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Case Studies of Aerosol Pollution in Different Public Transport Vehicles in Hungarian Cities

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Abstract: In this case study, aerosol pollution and passenger exposure were investigated while travelling on different public transport vehicles in Hungary. Two sampling campaigns were carried out: one in autumn 2012 and the other in spring 2014. Concentration, elemental composition and the size distribution of aerosol samples were determined in order to characterize the atmospheric particulate matter (APM) pollution inside the vehicles. The concentration of the PM_{coarse} fraction inside the different vehicles varied between 29 and 354 μg m^{-3}, while the $PM_{2.5}$ concentrations were found to be between 12 and 192 μg m^{-3}. This was significantly (2–19 times) higher than the outdoor concentration values. The main sources of the increased exposure were the resuspended mineral and road dust, including salt and fertilizers, and the direct exhaust of the vehicles. Rail abrasion and disinfectant and cleaning materials also contributed considerably to the aerosol pollution inside the vehicles. Moreover, organic fibrous particles were found in great number on the samples by single particle analysis using scanning electron microscopy (SEM).

Keywords: aerosol pollution; public transport vehicles; in-vehicle PM concentrations; composition and sources

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1. Introduction

Atmospheric particulate matter (APM) is one of the most serious pollutants in Europe in terms of adverse effects on human health [1]. According to a United Nations report, more than half of the world's population (55%) lives in urban areas [2], where currently the particulate matter (PM) pollution originates mainly from industrial emissions, local traffic and biomass burning [3,4], and it is the most appreciable environmental risk to human health [5]. The main causes of premature death from air pollution are heart disease, stroke and various lung diseases and lung cancer [3]. In 2013, the International Agency for Research on Cancer (IARC) classified air pollutants and found that PM, the main component of air pollutant mixtures, is carcinogenic [6].

In urbanized areas, transport plays a major role in people's lives. Public transport systems carry millions of passengers per day in numerous cities around the world. In Hungary, transportation is part of the daily routine, and it contributes significantly to aerosol pollution in the environment. Normally, concentrations of pollutants are significantly higher in traffic areas than in urban background sites [7]. In accordance with the Hungarian Central Statistical Office, 80% of the population travels on a daily basis, 20% of the people use public transportation and the average travelling time is approximately 1.5 h each day [8]. Most outdoor pollutants get into indoor spaces and can react with indoor pollutants. Therefore, harmful pollutants may be present in higher concentrations than outdoors [9].

Previous studies, when personal exposure from the perspective of drivers and passengers on different public transport modes were investigated, showed that people using or working in public transportation were exposed to elevated aerosol pollution levels [10–12]. In Hong Kong, a total of eight public transportation modes were investigated including bus, tram, and different railways. They found higher PM_{10} concentrations in non-air-conditioned vehicles than in air-conditioned vehicles [13]. The highest values were observed inside trams. Different types of public transport vehicles with different ages were compared in Helsinki, and the conclusion was that the newer technology eventuate less exposition for the commuters [14]. Numerous studies have been published so far about air quality in underground systems in Europe, Asia, America and Egypt, and PM was identified as the main pollutant in the metro [15]. Salma et al. showed that in underground railway stations in Budapest, Hungary, people are exposed to several times higher concentrations of heavy metals than when outdoors [16,17]. In our previous study on air pollution in trams in Debrecen, Hungary, we also found elevated levels of APM inside the vehicles; however, modern ventilation technology resulted in lower exposure [18]. In some studies [19–21], the impacts of air conditioning and air circulation were investigated through the indoor/outdoor (I/O) ratios and the ultrafine particulate matter (UFP) concentrations in cars. They found that closed air-circulation systems resulted in less exposure.

While metros and underground environments have been extensively studied, data on APM pollution in other types of public transportation are relatively rare, especially on the elemental and/or chemical composition of the in-vehicle pollution [22,23]. In addition, there is almost no information available from the middle-eastern European region.

Hungary lies in the Carpathian Basin in Central Europe, with 9.3 million inhabitants. With a population of 200,000, Debrecen is the second largest city in Hungary, and the transport network includes 2 tram-lines, 60 local buses and 3 trolleybus routes. In our research, we investigated the in-vehicle concentration and elemental composition of PM_{coarse} and PM_{fine} in different public transport vehicles under usual operating conditions. The local buses involved in this study have been running since 2009 and the trolleybuses since 2005. Both vehicles are equipped with air conditioning and heating systems with preprogrammed settings, and the buses operate with Euro 4 and Euro 5 diesel engines [24]. At the time of sampling, the air-conditioning system was not used in the buses and trolleybus; they drove with all windows open, which is the usual driving condition in good weather. Therefore, in the following, these buses were qualified as non-air-conditioned vehicles (i.e., non-A/C local bus and non-A/C trolleybus). Two types of tramcars are used in the public transportation of the city: KCSV (old type) and CAF (new type). The older ones have been in service since 1997, while the new ones since 2014 when the new tram-line was put into operation. The CAF trams have modern air-conditioning–ventilating and heating systems. The windows cannot be opened by the occupants, and the ventilation is provided by the tram's own system, which is preprogrammed depending on the weather and seasons. In contrast, the older models have central ventilation and a heating unit on top of the tram that is set to ventilate the vehicle 24 times per hour during the heating season and 37 times per hour during the summer season. Cooling is provided through the windows because this old model does not have an air-conditioning system [24]. The InterCity carriages were built between 1970 and 1971 and refurbished between 1994 and 2000. The railway carriages are equipped with air correction devices and air-conditioning systems. Air exchange is provided through the ventilation system, and the windows cannot be opened [25]. With 2 million inhabitants, Budapest is the capital city of Hungary. The transport system has several bus, trolleybus, tram and metro lines; however, in this paper, only the underground was engaged. Nowadays, the capital city has 4 metro lines: Metro 1 (M1), Metro 2 (M2), Metro 3 (M3) and the newest, Metro 4 (M4). The metro carriages involved in this study ran on the Metro 3 (M3) line, which is the longest line at 16.3 km, and they were in service between 1976 and 2016. The metro carriages were not equipped with an air-conditioning system, and the open windows ensured cooling and air exchange in the metro cabins. In addition, the tunnels had their own ventilation system [26].

In the present work, PM concentration, elemental composition, the size distribution and the possible sources of indoor APM pollution were determined in different public transport vehicles under normal travelling conditions in Debrecen, Hungary, and on intercity trains travelling between Debrecen and Budapest. As supplementary information, we collected aerosol samples while travelling on an underground train in Budapest in order to compare the characteristics of the APM pollution in public transport vehicles operating above ground. To our best knowledge, this is the first time that elemental size distribution has been reported from public transport vehicles.

2. Materials and Methods

2.1. Aerosol Sampling

Aerosol samplings were conducted while travelling in different vehicles: local buses, trolleybuses, old and new types of trams in Debrecen, an InterCity (IC) train between Debrecen and Budapest, and the M3 Budapest Metro. Figure 1 shows the investigated transport lines and the monitoring stations, while Table 1 includes the exact vehicle types, the sampling dates, the applied sampling devices, the line lengths and the measured size fractions. In-vehicle aerosol particles were collected with personal samplers equipped with Nuclepore two-stage sample head (NP two–stage sampler). The samplers were loaded with 2 Nuclepore polycarbonate filters: one with 8 µm and another with 0.4 µm pore sizes. With this method, the particles could be separated into two size fractions: the fine fraction ($PM_{2.5}$—particles with an aerodynamic diameter smaller than 2.5 µm) and the coarse fraction (PM_{coarse}—particles with an aerodynamic diameter larger than 2.5 µm). Buck Elite-5 personal pumps [27] were used to pump air through the system at an approximately 3 L min^{-1} flow rate. In 2014, besides the two-stage samplers, a Sioutas four-stage personal cascade impactor [28] was applied with an SKC Leland Legacy pump [29] at an 8.3 L min^{-1} flow rate. This allowed the separation of the aerosol within the size range of 0.25–10.0 µm into 4 fractions: 0.25–0.50, 0.50–1.00, 1.00–2.50 and 2.50–10.0 µm. Aerosol particles of different size fractions were gathered on metal-free paraffin coated thin Kapton foils 25 mm in diameter. The duration of the aerosol samplings was approximately four hours inside the different vehicles. Generally, the collection took place in the afternoons (1 p.m.–5 p.m.). In order to obtain enough samples quantity for the chemical analysis, 3–4 h long samplings were required. On the days of samplings, the weather was dry and warm. In 2012, the temperature was approximately 25–30 °C, while in 2014 it was 20–25 °C.

The results were compared to samples collected at the urban background (UB) station of the ATOMKI. In this UB location, PM_{10} and $PM_{2.5}$ samples have been collected two times a week since 1988 using a Gent-type sampler [30] equipped with 47 mm diameter Nuclepore polycarbonate filters with 8 and 0.4 µm pore sizes. Data obtained at this UB station were used as reference outdoor data in this study. Unfortunately, there was no sampling in autumn 2012 because of the renovation and isolation of the sampling sites.

2.2. Mass and Elemental Analysis

The total mass concentration of the PM samples was determined by gravimetric technique using a 6-digit microbalance (RADWAG MYA/5/2Y/F). The polycarbonate filters were conditioned at least 24 h before weighing in the weighing box at 25 °C and 55% relative humidity. An ionizer was applied during weighing in order to eliminate the static charge on the filters. The concentration, defined in µg/m^3, was computed from the volume of air and the mass load. In the case of Kapton filters, the uncertainty of the weighing was too high due to the very small PM mass and charging effects; therefore, no mass data were available for the impactor samples. Furthermore, there was no PM_{coarse} data available for the local bus in 2014.

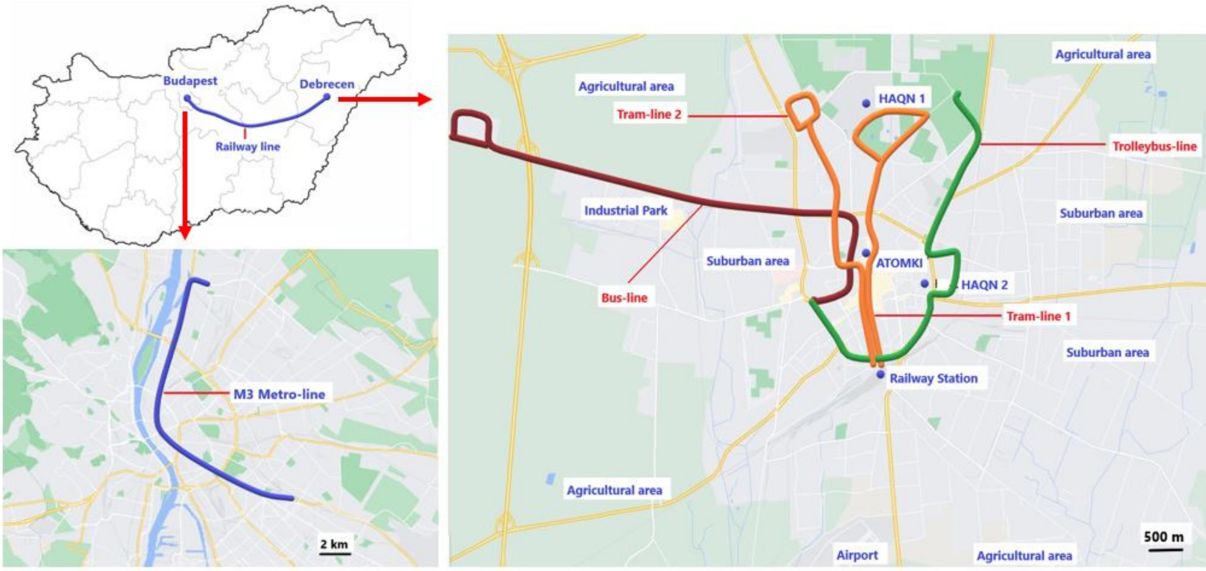


Figure 1. Map of the investigated transport lines: the location of the ATOMKI UB site and the Hungarian Air Quality Monitoring Network (HAQN) sites in Debrecen; the railway line between Debrecen and Budapest; the M3 metro line in Budapest.

Table 1. Vehicle types, sampling dates, the lengths of the lines (in km), the applied sampling devices and the measured size fractions.

Mode of Transport	Vehicle Type	Date	NP Two-Stage Sampler	PM_{coarse}; $PM_{2.5}$	Sioutas Four-Stage Impactor	Four Size Fractions *	Date	NP Two-Stage Sampler	PM_{coarse}; $PM_{2.5}$	Sioutas Four-Stage Impactor	Four Size Fractions
Local bus	VOLVO B9L—ALFA CIVIS 12–18	27 September 2012	X **	X	-	-	12 March 2014	X	X	X	X
Trolleybus	GANZ SOLARIS TROLLINO 12	25 September 2012	X	X	-	-	31 March 2014	X	X	X	X
IC train	BP 20–67	1 October 2012	X	X	-	-	20 March 2014	X	X	X	X
Old tram (Tram-line 1)	KCSV—6 1S	24 September 2012	X	X	-	-	2 April 2014	X	X	X	X
New tram (Tram-line 2)	CAF Urbos 3	-	-		-	-	13 March 2014	X	X	X	X
Metro	Ev3	-	-		-	-	20 March 2014	X	X	X	X

* Four size fractions: 0.25–0.50, 0.50–1.00, 1.00–2.50 and 2.50–10.0 µm. ** X means the sampler was applied in the campaign.

In order to define the elemental composition of the aerosol samples (13 ≥ Z), the particle-induced X-ray emission (PIXE) analytical method was used. The bulk measurements were implemented at the PIXE chamber installed on the left 45° beamline on the 5 MV Van de Graaff accelerator of the Institute for Nuclear Research (ATOMKI) [31]. A H^+ beam of 2 MeV energy and of 30–40 nA current was used for the irradiation. The accumulated charge on each sample was 40 µC, and the acquisition time was approximately 15–20 min. The beam spot had a diameter of 5 mm in the case of the filter samples. For the impactor samples, a 2 × 8 mm beam spot was applied, which covered the aerosol deposit created by the Sioutas impactor. The obtained X-ray spectra were evaluated with the PIXEKLM program package [32,33]. Concentrations of the following elements were assigned after blank correction: Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Cu, Zn, Br, Ba and Pb. The concentration values were given in ng m^{-3}. Depending on the element, the error of the determination of the elemental concentration varied between 5% and 20%, and the detection limit was between 2 and 0.1 ng m^{-3}. The minimum detection limit and the analytical error for all of the analyzed quantities are given in Table S1. Furthermore, PIXE spectra of selected samples are also provided in Supplementary Materials Figure S1.

Additional morphological and elemental analysis was conducted by electron microscopy using a Jeol JSM-IT500HR scanning electron microscope at the Laboratory for Heritage Science, ATOMKI. Backscattered (BS) electron images as well as secondary (SE) electron images and EDX spectra were collected on the filters at a 10 kV acceleration voltage and at a 50 Pa low-vacuum mode. All of the applied analytical techniques were non-destructive; therefore, the same samples were used for all of the analyses.

2.3. Data Analysis

2.3.1. Enrichment Factor (EF)

Enrichment Factors (EFs) were calculated in order to obtain information about the origin of the detected elements, using Ti as the reference element:

$$\mathrm{EF} = \frac{(^{\mathrm{X}}/_{\mathrm{Ti}})\mathrm{PM}}{(^{\mathrm{X}}/_{\mathrm{Ti}})\mathrm{Crustal}} \tag{1}$$

where X is the concentration of the element in question, and Ti is the concentration of the reference element. EF values of approximately 1 (less than 10) indicate a natural origin of the given element, while if an EF has a value of more than 10, it points to an anthropogenic origin. Enrichment factor calculations were based on the Mason's average crustal rock composition [34].

2.3.2. Indoor Enrichment Factor (Indoor EF)

In order to determine a difference between outdoor and "*indoor*" sources, *indoor*/*outdoor* EF ratios (in the following: indoor EF) were computed:

$$\text{indoor EF} = \frac{\left(\frac{(^{\mathrm{X}}/_{\mathrm{Ti}})\mathrm{PM}}{(^{\mathrm{X}}/_{\mathrm{Ti}})\mathrm{Crustal}}\right) indoor}{\left(\frac{(^{\mathrm{X}}/_{\mathrm{Ti}})\mathrm{PM}}{(^{\mathrm{X}}/_{\mathrm{Ti}})\mathrm{Crustal}}\right) outdoor} \tag{2}$$

By convention, an indoor EF ≤ 1 normally means an outdoor origin of the element. An *indoor* EF > 1 and significantly higher values suggest an *indoor* origin.

3. Results

3.1. Mass Concentration

Table 2 presents the PM concentrations measured inside the vehicles; $PM_{2.5}$, PM_{10} and PM_{coarse} were obtained at the ATOMKI UB station (from 24 h samplings) on the sampling days and PM_{10} concentrations were measured at an UB and at a traffic measurement site of the Hungarian Air Quality Network (HAQN) in Debrecen at the time of the samplings [35]. The automatic stations of the HAQN provided only PM_{10} mass data. The outdoor PM data show that the samplings were made in similar conditions: the outdoor pollution level was, on average, level in the city, and the meteorological circumstances were also similar for all days.

The PM_{coarse} concentration on the different vehicles varied between 29 and 354 µg m^{-3}, while the $PM_{2.5}$ concentration was found to be between 12 and 189 µg m^{-3}. The lowest concentrations were measured in the case of the new tram, while very high pollution levels were observed on the old tram, the bus, the IC train and the metro. The concentrations inside the vehicles were compared to the PM_{10} levels measured at the same time at two HAQN sites and the location of the ATOMKI. The results showed that in all cases, the PM pollution levels were significantly higher than for outdoors. The PM_{coarse} level was very high in both years in all vehicles, except the new tram, while in general, much higher $PM_{2.5}$ levels were measured in 2014 than in 2012. One exception is the local bus, where high

$PM_{2.5}$ concentrations were experienced in 2012. This bus line leads from the city center to a suburban area a few kilometers outside the city border using the main road No. 33. At the time of the sampling, due to the good weather, the bus was driving with all the windows down, and agricultural works were being conducted in nearby fields. Thus, the resuspended dust, the emission from the diesel engine and the field works together could cause the elevated PM pollution levels inside the bus. The differences between the $PM_{2.5}$ levels in the two campaigns can be explained by the different travelling conditions (in the warm weather windows are usually kept down) or the older age of the ventilating systems. We had no access to information about the date of maintenance, either. High concentrations were measured on the old tram in 2014. Roadworks next to the tramline were the most probable cause of the elevated pollution level, as we have shown in our study from 2017 to 2018 [18]. Significantly lower PM_{coarse} and $PM_{2.5}$ concentrations were observed in the new tram than in the old type in 2014, which is in accordance with our findings from 2017 to 2018 [18]. It is important to note that the two types of trams ran partly on different lines during the sampling period: the old ones ran on tram-line 1, while the new types on tram-line 2.

Table 2. The $PM_{fine+coarse}$, $PM_{2.5}$, and PM_{coarse} concentrations (in µg m^{-3}) in different vehicles and PM_{10} concentrations measured at two automatic measurement stations of the HAQN in Debrecen: HAQN 1 (urban background) and HAQN 2 (urban traffic).

Date	Location	PM_{coarse}	$PM_{2.5}$	$PM_{fine+coarse}$	HAQN1 PM_{10}	HAQN2 PM_{10}	ATOMKI $PM_{2.5–10}$	ATOMKI $PM_{2.5}$	ATOMKI PM_{10}
				(µg m^{-3})					
2012	Local bus	224	168	392	49	46	-	-	-
	Trolleybus	159	30	189	28	24	-	-	-
	IC train	354	19	373	-	-	-	-	-
	Old tram	180	12	192	34	31	-	-	-
2014	Local bus	no data	52	-	25	33	6	14	20
	Trolleybus	160	104	264	22	38	14	20	34
	IC train	182	66	248	-	-	-	-	-
	Old tram	248	145	393	24	30	13	23	36
	New tram	29	17	46	28	31	19	17	36
	Metro	288	189	477	-	-	-	-	-

In Table 3, the obtained PM concentrations are presented together with results from similar studies from Hong Kong [13], Munich [36], Barcelona [22], Lisbon [23] and our findings for the trams in Debrecen, 2017–2018 [18]. Basically, the measured concentrations were on the same order of magnitude as in other cities for similar vehicles; however, in most cases, we measured similar or higher concentrations. We note that in most of the other studies, the PM concentrations were obtained by real-time measurement using particle counters. The PM concentrations measured in the metro were in accordance with other results, e.g., Rome (407 µg m^{-3} on average) [37], London (800 µg m^{-3} on average) [38] or Stockholm (470 µg m^{-3} on average) [39]. In the case of the IC train, the observed PM_{10} concentrations were significantly higher than the findings in other studies [40–42], while the $PM_{2.5}$ concentrations were in good agreement with data from other countries [40,43,44]. More detailed information for comparison concerning the metro and trains can be found in Table S2 in the Supplementary Materials [17,18,37–46].

Table 3. Comparison of the PM_{10} and $PM_{2.5}$ concentrations with other studies.

	Vehicle Type	**PM_{10} ($\mu g\ m^{-3}$) ***	**$PM_{2.5}$ ($\mu g\ m^{-3}$)**
Present study *	Non-A/C Local bus (2012)	392	168
	Non-A/C Local bus (2014)	no data	52
	Non-A/C Trolleybus (2012)	189	30
	Non-A/C Trolleybus (2014)	264	104
	Old tram (2012)	192	12
	Old Tram (2014)	393	145
	New tram (2014)	46	17
	IC-train (2012)	373	19
	IC-train (2014)	248	66
	Metro (2014)	477	189
Hong Kong	Tram	110–240	68–163
	Non-A/C bus	80–161	78–109
	Railway	41–89	29–68
Munich	Bus	110–165	-
	Tram	av. 161 **	-
Barcelona	Metro	-	av. 37; 42
	Bus	-	av. 48; 49; 39
	Tram + walking	-	av. 27; 29; 35
Lisbon	Bus	-	av. 28
	Metro	av. 84	av. 38
Debrecen *	Old tram—heating s. (2017–18) ***	70–176	31–54
	Old tram—non-heating s. (2018) ***	69–152	29–50
	New tram—heating s. (2017–18)	36–153	17–49
	New tram—non-heating s. (2018)	46–71	22–49

* PM_{10} means in our results the amount of PM_{coarse} + PM_{fine} fractions. ** av. means the average concentration. *** Heating s. means heating season, non-heating s. means non-heating season.

3.2. Elemental Composition

The elemental concentrations of the measured 16 elements in different vehicles and size fractions, indoor/outdoor (I/O) ratios and EF values in 2012 and in 2014 are shown in Tables 4–6. In 2012, the observed elements exhibited high concentrations. EFs of Al, Si, P, K, Ca, V, Mn and Ba in all vehicles were below 10, which suggests soil resuspension as their source, whereas S, Cl, Cr, Fe, Cu, Zn and Pb had high EFs, indicating an anthropogenic origin. The enrichment factors of Mn and Fe were much higher in the case vehicles running on rails (i.e., tram and IC train) pointing towards an additional source of these elements.

Since the resuspension of soil was recognized as one of the main sources of PM pollution inside the vehicles, the contribution of mineral dust components was calculated using the formula: 1.9[Al] + 2.14[Si] + 1.4[Ca] + 1.3[K] +1.67[Ti] + 1.44[Fe] [47] (our assumption is that the elements occur in their common oxide form). For vehicles running on rail, the enrichment factors of Fe were higher than 1. Therefore, the following formula was used to estimate the amount of Fe from the Earth's crust [16]:

$$\text{PM Fe crustal} = (\text{Fe}/\text{Ti})\text{crustal} \times \text{PM (Ti) aerosol}, \tag{3}$$

where PM $(\text{Ti})_{aerosol}$ is the concentration of Ti in the aerosol sample, and $(\text{Fe}/\text{Ti})_{crustal}$ represents the concentration of X element in Mason's average crustal rock composition [34]. The calculations for mineral dust were based on the crustal Fe concentration.

Table 4. Elemental concentration, mineral dust concentration and enrichment factor (EF) in PM_{coarse} and $PM_{2.5}$ fractions in different vehicles in 2012.

	Local Bus				**Trolleybus**				**Old Tram**				**IC Train**			
Element	**PM_{coarse}**		**$PM_{2.5}$**		**PM_{coarse}**		**$PM_{2.5}$**		**PM_{coarse}**		**$PM_{2.5}$**		**PM_{coarse}**		**$PM_{2.5}$**	
	(ng m^{-3})	**EF**	**(ng m^{-3})**	**EF**	**(ng m^{-3})**	**EF**	**(ng m^{-3})**	**EF**	**(ng m^{-3})**	**EF**	**(ng m^{-3})**	**EF**	**(ng m^{-3})**	**EF**	**(ng m^{-3})**	**EF**
Al	5035	1	1390	2	3615	1	1130	3	2725	1	1350	5	2085	1	1790	10
Si	21,160	1	1570	1	12,320	1	680	1	16,650	1	1730	2	2310	0.4	755	1
P	140	1	<DL *	-	140	2	<DL	-	65	1	<DL	-	<DL	-	<DL	-
S	765	25	1020	540	750	45	1550	1300	765	55	1970	2045	885	165	945	1520
Cl	985	65	30	35	9295	1075	45	70	1225	178	50	105	915	340	120	395
K	3225	1	425	3	1640	1	245	3	1890	2	215	3	800	2	205	5
Ca	8815	2	600	2	6660	2	370	0.3	5150	2	305	2	2560	5	495	5
Ti	650	-	40	-	380	-	25	-	300	-	20	-	120	-	15	-
V	30	1	<DL	-	30	1	<DL	1	20	1	<DL	-	<DL	-	15	15
Cr	100	10	<DL	-	<DL	-	<DL	10	40	10	<DL	-	15	10	<DL	-
Mn	175	2	25	3	95	2	15	3	160	5	30	10	60	3	50	20
Fe	8435	1	1070	3	4960	1	600	2	13,860	5	2170	10	7250	5	5690	40
Nmd. Fe **	1050	-	615	-	640	-	315	-	10,450	-	1945	-	5885	-	5520	-
Cu	255	40	45	110	80	20	40	170	60	20	20	85	30	25	25	185
Zn	195	25	35	70	270	60	20	65	175	45	20	85	185	130	75	445
Ba	295	5	40	10	260	10	10	5	200	10	<DL	-	215	25	255	255
Pb	50	35	<DL	-	25	30	20	320	<DL	-	<DL	-	<DL	-	35	1155
PM ***	224	-	168	-	159	-	30	-	180	-	12	-	354	-	19	-
Mineral dust	83,250	-	8120	-	51,610	-	5345	-	55,980	-	7330	-	15,710	-	6235	-

* <DL: the elemental concentration was lower than the detection limit. ** Non-mineral dust Fe component. *** PM expressed in μg m^{-3}.

Table 5. Elemental concentration, mineral dust concentration and enrichment factor (EF) in PM_{coarse} fraction in different vehicles in 2014; outdoor concentration (ATOMKI monitoring site) with EF and indoor/outdoor (I/O) ratio.

Element	Local Bus	EF	Outdoor	EF	I/O	Trolleybus	EF	Outdoor	EF	I/O	Old Tram	EF	Outdoor	EF	I/O	New Tram	EF	Outdoor	EF	I/O	IC Train	EF	Metro	EF
	(ng m^{-3})		(ng m^{-3})			(ng m^{-3})		(ng m^{-3})			(ng m^{-3})		(ng m^{-3})			(ng m^{-3})		(ng m^{-3})			(ng m^{-3})		(ng m^{-3})	
Al	18,100	0.4	300	2	60	2785	0.3	520	1	5	2470	0.3	510	1	5	300	0.2	485	1	1	100	0.1	<DL	-
Si	88,200	1	395	1	223	15,690	1	1200	1	13	31,690	1	985	1	32	6665	1	1350	1	5	1380	0.4	1830	0.3
P	265	0.4	10	3	27	50	0.4	10	1	5	<DL	-	5	1	-	<DL	-	10	1	-	40	3	675	30
S	2130	14	60	96	36	715	24	320	176	2	610	25	215	117	3	125	28	365	150	0.3	280	95	<DL	-
Cl	4670	60	15	50	311	850	58	15	17	56	695	58	20	20	35	205	92	15	13	14	160	110	570	225
K	10,200	1	90	1	113	1945	1	240	1	8	1850	1	165	1	11	395	1	380	2	1	290	1	460	1
Ca	30,400	1	180	2	169	7520	2	600	2	13	6795	2	430	2	16	2255	4	580	2	4	1010	2	2385	3
Ti	2620	-	10	-	258	495	-	30	-	17	405	-	30	-	14	75	-	40	-	2	50	-	85	-
V	65	1	<DL *	-	-	<DL	-	<DL	-	-	<DL	-	<DL	-	-	<DL	-	<DL	-	-	<DL	-	<DL	-
Cr	115	2	<DL	-	-	<DL	-	<DL	-	-	65	7	<DL	-	-	<DL	-	<DL	-	-	<DL	-	120	60
Mn	685	1	5	3	137	100	1	10	2	10	250	3	10	1	25	40	2	15	2	3	70	5	2020	110
Fe	31,600	1	125	1	253	5740	1	530	2	11	19,220	4	415	1	46	2085	2	600	1	3	9230	15	207,000	215
Nmd. Fe **	1825	-	-	-	-	115	-	-	-	-	14,620	-	-	-	-	1235	-	-	-	-	8660	-	206,035	-
Cu	180	5	2	13	106	120	19	5	15	24	35	7	5	10	7	<DL	-	10	13	-	15	25	655	615
Zn	380	9	2	11	223	165	21	5	12	33	85	13	5	8	17	20	17	10	10	2	85	105	45	35
Br	35	24	<DL	-	-	<DL	-	<DL	-	-	<DL	-	<DL	-	-	<DL	-	<DL	-	-	<DL	-	85	1760
Ba	960	3	5	4	192	220	4	15	5	15	140	3	10	2	14	35	4	20	4	2	230	40	810	85
Pb	160	21	1	41	130	20	14	10	84	2	65	54	5	34	13	30	135	10	97	3	<DL	-	360	1435
PM ***	no data	-	6	-	-	160	-	14	-	11	248	-	13	-	19	29	-	19	-	1.5	182	-	288	-
Mineral dust	327,145	-	1985	-	-	61,035	-	5540	-	-	91,950	-	4550	-	-	19,900	-	6075	-	-	5865	-	9430	-

* <DL: the elemental concentration was lower than the detection limit. ** Non-mineral dust Fe component. *** PM expressed in µg m^{-3}.

Table 6. Elemental concentration, mineral dust concentration and Enrichment Factor (EF) in $PM_{2.5}$ fraction in different vehicles in 2014; outdoor concentration (ATOMKI monitoring site) with EF and indoor/outdoor (I/O) ratio.

Element	Local Bus	EF	Outdoor	EF	I/O	Trolleybus	EF	Outdoor	EF	I/O	Old Tram	EF	Outdoor	EF	I/O	New Tram	EF	Outdoor	EF	I/O	IC Train	EF	Metro	EF
	(ng m^{-3})		(ng m^{-3})			(ng m^{-3})		(ng m^{-3})			(ng m^{-3})		(ng m^{-3})			(ng m^{-3})		(ng m^{-3})			(ng m^{-3})		(ng m^{-3})	
Al	1370	0.5	330	2	4	305	1	450	2	0.7	615	1	145	1	4	145	1	315	2	0.5	455	1	<DL	-
Si	5380	1	180	0.4	30	1030	1	555	1	2	4140	1	265	0.3	16	1160	1	425	1	3	505	0.4	505	0.4
P	<DL *	-	<DL	-	-	<DL	-	<DL	-	-	<DL	-	<DL	-	-	<DL	-	<DL	-	-	30	6	<DL	-
S	580	65	345	765	2	1240	700	1185	1437	1.04	1500	565	1525	1821	0.9	290	325	445	720	0.65	580	490	410	345
Cl	260	55	10	40	26	35	40	5	12	7	65	50	1	2	74	20	45	10	35	2	30	50	230	390
K	840	1	385	10	2	355	2	445	5	0.8	340	1	395	5	0.9	220	2	475	8	0.5	115	1	95	1
Ca	2180	2	80	1	27	495	2	225	2	2	685	2	150	1	5	740	6	165	2	4	525	3	390	2
Ti	155	1	10	-	16	30	1	15	-	2	45	1	15	-	3	15	1	10	-	1.5	20	1	20	1
V	<DL	-	<DL	-	-	<DL	-	<DL	-	-	<DL	-	<DL	-	-	<DL	-	<DL	-	-	15	25	<DL	-
Cr	<DL	-	<DL	-	-	<DL	-	1	4	-	<DL	-	<DL	-	-	<DL	-	<DL	-	-	<DL	-	85	185
Mn	60	1	5	3	11	15	2	5	2	3	45	5	5	2	9	15	5	5	2	3	55	15	1050	245
Fe	3030	2	95	1	32	690	2	335	2	2	3580	7	260	2	14	545	3	300	3	2	6860	30	10,4700	460
Nmd. Fe **	1270	-	-	-	-	350	-	-	-	-	3070	-	-	-	-	375	-	-	-	-	6635	-	104,475	-
Cu	90	47	2	25	16	35	93	5	35	7	70	124	5	25	15	<DL	-	5	35	-	20	80	210	840
Zn	70	28	25	210	3	35	75	30	136	1.15	45	65	35	150	1.3	15	65	20	130	0.7	80	250	<DL	-
Br	<DL	-	2	555	-	<DL	-	3	355	-	<DL	-	5	530	-	<DL	-	2	360	-	<DL	-	<DL	-
Ba	90	5	5	7	18	30	10	15	11	2	<DL	-	15	10	-	<DL	-	10	8	-	295	130	505	220
Pb	<DL	-	<DL	-	-	<DL	-	<DL	-	-	<DL	-	<DL	-	-	<DL	-	<DL	-	-	<DL	-	130	2200
PM ***	52	-	14	-	4	104	-	20	-	5	145	-	23	-	6	17	-	17	-	1	66	-	189	-
Mineral dust	21090	-	1790	-	-	4990	-	3455	-	-	12,260	-	1980	-	-	4360	-	2820	-	-	3190	-	2115	-

* <DL: the elemental concentration was lower than the detection limit. ** Non-mineral dust Fe component. *** PM expressed in μg m^{-3}.

In the coarse fraction, the contribution of mineral dust varied between 30 and 50%. The exceptions were the new tram with 70% and the IC train with only 5% mineral dust contribution. In the fine fraction, the contribution of mineral dust ranged from 5% (local bus 2012, IC train) to 25% (trolleybus, new tram), with two exceptions: old tram—60%, local bus 2014—45%. For vehicles running on rails, the contribution of non-mineral dust Fe (Nmd. Fe) to the Fe concentration varied between 70 and 99% in both size fractions, indicating railway abrasion as a primary source of in-vehicle pollution.

We assumed that primarily carbonaceous particles were responsible for the unaccounted mass [48].

In 2014, the measured concentration data in the vehicles could be compared to outdoor data; hence, I/O ratios were calculated. We found that the elemental concentrations were significantly higher inside the vehicles than in the outdoor air, while the composition was similar. The highest I/O ratios were detected in the local bus, and the lowest in the new tram. Similar I/O ratios were found in the old tram and the trolley bus in both size fractions. In general, the I/O ratios were 5–10 times higher in the coarse fraction than in the fine fraction.

Nevertheless, the accumulation (I/O ratios) was higher for mineral dust elements, such as Al, Si, Ca, Ti and Ba, than for elements of anthropogenic origin (e.g., S, Zn, Pb or fine fraction K). The higher accumulation of mineral dust could be attributed to the resuspension of dust caused by traffic and passengers. The highest I/O ratios were measured in the bus and the lowest ones in the new tramcar. In the latter case, lower $PM_{2.5}$ concentrations of elements of anthropogenic origin were measured indoor than outdoor, indicating that the air filtering system of the modern tram worked very effectively. Lower PM and elemental concentrations, by at least a factor of two, were measured inside the modern CAF trams than in the old type of trams, which had already been running for 18 years. For Cl, the I/O ratios and, thus, indoor enrichment were all higher than all of the other elements for all vehicle types, indicating an indoor source of chlorine.

In terms of each vehicle type, we found at least one element that was characteristic to that type, and its origin was the vehicle itself or its infrastructure. These elements were identified by the higher EF and indoor enrichment values and high I/O ratios. High Fe and Mn concentrations in the trams, trains and the metro originated from rail abrasion. Copper and zinc could come from the overhead wires in the case of the trams, the trolleybus and the train.

Different aerosol compositions and very high concentrations for some elements were observed inside the metro carriage. Enrichment factors of Mn, Fe, Br, Ba and Pb were much higher than in the aboveground vehicles. This could be explained by the fact that the metro railway systems have a closed character, restricted ventilation and special emission sources [16]. Fe accounted for more than 50% of the measured mass, and other heavy metals, such as Cr, Mn, Cu, Br and Pb, were also present in remarkable concentrations. Their source could be rail abrasion. We note that these outdated Russian trains where we collected the samples are no longer operational. Very high PM_{coarse} concentrations were measured in the IC train on both occasions; however, in this case the measured elemental concentrations did not account for the elevated PM levels. Therefore, the samples were investigated by scanning electron microscopy to obtain better insight into the pollution in the trains (see Section 3.4).

Figure 2 presents the indoor EF ratios of the different vehicles in 2014. In most cases, the indoor EF values were approximately 1, as expected. Nevertheless, some elements had higher values. In the buses, the indoor EF values were all approximately 1 for both fractions, suggesting that the pollutants originated from outside. Chlorine had the highest indoor enrichment in the trolleybuses and old and new types of trams. In the fine fraction, the source of chlorine was assumed to be sodium hypochlorite, which is used to clean the interior of vehicles. The origin of the coarse fraction of Cl is a question to which single particle analysis by SEM could provide answers. The indoor EF of zinc was above 1 in the trolleybuses and the two trams; it could originate from the overhead wires. Moreover, high iron and silicon values were identified in the trams. The most probable source of iron could be the rail and tram wheel abrasion.

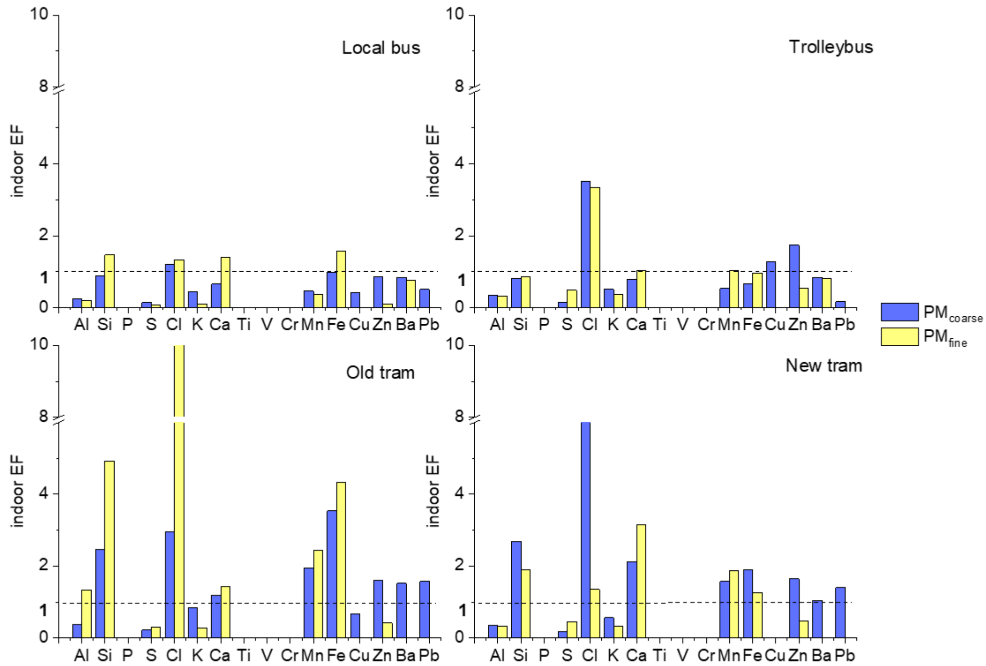
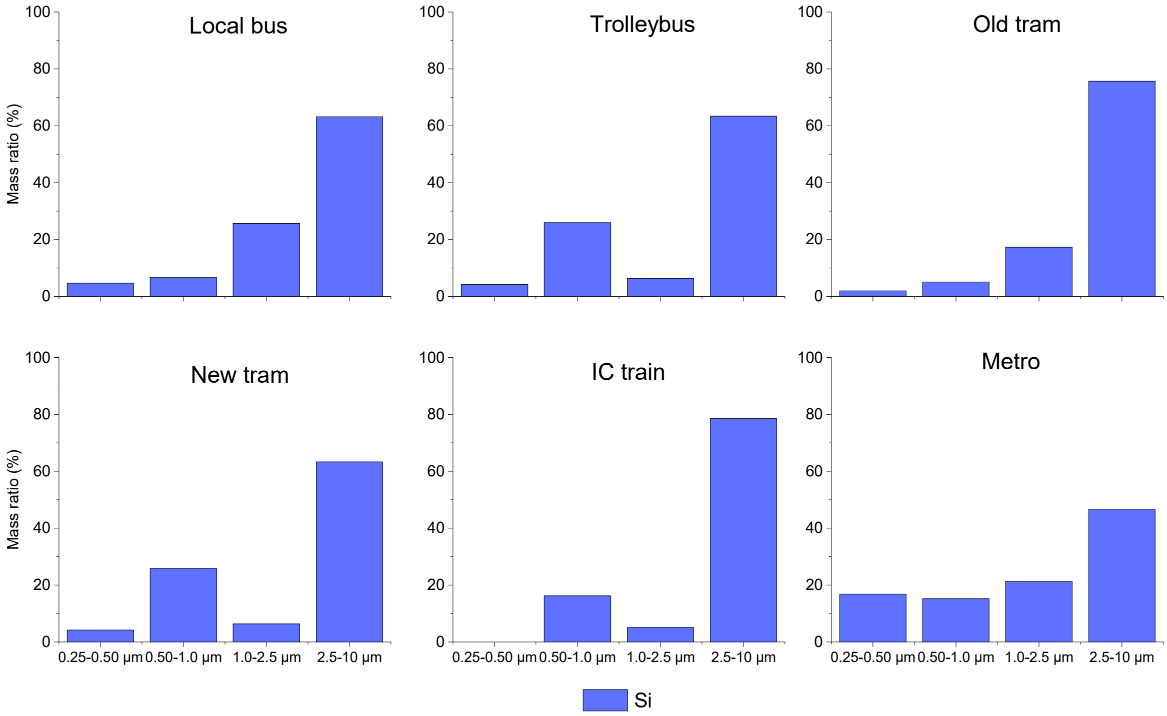


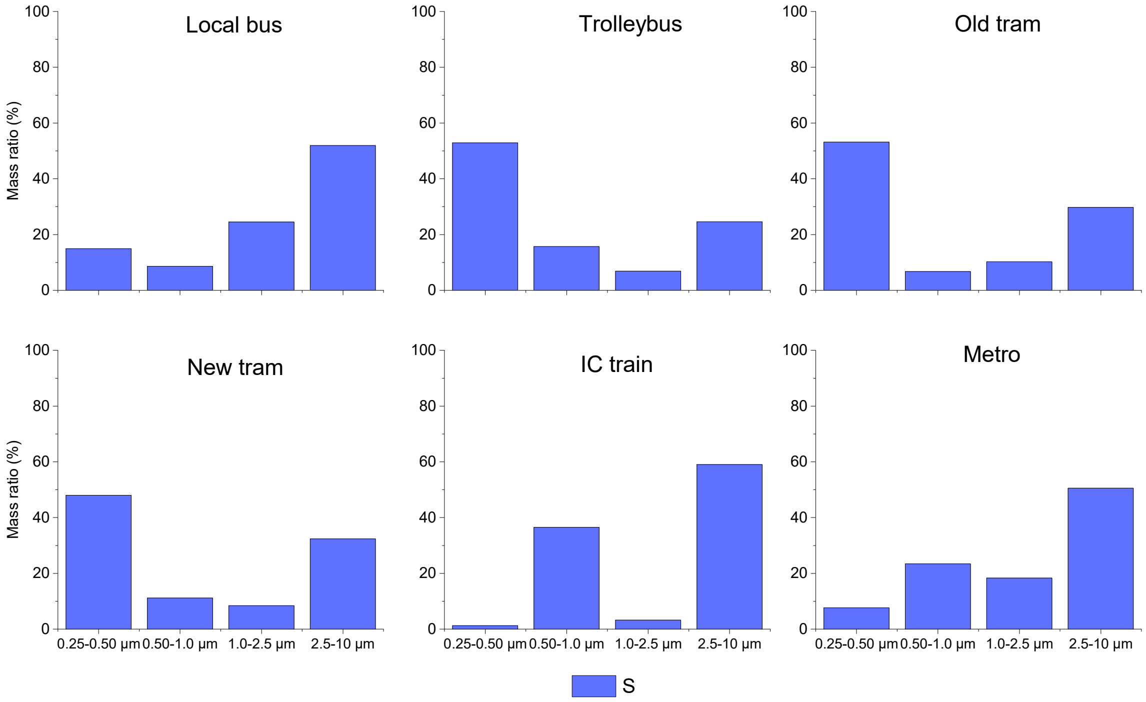
Figure 2. Indoor EF ratios in different vehicles in PM_{fine} and PM_{coarse} fractions in 2014.

3.3. Elemental Mass Size Distribution

The mass size distributions of three selected elements are shown on Figure 3. Silicon (Figure 3a) represents the crustal elements, sulphur is representative of anthropogenic elements of outdoor origin and chlorine is the element with an indoor source. The size distribution of all of the elements can be found in Supplementary Materials Figure S3. In the mass size distribution of the Earth's crust elements, the main peak appeared in the coarse fraction, as was expected. In the case of sulphur (Figure 3b), the distribution had a bimodal shape in most cases. One peak could be found in the condensation mode (0.25–0.50 µm) and another in the coarse mode. The mass size distribution of S in the trams and the trolleybuses showed similarity with a dominant peak in the condensation mode. The elevated PM_{coarse} concentrations could be responsible for the considerable peak in the coarse mode, especially in the case of the bus and the metro. In the IC train, the fine-mode S peak appeared in the droplet mode (0.5–1 µm). The IC train had a closed ventilation system, which could cause the shift in the size distribution. The mass size distribution of chlorine (Figure 3c) differed from vehicle to vehicle. Most of the Cl could be found in the coarse mode; however, in specific vehicles, considerable amounts of Cl were found in the 1–2.5 µm size fraction. One possible source of Cl is from the cleaning of the vehicles. However, it does not explain the very high concentrations in the coarse mode. The Metro carriage showed different features, which was expected due to the fact of its specific microenvironment.

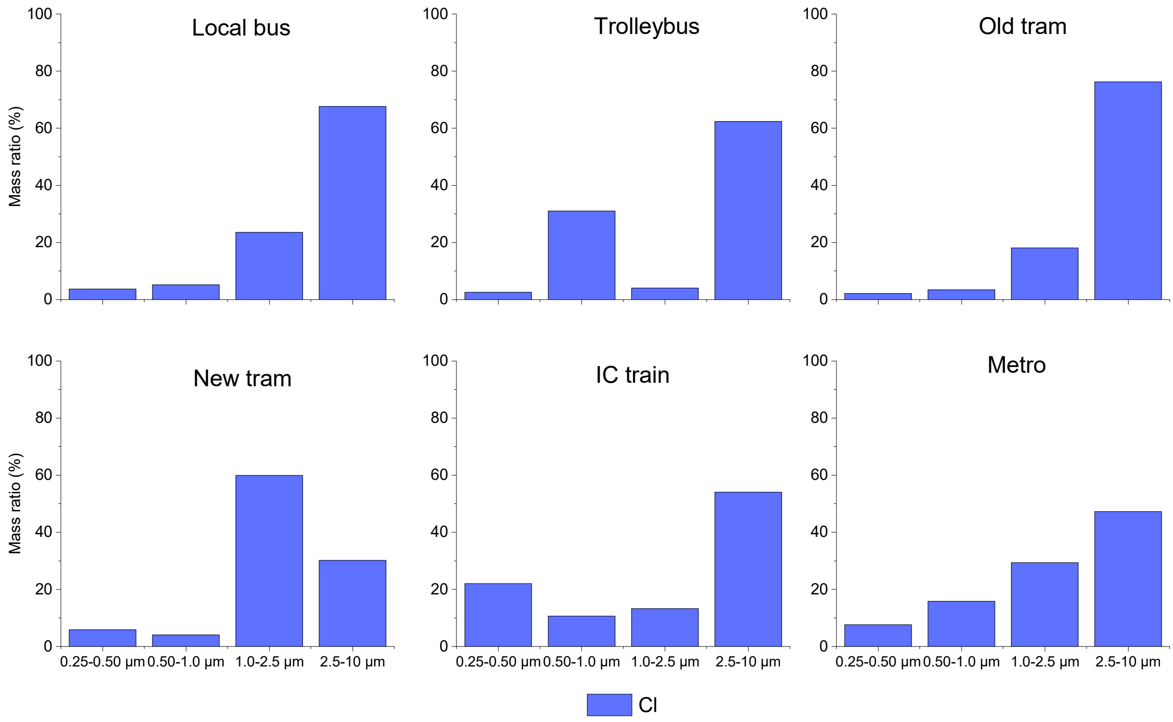


(**a**)



(**b**)

Figure 3. *Cont.*



(c)

Figure 3. Silicium (**a**); sulphur (**b**); chlorine (**c**) mass size distributions in different vehicles in 2014.

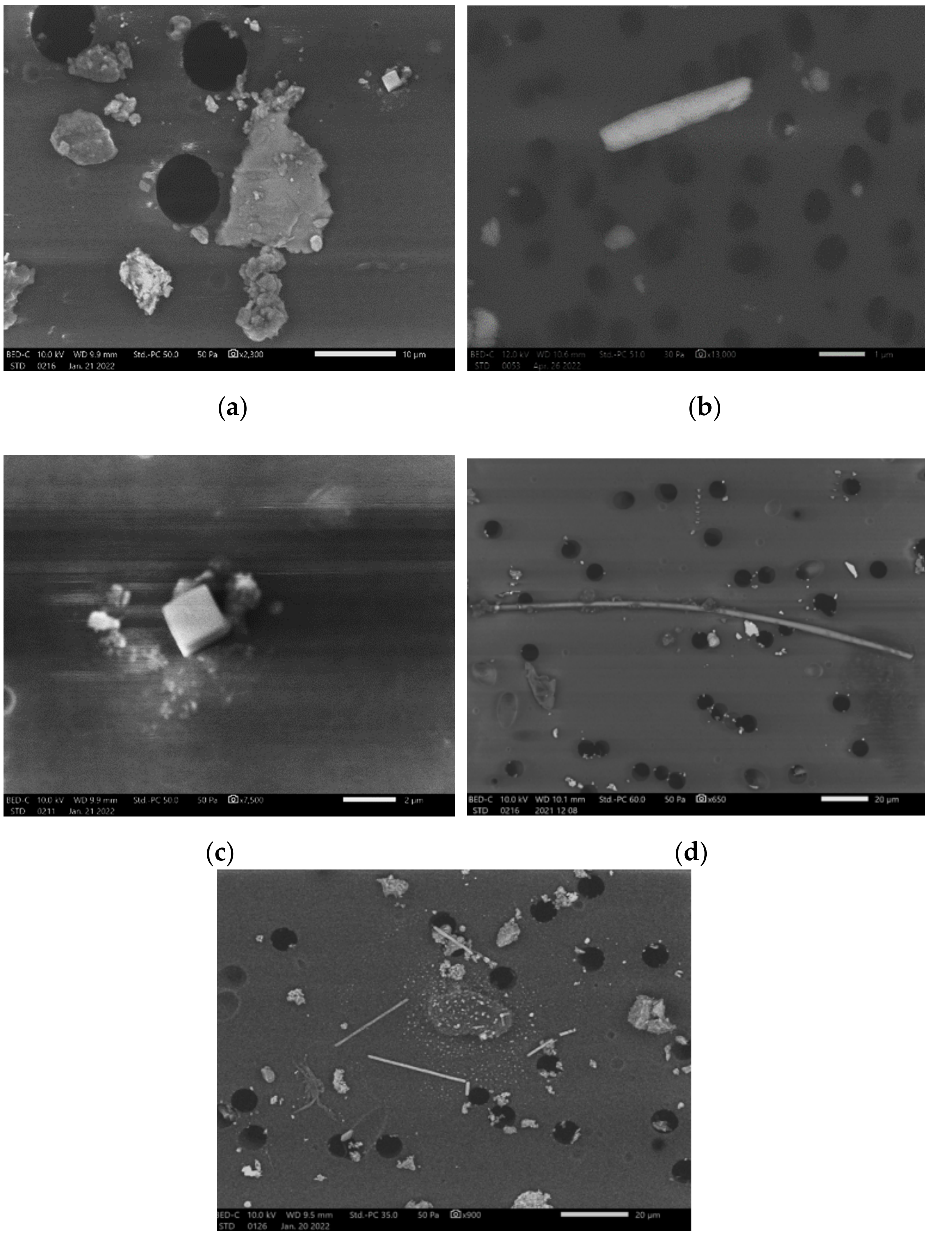
3.4. Single Particle Analysis by Scanning Electron Microscopy

As mentioned earlier, the bulk composition did not provide satisfactory explanations as to the origin of pollution in some cases. One interesting question was the enhanced PM_{coarse} levels in the IC train and in the old tram and another was the origin of the very high concentrations of Cl inside some vehicles. Therefore, single particle analysis using scanning electron microscopy was performed on all coarse mode samples from all of the vehicles. Morphology together with its chemical composition of an aerosol particle provides important information about its origin, formation, aging and its possible impact on the environment and human health. The following particle types were identified: amorphous (i.e., Al–Si rich, Al–Si–Fe rich, Si rich and Fe rich), fibrous (i.e., C–O rich), cubic (i.e., KCl and NaCl), fiber (i.e., K rich) and filings (Fe rich).

Figure 4a shows amorphous particles (i.e., Al–Si–K–Mg rich) identified as mineral dust [49]. This formation was found in all types of vehicles in great number.

Fe-rich filing particles (Figure 4b) were observed in the Metro, which was unique to this vehicle. Such particles were responsible for the very high iron concentration measured in the underground train.

Chlorine appeared in the form of NaCl and KCl crystal cubes. Its origin was presumably salt remaining from winter de-icing in early spring and fertilizer in autumn [50]. Mainly, KCl was found in the trolleybuses in September 2014 and NaCl in the case of the old tram in March 2012 (Figure 4c). Furthermore, fibrous material with C and O contents were found in all of the samples (Figure 4d), which suggests an organic origin. Such fibers occurred in a great number on the samples from the IC train. K-rich fiber particles were observed in 2014 in the old tram (Figure 4e). One possible origin of the fibers are the seats, another is the air ventilating system.



(**a**) (**b**) (**c**) (**d**) (**e**)

Figure 4. The determined particle types by scanning electron microscopy: (**a**) Al-, Si-, K- and Mg-rich amorphous particles (old tram, 2012); (**b**) Fe-rich filings (Metro, 2014); (**c**) cubic NaCl particle (old tram, 2012); (**d**) fibrous C–O-rich particle (IC train, 2014); (**e**) K-rich fibers (old tram, 2014).

4. Conclusions

In this work, APM pollution was characterized while travelling on different public transport vehicles in Hungarian cities. Considerable PM levels were observed, especially on the old vehicles. The measured concentrations in the different public transport vehicles were 2–19 times higher than the concentrations measured outside at a traffic site or at urban background sites at the same time. Consequently, significant aerosol exposure was experienced by passengers, even if they were exposed to this high pollution level only for a short time. With the help of elemental composition and enrichment factor analysis, the sources of PM pollution could be identified. The origin of the elevated pollution levels were the resuspension of the mineral and road dust, the emission of the engine and various other indoor sources. The identified indoor sources, such as cleaning and disinfectant materials, fibers from seats, railway and wheel abrasion and overhead wires significantly contributed to the PM pollution. On vehicles operating on railways, high amount of Fe, Cr and Mn concentrations were found, the source of which was obviously railway abrasion. We have shown that the air circulation systems can cause very high pollution levels without proper cleaning and maintenance. On electrical vehicles, the aerosol pollution was lower, and the exposure of the passengers and drivers could be further reduced by using modern air technology and sufficient maintenance.

We compared our results to other similar studies from all over Europe and the world. The concentrations and elemental composition measured inside the vehicles were in accordance with the results from other European cities.

The work presented here is the result of the first two sampling campaigns of a planned measurement series. The next step is a systematic study of air quality inside trams in Debrecen [19]. Unfortunately, an extensive and systematic campaign planned for 2020 had to be cancelled due to the COVID-19 pandemic, and it is still not clear when it can be carried out.

This is one of the first studies to report on the characterization of PM pollution inside public transport vehicles from the middle-eastern European region and the first time the elemental mass size distribution has been provided. Therefore, despite the limitation of the study due to the small number of samples, it can serve as a basis for comparison for further investigations. Most vehicles involved in this study were old, using outdated technology. Some of these vehicles are still operational. It will be interesting to see the improvement in air quality inside the different public transport vehicles when modern air circulation technology and "cleaner" transportation modes are applied.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/atmos13050692/s1, Figure S1. PMcoarse and PMfine PIXE spectra from different years and different transport vehicles; Figure S2. Mass size distributions for each element in different vehicles in 2014; Table S1. The detection limit and the error of the analytes.; Table S2. Comparison of mean PM_{10} and $PM_{2.5}$ concentrations inside in railway and metro microenvironments.

Author Contributions: E.P. and E.F.; were responsible for the sampling, E.P., A.A., Z.S., E.F., Z.T. and Z.K.; performed the experimental work, E.P.; performed the data analysis, A.A. and E.P. carried out the SEM analysis, E.P.; prepared the figures, E.P.; drafted the original manuscript, Z.K.; made the review of the original draft, A.A.; contributed to the review and editing, Z.K. was the supervisor of the project. All authors have read and agreed to the published version of the manuscript.

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