Toxicology Reports 6 (2019) 120–125

Contents lists available at ScienceDirect

Toxicology Reports

journal homepage: www.elsevier.com/locate/toxrep

Morphological and chemical composition of particulate matter in buses exhaust

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A R T I C L E I N F O

Keywords:
Bus exhaust gases
Particle size distribution
Environmental toxicity
Particulate matter
Air pollution
Solid particles

A B S T R A C T

This research article investigates the particulate matter originated from the exhaust emissions of 20 bus models, within the territory of Vladivostok, Russian Federation. The majority of evaluated buses (17 out of 20) had emissions of large particles with sizes greater than 400 µm, which account for more than 80% of all measured particles. The analysis of the elemental composition showed that the exhaust emissions contained Al, Cd, Cu, Fe, Mg, Ni, Pb, and Zn, with the concentration of Zn prevailing in all samples by two to three orders of magnitude higher than the concentrations of the other elements.

1. Introduction

Despite the fact that the bus is one of the most vital transportation means in urban areas playing a vital role in transportation of the public, particulate matter originated from their emissions act as a major pollutant of the atmosphere in the ambient air. Buses, along with the 2-vehicle transportation means, are one of the most significant sources of air pollution in modern cities [1]. While in operation, various concentrations of carbon dioxide, nitrogen oxides, particulate matter ($PM_{2.5}$ and PM_{10}), sulfur dioxide, carbon monoxide and volatile organic compounds (VOCs) are emitted into the atmosphere [2] causing a potential hazardous impact on human health [3–6]. Particulate matter originated from the exhaust gases of diesel engines is a complex mixture of organic molecules, insufficiently oxidized carbon, metal oxides, with the content of sulfate and nitrate groups [7]. The properties of these particles depend on the operating condition of the engine, the composition of the fuel, the lubricant oil, and the exhaust gas filtering equipment [8]. Soot can form in conditions of incomplete combustion due to lack of air, poor mixing of air with combustion gases and low combustion temperature. Under such conditions, hydrocarbon fragments have a high probability of collision with other fragments of hydrocarbons, and agglomeration occurs, instead of completely oxidizing them to CO, H_2, CO_2 and H_2O [9–11]. Substances including hydrocarbons (CHn) and their low boiling point derivatives are called volatile organic compounds (VOCs).

In addition to organic compounds and soot, particles in internal combustion engines exhaust, contain various chemical elements, including heavy metals, both in a free state and adsorbed on the surface of soot and ash particles. Bus exhaust gases can contain V, Cr, Al, P, Mn, Ni, K, Sr, Cd, Cu, Fe, Zn, Na, Ba and Zr [8,12,13] with the highest concentrations found for K, followed by Zn, Na, Fe, and P [13].

EGS are predominant constituents of $PM_{2.5}$ in urban areas [14] and can affect a variety of respiratory diseases. Experimental studies have recently reported that EGS exacerbate allergic asthma and neutrophilic lung inflammation which are related to bacterial endotoxin from gram-negative bacteria [15].

Abbreviations: PM, particulate matter; PM_{10}, particles with a diameter between 2,5 and 10 µm; VOCs, volatile organic compounds; EGS, exhaust gas suspension

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https://doi.org/10.1016/j.toxrep.2018.12.002
Received 18 October 2018; Received in revised form 3 December 2018; Accepted 7 December 2018
Available online 11 December 2018

Studies have shown that particulate matter from internal combustion engines exhaust can affect human health [16]. They can cause various respiratory and cardiovascular diseases, changes in the human immune system, damage to lung tissue, and carcinogenesis. Such effects on human health can be compared with toxic effects resulting from long term exposure to a complex mixture of pesticides, food additives and lifestyle products [17] all contributing to a premature death [18]. The World Health Organization (WHO, 2012) reported that chronic effects of air pollutants, including particulate matter (PM), lead to the premature death of 7 million people annually while air pollution is reported to be the most important environmental health risk in the world [19].

Bus passengers are exposed to high concentrations of particulate matter throughout their entire route [20]. The exhaust gas leaks into the cabin and passenger compartment of a bus during its parking, await in traffic jams, and upon various openings of doors and windows. This so-called "self-pollution" significantly increases the exposure of passengers, in this type of public transport, to particulate matter. It has been shown that up to 0.3% of cabin air of a motor vehicle, is its own exhaust gas [21]. In addition, an increased level of air pollution related to the exhaust gas was observed in the cabins of buses at route terminals and at long-term parking areas, where many buses simultaneously ran at idle speed [22]. It was shown that the $PM_{2.5}$ particles level in the bus parking areas was above the urban environment and averages at 32 mg/m^3 (the range is 22–52 mg/m^3) [12].

The aim of this research is to study the qualitative composition of particulate matter in the exhaust of buses running in Vladivostok city.

2. Materials and methods

In accordance with the Russian classification ON 025,270 66 [9] and the Classification of the Economic Commission for Europe, 20 types of buses representing the most significant ones regarding their environmental impact (by emissions) and bus types widely used in Vladivostok, have been selected (Table 1).

2.1. Sampling procedure for the particles from buses exhaust emissions

The research object was the exhaust gas suspension, obtained according to the procedure described below. The following equipment and materials were used to achieve the EGS and carry out the measurements: a 20 L plastic container, a polyvinyl chloride hose (1 m long and 50 mm in diameter for each measurement) and distilled water (10 L for each measurement). When the exhaust passes through the water, it cools down and traps most of the particulate matter contained in the exhaust gas. Prior to the measurement, the plastic container and hose was washed with deionized water from an ultra-pure water treatment system (SGWASSER UltraClear TWF/EL-ION UV plus TM, Siemens, Berlin, Germany).

Table 1
List of Buses used in the experiment in correspondence with the displacement, type of fuel, year of manufacture and passenger capacity.

No.	Bus model	Displacement (litres)	Fuel	Date of issue	Passenger capacity (number of seats)
1	FT	2.2	diesel	2012	17
2	G1614	2.8	diesel	2012	17
3	HC	4	diesel	2003	24
4	HC	4	diesel	2009	24
5	HAT	7.5	diesel	1998	33
6	HAT	7.5	diesel	1999	33
7	KLXMQ6127C	8.9	diesel	2014	51
8	HU	9	diesel	2013	43
9	DBS106	11	diesel	1993	91
10	DBS106	11	diesel	1995	91
11	DBS106	11	diesel	2005	91
12	DBS106	11	diesel	2006	91
13	DBS106	11	diesel	2010	91
14	DBH115	11	diesel	2003	45
15	HSAC	11	diesel	1993	75
16	HSAC	11	diesel	1994	75
17	HSAC	11	diesel	1995	75
18	HSAC	11	diesel	2010	75
19	KG	12	diesel	2011	45
20	HAQ	16	diesel	1996	49

2.2. Characterization of particulate matter from exhaust emissions

Laser particle size analysis of the obtained suspension was performed using a particle sizer ANALYSETTE 22 NanoTec plus (Fritsch GmbH, Germany) according to the vendor's recommendations using standard analytical software. The measurements were carried out in 'nanotec' mode with carbon/water 20 °C settings in three repeats. The results of particle size analysis of the bus EGS are presented in Table 2.

By static evaporation, a dry residue was obtained which was examined using an optical microscope combined with RAMAN spectroscopy Morphologi G3-ID (Malvern, Kaiser). RAMAN spectra were obtained using 785 nm laser at 8 Mega Watts, with the spectral resolution of 4 cm^{-1}.

In order to identify additional components contained in the particulate matter in the exhaust of internal combustion engines of buses, the elemental analysis of the EGS was carried out. The analysis metal content in the samples was performed using an atomic emission spectrometer with an inductively-coupled plasma ICE-9000 (Shimadzu). The results of the analysis of the content of metals are presented in Table 3.

2.3. Statistical data processing

All experiments were carried out 3 times, statistical processing of the results was done using soft Statistica 10 (StatSoft, Inc., USA). An assessment of the statistical significance of the indicators and differences of the samples were done under consideration according to the Student's *t*-test taking into account the characteristics of the distribution of quantities. A value of $p \leq 0.05$ was considered statistically significant.

3. Results

For sampling, we chose the idle mode. When the engine is idling, the content of products of incomplete combustion, mainly carbon monoxide and hydrocarbons, increase in the exhaust gases of automobiles, while the concentrations of nitrogen oxides remain low. When the engine is operating at medium and full loads, compared with idling, the concentrations of carbon monoxide (3–4 times) and hydrocarbons (5–7 times) decrease. One of the main problems for the truck industry is the problem of engine idling. After driving for a certain time period, drivers need to rest. During this time, drivers keep the engine idling to maintain the comfort of the cabin and to provide power, such as air conditioning (cooling and heating), refrigerators and microwaves. Studies show that long-distance trucks are idling between 6 and 16 h a day. When the engine is at idle, it consumes a rich mixture of air and fuel, so fuel consumption is high. In addition, during idling, the engine cannot operate at maximum operating temperature, and fuel combustion is incomplete, which leaves fuel residues in the exhaust consequently increasing emission levels. Therefore, we decided this as the most appropriate particular mode to study.

The results of particle size distribution analysis of all bus models EGS are presented in Table 2.

The analysis of the qualitative composition of particulate matter in the exhaust gase of buses has shown that the particulates upon entering the atmosphere are predominantly carbon (ash and soot) agglomerates having different geometric shapes, arithmetic mean size and specific surface area [23].

The results of the experiments in Table 2 show that the particle size

Table 2
Listed results of particle size distribution of the EGS of buses in the experiment.

Coded vehicle model	Displacement (litres)	Percent of fraction (distribution by number), μm							Mean diameter (distribution by number), μm
		≤1	1–10	10–50	50–100	100–400	400–700	≥700	
FT	2.2		0.1	0.2		13.4	42	44.3	671.48
G1614	2.8	0.5	1.3				2.5	95.7	1033.75
HC	4	0.1	0.1	0.3		19.6	40.2	39.7	642.67
HC	4					6.1	35.3	58.6	768.43
HAT	7.5	0.1		0.3		21.5	42.8	35.3	614.12
HAT	7.5	4.1	8.3				7.9	79.7	833.43
KLXMQ6127C	8.9	0.4	3.3	15.1		7.5	16.7	57	670.6
HU	9			0.3	13.4		38.4	47.9	685.69
DBS106	11	6.7	20.3				2.5	70.4	749.04
DBS106	11		0.1	0.3		10	33.1	56.4	736.4
DBS106	11						5.1	94.9	997.85
DBS106	11					12.4	38.2	49.4	700.13
DBS106	11	3.9	8.1	2.8	0.1	4.6	7	73.5	784.97
DBH115	11	22.7	77.3						1.55
HSAC	11	0.4	1.4	1.1		15.4	37.3	44.4	663.44
HSAC	11	7	25.6	4.7		18.8	15.9	28	410.87
HSAC	11	0.1		0.2		11	35.6	53.1	714.87
HSAC	11		0.1	0.4		19.3	42.5	37.7	631.61
KG	12	0.4	1.5	3.1		15.6	32.3	47.1	667.18
HAQ	16		0.1	0.4		18.2	41.3	40	643.59

distribution in most studied vehicles is divided into two separate groups, which are consistent with previous studies [24,7].

The first group consists of particles less than 50 μm in size. The second group consists of particles with sizes ranging from 100 to 1000 μm. The vast majority of the buses studied are the source of a high number of large particles of exhaust gas. Only in three cases, the proportion of particles from the potentially hazardous size class PM_{10} exceeded 27%. This is due to the large displacement of bus engines (from 4 to 16 L) and the type of fuel – all buses have diesel engines. In a previous publication of ours, we showed that diesel-fueled engines are the source of larger EGS particles [25].

The majority of experimental vehicles (15/20) emitting particles of the 2nd size class, constitute more than 80% of the total number of emitted particles. This demonstrates that buses running on diesel fuel are the source of emission of predominantly large particles (Fig. 1).

However, three buses (DBS 106 1993, DBH115, and HSAC 1994) emitted particles less than 10 μm (particles that adversely affect human health) at high concentrations of 27%, 100%, and 32.6%, respectively. Micro-sized particles, PM_{10} fraction, are potentially more hazardous to human health [26,27], and some of the buses studied were the source of only this fraction of EGS (Fig. 2).

Investigation of the dry residue of the EGS allowed us to define that for the most part, large particles according to RAMAN spectra are carbon agglomerates of irregular form. The most typical forms of particles in the exhaust gas of buses are shown in Fig. 3.

As shown in Fig. 3a–c, particulates having a mean diameter ≥ 5 μm represent an aggregate of several smaller particles from Fig. 3d, which agglomerate into larger particles.

Regarding the chemical structure, the soot particles of the exhaust of buses are represented by amorphous carbon (Fig. 4), which causes less stress on the human body than the particles of structured carbon. However, in 5% of all cases, structured carbon particles were found (Fig. 3), which have negative effects on human health due to higher lipid peroxidation at a lower ratio of reduced and oxidized glutathione [9].

Fig. 4 shows the spectrum of a particle, which was compared with the spectrum of carbon nanotubes. The graph shows that the particle spectrum has no congruent peaks and the particle structure is represented by amorphous carbon.

Table 3
Content of metals of the EGS of buses.

Sample No.	Al, mg/dm^3	Cd, mg/dm^3	Co, mg/dm^3	Cr, mg/dm^3	Cu, mg/dm^3	Fe, mg/dm^3	Mg, mg/dm^3	Mn, mg/dm^3	Ni, mg/dm^3	Pb, mg/dm^3	Sn, mg/dm^3	Zn, mg/dm^3
1	0.033	< 0.0001	< 0.0001	< 0.0001	0.0031	0.0062	0.0041	< 0.0001	< 0.0001	< 0.0001	< 0.0001	0.676
2	0.0329	< 0.0001	< 0.0001	0.0018	0.0029	0.0103	0.0499	< 0.0001	0.0008	< 0.0001	< 0.0001	0.137
3	0.0922	0.0022	0.0092	0.0157	0.0238	0.0129	0.0192	0.0004	0.0106	0.0002	< 0.0001	4.62
4	0.0776	0.0027	0.0075	0.0131	0.0212	0.013	0.17	0.0005	0.0105	< 0.0001	< 0.0001	2.05
5	0.0467	0.0001	0.0022	0.005	0.0114	0.0081	0.0291	< 0.0001	0.0033	< 0.0001	< 0.0001	3.29
6	0.0495	0.0017	0.0034	0.0068	0.0124	0.0084	0.0134	< 0.0001	0.0054	0.0019	< 0.0001	3.86
7	0.0491	0.001	0.0031	0.0069	0.0171	0.0167	0.0084	< 0.0001	0.0064	< 0.0001	< 0.0001	1.78
8	0.0468	0.0019	0.0033	0.007	0.0129	0.0088	0.0057	< 0.0001	0.0052	< 0.0001	< 0.0001	0.0817
9	0.0389	0.0014	0.0021	0.0048	0.0127	0.0103	0.0125	< 0.0001	0.0045	0.005	< 0.0001	8.32
10	0.0419	0.0006	0.0024	0.0053	0.0111	0.0321	0.0061	< 0.0001	0.0046	< 0.0001	< 0.0001	14.3
11	0.0413	0.0016	0.0026	0.0055	0.0116	0.013	0.0035	< 0.0001	0.0064	0.0019	< 0.0001	2.13
12	0.0416	0.0003	0.001	0.004	0.0101	0.0529	0.0116	< 0.0001	0.004	0.004	< 0.0001	12.4
13	0.0348	0.0008	0.0012	0.0036	0.0077	0.0099	0.004	< 0.0001	0.0038	0.003	< 0.0001	0.205
14	0.0284	< 0.0001	0.0001	0.0018	0.0078	0.0094	0.009	< 0.0001	0.0012	0.0024	< 0.0001	15.6
15	0.0333	< 0.0001	0.0011	0.003	0.0157	0.0153	0.0078	< 0.0001	0.0021	< 0.0001	< 0.0001	1.25
16	0.0387	0.0004	0.0011	0.0037	0.011	0.0536	0.0024	< 0.0001	0.0043	0.0015	< 0.0001	0.148
17	0.038	0.0012	0.0025	0.0052	0.0115	0.008	0.0019	< 0.0001	0.004	0.0017	< 0.0001	2.33
18	0.0385	0.0006	0.0022	0.0047	0.0109	0.0094	< 0.0001	< 0.0001	0.0036	0.0031	< 0.0001	2.36
19	0.0278	< 0.0001	0.0001	0.002	0.0096	0.0062	0.0071	< 0.0001	0.0019	< 0.0001	< 0.0001	5.17
20	0.0612	0.0004	0.0026	0.0552	0.0115	0.0155	0.38	0.0067	0.0109	< 0.0001	< 0.0001	4.67

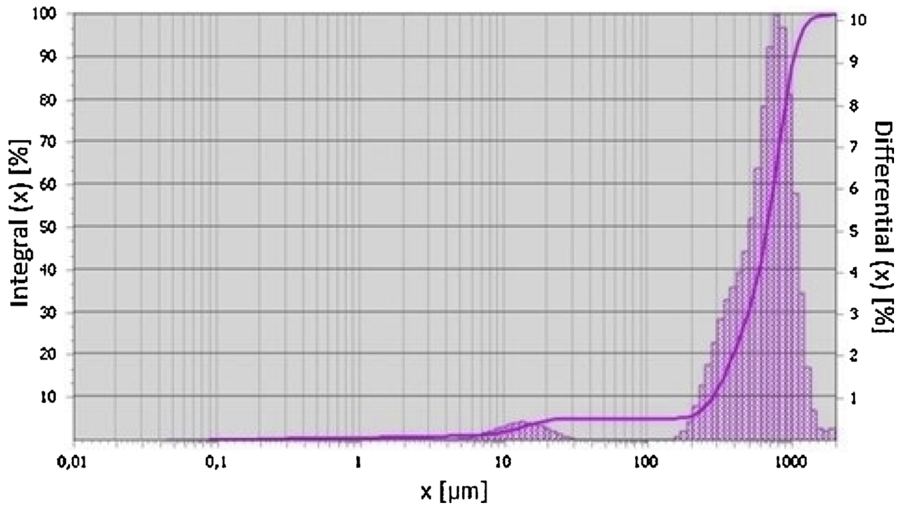

Fig. 1. Quantitative distribution of particles sizes in the exhaust gas in KGBird sample measured using laser particle size analysis.

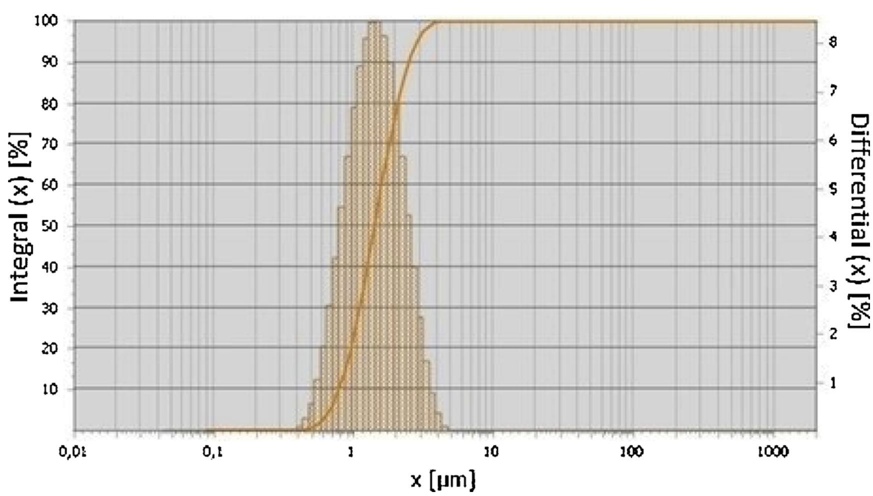

Fig. 2. Quantitative distribution of particle sizes in the exhaust gas in DBH115 sample measured using laser particle size analysis.

Fig. 5 shows the spectrum of a particle whose peaks coincide with the spectrum of carbon tubes related to functionalized carbon.

The elemental analysis of the exhaust gas has shown that the EGS particles include metal ions. In particular, the samples contained metals hazardous to organisms such as Al, Cd, Cr, Cu, Fe, Mg, Ni, Pb, and Zn (Table 3).

4. Discussion

The results of the experimental procedure from laser particle size analysis show that the particle size distribution in most studied vehicles is divided in two groups. The first group consists of particles less than 50 µm in size while the second group consists of particles within the

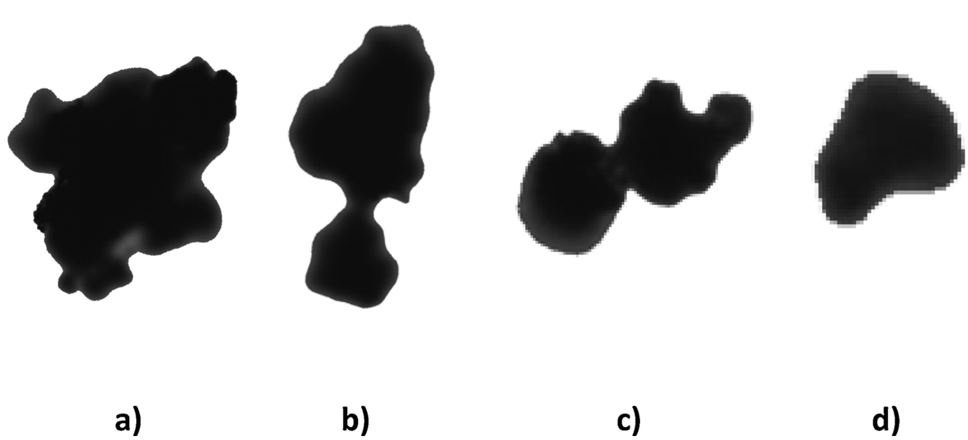

Fig. 3. Typical forms of particles in the exhaust gas of buses, magnification of 50 × . a) Mean diameter of particle (D_{mean}) – 79 µm, specific surface area $S_{mean.}$– 4912 μm^2; b) D_{mean}– 54.93 µm, S_{mean}– 2370.2 μm^2; c) D_{mean}– 11.5 µm, S_{mean}– 103.8 μm^2; d) D_{mean}–2.21 µm, S_{mean}– 3.85 μm^2.

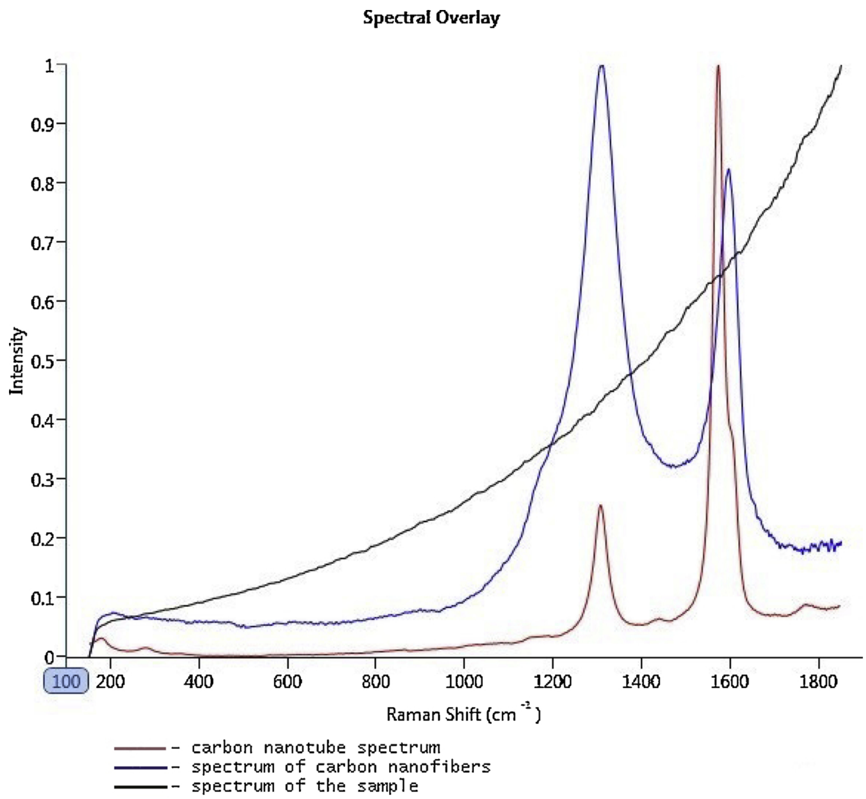

Fig. 4. RAMAN spectrum of a soot particle consisting of amorphous carbon.

range of 100–1000 μm. The vast majority of the buses studied are the source of a high number of large particles of exhaust gas. Only in three cases the proportion of particles from the potentially hazardous size class PM_{10} exceeded 27%. This is due to the large displacement of bus engines (from 4 to 16 L) and the type of fuel which is diesel in all cases. The majority of experimental vehicles (15/20) emitting particles of the

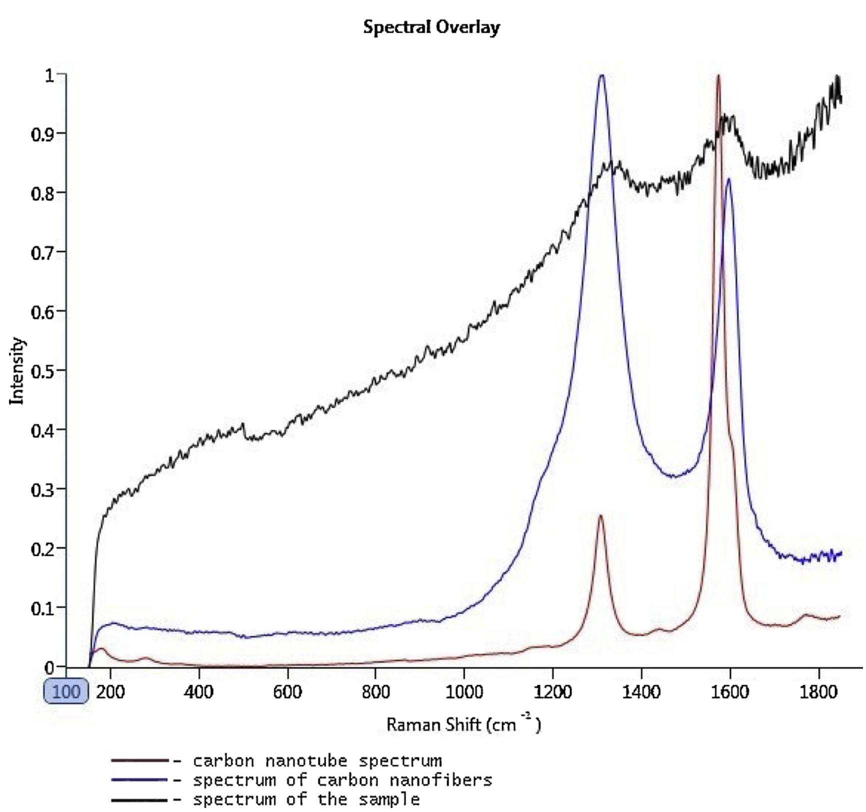

Fig. 5. RAMAN spectrum of a soot particle consisting of structured carbon.

2nd size class, constitute more than 80% of the total number of emitted particles. This demonstrates that buses running on diesel fuel are the source of emission of predominantly large particles. However, 3 out of 20 buses (DBS 106 1993, DBH115, and HSAC 1994) emit particles of potentially hazardous size class PM_{10} (that adversely affect human health) at high concentrations of 27%, 100%, and 32.6%, respectively. Micro-sized particles PM10 are one of the most hazardous particles to human health, along with PM1, PM2.5 and ultra-fine particles.

Investigation of the dry residue of the EGS allowed us to define that large particles according to RAMAN spectra are carbon agglomerates of irregular form.

The analysis of the qualitative composition of particulate matter in the exhaust gases of buses has shown that the particulates entering the atmosphere are predominantly carbon (ash and soot) agglomerates having different geometric shapes, arithmetic mean size and specific surface area.

Concerning the chemical structure, the soot particles of the exhaust of buses are represented by amorphous carbon, which causes less stress on the human body than the particles of structured carbon. However, in 5% of all cases, structured carbon particles were found, which have a negative effect on human health.

The concentrations of some elements (for example Zn) were two or three orders of magnitude higher than the concentrations of other elements. Zinc is an important element that is used for enzymatic purposes. Nevertheless, exceeding its level will interfere with the metabolism of iron and copper in the body. The larger doses of zinc mean the respectively lower levels of manganese, which increase the susceptibility of autoimmune reactions. Further symptoms may include anemia, growth inhibition, and an increase in low-density lipoprotein (LDL), leading to increased blood cholesterol levels [28].

This confirms that the issue of quality control of public transport should be taken very seriously [29], since the absence of control over the bus park can lead to a significant deterioration of the environment.

5. Conclusion

In this research study, particle size distribution and elemental analysis of particulate matter originated from the EGS of 20 bus models were examined by laser particle size analysis and atomic emission spectroscopy confirming the issue of quality control of public transport should be taken very seriously [29], since the lack of control over the bus park can lead to a significant deterioration of the environment.

According to the results of this study, the need for further research concerning the toxic effects of EGS particulate matter originated from exhaust emissions of the most popular models of buses in the territory of Vladivostok has proven to be imperative.

These results will help improve our understanding of risks that the emitted particles pose to human health and environment and may strengthen the need for the adoption of new standards for the efficient control of these hazardous particles.

Competing interests

AT is the Editor-in-Chief for the journal, but had no personal involvement in the reviewing process, or any influence in terms of adjudicating on the final decision, for this article.

Acknowledgements

The work was funded by the Russian Science Foundation (No. 15-14-20032).

The authors would like to thank the staff team of FEFU CCU (Center of Collective Use) ‘Interdepartmental center for analytical control of the environment’ and the Laboratory of Toxicology, University of Crete, Greece, for their dedicated involvement in this study.

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