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Evaluating exposure of pedestrians to airborne contaminants associated with non-potable water use for pavement cleaning

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Abstract Climate change and increasing demography press local authorities to look after affordable water resources and replacement of drinking water for city necessities like street and pavement cleaning by more available raw water. Though, the substitution of drinking by non-drinking resources demands the evaluation of sanitary hazards. This article aims therefore to evaluate the contribution of cleaning water to the overall exposure of city dwellers in case of wet pavement cleaning using crossed physical, chemical and biological approaches. The result of tracer experiments with fluorescein show that liquid water content of the cleaning aerosol produced is about 0.24 g m^{-3}, rending possible a fast estimation of exposure levels. In situ analysis of the aerosol particles indicates a significant increase in particle number concentration and particle diameter, though without change in particle composition. The conventional bacterial analysis using total coliforms as tracer suggests that an important part of the contamination is issued from the pavement. The qPCR results show a more than 20-fold increase of background genome concentration for *Escherichia coli* and 10-fold increase for *Enterococcus* but a negligible contribution of the cleaning water. The fluorescence analysis of the cleaning aerosol confirms the above findings identifying pavement surface as the major contributor to aerosol organic load. The physical, chemical and microbiological approaches used make it possible to describe accurately the cleaning bioaerosol and to identify the existence of significantly higher levels of all parameters studied during the wet pavement cleaning. Though, the low level of contamination and the very short time of passage of pedestrian in the zone do not suggest a significant risk for the city dwellers. As the cleaning workers remain much longer in the impacted area, more attention should be paid to their chronic exposure.

Keywords Street cleaning · Bioaerosol · Non-potable water uses · Airborne particles · Pathogens · Urban atmosphere · Paris

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Background

The city of Paris is one of the scarce cities in France owning a double water supply network. Besides drinking water network, it possesses a second one for the distribution of river water, the non-potable water network (RENP). This network, constructed at the end of the nineteenth century, is gravitational and uses principally water from the river Ourcq and the river Seine. It supplies in average 180,000 m^3 a day of roughly screened river water. As the water distributed is used exclusively for non-drinking purposes of the municipality: street and pavement cleaning, sewer flushing, and watering of green surfaces (APUR 2010) and does not correspond to reclaimed

water, it does not have to comply to any specific water quality standard.

On one hand, the use of non-potable water in urban areas can improve its ecological footprint by avoiding unnecessary water treatment and may mitigate the impact of climate change by diversifying its water supplies. But on other hand, it could bear health risks for city dwellers as water used in cleaning operations, generate water droplets, transporting pathogens and other contaminants issued from the cleaning water or issued from the pavement after resuspension. This issue has been addressed recently in the case of health risk assessment associated with crop or golf course irrigation with reclaimed water (Petterson and Ashbolt 2002; Salgot et al. 2012; Chen et al. 2013).

Bioaerosols and airborne particles have received much attention in indoor environment such as hospitals, public transport, food industry, etc. (Li and Hou 2003; Burfoot et al. 2003; Zhang and Li 2012) and in some outdoor environments such as organic waste treatment facilities (Pankhurst et al. 2011; Betelli et al. 2013) or wastewater treatment plants (Carducci et al. 2000; Upadhyay et al. 2013). However, poor attention has been paid to the impact of urban street and pavement cleaning operations on the quality (increase of particle number and composition change) of the local atmosphere. Amato et al. (Amato et al. 2010a) reviewed the effectiveness of street sweeping, as a method to control atmospheric particle matter (PM) in the city (Amato et al. 2010a, b). Their broad literature review focus principally on reduction of PM fractions and evaluation of the road dust removal. Very little information is given about transfer or microbiological risks. The work of Burfoot et al. (Burfoot et al. 2003; Burfoot and Middleton 2009) gives more details about dry cleaning process suggesting PM numbers as high as 10^7 m^{-3} and resuspension of particles up to tenth of microns.

The aim of this work was therefore to study in situ the levels of pedestrian exposure to aerosol generated during wet pavement cleaning and to define the contribution of non-potable process water to the overall exposure. Due to methodological constraints like short duration of cleaning events and small sample volume, the approach is here rather semi-quantitative, combining physical, chemical and biological tracers, based on non-destructive analysis like fluorescence, bacteria counting and particle observation.

Methodology

Field experiments

After preliminary tests on the university campus, the study of urban aerosols was conducted in Paris during real-time cleaning action. The Ledru Rollin Street (48° 50′ 58.88″ N, 2° 22′ 28.18″ E) was selected as representative of Paris average pavement. The section was characterized by seven floor high nineteenth century buildings, deserved by 5-m large pavements with trees, parking lanes and presence of shops and restaurants. The section used for measurements is wet cleaned twice a week by the municipal services. The wet cleaning consists of a manual sweeping of the pavement surface using a high-pressure water jet (20 bar). The jet can be concentrated on one point to dislocate the dirt or can be more diffuse to wet the pavement surface. The cleaning operator is progressing along the building sweeping with his jet papers, dead leafs, animal excreta, etc. from the pavement towards the street gully. The principal objective is to remove the coarse pollution from the pavement. The cleaning water is supplied by a small vehicle conducted by a second operator. The vehicle contains between 2 and 3 m^3 of non-potable water withdrawn from the RENP network (Fig. 1).

Five different measurement campaigns were conducted during pavement cleaning operations in the summer month of June and July. The sampling was done always in the morning after 2 days of non-cleaning, if no rain was observed. The tracer and basic microbiology experiments were done in the summer of 2013, while the qPCR and scanning electron microscope (SEM) analysis were done during the summer of 2014 in comparable meteorological conditions.

In outdoor environment such as urban environment with relatively low aerosol densities (Herr et al. 2003), the choice of an appropriate air sampler is of major importance in order to collect a sufficient amount of air to sample enough particles associated for all analysis needed, especially when taking in account the relatively short duration of processes such as pavement cleaning and the heterogeneity of urban space. The collection efficiency of a sampling method for bioaerosols depends on the cut-off particle diameter collection efficiency. Most portable impactors are used for indoor-contaminated environments and show cut-off sizes above 1 μm, meaning that the collection efficiency of single bacteria is not complete (An et al. 2004; Yao et al. 2009). Previous tests showed that sampling capacity of 5 m^3 h^{-1} is not sufficient in the above described experimental conditions. There exist only two marketed air samplers Coriolis (Bertin Technologies) and SASS (Micro Bio Detection) which are portable and able to collect more than 5 m^3 h^{-1} in a liquid medium (liquid impingement). The first one is much more affordable than second and that why it was used in this study.

The air contamination assessment was thus carried out using the Coriolis, high-volume air sampler equipped with cyclone-type device for collection and identification of airborne particles. In the sampler, the air is drawn into a conical vial of 30 mL filled up to 15 mL with collection liquid, in a whirling-type motion using suction. The particles are pulled against a wall by centrifugal force, separated from the air, and ultimately collected in the liquid medium (Carvalho et al. 2008). Sampling was carried out under controlled air flow rate

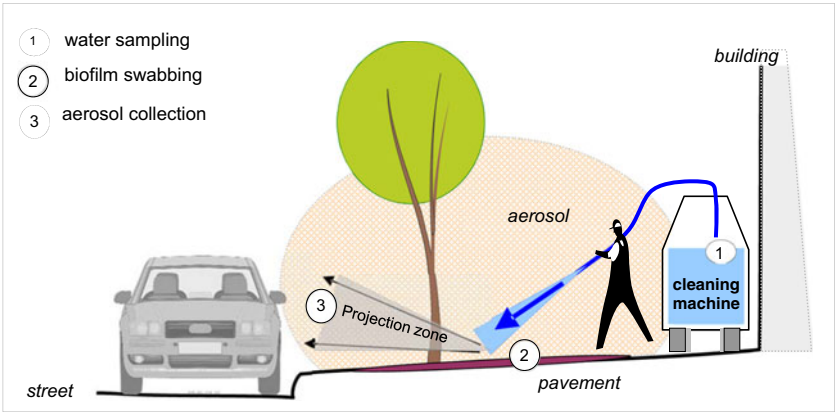

Fig. 1 Schematic representation of the cleaning process with the indication of sampling points

(300 L min^{-1}) during 3 min with 15 mL (initial volume) sterilized distilled water as collection liquid. The initial volume has to be corrected for the evaporation loss during the sampling. The sampling was repeated as often as possible.

To estimate the maximum exposure, the sampler was placed at 80 cm above the pavement level, corresponding to an average level of respiration of a child in pushchair, approximately 5 m in front of the zone of impact. The sampler was moved in front the projection zone as the cleaning machine progressed. Between three and five replicates were executed during the cleaning action of 30 min. One reference sampling (no cleaning) was done before the cleaning and if possible half hour after the cleaning at the same location and same sampler set-up.

The water used for cleaning was collected in a sterilized bottle before and after the cleaning directly from the jet point. Before the cleaning, surface samples were collected from the pavement to be cleaned. For more detail, see the microbiology section. After the sampling, the collection cones, the water samples and the contact petri dishes were transported in an icebox for analysis to the laboratory. The microbiological analyses were done the same day, the physicochemical analysis within 5 days and the particle analysis within 30 days after collection.

Fluorescein tracing

Two distinct tracer experiments were conducted. The first one has as objective the determination of the transfer coefficient between the cleaning water and the aerosol generated during pavement cleaning (liquid transfer coefficient (LTC)). For this purpose, the water of the cleaning engine was coloured with fluorescein (2.44 mg L^{-1}, sodium salt, Merck). Thereafter, samples of the aerosol were collected during a regular cleaning procedure as described above. To correct the values for eventual photodegradation, two bottles were filled with coloured cleaning water: one was exposed to light during the cleaning action and the second kept in dark aside. The samples were transported to laboratory in an icebox and kept in the dark prior to analysis the following day. The analysis was done using a JASCO FP-8300 spectrofluorometer (more details below) at the excitation wavelength of 490 nm (Agilent technologies 2014) and registration of emission at 512 nm.

The second tracer experiment was conducted to estimate the transfer between pavement and the cleaning aerosol. For this purpose, a stroke of 70 m of the pavement was sprayed with a fluorescein solution of 1.04 g L^{-1} using à small garden herbicide sprayer. The average surface charge obtained was 57±2.4 mg m^{-2}. After drying, the surface was cleaned in the usual manner by the municipality services. The aerosol collection and analysis were done as mentioned above. The surface transfer coefficient obtained is called STC.

The concentration of a component in the aerosol ($C_{aerosol}$) can be expressed as the sum of the contributions of the three compartments to the total concentration of this component: ambient air or background (C_{air}), cleaning water (C_{water}) and cleaned surface ($C_{surface}$):

$$C_{aerosol} = C_{air} + C_{water} + C_{pavement}$$
$$C_{water} = L\ T\ C \times C_L$$
$$C_{surface} = S\ T\ C \times C_A$$

For each physical, chemical and biological indicator, the aerosol concentration and the ambient air concentrations are directly measured, while the contribution of the cleaning water is dependent of the liquid concentration of the parameter measured, C_L. The same is true for the pavement contribution and the surface concentration C_A. The general equation using the transfer coefficients can then be applied to estimate the contribution of a specific compartment and to elaborate a pseudo balance.

Particle analysis

Particle size distribution measurements

An aerodynamic particle spectrometer (APS, TSI model 3321) was used to measure the time-resolved particle number,

size distribution in the range of 0.5–20 μm. The APS sampling pump flow rate was 5 L min^{-1}, including 4 L min^{-1} for sheath flow and 1 L min^{-1} for sample flow. Data were recorded at 1-s logging interval. To measure the particles generated by wet-pavement cleaning, the APS was placed 2 to 5 m away from the impact point of the water jet and at 80 cm above the pavement.

SEM analysis

The airborne particles collected with the Coriolis sampler during the last sampling campaign (July 2014) were analysed by SEM. Of the sample liquid, 1500 μL were filtered under slight vacuum on polycarbonate membrane (Nuclepore) of 25 mm diameter with a pore size of 0.2 μm.

The membranes were air-dried in laminar flow hood providing aseptic work area and then coated with a uniform platinum film by vacuum evaporation. The samples were observed and analysed by scanning electron microscopy (SEM Jeol6301F) fitted with an X-ray energy-dispersive spectrometer (Silicon Drift X-Max 80 mm^2 Detector and Aztec Advanced-INCA350 analyser, Oxford Instruments). The enumeration and X-ray analysis were realized at a magnification of×750 on two random transects, corresponding each to five successive images of 160×130 μm.

Fluorescence analysis

Naturally fluorescent organic matter like humic acids and some amino acids (Zsolnay et al. 1999; Nguyen and Hur 2011) were used as non-specific chemical tracers. Fluorescence measurements were made using a JASCO FP-8300 spectrofluorometer (Jasco analytical instruments) equipped with a 150-W xenon lamp. Slit widths for both excitation and emission monochromators were set to 5 nm, and a 0.1-s integration time was used. Analyses were done in a 1-cm quartz cuvette at temperature of 20 °C. The excitation-emission matrix (EEM) fluorescence was obtained by collecting a series of emission scans at 5-nm intervals between 250 and 600 nm for excitation wavelengths between 200 to 450 nm with 5-nm intervals.

UV-visible absorbance spectra were collected prior to the fluorescence scans using a double-beam Lambda-850 spectrophotometer (Perkin Elmer, Waltham, MA, USA) in a 1-cm quartz cuvette over the wavelength range of 190–750 nm with (sterilized) ultrapure water as the reference. Samples with optical densities greater than 0.4 at 240 nm were diluted prior to EEM fluorescence analysis. Spectral corrections for primary and secondary inner filter effects were made using the absorbance spectra (Lawaetz 2009). Raman scattering was mitigated by subtracting a blank EEM spectrum collected on (sterilized) ultrapure water from each corrected EEM. Rayleigh-Tyndall scattering effects were eliminated by differentiated spectra acquisition.

To compare the spectra, three commonly used fluorescence indexes were used: humification index (HIX), biological index (BIX) and fluorescence index (FI). The HIX (Zsolnay et al. 1999) corresponds to a ratio of the area under the emission spectra over 435–480 nm to that over 300–345 nm, obtained at excitation wavelength of 255 nm. The BIX is calculated for excitation wavelength of 310 nm, by dividing the fluorescence intensity emitted at emission wavelength of 380 nm, by the fluorescence intensity emitted at emission wavelength of 430 nm (Huguet et al. 2009). The FI is a ratio of the emission intensity at 450 nm to that at 500 nm, at excitation wavelength of 370 nm (McKnight et al. 2001).

The total organic carbon (TOC) content in the cleaning water and in the aerosol were measured according to the French norm NF EN 1484 (AFNOR 1997) using oven-cleaned glassware for sample and analysis.

Microbiological tracers

As biological tracer were used, the faecal indicators total coliforms, *Escherichia coli* and *Enterococcus* mentioned in the drinking water and reclaimed water standards.

The surface contamination was determined by two methods: by collection of microorganisms through simply contact and by swabbing. The first one should give an estimate for the easily remobilizable biological matter, while the second one should give the total amount of bacteria available in the pavement biofilm.

The contact or pad method is a classical approach used in the food industry (Scott et al. 1984) for detection of contaminated surfaces and consist of agar plates (Humeau VRBL ATL contact petri dishes), allowing direct application of the agar against the contaminated surface (10 s with 500 g lest). The total coliforms were determined by counting after 48 h of incubation at 35 °C. The thermo-tolerant coliforms were estimated by counting after 48 h of incubation at 44 °C (Leclercq et al. 2002). Each measurement was done in duplicate.

The swabbing method consists of a wet transfer of the biofilm present on the pavement surface using sterile nylon brush and sterile-distilled water. A representative surface of 50×50 cm were delimited and cleaned by means of repeated unidirectional brushing, washing the nylon brush after each movement in a beaker with 200 mL sterilized distilled water. The surface was cleaned by four perpendicular series to give about 800 mL sample.

The liquid samples (Coriolis, cleaning water and brushed surface water) were screened for total coliforms and *E. coli* using the US EPA-approved Colisure (IDEXX) substrate. Colisure® uses chlorophenol red β-D-galactopyranoside (CPRG) and 4-methyl-umbelliferyl-β-D-glucuronide (MUG) as specific substrate to simultaneously detect total β-

galactosidase-positive bacteria (counted as coliform according to the manufacturer) and β-glucoronidase-positive bacteria (counted as *E. coli*, even if it is well known that some *E. coli* strain does not express this biochemical characteristic. The samples were incubated, after appropriate dilution, during 48 h at 35 °C using micro trays according to Standard Methods for Water Examination 9223B (APHA et al. 2012). The most probable numbers and confidence limits were calculated using Excel according to Olstadt (Olstadt and Schauer 2007).

In 2014, a real-time polymerase chain reaction (qPCR) was used to precise the results obtained by classic methods mentioned above. The details of nucleic acid extraction and primers and probes are as follows:

In each sample, 500 living cells of *Streptococcus thermophilus* were added as internal inhibitor control. The water samples were filtered though 0.45-μm nitrocellulosic filters (Microsart-CN filters, Sartorius). The filters were then placed in 5 mL of sterile water containing 0.9 % of NaCl, vortexed and sonicated 20 min (Sonicator Bransan 8510). After sonication and final vortexing, 5 mL of sample were centrifuged at 13,000 RPM during 20 min, and then, the supernatant was eliminated. The pellet was resuspended in 400 μL of sterile water with 0.9 % NaCl, and nucleic acids were extracted using the Magna Pure Compact extractor (Roche Diagnostics) and the Magna Pure Compact Nucleic Acid isolation kit I (Roche Diagnostics). DNA was eluted in 50 μL of elution buffer (supplied in the kit).

The primers and Taqman® probes were designed to detect: *E. coli* (primers and probes designed in uidA gene adapted from Maheux (Maheux et al. 2011)), *Enteroccoccus* sp. (primers and probes designed tufA gene adapted from Ke (Ke et al. 1999) with Taqman probe) and the control *S. thermophilus* (primers and probes designed in epsA gene). These three sets of primers and probes were internally designed and are not published. *Legionella pneumophila* detection was made using SybrGreen® technology and mip1-A and mip2-A primers (Joly et al. 2006) in the PCR conditions described by the authors.

The Taqman® assays were performed with the Taqman Universal Mastermix 4× (Life Technologies) and performed on a Viia7 instrument (Life Technologies).

Results and discussion

Fluorescein tracing

The laboratory tests and in situ measurements with fluorescein allowed to determine LTC and its more known expression, the liquid water content (LWC). The LWC for the pavement cleaning bioaerosols gave an average of 0.24 g m^{-3}. The relation between LTC and LWC is the density; LTC is in millilitre of water per cubic metre of air, and the LWC is in grams of water per cubic metre of air.

The LWC makes it possible to compare the values obtained for the cleaning aerosol, with literature values for other type of liquid suspension in the atmosphere. The values reported in Table 1 show that the aerosol generated is comparable to fine rain or fog in terms of liquid water present in the air.

The transfer coefficients obtained are summarized in Table 2 below. The practical application of LWC is the possibility to estimate directly the probable air concentration by multiplying the water concentration in units per millilitre by LWC to obtain ultimately the air concentration in units per cubic metre of air.

Airborne particles

The aerosol size distribution of particles measured with the APS during our experiments is reported in Fig. 2.

Figure 2a shows the size distribution of the mean particle number concentrations as function of the aerodynamic diameter in the range of 0.5 to 20 μm. The figure shows that the size distributions of the aerosol before and 30 min after the cleaning are similar, and therefore, these two size distributions can be assimilated as an average background level of an urban atmosphere. As expected, the shape of the background aerosol

Table 1 Airborne liquid water contents of different type of aerosols as determined by fluorescein tracing, compared to literature data

Type of aerosol	Generator or reference	Set-up details	LWC (g m^{-3})
Pulverization	5 L herbicide sprayer	Laboratory 1-m distance	11.7
Projection	Karcher pressure washer	Campus, coarse street asphalt[a]	0.16
Projection	Pavement pressure washer	Paris, fine asphalt pavement[a]	0.24
Fog	Aikawa et al. 2006	Rural zone Japan	0.18
Fog	Gonser et al. 2011	Rural zone Taiwan	0.54
Fine rain	Vivekanandan et al. 2001	Colorado	0.15
Fine rain	Aikawa et al. 2006	Taiwan	0.36

[a] Collected at 80 cm above the ground at approximately 5 m from the point of impact

Table 2 Values of transfer coefficient as determined with fluorescein

Coefficient	Average	Unit	STD	*n*
LTC	2.40×10^{-7}	m^3 water per m^3 air	5.4 %	3
STC	1.23×10^{-5}	m^2 pavement per m^3 air	7.5 %	4

distribution is in accordance with previous studies conducted in outdoor urban environment (Whitby et al. 1972). For purpose of comparisons and discussion, the size distributions data were split into three subranges using the particle aerodynamic diameter d_{ae} subrange 1 $0.5<d_{ae}<2.5$ μm, subrange 2 $2.5<d_{ae}<10$ μm and subrange 3 $10<d_{ae}<20$ μm. The subrange 1, subrange 2 and subrange 3 accounted respectively for 99 % (8.38×10^7 m^{-3}), 0.62 % (5.27×10^5 m^{-3}), and 0.01 % (5×10^3 m^{-3}) of the total number of particles. The shape of the particle size distribution of the aerosol during pavement cleaning was comparable to the shape of the background aerosol, but the number concentration of particles during pavement cleaning was significantly higher.

The variation of differential size distribution concentration between background level (N_{bg}) and the concentration measured during the pavement cleaning (N_c) indicates a more important increase of particle concentration in the subrange 1 (with a mode at 1.4 μm) than in the subranges 2 and 3. Considering the time for a generated particle to enter the APS cell measurement, we assume that small particles in the subrange 1 might initially correspond to larger droplet which had sufficient time to evaporate and to turn into dry residues. The latter might stay airborne for long periods. Besides, a significant number of particles in the subrange 3 were detected (Fig. 2b), but such particles are significantly subject to fast settling. As reported in work of Xie et al. (2007) on respiratory diseases, large droplets will not totally evaporate into the air and could function as carriers of pathogens and thus infectious diseases. Only few studies published (Motzkus et al. 2011) treat the emission of fine droplets by liquid falls and jets. Therefore, the evidence of small size particle production during wet pavement cleaning presented in this study may be of major importance for hygiene workers.

The duration of the pollution peak in our experiment was only few minutes, comparable to the observation of Fitz and Bumiller (2000) for street sweeping in California. Far more, the pedestrians remained only few seconds in the contaminated zone lowering even more their exposure, contrary to the cleaning workers.

The origin of solids

Though the particle concentration in the air is underestimated due to imperfect sampling of small size particles by the Coriolis sampler (Langer et al. 2012), due to dissolution of salts and due to aggregation of particles during the impact in the liquid medium, the SEM analysis makes it possible to quantify the difference between background levels and the cleaning atmosphere. The number of particles sampled under the same experimental conditions is significantly higher for the cleaning atmosphere than for the background. This is the case for direct particle counting as well for total particle estimation calculated from surface and imaging software.

The distribution obtained from the SEM particle counting (Fig. 3) was corrected for under sampling according to Carvalho et al. (2008), who reported the Coriolis air sampler to have effective physical sampling efficiencies of 40, 84 and 92 % for respectively 1, 4.6 and 10 μm mass mean aerodynamic diameter.

The SEM observations confirm the results of APS, a substantial increase in particle number and particle diameter during the cleaning. The mean Feret diameter of particles rose from 2.8 to 5.0 μm while the particle number concentration rose from 3.3 to 4.5 million per cubic metre. As expected, these numbers are some lower than the APS due to the sampling bias.

The analysis made by Energy Dispersive X-ray Spectroscopy (SEM) shows that the background air samples and cleaning aerosol samples present great similarities in the characteristics of the particles they contain. In both cases can be observed a complex mixture of particles coming from various sources: (i) numerous isolated particles which size

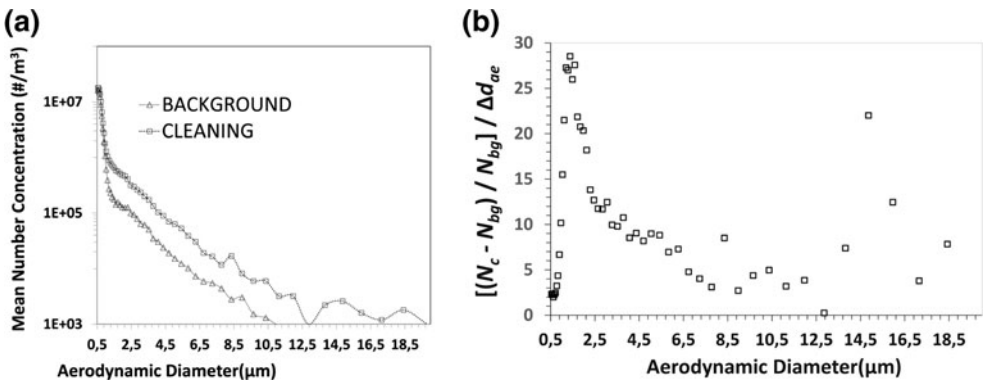

Fig. 2 The effect of wet pavement cleaning on the size distribution of the cleaning aerosol (**a**). Differential size distribution between cleaning and the background atmosphere (**b**). The latter is corrected for the difference in class size of the APS measurement

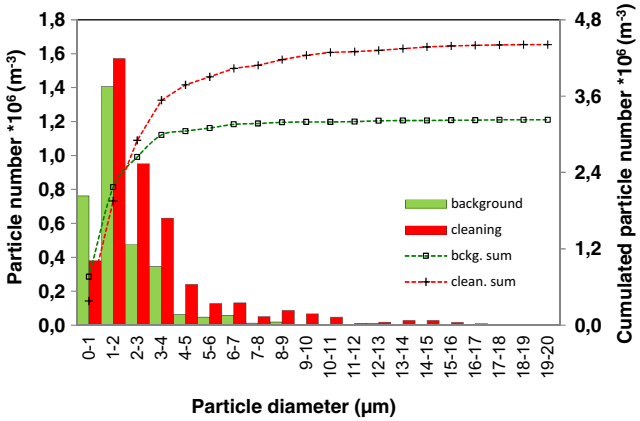

Fig. 3 Particle number distribution during the cleaning operation compared to the background level, corrected for under-sampling according to Carvalho et al. (2008). The distribution is based on 293 particles for the background level and on 447 particles for the cleaning run. The lower detection limit of SEM observation corresponds to a diameter of 0.5 µm. Remaining 2 % of particles, greater than 20 µm, are not shown

distribution extends from some nanometres to several tens of micrometres, (ii) numerous aggregates containing the particles previously quoted, (iii) isolated textile fibres and (iv) isolated biological particles of different origin. The first category is represented principally by minerals like silico-aluminates, feldspaths, quartz and carbonates issued from the erosion of urban surfaces. A relatively important part of particles contains iron and in less extend heavy metals issued from the traffic pollution. No specific tracer was identified being able to distinguish between the water, air or pavement components. As unique potential tracer for the cleaning water were identified the diatomea. As their structure is fragile and their initial number is low, they were not observed in the cleaning aerosol. No potentially present bacteria were identified, most probably due to their low numbers.

The principal qualitative difference between the compartments is the predominance of the calcium containing particles in the cleaning aerosol (Fig. 4). A more detailed work of the particles observed will be presented in short (Seidl et al. 2015).

Bacteriology

Figure 5 relates the stock of tracer bacteria present in the compartments involved in the cleaning aerosol production. The numbers are not easy to compare between them as they are expressed according to the volume or surface. The level of environmental surface and air contamination is difficult to evaluate as the literature data cover principally health care environments. The level of contamination of water used for cleaning can be defined as low. According to the water quality criteria defined by the French water agencies (MEDDE 2009), the water quality is classified as good or acceptable.

As the cleaning aerosol composition will depend on the existing air pollution, the resuspended pavement pollution and the cleaning water contamination, each of the compartments was assessed separately. As mentioned in the methodology, the average surface contamination was determined by two methods: simply contact and swabbing. We observe a fair linear correlation (>0.7) between the levels obtained for both methods as well for the total coliforms as for the *E. coli*. The Spearman correlation is only significant ($p>0.05$) for the total coliform data set of 2014. The weak slope or lack of strong correlation indicates that the amount of easily available (washable) coliforms will be almost independent of the total amount of coliforms present on the pavement surface (Fig. 5b).

Utilization of the LTC and STC ratio calculated previously can help to make an estimation for the aerosol contamination. We obtain, using the maximum surface load of Fig. 5, an aerosol concentration of total coliforms of 4 UFC m^{-3} (50 percentile interval between 2 and 24) instead of 12 UFC m^{-3} measured (50 percentile interval between 3 and 36) (Fig. 5). On one hand, the right order of magnitude of these concentrations validates our approach. On the other hand, this value indicates that the pavement contribution is probably underestimated, even using the maximum stock present. The maximum stock was measured on representative homogenous surface, though excluding birds and dog excreta. The presence of several points marked with animal faeces in the sample area could explain this difference as canine faeces are known to contain more than 10^8 coliforms a gram (Neidhardt 1996; Wright et al. 2009). As the pavement cleaning dislocate partly the present faeces, the average surface concentrations should be higher. If we take into account the absence of indicator organism in the ambient air and the measured water concentration, the overall pavement surface contribution would be about 67 %.

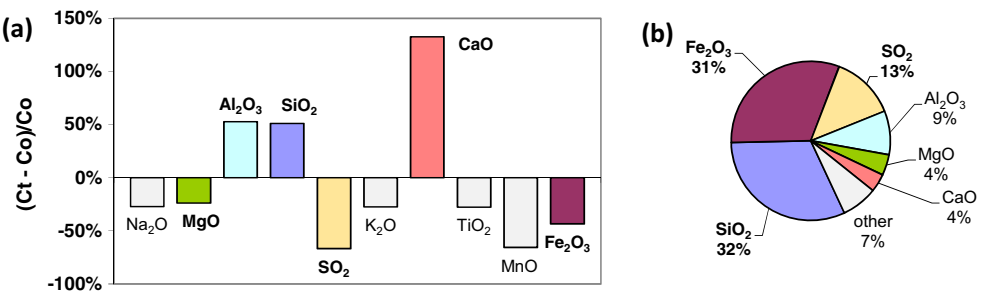

Fig. 4 **a** Difference between the background and exposure, expressed as percentage change of the background composition. **b** Average composition of the background particles

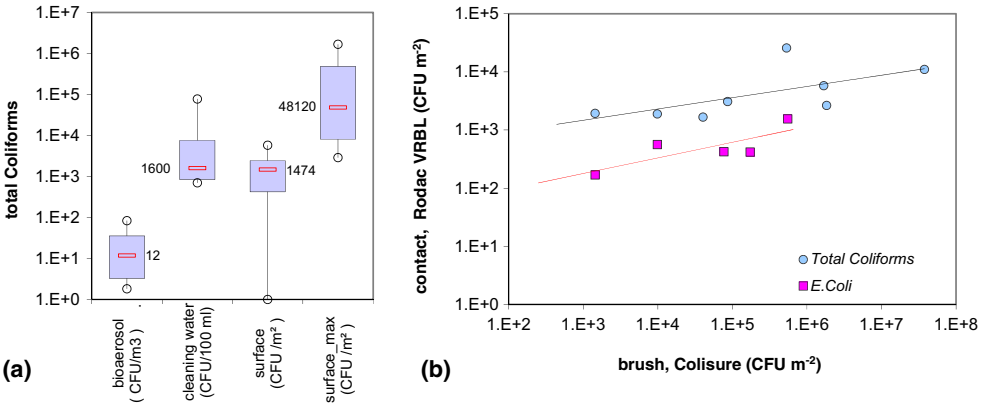

Fig. 5 **a** Boxplot (minimum, 25 %, median, 75 % maximum) of coliform contamination of the compartments involved in wet pavement cleaning resulting from three campaigns in the summer 2013 and 2014 in the avenue Ledru Rollin. The data for ambient air are zero and are therefore not shown. **b** Relation between the two sampling methods used for the pavement surface

Contrary to total coliforms, *E. coli* does not follow the model established above, which can mean that the approach is not adaptable to other bacterial tracers or that the methodology applied for *E. coli* is not adapted. An et al. (2004) et Langer et al. (2012) observed for bacteria a decrease to below 10 % when using Coreolis sampler in an outdoor environment and hypothesize that the test sampler's underperformance compared with the BioSampler (SKC) used as standard may be caused by the damage to sensitive microorganisms during the collection process, by sampler's wind sensitivity or by particle aggregation. Utilization of the qPCR method may help to overcome this effect.

The qPCR method (Fig. 6) shows presence of *E. coli* genomes in cleaning water and in ambient air contrary to the Fig. 5. No presence of *L. pneumophila* was detected. Using the background values, the LTC and the water concentration, we can estimate that pavement surface contribute over more than 90 % to the cleaning aerosol contamination ("Pies" in Fig. 6). These levels confirm the order of magnitude obtained with total coliform bacteria as tracer.

Even if the background concentration of genomes of pathogen organisms may increase more than ten times during the wet pavement cleaning, the short exposure time (an average of 7 s was observed) in the cleaning zone and the level of indicator organism measured in the aerosol, the risk of pathogenic bacteria inhalation for a randomly pedestrian, defined as product of concentration and respirated volume, appears relatively low, even if corrected for the Coriolis sampling inefficiency. Though, it must be taken into account that the cleaning operators are much more exposed to the contamination and that chronic exposure to low pathogens concentration can lead to health effects. Utilization of faecal bacteria genomes as an indicator of the origin of the contamination can be a useable tool for a quick analysis, but must be more assessed.

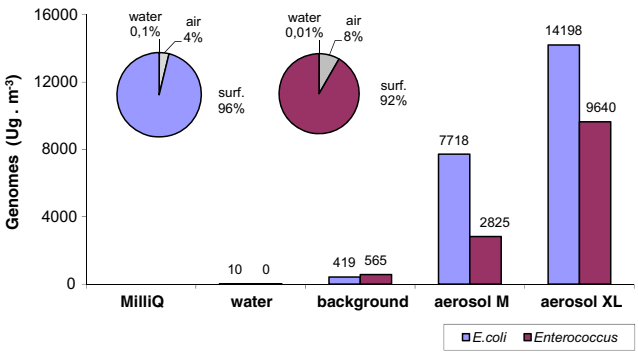

Fig. 6 Histogram showing the aerosol contamination as determined during the campaign of 2014 by qPCR. The values for cleaning water and MilliQ water are in Ug mL^{-1}. *Aerosol M* medium contaminated pavement, *aerosol XL* highly contaminated pavement. The *pies* show the contribution of each compartment to the total load during pavement cleaning. The water data represent the theoretical water contribution as calculated by LWC

Organic matter

Figure 7 shows an example of 3D emission spectra of (a) water used for the street cleaning, (b) ambient air and (c) aerosol generated during wet cleaning. All samples were collected under the same conditions, and the showed spectra should be considered as representative of the three campaigns executed.

The average TOC concentration of the cleaning water of the Ledru Rollin water network, as measured during 2014 at five different points by the water distribution company EDP, was 2.22 mg C L^{-1} (STD=0.56 mg C L^{-1} for n=49). The TOC content of ambient air liquor (Fig. 7b) was 1.41 mg C L^{-1} and that of cleaning atmosphere (Fig. 7c) was 0.58 mg C L^{-1}. If the concentrations of the liquor are recalculated for carbon concentrations in the air, we obtain 4.51 and 7.18 mg C m^{-3}, for respectively the background and the cleaning atmosphere, the cleaning atmosphere being 60 % higher than the background level. These concentrations are close to the value of 12 mg C m^{-3}, based on average composition and concentration of PM10 particles observed in the Paris region (Airparif 2012).

The 3D spectra (b) and (c) in the Fig. 7 show that the cleaning atmosphere contains more dissolved organic components than the ambient air. If we compare the contour plots of cleaning water and that of cleaning aerosol, some similitude appears like the position of maxima and the form of the plot.

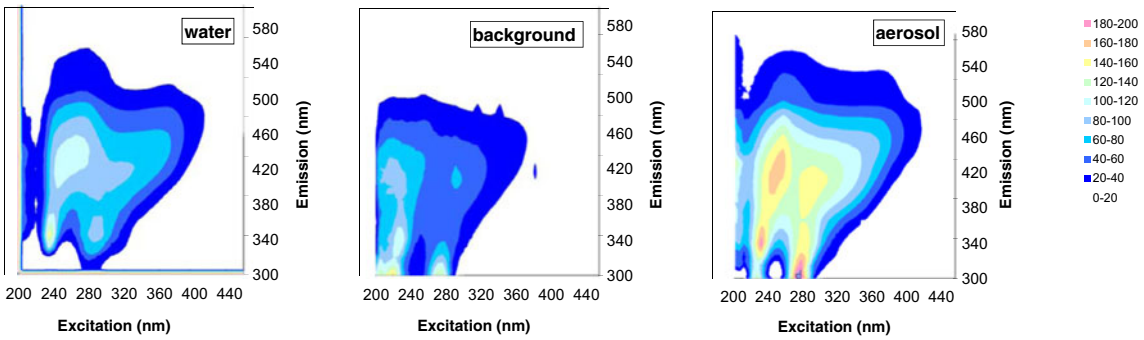

Fig. 7 Carbon normalized (1 mg TOC L^{-1}), 3D emission spectra for the sampling campaign of 26 June 2014. **a** Water used for street cleaning, **b** ambient air and **c** wet cleaning atmosphere (scale reduced by one third)

The cleaning aerosol shows a peak at 235 EX/340 EM, corresponding to the maximum of cleaning water and reinforced peak at 240 EX/440 EM. The first one is most probably due to biological substances like tryptophan, present in the river water used for cleaning (Fellman et al. 2010). The second maximum corresponds to the A/C region of Coble system (Coble et al. 1993; Coble 1996) representing humic-like terrestrial organic matter. It appears reinforced in the aerosol, most probably due to organic matter issued from fragments of tree leaves, being resuspended during the cleaning action. The spectra of the cleaning atmosphere (Fig. 7c) indicates a contribution of cleaning water to the aerosol. Though due to the important dilution factor (>10 times) and the important contribution of the cleaned surface to the aerosol composition, discussed previously, the "watermark" will not be very pronounced.

It has to be stressed that in our case, we study the overall organic composition of an artificial particle solution and not of an existing water sample (with exception of cleaning water). That's why our spectra have different shapes compared to spectra found in natural waters by other authors (Huguet et al. 2010; Valencia et al. 2014) and are closer to the findings of Birdwell and Valsaraj (2010) concerning fog water.

From the three indexes commonly used in the bibliography HIX (Zsolnay et al. 1999), FI (McKnight et al. 2001) and BIX (Huguet et al. 2009), only HIX appeared to be able to distinguish between the principal components of the cleaning aerosol: water, air and surface. A low BIX (below 1) would indicate a low presence of fresh biological material and a higher HIX rather old organic material like wood. Only the latter is showing significant differences between the cleaning aerosol and the ambient air (Table 3). The potable water, corresponding to raw river water should not contribute significantly to the increase of HIX, especially if we take into account the liquid water content of about 0.24 gm^{-3} (see paragraph 3.1).

Using the parameters H and L of HIX and the liquid water content of cleaning aerosol determined previously, we can estimate that the contribution of cleaning water to the overall load would be less than 5 % and that of cleaned surface would be preponderant. Using the total integrated fluorescence intensity of the EEM spectrum, we obtain a pavement surface contribution of 84 %. The contribution of the background pollution would be 14 %, while that of water only 2 %. These values corroborate with the findings obtained from other parameters studied.

Conclusions

The two transfer coefficients make it possible to estimate the contribution of cleaning water and cleaned surface to the aerosol generated, assuming that the substance followed behave in the same way as the fluorescein tracer used. For the soluble organic matter and the liquid—air transfer, this hypothesis appears to be right, though for the particulate matter including bacteria, this coefficient should be used with more precaution.

In situ particle analysis indicates a significant increase in particle number and particle diameter, though with the same

Table 3 Humic indexes for sterilized pure water (18 MΩ), cleaning water, background atmosphere Coreolis extract and cleaning aerosol Coreolis extract

	Beta	Alpha	BIX	H	L	HIX	470	520	FI
Pure water	8.87	6.55	1.35	580	7776	0.075	2.88	1.34	2.15
Cleaning water	71.3	76.9	0.928	4576	145	31.5	48.7	18.1	2.69
Ambient atmosphere	39.3	48.1	0.817	2468	1955	1.26	18.0	6.84	2.63
Aerosol	298	389	0.768	21,615	7748	2.79	200	73.6	2.71

The values are normalized for 1 mg TOC L^{-1}

chemical composition. The classical bacterial analysis using total coliforms as tracer suggests that an important part of the contamination is issued from the pavement. qPCR confirms the negligible contribution of the cleaning water and shows more over a significant increase of the background concentration (26 times for *E. coli* and 11 times for *Enterococcus*). No *L. pneumophila* was detected.

The three types of analysis, physical, chemical and biological identified the existence of significantly higher levels of all parameters studied during the wet pavement cleaning, compared to the background concentrations. Moreover, the analyses showed that the contribution of non-potable water used for cleaning, to the total pollutant charge of the cleaning aerosol is lower than the background level and that the contribution of the pavement surface to the cleaning aerosol contamination, is predominant. However, in the case of specific bacterial contamination of water like *Legionella*, water contribution could be more important.

The effect of wet pavement cleaning shows a rather low risk in terms of pedestrian exposure principally due to very short stay of pedestrian in the contaminated zone and the relatively low levels of aerosol contamination. As the duration of exposure of cleaning workers is much longer, more attention should be paid to their working atmosphere.

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