution is caused by road transport if the weather is sunny and air humidity is relatively low.

Ancupani forest has a border with Rezekne city in its northern part. It serves as a recreation area, although it is poorly equipped. It resembles a natural forest with a pronounced predominance of conifers. The measurements were carried out within the forest, where the impact of anthropogenic pollution is considered practically non-existent. The measurements did not reveal predominance of negative air ions over the positive ions, $K = 0.98$. The amplitude of fluctuations of air ion concentration was not as high as in Mezaparks.

Table 3.1

Summary of measurement results, ions per cm^3

Place	Max -	Min -	Average -	Max +	Min +	Average +	*K*
Hyytiälä	3520	590	1258	1980	360	984	0.78
Helsinki	690	340	519	1100	390	651	1.25
Riga	640	0	196	1010	0	129	1.39
Mezaparks	910	0	382	720	0	185	0.73
Rezekne	640	60	291	1030	120	591	2.03
Ancupani	530	210	372	580	20	365	0.98

Measurement results show that, in general, air ionisation level in Finland is higher than in Latvia. This is probably due to the radioactive emissions from the bedrock that, in Helsinki and at the research base Hyytiälä, is located near the Earth's surface. Average concentrations of negative ions in forest ecosystems are higher than in urban areas. Urban ecosystems are characterised by a clear predominance of positive cluster ions over the negative ones. This phenomenon is due to the fact that the mobility of negative ions is slightly higher than that of positive ions, which helps negative ions play a significant role in atmospheric pollution reduction processes, where air ions, with their charge, contribute to coagulation and sedimentation of atmospheric aerosol particles. The predominance of negative ions in forest ecosystems is less pronounced than the preponderance of positive ions in cities. For example, in Ancupani the unipolarity coefficient *K* is close to 1. In Mezaparks it was observed that ions of both polarities were subjected to a wider amplitude of concentration fluctuations. This may be related to the proximity of the city, which is constantly exposing this forest ecosystem to the impact of atmospheric pollution. From the point of view of human health, the air in the city's ecosystem is not favourable to the population because of the preponderance of positive ions.

3.5. Impact of road transport on air ion concentration in the city

Using the specific situation that arose due to the reorganisation of traffic flow during street repair works, air ion concentration was measured under conditions of a permanent traffic congestion when engine exhaust gases were being accumulated for a long period of time. The number of transport units crossing the street per day did not significantly change. In this case, however, it was not possible to observe the fluctuations of traffic intensity with expressed peaks in the morning and in the evening, which usually is a typical situation in city environment. The total ionisation level (total air ion concentration irrespective of the charge sign) in the evening equalled the ionisation level in the morning, while the unipolarity coefficient increased significantly because the concentration of negative air ions decreased dramatically, reaching only 58 % of the morning concentration. On the other hand, the concentration of positive ions increased by 37 %. At lunchtime, compared to the morning measurements, the total ionisation level had increased by 53 %. Besides, the concentration of positive ions had increased by 129 %, while the concentration of negative ions had slightly decreased. This means that the increase in the total ionisation level was observed only because of the increase in the number of positive ions.

Table 3.2

Average values of air ion concentration, traffic intensity and meteorological conditions

Time of measurement	Negative ions, cm^{-3}	Positive ions, cm^{-3}	Total level of ionisation, ions, cm^{-3}	K	Number of transport units	T, °C	W, %
Morning	202	150	352	0.74	1145	2	58
Afternoon	196	343	539	1.75	1127	6	41
Evening	117	206	323	1.77	1265	8	26

Under real-world urban conditions, it is not possible to express a direct relationship between instantaneous air ion concentration and pollution emitted by vehicles because of the importance of time in which accumulation of pollution takes place and because of complex chemical reactions that occur in polluting aerosols when they are not only in the gaseous phase but also when they are solid or liquid. Air ion concentration in proportions that are unfavourable for human health is, to a large extent, secondary effect of this chemical-physical transformation. However, during the measurements at midday, some individual sudden but temporarily peaks of positive air ion concentration were observed, which could be caused by different phenomena. For example, a sudden peak concentration was observed when two trucks stopped at the intersection and emitted diesel exhaust gases near the air ion counter. A similar but less pronounced peak was observed in the case of a passenger car with a diesel engine. During the measurements, several cars with gasoline internal combustion engines were periodically near the measuring device, but it did not result in any particular peak of ion concentration. Probably, the peaks of concentration of positive air ions in the presence of diesel engine exhaust gases can be associated with greater opacity compared to gasoline engines, especially if the engines are worn out and are not in good technical condition. It is known that exhaust gases emitted by diesel engines contain various aerosols, non-combusted hydrocarbon particles and some specific chemicals in elevated concentrations. Temporary peaks may sometimes be caused by various external factors, such as sudden wind gusts that mix the air layers and bring small aerosol particles, dust, or even fine sand grains into the measuring device. It should be noted that the aforementioned peaks of positive air ion concentrations caused by flue gases were related with minimum concentrations of negative air ions, which is not typical for all peaks of positive ion concentration.

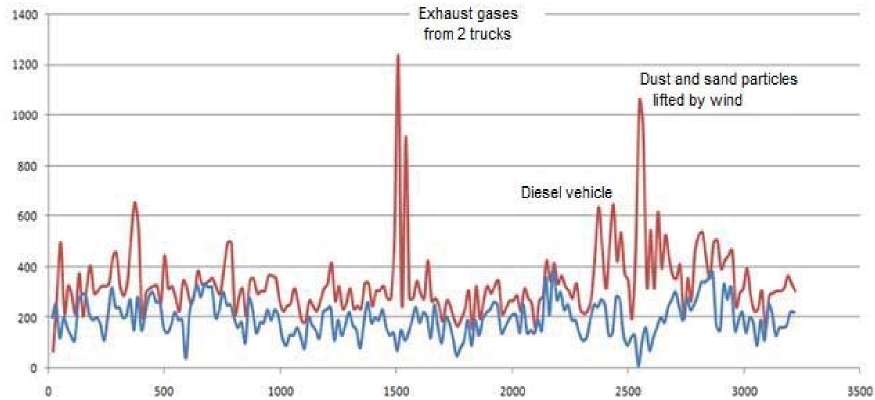


Fig. 3.4.Air ion concentration at an intersection with intense traffic:
a) measurements in the morning; b) at midday; c) in the evening. On the abscissa: time after the beginning of the measurement, s; on the ordinate – air ion concentration per cm^{-3}.

3.6. Characteristics of air ions in exhaust gases of an internal combustion engine

Although the measurement method allowed minimal mixing of exhaust gases with atmospheric air, it is reasonable to assume that most aerosols were formed directly in the engine during fuel combustion. Mathematical processing of experimental data was performed by dividing the whole set of measurements into three series: engine off, engine on and engine switched off again. Subsequently, the significance of difference between arithmetic means was checked by Fischer criterion in each two series. Such processing of the measurements obtained in Experiment 1 showed that the difference in ion concentration between the both cases when the engine was off and when it was turned on proved convincingly significant, but the difference between the beginning and the end of the experiment (when the engine was turned off) proved to be insignificant. This phenomenon was observed when analysing concentrations of both positive and negative ions.

Just like Experiment 1, the measurements performed during Experiment 2 reveal a significant difference between the arithmetic means of the sets when the engine is turned off at the beginning of the experiment and when it operates at 1000 rpm. A significant difference between arithmetic means in datasets when the engine is turned on and when it is stopped can be seen only in the area of positive ions. In the area of negative ions, it is not significant any more. Besides, Experiment 2 shows a significant difference between the arithmetic means in both cases when the engine is turned off, i.e., the background concentration of air ions after the engine is stopped is significantly different from the concentration before the engine is turned on.

In Experiment 3, when the engine was working at different speeds, no clear difference between series could be seen. Significant differences in the arithmetic means of concentrations occurred only in some cases, for example, between a disengaged engine and 1000 rpm. or between an engine operating at 1000 rpm. and an engine operating at 2000 rpm.; besides, this can only be seen in the area of negative ions. It cannot be excluded that the measurements of Experiment 3 were affected by some geophysical or meteorological factor that was more pronounced than the impact of car exhaust gases on ion concentration, thus mitigating the difference in the arithmetic means of air ion concentrations caused by different modes of the engine.

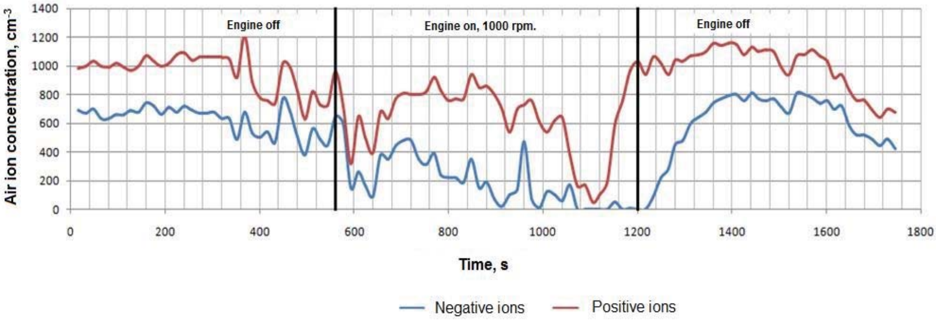


Fig. 3.5. Measurements of light air ion concentration in exhaust gases from motor vehicles, Experiment 1.

Although the differences of arithmetic means between different data series are not always significant, various engine operating modes are almost always associated with some short-term fluctuations in air ion concentration. As a result of turning on the engine, Experiment 1 showed a decrease in the concentration of both polarities, while in Experiment 2 the increase could be mainly seen in the area of positive air ions; thus there were obvious changes in both cases. The completely opposite changes in air ion concentration after the engine has been turned on in two seemingly identical experiments may be caused by the degree of warming of the engine. At the beginning of Experiment 1, the engine was still not heated to the nominal temperature, therefore a richer fuel mixture was supplied through the power system. The decrease in the concentration of air ions of both polarities may be related to the increased concentration of unburned hydrocarbon aerosols (including soot) in flue gases. In Experiment 1, after stopping the engine, air ion concentration returned rapidly to the previous level. On the other hand, when the engine was stopped during Experiment 2, no significant changes in air ion concentration could be seen, but there were visible differences between the two levels of background concentrations, which suggests that in this case the engine exhaust gases had had a longer-lasting effect on the air ion concentration in the ambient air.

In almost all cases, when the engine is turned on or accelerated, at least a temporary increase in air ion concentration can be observed, but stabilisation of the engine operating mode also stabilises the concentration of air ions. When the speed of the engine is decreased, air ion concentration is also lower, at least temporarily. As the engine operating mode changes, as it actually does when starting or accelerating the movement, the engine operates in a non-optimal mode for a short time and the fuel mixture is richer than normally required in stationary mode. Such changes in the engine operating mode are usually related to incomplete combustion of firelighter, which results in an increased concentration of CO and unburned hydrocarbons in the exhaust gases. As it can be seen, short-term fluctuations in the concentration of pollutants in exhaust gases also cause short-term fluctuations in the concentration of air ions. Changing the engine operating mode is a relatively short phenomenon compared to the overall operation cycle of the vehicle outside the city, but, in urban areas, due to the organisation of the traffic flow, the car engine is forced to operate in a non-optimal mode more often. The vehicle has to be stopped at the red traffic light more frequently, then the movement is started by acceleration or a sudden deceleration is needed to turn aside, followed by a rapid acceleration in order to enter timely the traffic flow, etc. In all these cases, the engine runs in a non-optimal mode, which results in more pollutants entering the urban air and, consequently, it affects the emitted air ions.

3.7. Determination of mathematical relationships using stepwise regression

The balance of air ions in the atmosphere depends on various and complex mechanisms of ion formation and extinction, therefore outdoor study of air ions is a complicated but necessary measure to gain new knowledge about the interaction of the atmosphere and its pollution with the ecosystem. The results obtained are often difficult to interpret.

A serious problem in the analysis of interaction between atmospheric pollution and air ions is multicollinearity (Fig. 3.6). There is a moderate or even a strong correlation between the components of chemical and physical pollution of the air. This is especially true in case of urban environment that has a high level of anthropogenic pollution. Many components of chemical and physical pollution of the atmosphere have similar sources of origin, such as industrial emissions, traffic, energy, etc., and therefore there is a close correlation between these components, which raises problems in the classic regression analysis. It is necessary to find and apply a mathematical model of regression analysis that would minimise the negative impact of multicollinearity on the interpretation of results and that could explain air ion concentration through analysis of changes in environmental factors.

	CO	NO	NO	NOx	O3	PM10	PM2,5	SO2	Rel. Hum.	Temp.	Wind Dir.	Wind Sp.	Cluster "-"	Cluster "+"	Middle "-"	Middle "+"	Heavy "-"	Heavy "+"	Total "-"	Total "+"
CO, mg/m3	1																			
NO, ug/m3	0,75	1																		
NO2, ug/m3	0,83	0,66	1																	
Nox,ug/m3	0,84	0,96	0,84	1																
O3, ug/m3	-0,52	-0,38	-0,64	-0,50	1															
PM10, ug/m3	0,48	0,35	0,48	0,43	-0,24	1														
PM2,5, ug/m3	0,15	0,09	0,15	0,12	-0,12	0,20	1													
SO2, ug/m3	0,41	0,31	0,40	0,37	-0,21	0,29	0,09	1												
Rel. Humidity, %	0,05	-0,01	0,05	0,01	-0,55	-0,20	-0,01	-0,07	1											
Temperature, ◦C	-0,44	-0,33	-0,45	-0,40	0,55	-0,12	-0,12	-0,39	-0,36	1										
Wind Dir., deg.	-0,15	-0,12	-0,15	-0,14	0,12	-0,21	-0,07	-0,11	0,06	0,05	1									
Wind Speed, m/s	-0,24	-0,18	-0,33	-0,25	0,32	-0,23	-0,02	-0,07	-0,08	0,01	0,03	1								
Cluster Ions "-"	-0,13	-0,07	-0,16	-0,10	0,11	-0,02	-0,03	-0,04	-0,10	0,32	0,02	-0,03	1							
Cluster Ions "+"	-0,11	0,01	-0,17	-0,05	0,10	0,00	-0,03	-0,11	-0,10	0,49	0,01	-0,09	0,80	1						
Middle Ions "-"	-0,08	-0,02	-0,07	-0,04	0,15	-0,03	-0,03	-0,02	-0,20	0,09	0,03	0,12	0,18	0,05	1					
Middle Ions "+"	-0,02	0,04	-0,02	0,02	0,12	0,02	-0,02	0,00	-0,23	0,08	0,03	0,09	0,11	0,08	0,41	1				
Heavy Ions "-"	0,16	0,19	0,18	0,20	-0,09	0,15	0,03	0,07	-0,06	0,02	-0,06	-0,08	0,08	0,10	0,14	0,10	1			
Heavy Ions "+"	0,48	0,49	0,49	0,53	-0,27	0,33	0,08	0,25	-0,09	-0,21	-0,09	-0,15	0,13	0,09	0,11	0,21	0,41	1		
Ions (Total) "-"	0,15	0,18	0,17	0,19	-0,08	0,14	0,02	0,07	-0,08	0,03	-0,05	-0,08	0,13	0,13	0,20	0,12	1,00	0,41	1	
Ions (Total) "+"	0,45	0,48	0,46	0,51	-0,24	0,32	0,08	0,23	-0,11	-0,16	-0,08	-0,15	0,19	0,17	0,15	0,31	0,41	0,99	0,42	1

Fig. 3.6. Correlation matrix of the data used for calculations.

To solve this problem, a multi-factor regression method "stepwise regression" was applied. Using the stepwise regression, it is possible to exclude insignificant factors and later analyse only the factors that actually affect the concentration of air ions. In statistics, the stepwise regression involves regression models in which the prediction of variables is carried out by an automatic procedure. It is usually accomplished by a sequence of F-tests or t-tests. It is also possible to apply other methods, such as adjusted R^2 value, Akaike information criterion (AIC), Bayesian information criterion (BIC), etc. (Social vocabulary ..., 2015). In this study, the author used the statistical software tool

"R". Regression coefficients were obtained by applying backward elimination and based on the Akaike information criterion. The backward elimination begins with a model that includes all 85 predicted variables, and the "step" function of the software "*R*" excludes the less important variable in each step. The "step" function stops when all variables in the model correspond to the value of Akaike information criterion that is less than or equal to the value of the given Akaike information criterion of the model.

Table 3.3

Characteristics of ion classes used in calculations

No. of the ion class	Name of the ion class	Size of ions,nm
1	Negative cluster ions	0.75 – 1.54
2	Positive cluster ions	0.75 – 1.54
3	Middle sized negative ions	1.54 – 7.50
4	Middle sized positive ions	1.54 – 7.50
5	Heavy negative ions	7.50 – 36.60
6	Heavy positive ions	7.50 – 36.60
7	Total negative ions	0.75 – 36.60
8	Total positive ions	0.75 – 36.60

Dependence of concentration of a particular air ion class on specific factors can be represented as a first degree polynomial. Argument *Y* is the concentration of air ions of the particular class designated by $N_{(i)}$, where *i* is the number of the corresponding air ion class (see Table 3.3). Chemical and meteorological parameters are independent variables that are represented in the polynomial with their coefficients. Designations of these parameters are shown in Table 3.4.

R^2 value of each regression model is given in the next line below the equation. The *p* value of the equation's coefficient significance level in all cases is less than 0.05.

$N_{(1)} = -0.008a + 0.222b - 0.216c + 3.136e + 0.140f + 0.645g + 2.321h + 0.022i + 2.557j$

$R^2 = 0.6734$

$N_{(2)} = 0.649b - 0.367c + 3.668e + 0.046e + 0.310f + 1.290g + 4.844h + 0.048i + 3.554j$

$R^2 = 0.8569$

$N_{(3)} = -0.023a + 0.169b + 0.639c + 0.391d - 0.244g + 0.405e - 0.280h + 0.041i + 9.489j$

$R^2 = 0.1985$

$N_{(4)} = -0.023a + 0.310b + 0.870c + 0.423d + 0.126f - 0.259g + 0.556h + 0.061i + 10.617j$

$R^2 = 0.274$

$N_{(5)} = -0.202a + 7.980b + 15.448c + 1.520d + 4.438f + 0.724g + 15.898h$

$R^2 = 0.1975$

$N_{(6)} = 10.851b + 16.680c + 2.124d + 8.769e + 5.087f + 0.214i$

$R^2 = 0.5679$

$N_{(7)} = -0.238a + 8.381b + 15.587c + 2.028d + 10.456e + 4.399f + 1.399g + 18.852h$

$R^2 = 0.2360$

$N_{(8)} = 11.655b + 17.254c + 2.318d + 13.398e + 4.480f + 0.693g + 6.264h + 0.356i + 26.890j$
$R^2 = 0.6246$

Table 3.4

Designations and units of measurements included in the regression equations

Number of the parameter	Designation	Parameter	Unit of measurement
1	*a*	CO	$\mu g \cdot m^{-3}$
2	*b*	NO	$\mu g \cdot m^{-3}$
3	*c*	NO_2	$\mu g \cdot m^{-3}$
4	*d*	O_3	$\mu g \cdot m^{-3}$
5	*e*	SO_2	$\mu g \cdot m^{-3}$
6	*f*	PM_{10}	$\mu g \cdot m^{-3}$
7	*g*	Rel. humidity	%
8	*h*	Temperature	°C
9	*i*	Wind direction	Degrees
10	*j*	Wind speed	$m \cdot s^{-1}$

It appears from the results obtained that CO is not among the main factors influencing air ion concentration,its coefficients are small. In some cases, the stepwise regression even rejects the effect of CO. Whenever it has any influence, its impact on air ion concentration is negative.

Nitrogen oxides, on the other hand, have relatively high impact on air ions. They have a strong positive effect on heavy ions, but do not have significant influence on cluster ions and in the area of middle-sized ions. NO_2 has a weak negative impact on cluster ions, moderately strong positive effect on middle-sized ions and strong positive effect on heavy ions and on ions in general. This suggests that nitrogen oxides play an important role in the formation of the chemical composition of heavy ions.

Basing on the stepwise regression analysis, the impact of O_3 on air ions is ambiguous. On the one hand, O_3, due to its oxidative properties, can reduce the total air pollution level and thus increase the life cycle of air ions, but, on the other hand, it can also reduce the amount of positive air ions. However, the equation coefficients show that O_3 has a very weak impact on air ion concentration irrespective of the air ion class.

SO_2 has a moderate positive impact on the concentration of cluster ions (mainly the negative ones); besides, it significantly increases the concentration of positive heavy ions. This influence is also reflected in the area of total ion concentration. In the area of other ions the impact of SO_2 is not significant.

PM 10 mainly affects heavy ions and thus the total concentration of air ions. Heavy ions consist of small aerosol particles, on which cluster ions have been condensed.

Under natural conditions, small aerosol particles have a positive correlation with the dustiness PM 10.

The impact of meteorological factors on ions varies. Relative humidity does not have any significant or decisive influence. Air temperature in all cases slightly increases the air ion concentration. No dependence of air ion concentration on the wind direction was detected, however, in some cases, its speed had a positive effect on air ion concentration.

In general, air pollution by chemical substances has a greater effect on positive air ions of all classes than on the negative ones. In cases where potentially secondary factors, such as wind direction or speed, play an important role in the regression equation, it is evident that the regression model lacks a significant parameter that was not present among the raw data. For example, the mathematical model obviously overestimates the impact of wind direction on middle-sized negative ions, and, at the same time, R^2 = 0.1985, which indicates that the given regression model does not sufficiently characterise regularities of the raw data.

Basing on the coefficient of determination R^2, the best regression model was found for positive cluster ions ($R^2 \approx 0.86$), negative cluster ions, positive heavy ions and positive total ions, where $R^2 > 0.5$.

3.8. Impact of increased background radiation on air ion concentration

3.8.1. Experiment with ^{239}Pu isotope

Comparativemeasurements of air ion concentration with two measuring devices allow evaluating the correlation between the results of both apparatuses. The portable air ion counter “Sapfir-3M” measures the concentration of air ions whose mobility is $k \geq 0{,}4$ $cm^2 \cdot V^{-1} \cdot s^{-1}$, which is very close to the class of cluster ions (see Chapter 1.2), while a special mode of the air ion spectrometer “SIGMA” allows measuring the total concentration of cluster ions.

The experiment with ^{239}Pu isotope made it possible to compare the results of the two devices (see Fig. 3.7). The operating principle of both apparatuses is different, and the airflow through “SIGMA” is almost 10 times larger than that through “Sapfir-3M”. This is probably the reason for the faster response by “SIGMA” to drastic changes in air ion concentration, while the reaction of “Sapfir-3M” is slightly late. Under different conditions this delay can reach 2 to 6 s. However, despite these differences and individual features of the two measuring devices, they both show the same tendency and changes in air ion concentration fixed by both apparatuses correlate with each other very well.

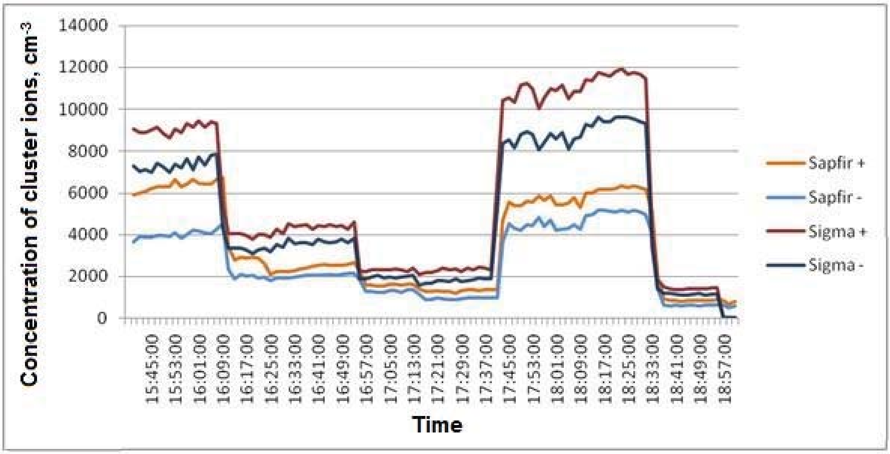


Fig. 3.7. Ionisation measurements with “Sapfir-3M” and “Sigma” under conditions of ^{239}Pu radiation of various intensity.

When air ion concentrations are relatively low, such as the natural level of air ionisation in a city or in living spaces, the difference between the two measuring devices is not significant. The average background concentration of cluster ions at the Laboratory of Atmospheric Physics of the University of Tartu was 705 positive and 413 negative ions per cm^3.

Subjected to artificial ionisation, sending a flow of compressed air through the ^{239}Pu isotope at a different speed resulted in a level of air ionisation that was directly proportional to the airflow. When the airflow is increased two times, air ion concentration becomes two times higher, too. By studying the distribution of cluster ions over fixed airflow velocities, it can be seen that linear regression coefficients in each case show a close correlation between two parameters, which indicates that even small radiation emissions in a closed space greatly increase the concentration of cluster ions. Air ion measurements may serve as a good indicator for even a small amount of radioactive contamination.

Table 3.5

Measurements of air ion concentration under conditions of artificial ionisation, using “SIGMA” and “Sapfir-3M”

Time	Air flow through the ^{239}Pu element, $l \cdot h^{-1}$	Average concentration of air ions, cm^{-3}			
		Sapfir-3M		SIGMA	
		Positive	Negative	Positive	Negative
15:39 – 16:09	600	6384	4022	8865	7163
16:09 – 16:54	300	2746	2123	4199	3468
16:54 – 17:41	150	1437	1088	2472	1991
17:41 – 18:33	600	5653	4531	10880	8753
18:33 – 18:57	0	850	601	1439	1152

When concentrations of cluster ions exceed 1000 ions per cm^3, differences in the readings of both devices begin to appear. Readings of “SIGMA” are almost always two times higher than those of “Sapfir-3M”. The amplitude of the value of K in the measurements taken by “SIGMA” is relatively small, i.e. 1.21 to 1.25, while for “Sapfir-

3M” it varies from 1.25 to 1.59. This indicates a greater dispersal of the results obtained by “Sapfir-3M”, which in turn suggests that “SIGMA” is more precise than “Sapfir-3M”.

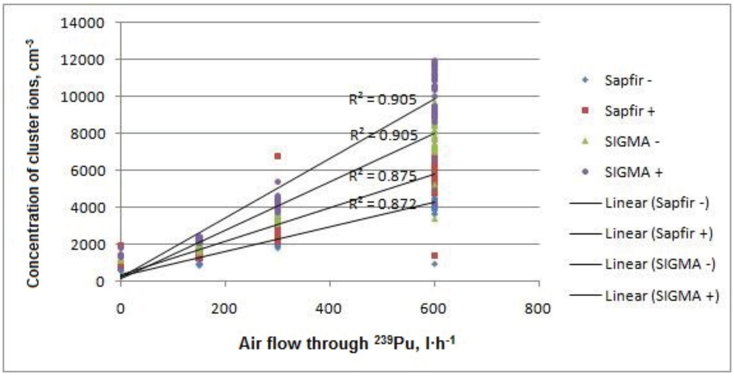


Fig. 3.8. Dependence of cluster ion concentration on the speed of the airflow through ^{239}Pu isotope, comparative measurements by “SIGMA” and “Sapfir-3M”.

Although the measurements carried out with both devices indicate different levels of ionisation, the correlation of the measurement results should be considered sufficiently good, which means that both apparatuses are sufficiently sensitive to changes in air ion concentration and reflect actual fluctuations of their concentration.

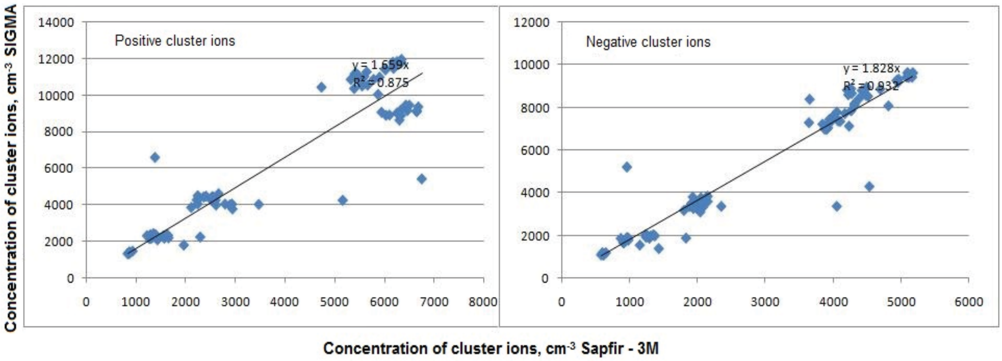


Fig. 3.9. Mutual correlation of measurements performed by “Sapfir-3M” and “SIGMA”.

The question why there are such significant differences in concentration levels remains unanswered. It is possible that this is due to the different structure and operating principles of the measuring devices. It can also be suggested that “Sapfir-3M” has a higher sensitivity threshold for measuring air ion mobility, which is not specified in the technical documentation.

3.8.2. Impact of radon on air ion concentration

Radon gas emanations from rocks are a topical problem in many parts of the world. In particular, it tends tobe accumulated in closed cellars with poor ventilation during the winter months, when the soil is covered with snow and the release of radon into the atmosphere is difficult. Radon gas is 7.5 times heavier than air. Accumulation of radon is endangering people living at the basement level or in similar premises, given that radon, in long term, has a carcinogenic effect and that this gas cannot be feltorganoleptically. Radon degradation products are reactive metals that form oxides and other compounds that deposit in human body. These compounds are unstable too and decompose further, thus subjecting the human body to internal radiation. The result of such exposure isoften development of malignant tumours (primarily lung cancer) (Kļaviņš et al., 2010). Negative consequences can be caused by prolonged stay in premises with radon concentration above 40 $Bq \cdot m^{-3}$ and maximum allowable concentration of 200 $Bq \cdot m^{-3}$.

At the Atmospheric Physics Laboratory of the University of Tartu the average daily concentration of radon was 3.05 $Bq \cdot m^{-3}$, but in the basement (in autumn), when it had not been ventilated for 48 hours, it reached 76.51 $Bq \cdot m^{-3}$. Maximum recorded 15 min concentration in the basement amounted to 157.80 $Bq \cdot m^{-3}$.

The concentration of radon, but especially the resulting air ion concentration, is very sensitive even to a slight air exchange in the room. Each time when someone opened the door or entered the room was also reflected in the concentration measurements (Fig. 3.10). For example, the door was opened at the 20th and 27th hour of measurements to enter and check the state of measuring devices. Starting from the 42nd hour up to the end of measurements, the door was opened several times, which can be seen in the graph as a decrease of both radon and cluster ion concentration (Fig. 3.10).

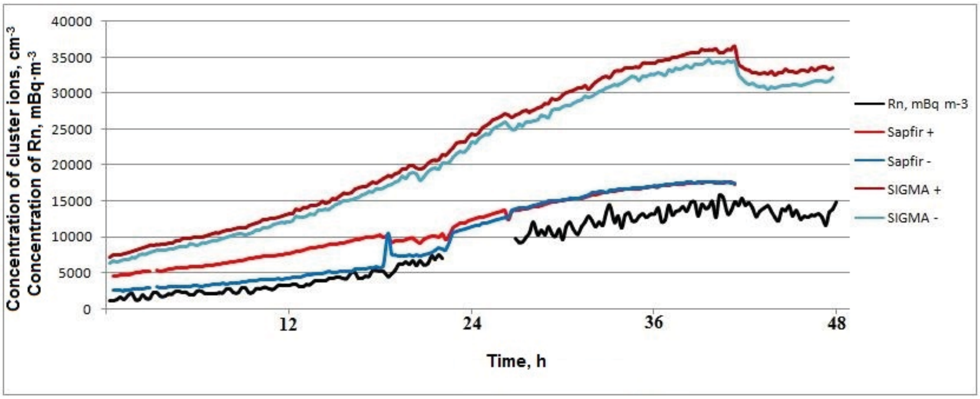


Fig. 3.10. Measurements of radon and cluster ion concentration in a closed space.

Between the radon concentration and the concentration of cluster ions there is a very close correlation with low dispersion. This means that cluster ions are a good indicator of radon gas in a closed space. Basically, the concentration of cluster ions, due to its linear relationship with radon, can be used as a radon indicator in a closed room. The level of ionisation caused by radon is sufficiently high so that the impact of all other possible ionisers or air ion concentration reducers becomes insignificant.

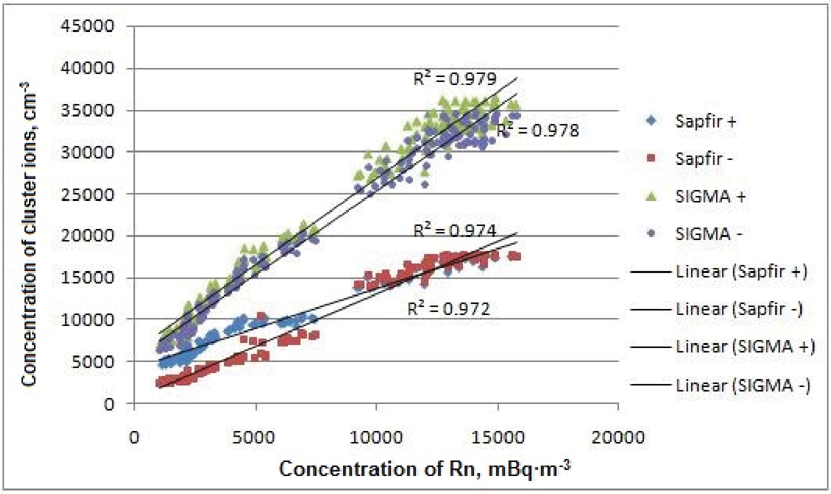


Fig.3.11. Dependence of cluster ion concentration on Rn concentration.

4. ASPECTS OF PRACTICAL APPLICATION OF RESEARCH RESULTS

Understanding atmospheric ionisation processes provides extended opportunities for monitoring environmental quality. Issues related to the content and composition of air ions in the ground-level atmosphere are topical mainly when assessing the quality of the human environment. The content of both positive and negative air ions in the air has a significant impact on physiological processes in human body, thus affecting the state of human health, mental and physical capacity, as well as well-being. Scientific findings related to air ion concentration optimal for improving working capacity (Рейнет, 1983; Hawkins & Barker, 1978; Hawkins, 1981) have already been incorporated into the national legislation of several countries. Studies on interactions between air ions and indoor air pollution by aerosols have been described in numerous publications (e.g. Priyman, 1984); besides, air ionisation technologies are widely used in purification of indoor air from fine aerosols (e.g. Shiue & Hu, 2011). Air ions are used to improve the state of health and well-being, as well as to treat burns, injuries, and other diseases (Герасимова et al., 1996).

On 16 June 2003, sanitary and epidemiological rules and regulations "Hygienic Requirements for the Air-Ion Level of Industrial and Public Facilities SanPin 2.2.4 1294-03" (Санитарно-эпидемиологические правила и нормативы "Гигиенические требования к аэроионному составу воздуха производственных и общественных помещений СанПин 2.2.4 1294-03") entered into force in the Russian Federation. Similar regulations have recently been adopted in Ukraine and Belarus (Skromulis & Noviks, 2012). "SanPin 2.2.4 1294-03" defines requirements for air ion composition in indoor environments where, for various reasons, too low or too high air ion concentration inappropriate for human organism may occur. The lowest air ion concentration that is suitable for human beings is 400 ions per cm^3, but the highest limit is 50,000 ions per m^3, while the value of unipolarity coefficient should be in the range of $0.4 \leq K \geq 1.0$.

Many studies describe the significant impact of some specific impurities, such as iodine and diethylamine vapours, on changes in the mass and mobility of air ions (Luts et al., 2011), but so far little research has been carried out on the influence of major anthropogenic air pollutants such as NO_X, CO, SO_2, etc. on air ion concentration in the ground-level atmosphere.

Referring to the research carried out, significant impact of transport pollution on changes in natural background concentrations of air ions is evident. Continuous measurements of air ion concentration in different parts of the city allow detecting the level of specific air ions in each place and finding areas with potentially better or less favourable living conditions. This opens up the opportunity to use the data obtained when planning the development of urban infrastructure and to channel the main road flows away from the most polluted areas, thus improving the air quality and increasing the air ion content to the required level. It is possible to widen green zones and install water fountains in the areas where air ion concentration and value of *K* does not meet the requirements. This also implies the need to protect the natural areas of the city where the level of air ion concentration is favourable to human health, i.e. to restrict construction and traffic therein, to improve these territories as recreation areas. For example, in Rezekne it is highly recommended to preserve the park and greenery in the vicinity of Miera Street, Tuberculosis Hospital and Secondary School No. 5. In case of necessity, these territories would be appropriate for construction of kindergartens, while the deployment of serious industrial centres or transport nodes should not be allowed there.

For the above reasons, it is necessary to include measurements of air ion concentration in the range of parameters to be measured by urban air monitoring stations. In addition to chemical, meteorological and physical air pollution, concentration of air ions and unipolarity coefficient *K* has an unintended additional impact on the environment and, in particular, on human health. When $K \geq 1$, air ionisation, in combination with polluting gases, aerosols and dust, is an additional factor affecting harmfully human health and well-being. Under conditions of intense air pollution, ion chemical transformations can result in the formation of large amounts of heavy aerosol ions that have a pronounced negative impact on human health. If the ionisation affects the molecules of pollutant gases, they have a more severe negative impact on both nature and human body.

From the point of view of human health, it is very important to understand the total effect of air ion concentration and various components of air pollution on the body. Basing on a large number of studies, including those mentioned above (Reinet, 1983; Hawkins, 1981), air ions in appropriate proportions and at the required concentration level are able to compensate for symptoms caused by atmospheric anthropogenic contamination. Air ion therapy contributes to the treatment of respiratory and pulmonary diseases, hypertonia, tachycardia, excessive nervous excitement, spasms, and other diseases. This suggests that there is a zone in which the beneficial or curative impact of air ions on human body is able to compensate for the negative effects of pollution. For example, an increased concentration of CO_2 triggers accelerated pulse, respiration and sweating, but it can be partly compensated by increasing the concentration of light air ions.

On the other hand, ionisation as such can increase the harmful effects of pollutants, because, when entering body in ionised state, the harmful substance is involved in metabolic processes faster and more efficiently. Toxic effects of pollutants, both aerosols and gases, in ionised state grow from a few percent to several times depending on the nature of the substance and on the degree of ionisation (Milov, 2004), for example, the toxicity of Mn dioxide aerosol emissions in ionised form increases several times (Pasynich et al., 1988). When investigating harmful effects of vanadium oxides on the body, it has been found that bipolarly ionised aerosols are less harmful than the unipolarly ionised ones. Besides, negative heavy ions areless harmful to the body than the positive ions (Пазынич, 1975).

Air ions probably have a positive effect if the concentration of pollutants does not exceed the maximum allowable concentration. Otherwise, there are conditions that contribute to the formation of heavy aerosol ions at high concentrations. For example, in order to maintain the necessary air ion concentration in classrooms with artificial ionisation, the total concentration of aerosol particles (PM 10) in premises should not exceed 0.1 mg/m^3, otherwise the generated light air ions would transform into heavy ions and become harmful to students' health (Милова, 2004). By measuring aerosol PM 10 concentrations in the air, it is not possible to prove directly the presence of smaller aerosol fractions in the air, because heavy air ions are formed primarily from aerosol particles of 10 to 100 nm (the so-called Aitken mode), but it has been proved that fractions of anthropogenic aerosol pollution of different size mutually correlate well enough (Pirjola et al., 2006) to predict the presence of finer fractions in the air by measuring the concentration of bigger particles. When the concentration of aerosols with average size of 10 nm is 0.1 mg/m^3, the concentration of light air ions in the indoor air is close to zero. It was also found that in the indoor air where tobacco smoke with particle size 10 to 100 nm reaches concentration of 0.1 mg/m^3 the concentration of heavy ions was 10^5–10^6 per cm^{-3}, which is considered harmful to health (Дударев & Турубаров, 2002).

Due to the lack of convincing data on the maximum allowable concentration of heavy air ions for the protection of human health and as there are no regulations for it, it is difficult to determine the concentration threshold beyond which negative impact begins. Heavy air ions can always be found in nature, as they naturally occur due to the aging of cluster ions. Basing on long-term measurements in areas free from anthropogenic contamination, it can be considered that the background concentration is 1200 heavy air ions per cm^3 (Hõrrak, 2001). It cannot be stated that the exceedance of this concentration has negative consequences, but it can be considered that heavy ions, if they are at the level of background concentration, do not have any effect on human health. The chemical composition of heavy air ions is another factor that requires further research, because the negative impact on health greatly depends on the chemical composition.

The level of ionisation is an important additional factor affecting the human body, along with the chemical and physical contamination of the air. Circumstances with high level of air pollution in anthropogenic environment are often associated with a rapid decrease in the concentration of light air ions (i.e. cluster ions), thus the human body is affected by the summary negative impact of air pollution and deionised environment (see Table 4.1, examples 5 and 6). In cases where the level of air pollution is close to or slightly exceeds the maximum allowable concentration and the concentration of light air ions is optimal or slightly elevated (mainly indoors, using artificial aeroionisation), the positive effect of air ions can, theoretically, compensate for the negative impact of pollution (see Table 4.1, examples 8 and 9). In order to define adequate levels of chemical pollution above the maximum allowable concentration and their intervals allowing compensation for the harmful effects, further studies in the field of medicine and hygiene are necessary. For this reason, if the value of at least one parameter of the maximum allowable concentration is exceeded, hygienists do not recommend the use of artificial ionisation in premises when people are present therein. Therefore, air purification with ionisers should be carried out when people are not present (e.g. Милова, 2004). It follows from the above assertions that the compensation zone exists only when the maximum allowable concentration of PM 10 (i.e. 40 $\mu g \cdot m^{-3}$) is exceeded, because a stable harmful level of heavy air ion concentration is reached only when the concentration of PM 10 is 0.1 $mg \cdot m^{-3}$.

Table 4.1

Impact of various combinations involving air ionisation and chemical / physical pollution on human body

No.	Ionisation degree (cluster ions)*	Degree of chemical pollution, basically CO, NO_X, SO_2	Air ion concentration, including PM 10	Impact on body
1	Optimal	No	No	Optimally favourable
2	Increased	No	No	Curative impact of air ions
3	Very high	No	No	Harmful impact of increased ionisation
4	Very low or non-existent	No	No	Harmful impact of deionised environment
5	Very low or non-existent	Exceeds the MAC	Does not exceed the MAC	Harmful impact of chemical pollution and deionisation
6	Very low or non-existent	Does not exceed the MAC	Exceeds the MAC	Harmful impact of aerosol pollution and deionisation
7	Very low or non-existent	Exceeds the MAC	Exceeds the MAC	Total harmful impact of chemical pollution, deionisation and aerosols
8	Optimal or increased	Does not exceed the MAC	No	Partial compensation of the negative impact of chemical pollution
9	Optimal or increased	No	Does not exceed the MAC	Partial air purification from aerosols – favourable impact
10	Optimal or increased	Does not exceed the MAC	Does not exceed the MAC	Partial compensation of the negative impact of chemical and aerosol pollution
11	Optimal or increased	Exceeds the MAC	Does not exceed the MAC	Additional negative impact of partially ionised pollution
12	Optimal or increased	Does not exceed the MAC	Exceeds the MAC	Formation of heavy air ions, partially negative impact
13	Optimal or increased	Exceeds the MAC	Exceeds the MAC	Negative impact of ionised pollution and heavy ions
14	Very high	Exceeds the MAC	Exceeds the MAC	Strong negative impact of increased ionisation, heavy air ions and ionised pollution

*0...100 ions per cm^3 – very low or non-existent
400...3000 ions per cm^3, $0.4 \leq K \geq 1.0$ – optimal
3000…50000 ions per cm^3, $0.4 \leq K \geq 1.0$ – increased
above 50000 ions per cm^3 – very high (harmful to health)

Table 4.2

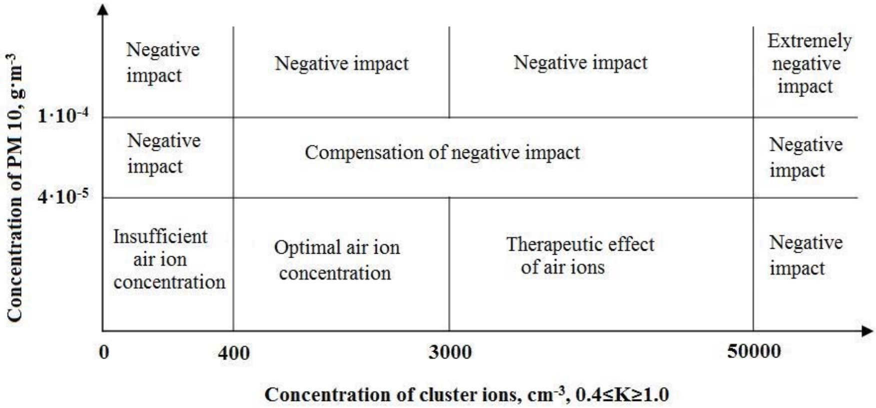
Maximum allowable concentration (MAC) of air pollution in accordance with the Cabinet Regulation No. 1290 on the Air Quality (03/11/2009)

Parameter	Type of the air pollution						
	CO, $mg \cdot m^{-3}$	NO_X, $\mu g \cdot m^{-3}$	SO_2, $\mu g \cdot m^{-3}$	O_3, $\mu g \cdot m^{-3}$	C_6H_6, $\mu g \cdot m^{-3}$	PM 2.5, $\mu g \cdot m^{-3}$	PM 10, $\mu g \cdot m^{-3}$
Maximum allowable concentration	10	40	125	120	5	25	40
Critical pollution level	–	30	20	180	–	–	–
Alarm	–	400	500	240	–	–	–

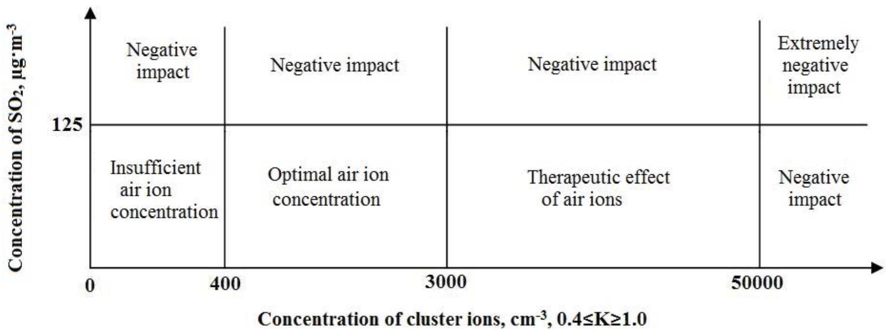
The value of unipolarity coefficient K of both light and heavy air ions is a significant additional factor. From the point of view of medicine and air quality standard, this value should be in the range of $0.4 \leq K \geq 1.0$, but it is clear from the research carried out in this work that it often exceeds the indicated values as a result of daily variations, and in places with pronounced atmospheric pollution the long-term value of K exceeds 1. Table 4.1 represents various theoretical cases when the value of K complies with the requirements, but it should be taken into account that increased or reduced air ion concentration, if the value of K highly exceeds 1, in combination with chemical and physical air pollution of the air results in even more negative impact on the human body.

Another additional circumstance is the different chemical composition of air ions with different polarities. Some studies (e.g. Patil et al., 2012) have found that, during the process of air ion formation or artificial generation, oxygen molecules usually form negative ions, while molecules of different pollutants mainly get positive charge. Overall, the predominance of positive air ions is a negative factor for human health, but their chemical composition determines the level of additional toxicological impact.

Air ion concentration in the atmosphere has an extremely variable character in time and space (Сидоренко et al., 1998). Relatively large variations were also observed in measurements in Rezekne and other places (see Chapter 3.2). Due to the fact that air ion concentration is one of the factors influencing human health and well-being and because of its high variation under natural conditions, this is one of the parameters to be registered in the atmospheric air monitoring program. Usually, air quality monitoring station constantly receives data on chemical and physical pollution of the air that are used to calculate pollution values of 1 hour, 8 hours, 24 hours or other values required by Cabinet Regulation No. 1290; these values help in assessing the level of pollution. It is necessary to measure concentrations of light air ions or cluster ions (0.36–1.6 nm) and heavy ions or aerosol ions (7.1–79 nm) in a similar way. In the first case, it primarily ensures the control of air ions as a beneficial factor, while, in the second case, it controls the unhealthy factor. Continuous measurements of concentration should be carried out with a 5 minute interval, while, taking into account the daily fluctuation cycle of air ions, 1 hour and 8 hour average values play an important role in further interpretation of the data, too. During the daily mode of air ion measurements, 8-hour cycles succeed each other, and the delay of any extreme air ion concentration value for more than 8 hours would indicate a deviation from the usual pollution and air ion cycle. NAIS spectrometer is suitable for such control of the level of air ion concentration (see Chapter 2.1), and it would also replace the measurements of some aerosols and meteorological parameters.



a)



b)

Fig. 4.1. Area of the summary impact of atmospheric pollution and concentration of light air ions: *a)* with compensation of the negative impact; *b)* without compensation of the negative impact.

Table 4.3

Parameters to be measured in the air monitoring station

Type of parameters	Parameter
Chemical	NO_x, SO_2, O_3, CO, CO_2, C_6H_6
Physical	PM 2.5, PM 10, radioactive background, air ion concentration (0.36–1.6 nm and 7.1–79 nm)
Meteorological	Temperature, relative humidity, atmospheric pressure, wind speed and direction

Comprehensive air monitoring system is able to provide the fullest possible assessment of the city's air quality from the point of view of chemical, physical and meteorological factors. From all of these parameters, relative humidity, wind direction, radioactive background, nitrogen and carbon oxides are the main factors affecting air ion concentration, therefore, a common monitoring of all these parameters is necessary to properly analyse the reasons of changes in air ion concentration and, if necessary, to be able to correct them timely.

Information on the concentration of light air ions and its assessment (insufficient, optimal or increased), as well as the value of unipolarity coefficient K should be included in the daily weather assessment, similarly to the way people are informed about atmospheric pressure, temperature and other parameters. In cases where the concentration of light air ions ranges from 100 to 400 ions per cm^3 (i.e. it is considerably reduced, but not catastrophically low), it is recommended to compensate air ion deficiencies by artificial air ionisers at home or working premises, which would help in maintaining appropriate air ion levels.

In accordance with the procedure provided by current laws and regulations, to improve the air quality in agglomerations or areas where the level of air pollution exceeds or may exceed one or more alert levels, the local authority shall develop and implement a short-term action plan including emergency measures to reduce air pollution. The public shall be informed if the respective pollution levels are exceeded. Depending on each specific case, steps may be taken to control or to interrupt activities that increase the possibility of exceeding the relevant threshold or alert level (Cabinet Regulation No. 1290 on the Air Quality, 03.11.2009). Similarly, the public should be informed about extreme values of air ion concentration and their impact on human health of the population.People should be informed in the following cases:

1) when extreme values of light air ion concentration (less than 100 or over 50,000 per cm^{-3}) have been detected for at least 8 consecutive hours;
2) when maximum allowable concentration is exceeded in combination with a large (over 50,000 per cm^{-3}) or very small (under 100 per cm^{-3}) ionisation due to the accumulation of negative effects;
3) whenthe ratio of positive and negative air ions is harmful to human health and well-being, $K \geq 1.5$ in itself or in combination with the above-mentioned cases for at least 8 consecutive hours.

In order to eliminate extreme concentrations of light air ions, if the cause is ascertained, responsible authorities should take the necessary measures, such as redirecting or stopping traffic flows, stopping industrial plants, eliminating consequences of accidents, etc. In case of increased concentrations of heavy air ions, measures should be taken to prevent the spread of fine aerosols in the atmosphere and, if necessary, inform or warn the public.

Air ionisation based on the balloelectric process can be used for the improvement of outdoor air quality in contaminated urban areas. It involves installation of artificial waterfalls and fountains that can purify the air while enriching it with negative cluster ions. In this way, fountains and waterfalls have not only aesthetic, but also a curative function. Such use of artificial ionisation can also add value to the positive impact of natural resorts on human health.

Artificial ionisation is most commonly used in purification of indoor air from aerosols and dust, as well as for disinfection. In specific cases, air ionisers are used to provide appropriate air quality in airtight premises such as underground bunkers, submarines or spacecrafts, along with technologies that simultaneously provide air purification from gases such as CO_2 and other impurities such as aerosols. Nowadays, in the market there is a large variety of equipment suitable for indoor air ionisation; advertisers highlight the positive effects of air ions on human well-being, but none of them provides simultaneous control of air ion concentration that is likely to be quite variable due to various chemical and physical factors. Besides, the level of air ion concentration and unipolarity coefficient is very often in the range that is unfavourable for human health and well-being; therefore, it is recommended to use air ionisers that can stabilise the level of air ion concentration and prevent its natural fluctuations. At the same time, there is a need for daily control of the level of air ion concentration and for regulation of ionisation intensity in order to maintain it within the required limits, because various electronic devices in offices can change the level of air ion concentration, for example intensive use of copying machines significantly increases the concentration of air ions in the ambient air.

Application of mathematical relationships between the concentration of positive and negative air ions and chemical pollution that have been found during this research can help in creation of a methodology for assessing the composition and concentration of pollutant gases under given meteorological conditions, basing solely on the measurements of air ion concentration. These relationships can be used to predict the probable future levels of air ion concentration basing on measurements of some meteorological and chemical atmospheric components.

The primary significance of the mathematical relationships found in this research is scientific and, for greater accuracy, an analysis of measurements taken over several years is necessary. It is also possible to evaluate the environmental security of different technological processes in a similar way. Various combustion processes, as well as activities involving electrostatic fields greatly change the natural level of air ion concentration, thus endangering the personnel involved in the management of these processes.

CONCLUSIONS

1. Previous studies do not clearly answer the questions related to the impact of atmospheric pollutants on air ion concentration. Studies on air ions in Latvia are rare, mainly performed in 1960s, there is a lack of more topical information. There are no data on air ion concentration in urban and rural areas of Latvia.

2. The author has analysed the variations of natural air ion concentration in urban areas with different levels of anthropogenic air pollution. Speaking about cluster ions, the value of unipolarity coefficient K in the most polluted areas of Rezekne city ranges from 0.99 to 1.12, while in urban areas with the lowest level of air pollution the value of K is 0.77. Daily fluctuations in atmospheric pollution of urbanized areas depend mainly on changes in traffic intensity. The level of atmospheric pollution affects the level of air ion concentration. As the concentration of pollutants increases over 24-hour period, the concentration of cluster ions is reduced proportionally (by 30 % on average).

3. The impact of meteorological factors on air ions is variable. Relative humidity does not have large and decisive influence. Air temperature always slightly increases the concentration of air ions. No dependence of air ion concentration on the wind direction was detected, however, in some cases, wind speed has had a positive impact on air ion concentration. Seasonal fluctuations are also typical for air ion concentration. The lowest levels of air ion concentration were established in winter, but the highest concentration was found in spring and autumn.

4. The author has analysed air ion emissions in exhaust gases from vehicles when burning different types of fuel. The lowest level of light air ions is emitted by combustion of liquefied petroleum gas, while the diesel fuel results in the most significant emissions. Internal combustion engine emits light air ions at high concentrations and in varying proportions, but the aggregate negative effect of exhaust gases on the concentration of light negative air ions can be felt after a certain amount of time.

5. The paper studies the dependence of the concentration of cluster ions, middle-sized and heavy ions on chemical air pollutants and meteorological conditions, using a statistical analysis method which is called stepwise regression. The obtained regression model best describes the concentration of negative and positive cluster ions, i.e. R^2 is 0.67 and 0.86 respectively, as well as the concentration of heavy positive ions and the total positive ion concentration, i.e. R^2 is 0.57 and 0.62, respectively. For middle-sized ions, as well as for heavy negative and total negative ions, R^2 of the given model ranges from 0.20 to 0.27.

6. It follows from the obtained regression model that the chemical contamination of air mainly affects cluster ions, as well as heavy positive ions and positive ions in general. Middle-sized ions that can be considered transitional form from cluster ions to aerosol ions are naturally occurring at relatively low concentrations (on average some dozens of ions per cm^3), and they do not strongly depend on chemical pollution, but are affected by other factors, such as wind speed.

7. Basing on the stepwise regression model, cluster ions are mostly affected by NO_2 and SO_2 that increase the number of positive cluster ions. NO, NO_2 and O_3, SO_2 (for positive ions only), as well as PM 10 are the main factors influencing heavy air ions. NO, NO_2, O_3, and PM 10 are the main factors affecting the aggregate air ion concentration. The results suggest that nitrogen oxides play an important role in the formation of chemical composition of air ions.

8. Scientific novelty of the work: this was the first time when the influence of air chemical pollution and physical factors on air ion concentration was analysed basing on outdoor measurements. New mathematical relationships have been found for calculating the concentrations of different ion classes depending on the physical and chemical parameters of the air. It has been found out that stepwise regression is the method of analysis that allows avoiding multicollinearity among the parameters.

9. Possibilities of practical application of the work. The obtained information on air ion levels and unipolarity coefficient in urban and natural environments is useful for planning urban infrastructure, optimisation of traffic flows and improvement of recreation areas. Monitoring of air ion concentrations should also be included in the air pollution monitoring system because air ions have a significant impact on human health.

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