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Exposure to air pollution inside electric and diesel-powered passenger trains

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Keywords

Diesel exhaust, diesel train, personal exposure, ultrafine particles, black carbon, nitrogen oxides, underground station, commuter

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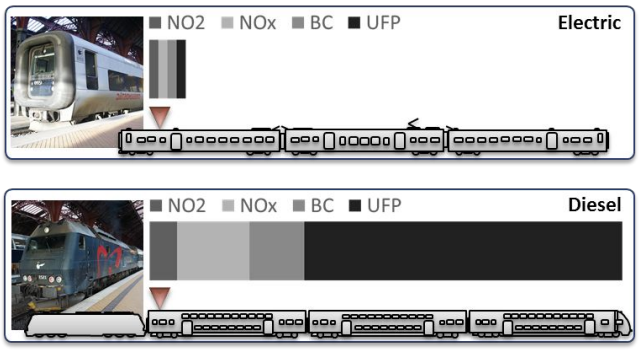


Abstract

Diesel-powered trains are used worldwide for passenger transport. The present study aimed to assess air pollution concentrations in passenger cars from diesel and electric trains. Personal exposure monitoring (6-7 hours per day) was carried out for 49 days on diesel and 22 days on electric trains. Diesel trains had higher concentrations of all the assessed air pollution components. Average increases (and fold differences) in passenger cars of diesel trains compared with electric trains were for ultrafine particles 212,000 particles/cm^3 (35-fold), black carbon 8.3 µg/m^3 (6-fold), NO_X 316 µg/m^3 (8-fold), NO_2 38 µg/m^3 (3-fold), $PM_{2.5}$ 34 µg/m^3 (2-fold) and benzo(a)pyrene 0.14 ng/m^3 (6-fold). From time-series data, the pull and push movement modes, the engine in use and the distance to the locomotive influenced the concentrations inside the diesel trains. In conclusion, concentrations of all air pollutants were significantly elevated in passenger cars in diesel trains compared to electric trains.

Introduction

Railway passenger and freight transport activity have been growing globally in the last 40 years[1]. Electricity is the power supply of choice in trains in many areas, but a significant proportion of commuter and freight trains worldwide are still powered by diesel fuel. It has been shown that

diesel-powered trains increase the concentrations of fine particulate matter ($PM_{2.5}$) and ultrafine particles (UFP; <0.1 μm) in the open air close to the railway[2, 3]. However, PM has been shown to also penetrate into passenger cars of the train[4, 5]. A review of commuter exposure to particles reported three times higher UFP concentrations in passenger cars of diesel than electric trains, with highest concentrations when diesel-propelled locomotives operate in pull mode[6]. Two more recent studies also reported significant differences of UFP concentrations inside passenger cars of diesel trains depending on position of locomotive[4, 7]. In general, high variability of UFP concentrations has been reported inside diesel trains. More knowledge on seasonal variation and other components of the complex diesel exhaust mixture penetrating to train cars is needed. Electric trains generate only particle emissions from non-exhaust sources such as wear and mechanical friction on rail tracks, wheels and brakes, as well as friction and arcing of the electrical wires. The majority of the studies on electric trains have addressed particle exposure on train platforms and tunnels[8], although a few recent studies assessed exposures and factors affecting exposures inside passenger cars[5, 9].

High concentrations of diesel exhaust in passenger cars are worrying because it is associated with increased risk of lung cancer as well as other lung and cardiovascular diseases[10-12]. In addition to passengers, train conductors, engine drivers, maintenance staff, platform staff and other train personnel can be exposed. Our study was initiated because of concern about potential health effects in workers and passengers who commute in diesel-powered trains. In Copenhagen, Denmark, the urban train system is electrified, but co-exists with diesel-powered trains that travel further on intercity routes. A recent report showed that two types of diesel rolling stock used in Denmark gave rise to higher UFP exposure to staff working in the passenger cars than what would occur on the most busy street in Copenhagen[13]. The aim of the present study was to perform a

comprehensive comparison of the concentration levels inside passenger cars of diesel-powered and electric trains including both particulate and gaseous air pollutants.

Methods

Study design

Twenty-nine volunteers were recruited to sit inside the front car of Danish State Railways trains running in the Zealand region, carrying portable measuring devices and samplers. The concentrations characterization was performed in the same type of trains on three consecutive days each week (Tuesday, Wednesday and Thursday) for 6-7 hours per day. A pilot study was conducted during two days in March in order to characterize the size distributions, test the concentration contrast and test the chosen routes. The main study period covered a total of 23 weeks from May to December 2017. Out of the 23 monitoring weeks, seven were conducted in electric trains and 16 in diesel trains. In addition to measurements in trains, two more days of measurements were done on a busy multimodal underground train platform in Copenhagen, as a worst-case scenario used opportunistically to assess the contribution of the studied diesel trains in the underground environment.

Trains and train routes

The electric trains operated on a route from Østerport station to Elsinore station (46 km). The train route was taken forward and back 3 times per day, for a total of 5-hours and 37 minutes per day, including waiting time at Østerport and Elsinore stations, starting at 9h39 and ending at 15h16. The rolling stock is termed Litra ET and runs on alternating current in overhead power lines (25 kV and 50 Hz).

As the diesel train exposure scenario, a double deck train propelled by a diesel locomotive (Litra ME, from 1981-85, 33 units in circulation) was chosen in a route from Østerport to Kalundborg station (114 km), return to Copenhagen Central Station, and from there to Holbæk (part of Kalundborg line, 68 km) and back to Copenhagen Central Station. The route took a total of 7 hours per day, including waiting time at the stations, starting at 8h34 and ending at 15h32. This route was chosen because the same locomotive was used all the day (defined as "diesel A"). For the measurements in October and November, the trip was firstly heading to Holbæk and then to Kalundborg, driven with three different ME locomotives and saving one hour of waiting time at the station (defined as "diesel B"). The driving locomotive code was noted every day for the diesel scenario. The diesel routes include a passage at one underground station in the Copenhagen city centre (Nørreport Station). The plan for the different weeks and schedules are available in supporting information (SI, Tables S1 and S2).

The windows in both passenger train cars (electric and diesel) were sealed and could not be opened. Furthermore, the ventilation system was not passenger-adjustable.

Underground station

The underground train platform at Nørreport station is a 200 m long single platform positioned between two tracks. It is equipped with a modern ventilation system (from 2014) and serves both electric and diesel regional and intercity trains. Measurements were carried out on two different days: on the first day (Friday January 26th) all train types were running as usual, but on the second day (Friday February 2nd) all ME locomotives had been taken out of circulation for maintenance, providing a unique opportunity to perform measurements at the underground station both with and without the ME-diesel locomotives. The monitoring instruments were maintained in the same

118 position on the train platform for 4 hours. The train passages were noted for both tracks in both
119 days.
120
121 ***Measurements, instrumentation and chemical analyses***
122 All air pollution measurements were performed as personal monitoring with the volunteers
123 carrying the equipment. In the diesel trains the volunteers were sitting in the compartment in the
124 front section of the first passenger car (Figure S2) and in the electric trains they were sitting in the
125 first passenger car. Black carbon mass concentration (BC) and UFP number concentration were
126 measured on each monitoring day during the total period of the study, both on-board of the diesel
127 and electric trains and on the platform of the underground station. Nitrogen oxides (NO_2 and NO_x;
128 whole period), aldehydes (10 weeks from May to September), $PM_{2.5}$ and polycyclic aromatic
129 hydrocarbons (PAHs; 9 weeks in October and November) were sampled on-board of the electric
130 and diesel trains (Table S1). Additionally, PM was collected by means of a mid-volume
131 electrostatic sampler, described elsewhere[14], in the compartment in the front train car during the
132 monitoring weeks in December. Measurements to investigate concentration gradient inside the
133 trains were performed for three days, where one person sat in the compartment in the front section
134 of the first passenger car, while another person sat each day at a different position in the lower
135 deck of the train (for entire day trip), each of them carrying one BC and one UFP measuring
136 device. A summary of the measurements, instruments, periods and sites monitored as well as time
137 resolutions and flows used is presented in SI (Table S3).
138
139 *Black carbon*
140 Black carbon was measured with 1-minute resolution using two MicroAeth AE51 aethalometers
141 (Aethlabs, San Francisco, CA) with high degree of comparability (correlation coefficient, r=0.95).

The filters inside the aethalometer were changed after each monitoring day in the diesel trains, and after the three monitoring days in the electric trains.

Ultrafine particles

UFP concentrations were measured using both diffusion chargers DiSCmini (DM; DiSCmini, Matter Aerosol AG, Wohlen, Switzerland) and NanoTracer (NT; Aerasense NanoTracer, Oxility, Eindhoven, the Netherlands). For the two-day pilot study the NanoScan SMPS model 3910 (TSI, Shoreview, MN, USA) was also used. The devices were synchronized with a working computer's clock. Two DM devices were used for the gradient measurements with high degree of comparability (r=0,96). Measurement time resolutions were 1-second for DM, 16-seconds for NT and 1-minute for NanoScan SMPS (Table S3).

Nitrogen oxides

Passive Ogawa samplers (Ogawa, FL, USA) were used for personal sampling of nitrogen oxides. Two samplers were used over the three consecutive days in each exposure scenario and kept in sealed plastic bags overnight at 4°C (and after completed sampling). Samplers were analysed by ion chromatography and corrected with field blanks[15]. The limit of detection (LOD) for the sampling time of 18-21 hours was 0.7 $\mu g/m^3$ for NO_2 and 1.4 $\mu g/m^3$ for NO_x. None of the samples were below the LOD.

Aldehydes

Aldehydes were measured with Sep-Pak XPoSure aldehyde samplers (Waters, MA, USA) connected to pumps. The samplers were closed with caps and kept overnight at 4°C during the three cumulated sampling days for each exposure scenario and in the end kept at -20°C. The samplers were analysed for 13 different aldehydes and one ketone using high-performance liquid

chromatography with UV detection. The sampled air volume was 0.21-0.27 m^3 and the LOD was 0.5 μg/sample for each substance.

$PM_{2.5}$ and polycyclic aromatic hydrocarbons

$PM_{2.5}$ was collected on filters (Teflon w/ring PALL 2.0 μm, 37 mm) using a cyclone (Triplex cyclone, BGI, USA) at 1.5 L/min. A sequence set of two XAD-2 tubes (sample + backup tube) for sampling of gaseous PAHs were attached after the cyclone (Figure S1). After the sampling week was completed, the XAD-2 tubes were wrapped in aluminium foil and kept at -20°C. The filter cassettes were wrapped in aluminium foil and kept in the dark, dry and at room temperature before and after use. The Teflon filters were weighed before and after sampling in controlled climate conditions. LOD for particle mass was 29 μg. Two filters were excluded due to sampling failure and/or failure in the chemical analysis.

Particles on filters were extracted and analysed for the content of 16 US EPA priority PAHs (Table S5) using high-resolution gas chromatography/low-resolution mass spectrometry[16]. Field blanks were analysed along with the samples, and certified reference material was used for quality control. The XAD-2 from the sample and backup tube were combined and sonicated with 5 mL cyclohexane for 60 minutes and analyzed by gas chromatography-mass spectrometry for the same PAHs as in filters, analyzed in a different laboratory as previously described[17]. LODs for gaseous PAHs were 0.1-0.6 pg/μL extract depending on the individual PAH.

PM metal content

PM collected with the electrostatic sampler in weeks 23 and 24 in diesel A scenario was analysed for the content of 24 elements by inductively coupled plasma mass spectrometry. Detailed information is available in SI, Table S6.

Time-series data analyses

The time-series data from BC, DM and NT (also from NanoScan, for the pilot study) were synchronized in start and ending times and grouped by scenario (diesel A, diesel B and electric). The BC data were collected with two duplicate devices, which were averaged for each measured resolution unit (minute). Data for each monitoring day from each device were aggregated in 10 minutes intervals and the time slots of the different days were averaged. The averages and percentiles for each 10-minute average concentration over time of the day within each scenario were then plotted, as well as 10-minute maximum values using R statistical language[18]. The BC and DM time-series from the concentration gradient measurements and from the underground station were kept and re-sampled in the smallest time resolution possible for comparison (i.e. 1 minute). Some of the monitoring days were excluded from the analysis (Table S4).

Statistics

The Welch t-test was used to compare the concentrations in diesel and electric scenarios using R statistical language[18]. The average of daily 10-minute slots for all days measured was used for calculating the mean, SD and range for time-series data from BC and UFP. The average concentration of the two duplicate personal samplers used per week was used for calculating the mean concentrations, SD and the range in concentrations for NO_x, NO_2, aldehydes, $PM_{2.5}$ and the PAHs.

Results

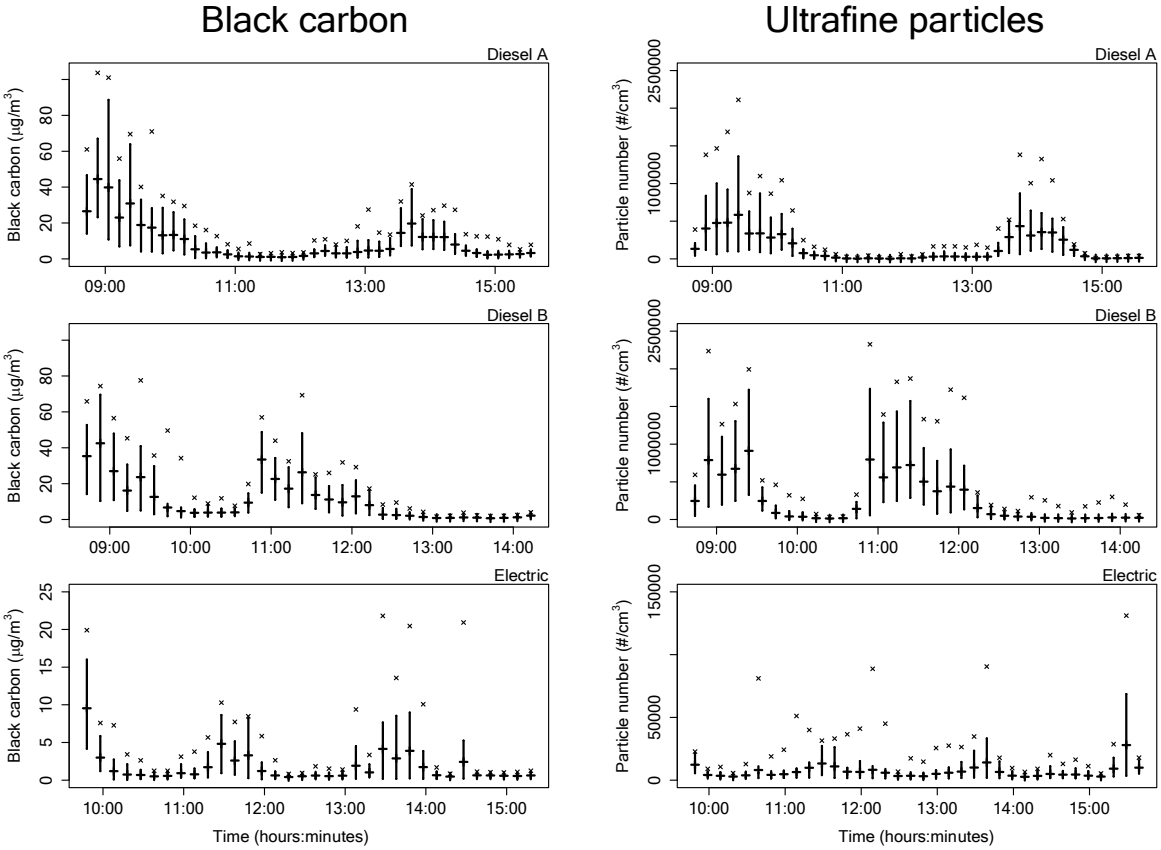
On-board the trains

Black carbon and ultrafine particles

Total particle number concentration and average size of UFP were measured in a pilot study in Diesel A scenario with three different devices, showing similar readings of particle number concentrations between devices (Figure S3). Detailed output figures from the NanoScan SMPS device from both electric and diesel pilot measurements are in SI (Figures S4 and S5). For personal monitoring in the main study the NanoScan device was not used since it is less portable.

Figure 1 shows the mean daily particle concentration patterns inside the train in the Diesel A, Diesel B and Electric exposure scenarios. The patterns for BC mass concentrations and UFP number concentrations (measured by DM equipment) showed similar trends (with daily averages from both devices with r= 0.75). The NT measurements were similar to the DM data and are shown in SI (Figure S6). For both BC and UFP in the Diesel A scenario, a large increase in concentrations was observed between 8h30 and 10h, and a small increase between 13h30 and 14h30. A similar trend was observed for the Diesel B scenario, with the second peak appearing earlier in the day. The increased concentrations of BC and UFP in both diesel scenarios occurred when the locomotive was pulling the train (leaving Copenhagen) and the lower concentrations when the locomotive was pushing the train (returning to Copenhagen). When the locomotive was in front, higher concentrations of BC and UFP were measured inside the passenger car with average particle number concentrations of around 400,000 particles/cm^3 and BC mass concentration exceeding 20 $\mu g/m^3$. For the 10-minutes average maximums, the UFP concentrations exceeded 2 million particles/cm^3 and BC mass concentration was 70-100 $\mu g/m^3$ (Figure 1). When the locomotive was in push mode the averaged concentrations were 10,000-30,000 particles/cm^3 and 2-3 $\mu g/m^3$ BC. The concentrations of BC and UFP inside the electric trains were considerably lower than in the diesel trains. A small increase in concentrations could be observed when the train was at Østerport station in Copenhagen, in the beginning and end of the measurements, and also in the time intervals 11h16-11h39 and 13h16-13h39 when the

241 volunteers waited on the Østerport station platform to change trains. At the terminal station for
242 electric trains in Elsinore, the concentrations were at the same level as when the train was in
243 movement (between 3,000-7,000 particles/cm^3 and 0.5-4 µg/m^3 BC), while at Østerport station,
244 shared with diesel trains, the averaged concentrations were 10,000-13,000 particles/cm^3 and 3-5
245 µg/m^3 BC. Table 1 summarizes the daily averaged concentrations of BC and UFP for diesel (both
246 diesel A and B) versus electric scenario and Table 2 shows the differences between movement
247 mode (pull/push, forward/backward, for diesel and electric) and for the different train scenarios
248 (diesel A, diesel B and electric). For BC, concentrations were 6-fold higher in the diesel compared
249 to the electric scenario and for UFP 35- and 20-fold higher (measured with DM and NT,
250 respectively).
251



252
253 Figure 1 - Daily temporal BC mass concentrations and UFP number concentrations (measured by
254 DiscMini) in the defined scenarios (Diesel A, Diesel B and Electric). The symbols represent the

255 arithmetic mean (horizontal lines), the 10th to 90th percentile distributions (vertical lines), and the
256 maximum values (point crosses) for the 10 minutes averaged data. BC data were collected from 28
257 monitoring days for the Diesel A scenario, for 17 days for the Diesel B scenario and 18 days for
258 the Electric scenario. UFP data were collected from 21 days for Diesel A, 18 days for Diesel B
259 and 17 days for Electric (Table S4 presents missing data). Note the different y-axis ranges for
260 diesel and electric trains.
261
262 Table 1 – Black carbon, ultrafine particles, nitrogen oxides, aldehydes, $PM_{2.5}$ mass concentration
263 and PAHs in particles mass and gas phase, respectively.

	Diesel train			**Electric train**			**Fold difference** (diesel vs electric)	**Mean difference (CI 95%)**
	n	**Mean±SD[1]**	**Range[1]**	**n**	**Mean±SD[1]**	**Range[1]**		
Black carbon ($\mu g/m^3$)	21	10.0 ± 3.3	3.8-15.1	7	1.7 ± 0.6	1.2-3.2	6	8.3 (7.3; 9.3)***
UFP from DM ($\#/cm^3$)	15	218,000 ± 140,000	31,700-560,000	6	6,150 ± 2,310	4,100-12,700	35	212,000 (167,000; 257,000)***
UFP from NT ($\#/cm^3$)	17	153,000 ± 81,000	29,200-425,000	7	7,760 ± 4,200	3,670-22,600	20	145,000 (121,000; 169,000)***
NO_x ($\mu g/m^3$)	14	364 ± 81	275-531	7	48 ± 16	25-65	8	316 (268; 365)***
NO_2 ($\mu g/m^3$)	14	53 ± 16	35-99	7	16 ± 10	6-31	3	38 (26; 49)***
Aldehydes (sum) ($\mu g/m^3$)	6	50 ± 8	37-59	4	41 ± 2	39-44	1.2	NC[2]
Formaldehyde ($\mu g/m^3$)	6	9.0 ± 2.1	6.4-11.4	4	5.2 ± 0.3	4.8-5.5	1.7	NC[2]
$PM_{2.5}$ ($\mu g/m^3$)	6	68 ± 15	51-96	3	32 ± 7	24-37	2	NC[2]
∑PAH in $PM_{2.5}$ (ng/m^3)	6	1.7 ± 0.6	1.0-2.5	3	0.31 ± 0.13	0.21-0.45	6	NC[2]
BaP (ng/m^3) in $PM_{2.5}$	6	0.17 ± 0.09	0.08-0.30	3	0.03 ± 0.003	0.028-0.034	6	NC[2]
Σ PAH XAD-2 tubes (ng/m^3)	6	165 ± 56	110-261	3	146 ± 48	116-201	1.1	NC[2]
Naphthalene XAD-2 tubes	6	134 ± 26	99-170	3	108 ± 55	67-170	1.2	NC[2]

(ng/m^3)								

n, number of weeks sampled (3 days per week); SD, standard deviation; CI, confidence interval; UFP, ultrafine particles; DM, DiscMini; NC, not calculated; NT, NanoTracer; ∑PAH, total amount of PAH in particles collected on filter or collected in the XAD-2 tubes; BaP, benzo(a)pyrene. [1]Mean and Range determined from values over daily averages of 10-minute time periods; [2] Limited number of samples to calculate a reliable mean difference. Diesel trains include both scenarios A and B; *** $p<0.001$

Table 2 – Black carbon and ultrafine particle concentrations for movement mode and scenario

		Pull mode/forward		**Push mode/backward**		**Fold difference** (pull vs push)	**Mean difference** (CI 95%)
		Mean±SD[1]	**Range[1]**	**Mean±SD[1]**	**Range[1]**		
Scenario A	Black carbon (µg/m^3)	19.8 ± 10.7	8.0-44.5	2.0 ± 0.9	0.9-3.3	10	17.8 (12.1; 23.5)***
	UFP from DM (#/cm^3)	347,600 ± 111,200	132,400-583,500	10,400 ± 7,800	3,300-34,000	33	337,200 (278,000; 396,600)***
	UFP from NT (#/cm^3)	279,500 ± 88,000	128,000-500,200	13,100 ± 12,000	5,200-53,800	21	266,400 (219,000; 313,700)***
Scenario B	Black carbon (µg/m^3)	20.8 ± 10.4	8.0-42.5	2.5 ± 1.8	0.7-6.7	8	18.3 (12.5; 24.2)***
	UFP from DM (#/cm^3)	539,600 ± 228,000	152,400-912,300	27,600 ± 18,700	13,100-87,400	20	512,000 (385,500; 638,600)***
	UFP from NT (#/cm^3)	400,400 ± 130,800	185,200-656,300	26,000 ± 23,000	11,500-103,600	15	374,400 (301,200; 447,500)***
Electric	Black carbon (µg/m^3)	1.1 ± 0.8	0.4-3.0	0.9 ± 0.5	0.5-1.9	1.2	0.3 (0.4; 0.9)
	UFP from DM (#/cm^3)	4,500 ± 1,900	2,700-8,300	5,500 ± 2,200	2,800-9,900	0.8	960 (900; 2,900)
	UFP from NT (#/cm^3)	5,200 ± 1,400	4,000-8,000	7,400 ± 5,700	3,900-24,000	0.7	2,200 (1,500; 5,900)

SD, standard deviation; CI, confidence interval; UFP, ultrafine particles; DM, DiscMini; NT, NanoTracer; [1]Mean and Range determined from values over daily averages of 10-minutes data collected with trains in movement, excluding time at station. The 10-minute resolution averages were not completely synchronized with the real position of the train, but buffered with time at station; *** $p<0.001$

Nitrogen oxides

NO_x on board the diesel trains was nearly 8 times higher than on the electric trains (Table 1 and Figure 2). NO_2 concentrations were about 3-fold higher on the diesel trains compared with the electric trains. There was a trend of increasing concentrations of NO_2 during the two last weeks of the project (end of November).

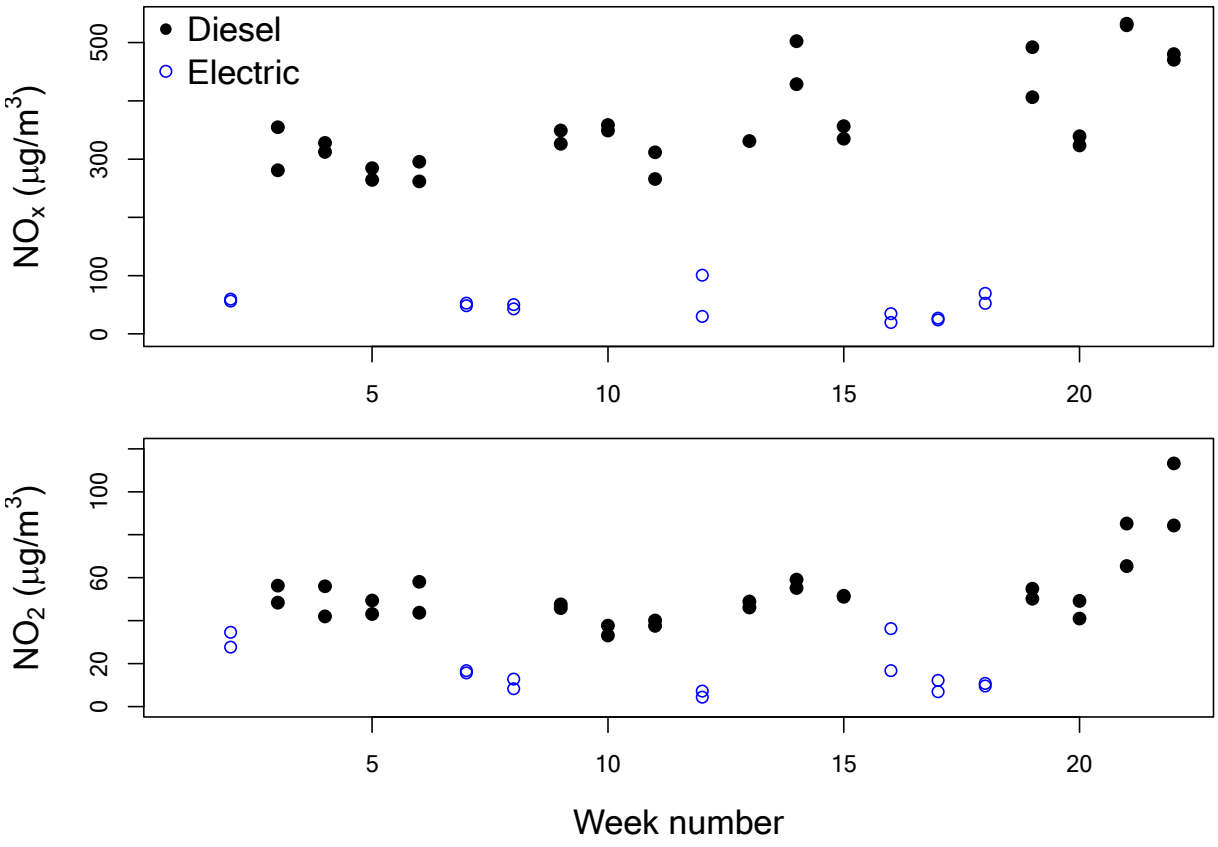


Figure 2 - NO_x (upper) and NO_2 (lower) air concentrations on board the diesel and the electric trains. Each point represents the average air concentration over the three consecutive days,

collected with two personal samplers per week. Week numbers are reference numbers for dates presented in supporting information (Table S1)

Aldehydes

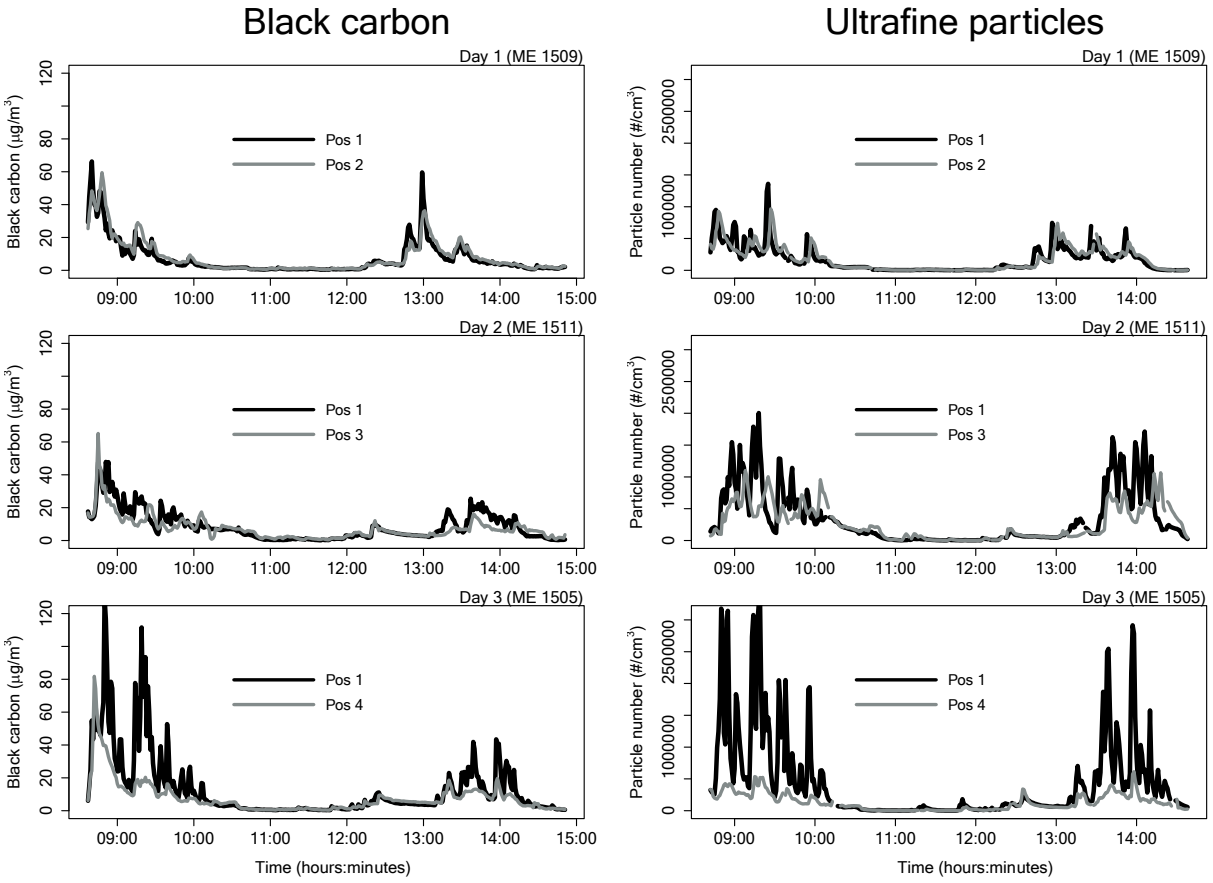
The concentrations of aldehydes were very low, and therefore these samplings were discontinued after 10 weeks. For eight of the 13 analysed aldehydes (and acetone), all or the majority of the samples were below the LOD. The other six (acetaldehyde, acetone, decanal, formaldehyde, hexanal and nonanal) are presented as aldehyde total sum (Table 1). The mean concentration of formaldehyde was 1.7-fold higher in the diesel than electric trains (Table 1).

$PM_{2.5}$ and polycyclic aromatic hydrocarbons

Five filter samples had mass levels below LOD (all corresponded to the electric scenario). The mean concentrations of $PM_{2.5}$ inside the diesel trains were 2-fold higher than inside the electric trains. Concentrations of benzo(a)pyrene (BaP) and the sum of particle-bound PAHs were 6-fold higher in diesel than in electric trains (Table 1). Among the 16 PAHs analysed in the particle mass, seven were quantified in all filters from both diesel and electric trains (pyrene, benz(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-cd)pyrene and benzo(g,h,i)perylene), whereas four were only found on filters from the diesel scenario (acenaphthylene, fluoranthene, chrysene and dibenz(a,h)anthracene). Among the PAHs in gas-phase (XAD-2), naphthalene was the only compound measured in all samples. Six other PAHs were detected in some of the samples, both from diesel and electric scenarios, with pyrene only detected in diesel scenario. PAHs with molecular weight higher than pyrene (MW=202) were not detectable in any of the XAD-2 tubes. Results in SI Table S5.

PM metal content

311 The results from elemental analysis of acid extracts from PM collected in diesel trains during 6
312 days in December are presented in SI (Table S6) showing iron as the most abundant element
313 measured (~45 mg/g), followed by zinc (~3 mg/g), magnesium (~2 mg/g) and copper (~1.5 mg/g),
314 and with higher metal contents than the standard diesel exhaust particle SRM2975. The analysis
315 was made using 2.35 mg PM of 100 mg collected in total.
316
317 ***Gradient on-board the diesel train***
318 Figure 3 shows the concentrations of BC and UFP at four different positions inside the train.
319 Position 1 was used as the reference position, where the volunteers were sitting in the main study
320 (Figure S2). The measured concentrations at this first position and at position 2, just next to the
321 first compartment, were similar, with a slightly lower average of the 6-hours of measurements on
322 position 1, although the maximum values corresponded to position 1, both for particle number and
323 BC mass concentrations (Tables S7 and S8). During the gradient measurements, an electrostatic
324 sampler was placed at position 1 for the collection of PM. Even though the electrostatic sampler
325 might have had an air cleaning effect at position 1, the peaked data reflected the proximity of
326 position 1 to the source. Position 3 was in the end of the first train car and position 4 was in the
327 beginning of the second car. The figure shows some day-to-day variation in concentrations, which
328 is attributed to different engines, but overall it demonstrates a decrease in particle concentration
329 with increasing distance from the engine.



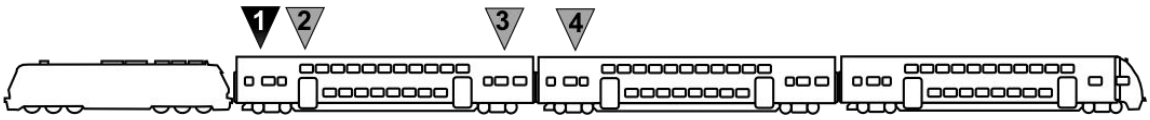
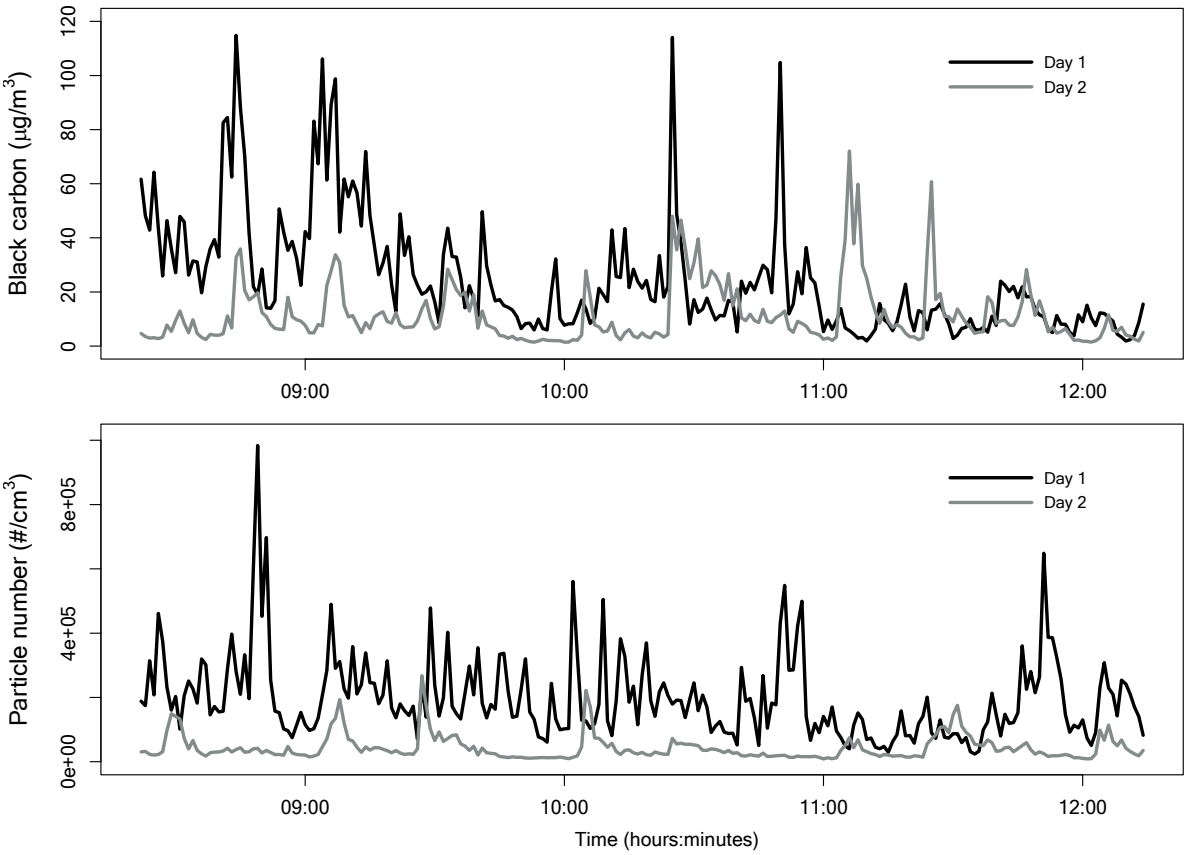


Figure 3 – Time-series data from BC and UFP concentrations (measured with DM) at four positions inside the train during three days in December 2017 (Day 1, 13-12-2018; Day 2, 06-12-2018; Day 3, 07-12-2018). Position 1 was in the front section of the first passenger car of the diesel train, closest to the engine (Figure S2); position 2, was still in the first passenger car but partially separated from position 1 with a glass that partially confines the front section (without being closed); position 3, in the end of the first car; position 4, in the beginning of the second car. Some short interruptions in the position other than position 1 correspond to device restarts performed at stations.

Underground station

342 Figure 4 shows time-series of BC and UFP concentrations for two different days at Nørreport
343 station underground train platform. The BC mass and UFP number concentrations were 2.2-fold
344 and 4.5-fold higher, respectively, on the day the ME trains were in circulation (day 1) as compared
345 to the day when the ME trains were taken out due to maintenance (day 2). Data for both days were
346 recorded for the same time period and weekday, with slightly fewer trains in traffic in day 2
347 (Figure 5). For day 2 the "other diesel" train passages were the ones that seemed to contribute the
348 most to both mass and number concentrations (Table S9 and Figures S7 and S8).



349

350 Figure 4- Time-series data from BC and UFP measured on underground train platform on two
351 different Fridays, with and without ME trains in circulation. Day 1, with ME trains in circulation
352 is represented in black colour and day 2, without the ME trains is represented in grey, both for
353 time-series data with 1-minute resolution.

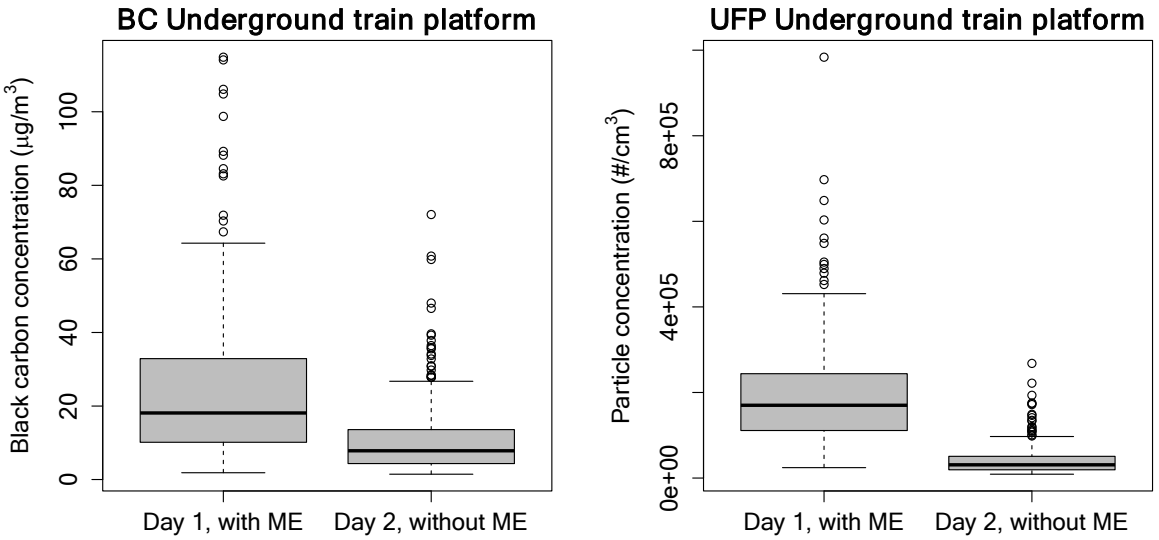


Figure 5- Underground train platform concentrations of black carbon mass (BC) and particle number (UFP) on two different days with and without ME trains in circulation. Day 1 (26/01/2018) had 111 train passages during the 4 h of measurements (37 ME, 14 other diesel and 60 electric trains). Day 2 (02/02/2018) had 89 train passages during the 4 h of measurements (20 other diesel and 69 electric trains).

Discussion

Concentrations of UFP, BC, NOx, NO_2, $PM_{2.5}$ and BaP were higher in the passenger cars of diesel trains as compared to electric trains. Considerably higher concentrations of UFP and BC were measured when the locomotive was in pull mode. Our study presents a larger data set (more trips), covers a longer time period and measures more components of DE than previous studies[4, 7, 19]. It is also clear that older diesel trains contributed substantially to UFP and BC concentrations in an underground station in Copenhagen.

The average increase in air concentrations of UFP inside the diesel train (around 200,000 particles/cm^3) was considerably higher than the personal exposure measured during passive

371 transport (car, bus or train), average 17,000 particles/cm^3, in a study of subjects in the general
372 population in Copenhagen[20]. For a daily commuter in diesel trains it would give a significant
373 contribution to the total exposure, depending on the time spent in transit. For train staff, the
374 average excess BC exposure of 8 μg/m^3 would, if assumed it is equal to elemental carbon, for 8
375 hours workday give rise to around 160 excess lung cancer deaths per 10,000 individuals over a
376 lifetime, according to published exposure-response estimates[12]. Furthermore, environmental
377 exposure to elemental carbon has been shown to increase the overall long-term mortality with
378 around 6% per 1 μg/m^3 increase at the residence[19].
379
380 BaP is a potent carcinogen and air concentrations of BaP were 6-fold higher in diesel trains
381 compared with electric trains (the same fold difference as for BC). The average concentrations of
382 BaP in diesel trains (0.17 ng/m^3) were higher than the reference level of 0.12 ng/m^3 (lifetime
383 exposure) based on the World Health Organization (WHO) unit risk for lung cancer for PAH
384 mixtures (additional lifetime cancer risk of 1 per 100,000)[21]. The average concentrations of $PM_{2.5}$
385 and NO_2 were 68 μg/m^3 and 53 μg/m^3, respectively, inside the diesel trains. For a commuter who
386 spends two hours per day travelling inside the first car of the diesel train, the average daily (24
387 hour) personal exposure to $PM_{2.5}$ would increase from 15[22] to 19 μg/m^3 and for NO_2 from 20[22] to
388 23 μg/m^3. The WHO air quality guidelines for $PM_{2.5}$ is 25 μg/m^3 for 24 h and 40 μg/m^3 annual for
389 NO_2.
390
391 The results from the real-time instruments (UFP and BC) showed that diesel exhaust enters the
392 train passenger cars especially when the locomotive is in pull mode. It means that exposure to
393 diesel exhaust in train passenger cars can be reduced considerably by using push rather than pull
394 mode. Dramatic increases of UFP in pull compared with push mode have also been observed
395 inside passenger cars of diesel trains in Israel (3 to 43 fold higher concentrations, measured in

different trains)[4] and in Canada (18 times higher concentrations, average across 28 trips)[7]. From the USA, the difference between pull and push mode was reported to be 3- and 9-fold in trains from New York and Boston, respectively[19]. Overall, the mean concentrations of UFP inside diesel trains in the present study were 20 and 35 times higher than in electric trains (measured with NT and DM, respectively), which is a larger difference than reported from previous studies[6]. Average concentrations of UFP and BC in first passenger car when locomotive was in pull mode in our study were about 450,000 particles/cm^3 and 20 μg/m^3, respectively, which is higher than reported from Canada (UFP: 126,000 /cm^3 and BC: 17.8 μg/m^3)[7].

Another observation from our study is the elevated BC and UFP concentrations in the first morning trip, demonstrated in the diesel A scenario time-series where the same locomotive was used all day (Figure 1). Higher emissions in the morning might be attributable to a "cold" engine having non-optimal combustion efficiency. The study also showed a decline in BC and UFP concentrations from the passenger car proximal to the locomotive to the more distal train cars, suggesting that infiltration of diesel exhaust in the first car is also spreading to other cars in the train, however being diluted along the way. The gradient measurements were performed in different days (different locomotives), which constitutes a limitation for the quantification and comparison of concentrations at each position. This exposure study was carried out in relatively old diesel trains from the 1980s and it may not be possible to extrapolate the observed results to other diesel trains.

Cha and co-workers have reported a gradient in particle exposure inside electric railcars and noted the importance of interior ventilation for indoor air quality[5]. For the trains in our study, ventilation systems differ with season. In addition, there are two types of first passenger car in the double deck rolling stock, as well as two types of diesel fuel (winter and summer diesel). We did not have

421 enough information regarding ventilation to assess the impact on concentration levels, however
422 during the 49 monitoring days on diesel scenarios we covered both ventilation systems, double
423 deck cars and fuel type, and we always observed higher personal concentrations inside the
424 passenger car for diesel compared to electric trains.
425
426 We observed a tendency of increased NO_2 concentrations in the end of the study, during winter, as
427 might be expected[23, 24], however we cannot conclude about any seasonal differences in relation to
428 other parameters. The particle number concentration also appeared to increase in the colder
429 months, but the contribution of the specific engine in use might be more important. As the trains
430 move as far as 114 km from Copenhagen, local precipitation and wind information have also been
431 difficult to investigate.
432
433 The aldehyde concentrations inside the trains were very low; however, the mean concentrations
434 were slightly elevated in diesel compared with electric trains. The formaldehyde concentrations
435 inside the trains were lower than the average personal exposure found among the general
436 population in Sweden (19 $\mu g/m^3$)[25], and lower than average concentrations found in Danish
437 dwellings (20 $\mu g/m^3$)[26].
438
439 The metal and metalloid elemental composition of the PM collected inside diesel trains showed
440 enrichment especially in iron, with other major elements being zinc, magnesium and copper.
441 Although with higher concentrations, the pattern is similar to the standard reference material 2975,
442 (Table S6) and also in comparison with other DE particulates[27]. The presence of silver was likely
443 attributed to the electrostatic sampler silver-coating[28] .
444

The ME-locomotives will be replaced with new electric locomotives in 2021, according to a public announcement[29], nevertheless abatement actions should be put on place until then.

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Supporting information

Measurement details; detailed output of NanoScan SMPS; UFP measured with NT; detailed results from PAHs; detailed results from elemental analysis; and details of gradient and underground measurements. This information is available free of charge via the Internet at http://pubs.acs.org.

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