Estimation of the Size Distribution of Suspended Particulate Matters in the Urban Atmospheric Surface Layer and Its Influence on Bronchopulmonary Pathology

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Abstract: The surface layer of the urban atmosphere significantly affects human health. Its quality depends on the level of air pollution. The aim was to determine the size distribution of suspended particulate matters (SPMs) in the atmospheric surface layer of Vladivostok city and to assess the response of the blood immune and metabolic parameters of the population with bronchopulmonary pathology. Sampling of SPMs was conducted in the continental zone of the city with a high level of technogenic pollution (138 samples) and the island part of the city with an insignificant level of technogenic pollution (132 samples). The SPM fractional composition was analyzed by laser granulometry. We examined patients with bronchopulmonary pathology living in a one-kilometer zone centered on sampling area for at least 5 years (continental territory—220 patients, island territory—176 patients). We calculated the D% index characterizing the integral response of the blood immune and metabolic parameters to the exposure of dust fractions. It was found that PM > 10 (mainly of a natural origin) predominate in the island zone with insignificant level of technogenic pollution. The PM10 fraction prevails in the area with a high level of technogenic pollution. The response of the immune and metabolic systems to the exposure to microparticles in population living in the marine and forest/park zone of the island indicates a health-improving effect of the area. Low values of D% were detected for the patients living in the area with a high air pollution level, indicating a pathogenic reaction of immune and prooxidant-oxidative systems.

Keywords: suspended particulate matter; atmospheric surface layer; immune system; lipid peroxidation-antioxidant defense system; bronchopulmonary pathology

1. Introduction

The promotion of a healthy living environment in industrial cities is an urgent problem of our time [1–3]. Currently, urban air pollution by technogenic emissions from vehicles, energy enterprises, and other industrial facilities is an acute problem. Suspended particulate matters (SPMs), and chemical and toxic components of the urban atmosphere affect the health and quality of life of the population [4–7]. A significant dangerous factor affecting the human respiratory system is the accumulation of SPMs in the surface layer of the atmosphere (breathing zone up to 2.5 m). The tracheobronchial (particles with an aerodynamic diameter of less 10 µm, PM10) and respirable (particles with an aerodynamic diameter of less 2.5 µm, PM2.5) SPM fractions are of particular interest to researchers [8–11]. Ultrafine particles (UFPs) less than 0.1 µm in diameter have a significant negative impact on human health. The incorporation of UFPs leads to the disruption of functioning and energy state of cells [12]. The impossibility of quickly removing micro-sized SPMs from the body leads to their accumulation, which may contribute to the development of bronchopulmonary diseases [13–15]. Oxidative stress and systemic inflammatory response play an important role.
in the pathogenesis of bronchopulmonary diseases [16]. These two processes are mutually influential and also represent the key targets of exposure to fine and ultrafine dispersed air toxicants, which entails negative consequences for the respiratory system and the whole organism. The response of the blood immune and metabolic parameters is a diagnostic and prognostic criterion for the health status of the population in an urbanized area.

The aim of the study was to determine the fractional composition of suspended particulate matters (SPMs) in the atmospheric surface layer of Vladivostok and to assess the response of the blood immune and metabolic parameters of the population with bronchopulmonary pathology.

2. Materials and Methods

The continental (with a high level of technogenic air pollution) and island (with a relatively low technogenic air pollution) territories of Vladivostok city were included in the study.

The collection of atmospheric suspensions for the determination of suspended particle size distribution was carried out in the “breathing zone” (h = 1.5–2.5 m) based on the technique developed by Vladivostok Branch of Federal State Budgetary Science Institution “Far Eastern Scientific Center of Physiology and Pathology of Respiration”—Institute of Medical Climatology and Rehabilitative Treatment [17].

Sampling of SPMs was carried out in the continental (138 samples) and island parts (132 samples) of Vladivostok during the hours of intense technogenic air pollution (10 a.m.–1 p.m.) in the period of 2013–2018 [17] (Figure 1).

Figure 1. Location of the sampling sites of the atmospheric surface layer in two districts of Vladivostok with different levels of technogenic pollution.
The atmospheric surface layer at the sampling point of the continental part of the city is characterized by a high level of technogenic pollution associated with the proximity of the incineration plant (500 m from the sampling point, chimney height—98 m) and heating plant powered by gas during the heating period (200 m from the sampling point, chimney height—30 m) [18]. However, the main impact on pollution of the atmospheric surface layer in this area is caused by high intensity of vehicle traffic (2400–3000 vehicles per hour). The residential development is dense, with a lot of 5–8-storey buildings and few green spaces.

There are no large enterprises near the sampling point in the Russkiy Island, where a relatively low level of technogenic pollution is observed for the atmospheric surface layer. A low traffic intensity (40–60 vehicles per hour) is observed on the dirt roads of the island. The quality of air environment in this zone is predominantly influenced by dirt roads as a source of dust emissions. The territory is located close to sea and large areas of broadleaf forests. The residential development at the sampling point is insignificant, with only sparse buildings.

Atmospheric air samples were collected into a liquid absorption medium (highly purified water) and a high-speed Richter’s absorber at a rate of 10 l/min using a PU-4E electric aspirator (KHIMKO, Moscow, Russia) [17] (Figure 2). Richter’s absorber No. 1 contained a liquid medium for collecting SPMs. Richter’s absorber No. 2 (empty) located between the Richter’s absorber No. 1 and the inlet connector of the corresponding channel was used to prevent the liquid absorber from getting into the aspirator. Simultaneously, a sample was absorbed on a filter (AFA-VP-20) at a rate of 35 l/min. Then we determined the total mass of suspended particles and calculated their concentration per unit volume.

![Figure 2. Scheme of apparatus for collecting atmospheric air samples into a liquid absorption medium: (1)—Richter’s absorber No. 1 with a liquid medium; (2)—Richter’s absorber No. 2 without medium; (3)—PU-4E aspirator: (a)—channel No. 2, air flow rate of 10 l/min; (b)—channel No. 4, air flow rate of 35 l/min; (4)—AFA-VP-20 filter; (5)—battery (12 V).](image)

The atmospheric air-sampling procedure consisted of six successive cycles of 30 min each (the time of one cycle corresponds to the sampling time for calculating the short-term exposure limit value) with an interval between cycles of 5–10 min. The meteorological indicators (air temperature and atmospheric pressure) were estimated at the beginning of each cycle for standardizing the method of evaluation of SPM concentration [17].

Analysis of the SPM size distribution expressed in mass fractions was carried out using Analysette 22 NanoTech laser analyzer (Fitsch, Germany).

SPMs were classified into the following size ranges: <0.1, 0.1–1, 1–2.5, 2.5–10, 10–50, 50–100, 100–400, 400–700, 700–2000 μm. Their percentages of the total mass of dust fractions were determined. The results are presented as the mean values and mean error (± m). Comparative analysis in different groups was carried out using the t-test. Results were reported as significant when p < 0.05.

For the period of 2013–2018, we examined 396 people living in the continental zone (170 patients with respiratory diseases, 50 healthy volunteers) and on Russkiy Island (131 patients with respiratory diseases, 45 healthy volunteers). Asthma and chronic obstructive pulmonary disease (COPD) were diagnosed in accordance with the Global Strategy
for Asthma Management and Prevention, Global Strategy for the Diagnosis, Management, and Prevention of Chronic Obstructive Lung Disease, and the International Classification of Diseases (10th Revision). The control group included healthy volunteers. The study was conducted in accordance with the requirements of the Declaration of Helsinki (2013) and approved by the Ethics Committee of Vladivostok Branch of Federal State Budgetary Science Institution “Far Eastern Scientific Center of Physiology and Pathology of Respiration”—Institute of Medical Climatology and Rehabilitative Treatment. Voluntary informed consent to the examination was obtained from each patient. The study included subjects living for at least five years in the studied area (within a radius of 1 km from the sampling point). The exclusion criteria were acute infectious diseases, chronic diseases of internal organs in the acute phase, decompensated heart failure.

The assessment of the health status of patients included general clinical and laboratory examination, which was carried out on the following day after the date of sampling. The venous blood was used as the biological material. Fourteen key immune parameters were estimated. Parameters of cellular immunity (CD3+, CD4+, CD8+, CD4+/CD8+, CD19+, CD16+56+; BD Multitest 6-color TBNK, BD Biosciences, San Jose, CA, USA), the levels of serum cytokines (tumor necrosis factor α (TNF-α), interferon γ (IFN-γ), interleukin 4 (IL-4), IL-6, IL-10, IL-17A; Cytometric Bead Array System, BD Biosciences, San Jose, CA, USA) were measured by flow cytometry (BD FACSCanto II, BD Biosciences, San Jose, CA, USA). Phagocytic activity of neutrophils was determined according to Mayansky et al. [19]. Oxygen-dependent mechanisms of bactericidal activity of peripheral blood neutrophils were evaluated by the method of Shmelev et al. [20]. The assessment of the lipid peroxidation-antioxidant defense (LPO-AOD) system included five parameters: total antioxidant activity (AOA) (Randox Laboratories Ltd., County Antrim, UK), the blood level of malondialdehyde (MDA) (Next Level Security Systems, Carlsbad, CA, USA), MDA/AOA ratio, the blood level of antioxidant enzymes—glutathione peroxidase (GP), glutathione reductase (GR), and reduced glutathione (GSH) (MyBioSource, San Diego, CA, USA).

The integral response characterizing the intensity of exposure to various fractions of dust particles in atmospheric surface layer on the blood immune (14 parameters) and metabolic (5 parameters) parameters of the subjects was determined using the statistical module “Multiple Pearson’s correlation” (STATISTICA 8). Intersystem (subject—environment) rectangular matrices of 7 × 5 parameters (fractions of dust particles—LPO-AOD system) and 7 × 14 parameters (fractions of dust particles—immune system) were formed, in which statistically significant paired correlations (r, p < 0.05) were selected. As the result, the integral indicator (D%) of the response was calculated by dividing the sum of statistically significant relations by the sum of hypothetically possible values in rectangular matrix (R = 1.0): D% = Σ absolute values of r/ΣR x 100.

3. Results

The particle size distribution in air suspensions of the atmospheric surface layer in the territories with various levels of technogenic air pollution (the continental and island parts of Vladivostok city) was determined. The analysis showed that 10–50 µm SPMs predominated in the island part of the city. The most pronounced differences in the content of dust particles between studied territories of the city were noted for the 0.1–10 µm and 400–700 µm fractions (Table 1).
Table 1. The content of atmospheric surface layer SPMs (%) of different size ranges in the continental and island parts of Vladivostok.

| SPMs (%) | Russkiy Island | Continental Part of the City |
|----------|----------------|-----------------------------|
| <0.1 µm  | 0.044 ± 0.005  | 0.764 ± 0.029 (p = 0.002)  |
| 0.1–1 µm | 0.612 ± 0.065  | 1.577 ± 0.262 (p = 0.014)  |
| 1–2.5 µm | 7.388 ± 0.713  | 11.955 ± 0.533 (p = 0.037) |
| 2.5–10 µm| 26.188 ± 2.513 | 45.721 ± 1.872 (p = 0.021) |
| 10–50 µm | 34.889 ± 3.869 | 11.034 ± 2.713 (p = 0.046) |
| 50–100 µm| 0.878 ± 0.045  | 0 (p = 0.009)               |
| 100–400 µm| 4.276 ± 0.236  | 0.688 ± 0.028 (p = 0.003)  |
| 400–700 µm| 3.652 ± 0.605  | 8.873 ± 0.310 (p = 0.024)  |
| 700–2000 µm| 21.669 ± 1.988 | 19.383 ± 1.111             |

The differences in the level of finely dispersed fractions (PM2.5) between the studied areas were highlighted. The content of ultrafine particles (<0.1 µm) in the area with a significant technogenic air pollution (the continental area) was 17.4 times higher than in the area with an insignificant technogenic load (Russkiy Island). The levels of 0.1–1 µm and 1–2.5 µm SPM fractions in the continental area of Vladivostok exceeded 2.6 and 1.6 times, respectively, compared to the island territory. The percentage of particles with an aerodynamic diameter of 2.5–10 µm in the continental zone was 1.7 times elevated relative to the area with an insignificant technogenic load.

At the same time, the content of PM > 10 particles with size ranges of 10–50, 100–400, 700–2000 µm in the island territory increased 3.1, 6.2 and 1.1 times, respectively, compared to the continental area. The prevalence of the 50–100 µm PM fraction was the most pronounced. The percentage of particles with an aerodynamic diameter of 400–700 µm in the continental zone was 2.4 times higher than in the island territory.

When comparing the pollution of the atmospheric surface layer by SPMs in the areas with different technogenic load, it was revealed that the percentage of particles of the most pathogenic fractions PM2.5 and PM10 in the continental technogenically unfavorable zone of Vladivostok was 1.8 times higher than in the territory of the relatively favorable city zone (Russkiy Island).

The determination of the effect of dust fractions on the blood immune and metabolic parameters in patients with bronchopulmonary pathology was carried out using D% index. For two areas (continental and island), the correlations were grouped by seven SPM size ranges for each group of blood parameters (Table 2).

The healthy residents of the continental territory exhibited a more intensive response of the immune and LPO-AOD systems (more than 1.5 times for each system) to the exposure to SPMs of the atmospheric surface layer than the residents of the island territory. The greatest response was observed for the particles with an aerodynamic diameter from 1 to 10 µm. In healthy subjects living on the island territory, there was no marked reaction of the LPO-AOD system to specific dust fractions, and the immune parameters reacted to SPMs over the ranges of 0–10 µm and 50–100 µm. The highest total values of D% index in healthy volunteers were found for the lipid peroxidation-antioxidant defense system.
Table 2. Effect (D%, p < 0.05) of atmospheric surface layer SPMs of different size ranges on the blood immune and metabolic parameters in healthy subjects and patients with bronchopulmonary pathology living under different environmental conditions.

| Size Range of SPMs (µm) | Island Part of the City | Continental Part of the City |
|------------------------|-------------------------|-----------------------------|
|                        | LPO-AOD System          | Immune System               | LPO-AOD System | Immune System |
|                        | Control group           |                            |                |               |
| 0–1                    | 0.28                    | 0.32                       | 0.72           | 0.43          |
| 1–10                   | 0.29                    | 0.36                       | 0.92           | 0.41          |
| 10–50                  | 0.32                    |                            | 0.39           | 0.31          |
| 50–100                 | 0.21                    |                            |                |               |
| 100–400                | 0.19                    |                            |                |               |
| 400–700                | 0.24                    |                            |                |               |
| >700                   |                         |                            |                |               |
| Total                  | 1.53                    | 0.91                       | 2.57           | 1.52          |

On the territory with low air pollution by SPMs, the population with bronchopulmonary pathology had a slight increase in the total value of response in comparison with the control group (Table 2). The intensity of cumulative response of both the immune and LPO–AOD system parameters in patients with bronchopulmonary pathology living on the continental territory was reduced in comparison with healthy individuals. The patients were found to exhibit the response of the LPO-AOD and immune systems to SPMs with an aerodynamic diameter 0–400 µm and 0–100 µm, respectively. The total D% values of trigger systems in patients with bronchopulmonary pathology living on the continental territory were lower than in the healthy population.

4. Discussion

Vladivostok city is characterized by a high level of air pollution associated with vehicle traffic (>500 vehicles per 1000 residents) and man-made emissions. Monsoon climate with characteristic seasonal variability of wind directions, dissected relief of the city, and the coastal location of residential areas cause the accumulation of surface fine dust with pathogenic properties, which ambiguously affect various systems of human body, in the inter-hill valleys [13,21,22].

Differences in the SPM concentration in the urban air environment of the continental and island parts of Vladivostok are caused by the level of technogenic load on the territories. Traffic-related air pollution has the most negative impact on respiratory function [23].

The analysis of SPM content in the atmospheric surface layer of areas with different levels of air pollution has shown a significantly lower content of ultrafine particles (UFP) and suspended particles with an aerodynamic diameter from 1 to 10 µm on Russkiy Island compared to the continental part of the city (Table 1). Given the insignificance of vehicle traffic on Russkiy Island as the main source of airborne fine PM2.5, the natural origin of these particles can be assumed (sea aerosols, aerosols formed in the result of soil erosion, forest biological aerosols containing fungi, bacteria, and pollen) [24–26]. The presence of fine PM2.5 fraction in the island part of the city may be slightly related to the use of wood and coal during the heating season [27]. The high levels of particle fractions of 10–50 µm and 700–2000 µm in this territory may be explained by the increased dustiness...
of the surrounding area as the result of dirt road operation (Table 1) [28]. It should be noted that PM > 10 remain in the atmospheric air for a short period due to gravitational sedimentation. When entering the respiratory tract, these particles are sedimented in the nasal cavity and excreted from the body.

In the polluted urban environment (continental area), the source of fine and ultrafine SPMs is mostly man-made emissions (road dust, construction waste, products of oil combustion, and operational wear of components of the roads and automobiles) [29]. When considering the content of different dust particle fractions in the air of this territory, we detected the prevalence of SPMs of the size range from ultrafine particles to particles with diameter 2.5–10 µm that are directly related to road and roadside dust, year-round operation of an incineration plant, and operational wear of components of the roads and automobiles (Table 1). These particle fractions (especially 0–2.5) have the most negative effect on the respiratory organs.

The analysis has shown a higher level of systemic compensatory capabilities in healthy subjects living in the unfavorable continental zone compared to the population with bronchopulmonary pathology. The reaction of the lipid peroxidation-antioxidant system was the most indicative. The immune system also actively responds to the exposure, but its response was characterized by a lower intensity of reaction (Table 2). The reaction observed among healthy population of the island area can be associated with the sanogenic properties of this environment, which can be used in health-improving and preventive healthcare activities.

In the population with bronchopulmonary pathology of the continental territory, an anergic reaction associated with oxidative stress, systemic inflammation, and impaired adaptive mechanisms was detected. Fine and ultrafine SPM fractions presented in the polluted air environment of the continental area had predominantly technogenic origin, therefore the lower D% values in patients with bronchopulmonary pathology compared to healthy subjects living on this territory imply pathogenic effect of these particles [30,31]. It is known that the respirable fractions (PM2.5) penetrating the most deeply into the respiratory system lead to the disruption of the functioning and energy state of cells [12,15,16]. Inhaled PMs induce oxidative stress in the lungs, as well as interact with various components of the immune system and increase allergic inflammatory response. Moreover, the particles not only enter the circulatory system, but can also absorb many airborne toxic substances, such as heavy metals, PAHs, and organic/inorganic ions. It has been hypothesized that PMs induce oxidative stress through several different mechanisms. Firstly, the oxidation-reduction cycle of some components absorbed on the particle surface, such as iron or quinones, leads to the generation of ROS, hydrogen peroxide, and a damaging hydroxyl radical in the lungs [32]. Secondly, particle-attached bacterial endotoxins can cause inflammation [33,34]. PMs enhance airway inflammation by interacting with innate and adaptive components of the immune system. SPMs activate neutrophils and eosinophils by increasing the levels of proinflammatory cytokines, most notably interleukin 6 and 17. PMs induce inflammatory reactions mediated by antigen-presenting cells, as well as an imbalance of T helper (Th) cell subpopulations characterized by an increase in the number of Th2 and Th17 cells and a suppression of Th1 cells. Additionally, UFPs penetrate cell membranes and directly interact with intra-cell structures [35,36]. Mills et al. demonstrated that UFPs were found in the blood immediately after inhalation and remained within the lungs up to 6 h after the exposure. UFPs can cause severe eosinophilic inflammation and epithelial damage [37]. Long-term chronic systemic inflammation in patients with bronchopulmonary pathology is characterized by the impairment of adaptive processes. Therefore, the air environment contaminated with microtoxicants induces the response of the trigger systems of the body, the immune system and the lipid peroxidation-antioxidant defense system, in the population of the unfavorable territory (the activation of these systems in healthy people and their anergy in people with bronchopulmonary pathology).

The strength of the study is the assessment of SPM pollution of the atmospheric surface layer, which corresponds to the human breathing zone and has the greatest impact.
on the human respiratory system. The study is focused on analysis of air pollution by different size ranges of particles and their integral effect on the trigger parameters of the human body. The limitation of the study is the fact that some of the identified effects may be region-specific, i.e., associated with the regional characteristics of the studied areas. It is necessary to expand understanding of particular nosological forms and to develop quantitative criteria for the SPM exposure, taking into account climatic factors in order to predict the development and progression of ecology-dependent respiratory pathologies.

5. Conclusions

Urban air pollution depends on an intensity of emissions from motor vehicles and energy enterprises, distance to the water surface, and area of green space. In Vladivostok city, it was found that PMs >10 of natural origin prevail in the atmospheric surface layer in the green coastal zone with insignificant technogenic pollution. The evaluation of the effect of SPMs on human health and its adaptive potential is based on the origin (natural or man-made) of dust particles and their ability, depending on their aerodynamic diameter, to penetrate into the body and cause the disruption of functioning of the immune and antioxidant systems. The healthy population in the urban environment exhibits an adequate compensatory response. The D% index calculated for patients with bronchopulmonary pathology has indicated the inability of the body to develop a defense reaction. The pathogenic effect observed in the urban zone with intense air pollution is mainly associated with technogenic fine particles.

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