

Variations of Particulate Matter Concentrations and Particle-Associated Substances in Latvia

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Abstract – Concentrations of air pollutants mainly depend on meteorological conditions and pollutant emission levels. Airborne particulate matter (PM_{2.5} and PM₁₀) concentrations were measured in Riga (Latvia) from the end of April to the end of December 2007. The collected particulate matter was analyzed for the anion, cation and heavy metal content. The seasonal variations of particulate matter concentrations were investigated, together with their relationships with meteorological parameters and other pollutant gases. The maximum daily concentration of PM_{2.5} and PM₁₀ reached 52.5 µg/m³ in June and 83.4 µg/m³ in December, respectively.

Keywords – Particulate matter, PM_{2.5}, PM₁₀, Riga

I. INTRODUCTION

Just about 30 years ago British epidemiologists reported that there was no evidence for negative health effects from particulate matter pollution levels [1]. Nonetheless, in 2009 the Environmental Protection Agency of the U.S. reported that the daily pollution of particulate matter, which is experienced by most Americans, is associated with premature deaths [2].

Ambient air pollution has adverse effects on respiratory and cardiovascular systems influencing lung functions, increasing risk of pneumonia, etc. [3].

Particulate matter is a complex mixture of suspended solid and liquid particles. These particles come in many different size ranges, such as coarse, fine and ultra fine. Particles with diameters between 2.5 and 10 µm can enter lungs, however, those with the diameter of less than or equal to 2.5 µm can reach alveolus and from there enter the blood-stream. But there is still a fundamental lack of understanding underlying the mechanisms of their toxicity; one of the widely accepted hypotheses is that the toxicity of particulates depends not only on their size, but also on their composition, both of which depend on location, time of year and meteorological conditions [4].

The combustion of fossil fuels, such as coal, oil and petrol, can produce coarse particles, fly ash, fine particles and secondary particles.

Some studies show that water soluble ions, such as sulfate and nitrate, accounted for one-third or more of the aerosol particles in some urban regions [5, 6, 7]. Moreover, the relative abundances of the above mentioned ions play an important role in determining the hygroscopicity of atmospheric aerosols [8]; and so their tendency to absorb

water at high relative humidity leads to a decrease in visibility [9, 10, 11].

The interest of this research is to find relationships between particulate matter concentrations, their composition and meteorological conditions. Some studies performed on airborne particulates have revealed that heavy metals like Pb, Ni, Cd, As adsorbed by particulate matter may play an important role in generation of adverse health effects.

II. METHODOLOGY

A. Study area

During the last 15 years in Latvia, a significant decrease in total emission of gaseous pollutants has been detected, e.g. the total SO₂ emissions decreased by 96.1 %, NO_x emissions by 42.9 % and NH₃ emissions by 72 %. The reduction is mainly due to the use of fuels with lower content of sulphur, as well as switching from solid and liquid types of fuel to natural gas and biomass, due to fuel consumption and the increasing use of catalyst cars. The main sources of the particulate matter emission are the commercial and residential sectors. Combustion of wood and wood products account for 71.7 % of total PM₁₀ emission, against industry - 10.2 %, traffic - 8.9 % and agriculture - 5.8 % [19].

The long-range pollution transport significantly contributes to the PM₁₀ pollution level caused by the geographical position and close proximity of the sea. Secondary particles are mainly formed by oxidation of SO₂ and NO_x and reaction with ammonia. Long-range pollution transport contributes to about 80 % for oxidized sulphur, 75 % for oxidized and reduced nitrogen from the total deposition in Latvia [20].

In addition, there is a large contribution from natural sources, e.g. soil dust and sea spray.

The study area is situated in the city of Riga (Latvia). Riga is the capital of Latvia and the biggest city in the country (about 0.7 millions inhabitants, accounting for 32 % of the Latvian population) [21].

The monitoring station type is classified as urban-industrial, and it is located close to the Freeport of Riga territory. Freeport of Riga is a significant global and regional cargo supplier. In 2010 the volume of the transhipped cargoes has reached 30.5 million tons, 4040 vessels were mounted. The main types of cargo handled are containers, various metals, timber, coal, mineral fertilizers, chemical cargoes, and oil and food products [22].

TABLE 1

MEASUREMENT METHODS, ACCURACY OF PARTICULATE MATTER RELATED SUBSTANCES

Parameter	Methodology	Accuracy, %	Period
SO ₄	LVS EN ISO10304-1:2004	3.2	03.08-1.09, 03.12-31.12
NO ₃	LVS EN ISO10304-1:2004	2.7	03.08-1.09, 03.12-31.12
Cl	LVS EN ISO10304-1:2004	5	03.08-1.09, 03.12-31.12
Ca	LVS EN ISO 17294-2:2005	2	01.06-30.06, 02.10-02.11
Mg	LVS EN ISO 17294-2:2005	3	01.06-30.06, 02.10-02.11
Na	LVS EN ISO 17294-2:2005	4	01.06-30.06, 02.10-02.11
K	LVS EN ISO 17294-2:2005	3	01.06-30.06, 02.10-02.11
As	LVS EN ISO 17294-2:2005	6	28.04-24.05, 04.07-30.07, 05.09-01.10, 07.11-30.11
Cd	LVS EN ISO 17294-2:2005	5	28.04-24.05, 04.07-30.07, 05.09-01.10, 07.11-30.11
Ni	LVS EN ISO 17294-2:2005	4	28.04-24.05, 04.07-30.07, 05.09-01.10, 07.11-30.11
Pb	LVS EN ISO 17294-2:2005	3	28.04-24.05, 04.07-30.07, 05.09-01.10, 07.11-30.11

B. Measurement methods

The measurement campaign for particulate matter and related substances composition was held from April 28 to December 31, 2007. The particulate matter measurements were done by the beta gauge method, SM200 equipment.

Measurement methods used for heavy metals; anions and cations concentration detection and accuracy is given in Table 1.

The measurement campaign was organized to cover as wide spectrum of substances as possible. An illustrative measurement campaign is given in Table 1 as well.

The field campaign (measurements) and chemical analysis were performed by the Latvian Environmental, Geology and Meteorology Agency.

III. RESULTS AND DISCUSSIONS

Descriptive statistics for particulate matter and related substances during the sampling period is given in Table 2.

A. Variations of PM_{2.5} and PM₁₀ concentrations

The concentrations of PM_{2.5} and PM₁₀ fractions of ambient airborne particulates were measured daily. The highest daily concentrations of PM_{2.5} reached 52.5 µg/m³ (detected on June 12, 2007), but the lowest 1.3 µg/m³ (detected on October 13, 2007). The monthly concentration changes are shown in Fig. 1.

TABLE 2

DESCRIPTIVE STATISTICS FOR PARTICULATE MATTER AND RELATED SUBSTANCES DURING THE SAMPLING PERIOD

	Maximum	Minimum	Median	Mean	Standard deviation	Sampling period
PM _{2.5} , µg/m ³	52.5	1.3	18.0	20.1	10.1	April – December 2007
PM ₁₀ , µg/m ³	83.4	3.5	24.0	27.5	15.4	April – December 2007
SO ₄ , µg/m ³	2.08	0.11	0.76	0.87	0.53	April – December 2007
NO ₃ , µg/m ³	0.95	0.08	0.17	0.29	0.28	April – December 2007
Cl, µg/m ³	0.13	0.05	0.08	0.09	0.03	April – December 2007
Ca, µg/m ³	0.22	0.07	0.15	0.14	0.06	April – December 2007
Mg, µg/m ³	0.07	0.02	0.03	0.04	0.02	April – December 2007
Na, µg/m ³	0.19	0.07	0.12	0.13	0.05	April – December 2007
K, µg/m ³	0.38	0.04	0.12	0.16	0.11	April – December 2007
As, ng/m ³	2.14	0.13	0.31	0.44	0.47	April – December 2007
Cd, ng/m ³	4.35	0.03	0.29	0.64	1.09	April – December 2007
Ni, ng/m ³	4.29	0.24	2.09	2.10	1.06	April – December 2007
Pb, ng/m ³	25.15	1.14	13.31	13.33	7.70	April – December 2007

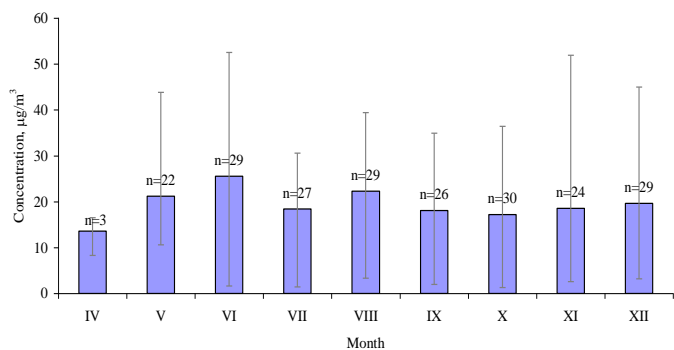


Fig. 1. Monthly variations of PM_{2.5} concentrations during April 28 - December 31, 2007 (n indicated number of observations).

The highest monthly variations of PM_{2.5} were detected in June and November. Unfortunately, no substantial seasonal variations have been found. For all periods of measurements, PM_{2.5} concentrations dominated (42 % of all cases) at a range from 11-20 µg/m³ (see Fig. 2).

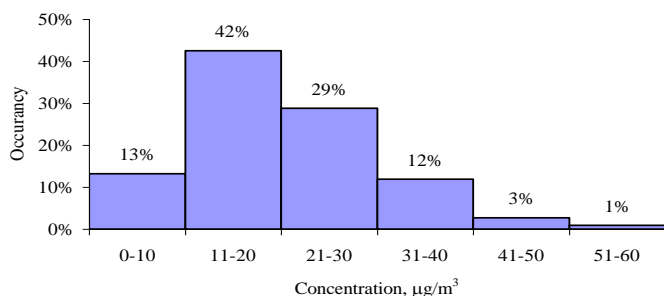


Fig. 2. Distribution of PM_{2.5} concentrations during April 28 - December 31, 2007.

Analysis of the PM_{2.5} monthly distribution shows that practically in all cases the dominating concentrations are at the range from 11-30 µg/m³, and no seasonal differences are found.

The highest daily concentrations of PM₁₀ reached 83.4 µg/m³ (detected on December 27, 2007), but the lowest 3.5 µg/m³ (detected on April 20, 2007). The monthly concentration changes are shown in Fig. 3.

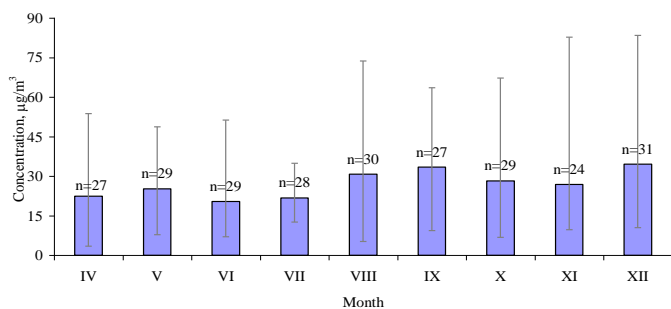


Fig. 3. Monthly variations of PM₁₀ concentrations during April 1 - December 31, 2007 (n indicated number of observations).

The highest concentration variations are observed in the winter period, but the lowest - in the late spring - early summer period. For all period of measurements, PM₁₀ concentrations dominated (58 % of all cases) at a range from 11-30 µg/m³ (see Fig. 4).

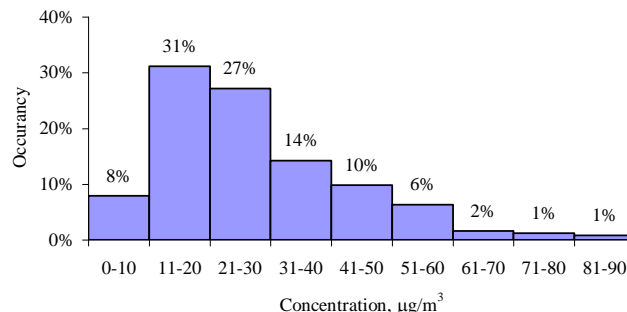


Fig. 4. Distribution of PM₁₀ concentrations during April 1 - December 31, 2007.

According to the measurements and analysis of PM₁₀ concentrations month by month, it has been found that during the summer time concentrations are very stable, they are changing very slowly, they do not split, which means that the influence of local sources is not so important, and the PM₁₀ pollution level mainly originates from the long-range transboundary and background pollution.

Relationships between PM_{2.5} and PM₁₀ mass concentrations were analyzed in terms of the linear regression, and the results revealed that the coefficient of determination is 0.3957 (see Fig. 5). The relatively middle coefficient shows that PM_{2.5} and PM₁₀ could have both similar and different emission sources, and they could be influenced by the same local conditions. Such a relatively low coefficient of determination indicates significant contributions from primary sources, such as re-suspended soil/road dust and other mechanical activities [12, 13, 14].

Some differences were found in particulate matter concentrations during work days and weekends. In case of PM_{2.5} concentrations, it was determined that pollution levels are on average by 10 µg/m³ lower on weekends, in comparison with working days.

B. Relationships between concentrations of particulate matter, meteorological parameters and associated components

Some analogies found in the heavy metals analysis included: (a) concentrations Ni are quite variable, (b) analogies found in Cd and As concentrations variations that could be explained by origin of the same source. No relationships were found between Pb, Cd and Ni variations.

Changes of Ca, K and Mg had some similarities that could be explained by one group of source or, obviously, the reason of such a changes are resuspension and erosion processes (see Fig. 6, 7).

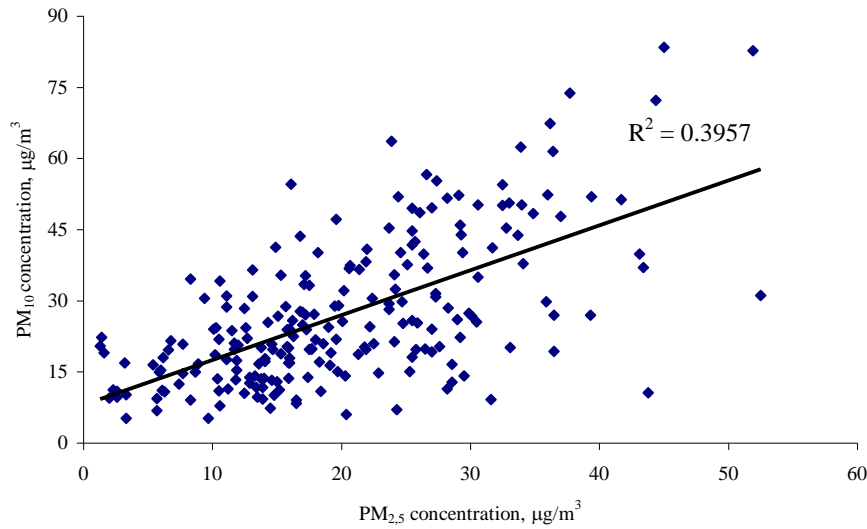


Fig. 5. PM_{2.5} and PM₁₀ mass concentration ratio.

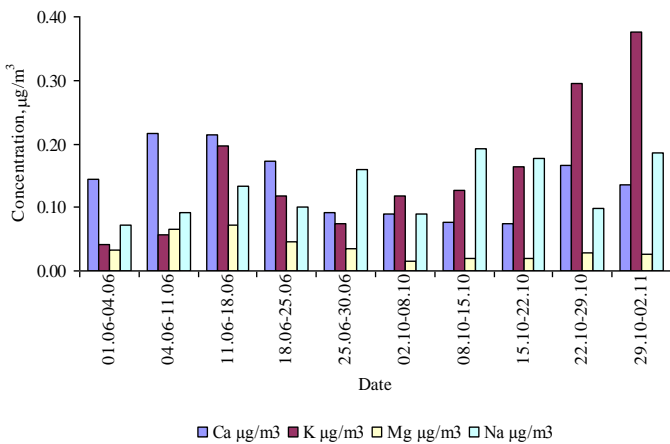


Fig. 6. Concentrations of anions and cations in the PM_{2.5} fraction.

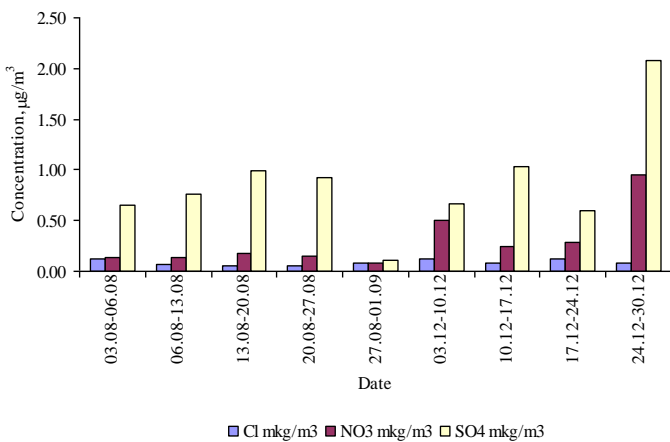


Fig. 7. Concentrations of anions and cations in the PM_{2.5} fraction.

The relationships between PM, heavy metal, cations-anions concentrations and meteorological parameters (temperature, wind speed, relative humidity and precipitation) were investigated by Pearson's correlation analysis. The meteorological data, which were found to be significant in describing the variation in the 24-h averaged PM concentrations utilized in the present work, are: (1) air temperature (°C), (2) relative humidity (%), (3) wind speed (m/s) and (4) precipitation (mm). The correlation coefficients between the daily average (24 h) PM, particle-related substances and meteorological parameters are shown in Table 3. The correlation coefficients were marked in the Table to indicate the significance levels ($P < 0.01$ or < 0.05).

The statistically strongest correlations were detected for PM_{2.5}-Temperature, PM₁₀-precipitation, PM₁₀-Ozone.

Wind speed has long been recognized as an important controlling factor on concentrations of air pollutants [15, 16, 17, 18]. Pursuant to this study, there is no statistical relationship between wind and particulate matter pollution.

The effects of rainfall on particulate concentrations are detected. Some evidences have been found that after longer periods without rainfall particulate matter concentrations are raising, which means that PM have a tendency to accumulate in the atmosphere. Some of the above mentioned accumulation episodes have been identified, e.g. early May and July, late September, October, November and December (see Fig. 8). Moreover, precipitation is a much more effective mechanism for particulate removal from the atmosphere, in comparison with the wind speed.

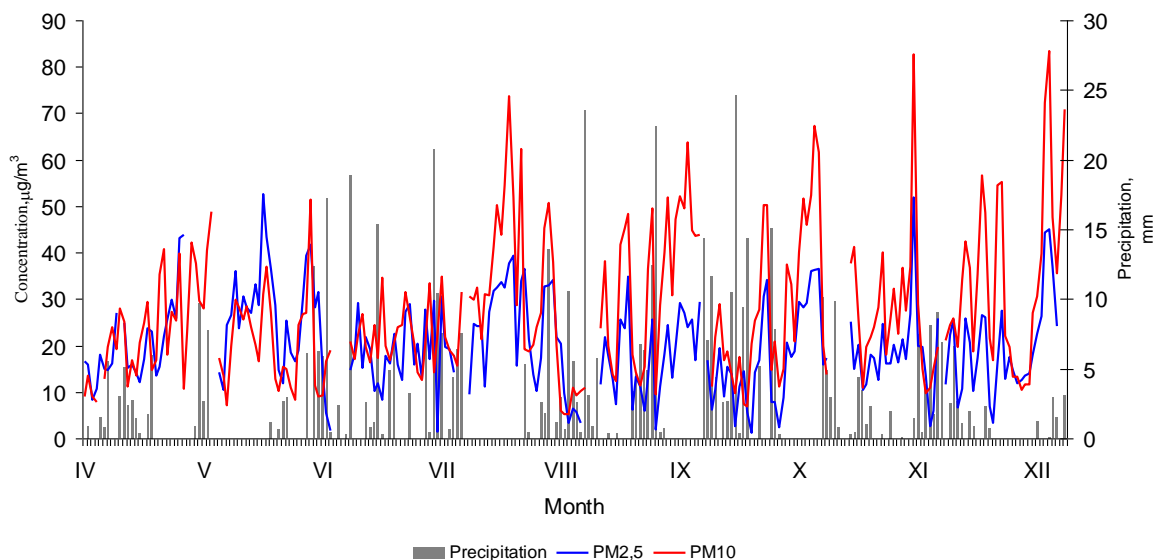


Fig. 8. Particulate matter concentration and precipitation.

TABLE 3

CORRELATION MATRIX OF PARTICULATE MATTER, PARTICLE-RELATED SUBSTANCES AND METEOROLOGICAL PARAMETERS (SIGNIFICANT CORRELATIONS AT P<0.05 LEVEL ARE MARKED IN BOLD)

	PM _{2.5}	PM ₁₀	PM ₁₀ /PM _{2.5}	O ₃	SO ₂	NO ₂	Precipitation	Temperature	Wind speed	Relative humidity
PM _{2.5}	1									
PM ₁₀	0.63	1								
PM ₁₀ /PM _{2.5}	-0.39	0.066	1							
O ₃	0.09	-0.24	-0.07	1						
SO ₂	0.05	0.166	-0.03	-0.089	1					
NO ₂	0.07	0.065	-0.08	-0.344	0.352	1				
Precipitation	-0.16	-0.27	0.05	0.0764	-0.082	0.074	1			
Temperature	0.28	0.008	-0.10	0.4902	-0.168	-0.251	0.16156	1		
Wind speed	-0.03	-0.07	0.06	-0.141	-0.008	-0.162	0.02691	-0.257	1	
Relative humidity	-0.13	0.122	0.04	-0.662	0.175	0.289	0.22735	-0.559	0.171	1

IV. CONCLUSIONS

Measurement of PM_{2.5} and PM₁₀ fractions of ambient airborne particulates and the determination of particulate related substances have been performed in Riga (Latvia).

No seasonal variations were detected for PM_{2.5}, but for PM₁₀ seasonal unstable and higher concentrations were detected in the winter period.

Quite stable and inert PM_{2.5} pollution levels prove that no strong impact of local sources and PM_{2.5} pollution levels could be explained by long-range transboundary pollution and background pollution.

In case of PM_{2.5} concentrations, it has been determined that the pollution levels are on average lower by 10 µg/m³ on weekends, in comparison with work days.

The relatively middle coefficient shows that PM_{2.5} and PM₁₀ could have both similar and different emission sources, and they could be influenced by the same local conditions. Such a relatively low coefficient of determination indicates significant

contributions from primary sources, such as re-suspended soil/road dust and other mechanical activities.

Variable concentrations of Ni ions indicate a pollution source with the periodical operational regime, which could be liquid petroleum furnaces or suchlike.

Similar variations of Cd and Ni ions indicate a legal/non-legal waste incinerator.

Quite high concentrations of Pb ions could be associated with the intensive traffic flow.

Similar variations of Ca, K and Mg ions could be associated with one type of source or the reason for such high concentrations must be resuspension and/or erosion processes.

The statistically strongest correlations were detected for PM_{2.5}-Temperature, PM₁₀-precipitation, PM₁₀-Ozone.

The effects of rainfall on particulate concentrations are detected. Some evidences have been found that after longer periods without rainfall particulate matter concentrations increase, which means that PM have a tendency to accumulate in the atmosphere.

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Iveta Šteinberga. Cieto daļiņu un to sastāva izmaiņu variāciju novērtējums

Cieto daļiņu (PM_{2.5} un PM₁₀) piesārņojuma līmeņa un to sastāva mērījumi veikti Rīgā laika posmā no 2007. gada aprīļa līdz decembrim. Piesārņojošo vielu koncentrācijas gaisā galvenokārt ir atkarīgas no to apjoma un meteoroloģiskajiem apstākļiem. Īpaši dažādu meteoroloģisko faktoru loma ir nozīmīga sekundāro cieto daļiņu rašanās procesā. Cieto daļiņu koncentrāciju mērījumi veikti izmantojot Beta-radiācijas metodi (iekārta SM200), anjonu koncentrācijas noteiktas atbilstoši LVS EN ISO10304-2004 standartam, katjonu un smago metālu – LVS EN ISO 17294-2-2005 standartam. Mērījumi veikti Latvijas Vides, Ģeoloģijas un Meteoroloģijas centrā. Galvenie secinājumi: (1) novērtēto PM_{2.5} koncentrācijas, sezonālas variācijas netika konstatētas, augstākās PM₁₀ koncentrācijas konstatētas ziemas periodā, bez tam, tām novērota augsta mainība (nestabilitāte); (2) salīdzinoši inertās un stabilās PM_{2.5} koncentrācijas nav saistāmas ar lokālu piesārņojuma avotu ietekmi, bet gan ar pārrobežu un fona piesārņojumu; (3) novērtēto PM_{2.5} koncentrācijas konstatēts, ka brīvdienās to piesārņojuma līmenis ir vidēji par 10 µg/m³ zemāks nekā darba dienās; (4) līdzīgā PM_{2.5} un PM₁₀ mainība norāda uz kopēja piesārņojuma avota eksistenci, kā arī līdzīgu meteoroloģisko faktoru ietekmi sekundārā PM rašanās mehānismā; (5) lielā niķeļa jonu koncentrācija norāda uz lokālu piesārņojuma avotu, piemēram, šķidrā kurināmā sadedzināšanas iekārtu, (6) niķeļa un kadmija jonu izmaiņu līdzība norāda uz tuvumā esošu legālu/nelegālu atkritumu sadedzināšanas iekārtu, (7) līdzīgās kalcija, kālija un magnija jonu koncentrāciju izmaiņas norāda uz kopēju piesārņojuma avotu, visticamāk resuspenijas vai erozijas procesiem; (8) statistiski būtiskākās korelācijas konstatētas starp PM_{2.5}-gaisa temperatūra, PM₁₀-nokrišņi, PM₁₀-ozons.

Ивета Штейнберга. Оценка измененной вариации аэрозолей и их состава

Измерения уровней загрязнения аэрозолей (PM_{2.5} и PM₁₀) и их состава проводились с апреля по декабрь 2007 года в Риге. Концентрации загрязняющих веществ в воздухе в основном зависят от их объема и метеорологических условий. Разные метеорологические факторы особенно важны в жизненном цикле аэрозолей. Измерения концентрации аэрозолей проводились с использованием метода бета-радиации (прибор SM200), концентрации анионов определены соответственно стандарту LVS EN ISO 10304-2004, катионов и тяжелых металлов – стандарту LVS EN ISO 17294-2-2005. Измерения проводились в Центре окружающей среды, Геологии и метеорологии Латвии. Главные выводы: (1) оценивая концентрации PM_{2.5}, сезональные вариации не констатированы, самые высокие концентрации PM₁₀ констатированы в зимний период, кроме того наблюдалась высокая их изменчивость (нестабильность); (2) относительно инертные и стабильные PM_{2.5} концентрации связаны не с локальными источниками загрязнения, а с трансграничным и фоновым загрязнением; (3) оценивая концентрации PM_{2.5} констатировано, что в выходные дни уровень их загрязнения в среднем на 10 µg/m³ ниже, чем в рабочие дни; (4) похожая изменчивость PM_{2.5} и PM₁₀ указывает на общий источник загрязнения, а также на влияние похожих метеорологических факторов в механизме появления PM; (5) большая концентрация никелевых ионов указывает на локальный источник загрязнения, например, устройства сжигания жидкого топлива; (6) сходство изменения никелевых и кадмиевых ионов указывает на находящейся по близости легального/незаконного устройства сжигания отходов; (7) сходство изменения концентрации ионов кальция, калия и магния указывает на общий источник загрязнения, скорее всего на процессах ресуспенсии или эрозии; (8) статистически существенные корреляции обнаружены между PM_{2.5}-температура воздуха, PM₁₀-осадки, PM₁₀-озон.