Intra- and inter-site variability of soil contamination in road shoulders – Implications for maintenance operations

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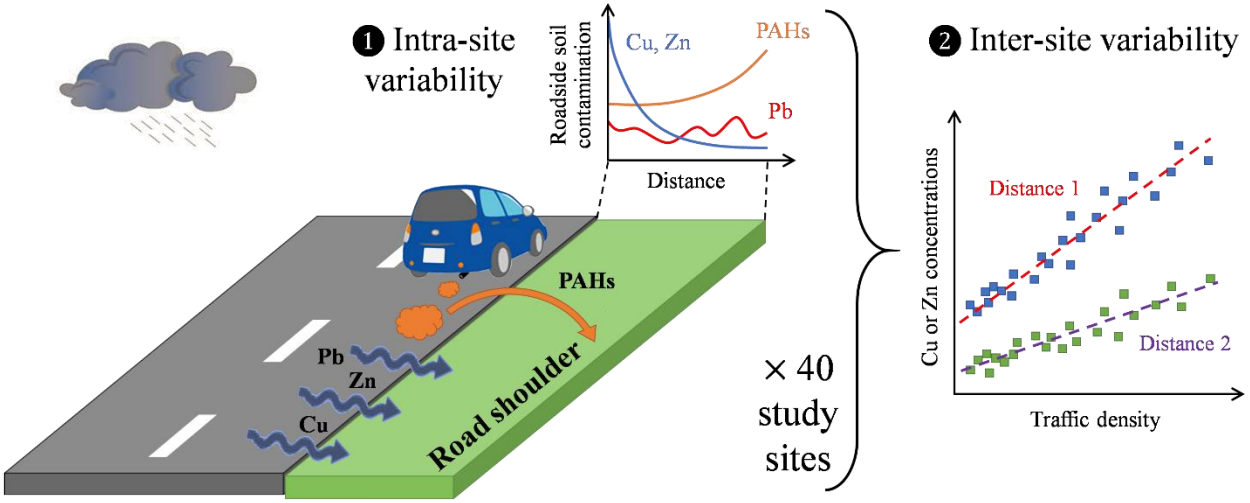
ABSTRACT

This study addresses soil contamination in vegetated road shoulders with diffuse inflow of runoff. It aims (i) to characterize the spatial distribution of three metals (copper, lead, zinc) and PAHs, and (ii) to identify influencing factors for the inter-site differences. An extensive sampling campaign was carried out on forty road segments in the Paris region, targeting various distances and soil depths. Copper and zinc contamination was found to be mainly restricted to the first 30 cm from the road, where their concentrations ranged respectively between 40-270 $mg.kg^{-1}$ and 150-950 $mg.kg^{-1}$ (with a few additional extreme values related to the presence of a galvanized steel guardrail). Lead contamination was moderate (< 100 $mg.kg^{-1}$ in more than 75% of the sites) and relatively uniform across the width of the shoulders. Conversely, highest PAH concentrations were found further from the road. These differences between contaminants were likely due to the varying importance of wet weather-related processes and atmospheric transport in their dispersion mechanisms. Copper and zinc concentrations correlated well with the traffic density, which alone explained $\geq 69\%$ of the inter-site variability, whereas lead and PAHs did not exhibit such dependence. Soil organic matter was found to control the "ceiling" concentration of metals, thus limiting the maximum amounts that can be intercepted from road runoff. These results illustrate the feasibility of estimating contamination levels on road shoulders, and thus offer interesting perspectives for better consideration of the infrastructures' maintenance needs and improved routine operations.

KEYWORDS

Metals, Polycyclic Aromatic Hydrocarbons (PAHs), Road runoff, Roadside soil, Stormwater management, Traffic

GRAPHICAL ABSTRACT



1 INTRODUCTION

In road environments, especially medium-sized roads, stormwater management is generally achieved *via* a roadside ditch, connected to detention/infiltration basins or other structural Best Management Practices. However, where the road shoulder – *i.e.*, the space between the roadway and the ditch – is not covered with asphalt (Fig. 1), it is subject to the progressive formation of a humus and sediment layer, due to vegetation growth and suspended solid accumulation (Tedoldi *et al.*, 2018). This layer eventually constitutes an obstacle that compromises the diffuse evacuation of road runoff, and may cause safety hazards during wet weather (flooding of the road, water ponding at low points). Thus, a regular maintenance of the shoulder – mostly consisting of surface soil scraping – is required in order to preserve the infrastructure's hydraulic functions. These operations generate large amounts of scraped soil that need to be subsequently managed.



Fig. 1 – Typical French configuration of a vegetated road shoulder followed by a longitudinal ditch.

On the other hand, it has been widely documented that road traffic generates ubiquitous and persistent compounds such as trace metals or polycyclic aromatic hydrocarbons (PAHs), which accumulate durably in roadside soils as a result of runoff spreading and infiltration, and atmospheric deposition (Dierkes and Geiger, 1999). This pervasive contamination brings about several questions regarding the specific requirements for the disposal, treatment or direct reuse of the scraped materials during maintenance operations. This requires addressing (i) the spatial extent of the contaminated area within each road shoulder – providing insight into the feasibility and benefit of implementing differentiated management for "contaminated" and "non-contaminated" soil – and (ii) the predictability of the contamination levels from site-specific characteristics.

Despite a relatively abundant literature about roadside soil contamination (Kaur and Katnoria, 2014; Werkenthin *et al.*, 2014), focusing on either metals – the most widely studied ones being copper, lead and

zinc – or PAHs, it is still difficult to identify pragmatic responses to these concerns. First, the intra-site variability of concentrations has rarely been characterized at resolutions fine enough to be of interest regarding the present objective. The number of sampled distances within two meters from the road is often limited to 1 or 2, while the investigated zone reaches up to 10-50 m (Chen *et al*., 2010; Wang *et al*., 2017; Yang *et al*., 1991). When finer sampling grids were used, however, it was evidenced that metal concentrations sharply decrease with increasing distance from the inflow zone (Legret and Pagotto, 2006; Tedoldi *et al*., 2017a), so that their accumulation mostly occurs within less than 3 meters from the road (Steiner *et al*., 2007). Hence, the concentrations measured outside this 3-meter-wide strip are likely to show more uniform distributions (Aljazzar and Kocher, 2016; Jaradat and Momani, 1999) and weaker correlations with the site-specific sources of contamination (Bernardino *et al*., 2019).

Spatial patterns of PAHs, however, do not appear to be as systematic as for metals: while Yang *et al*. (1991) observed a consistently decreasing trend with increasing distance from the road, in other studies, the highest PAH concentrations were not always measured close to the road edge (Dierkes and Geiger, 1999; Marusenko *et al*., 2011; Piguet *et al*., 2008). Kumar and Kothiyal (2012), who analyzed soil samples collected at 1, 2, and 3 m from various roads, even found different patterns depending on the considered compound. For both metals and PAHs, it appears necessary to better characterize the spatial distribution of soil contamination in the first 2-3 meters from the road, so as to identify, where applicable, the area subject to the highest accumulation.

As regards the inter-site variability of roadside soil contamination, many governing factors have been proposed and investigated so far, but there is no consensus over their effects or relative importance. For metals, traffic density and composition were identified as key factors by Assirey *et al*. (2015), Horstmeyer *et al*. (2016) and Yassoglou *et al*. (1987), so was the vehicular speed (De Silva *et al*., 2016), whereas Carrero *et al*. (2013) and Franco (2005) found metal concentrations to be mostly dependent on the road age. Hiller *et al*. (2020) proposed a double linear regression to predict copper, lead and zinc concentrations from 39 crossroads as a function of both annual average daily traffic and road age (R^2 = 0.17-0.32). Traffic fluidity has also been proposed as an additional factor since some metals are released to the environment during braking mechanisms (Davis *et al*., 2001). On one hand, Horstmeyer *et al*. (2016) observed higher contamination on road crossings and transects that witness *stop-and-go* traffic, which was also noted by Pérez *et al*. (2008). On the other hand, factors such as the presence of road signs (Hjortenkrans *et al*., 2006) and the number of traffic lights (Aslam *et al*., 2013) have also been deemed significant. Additionally, several authors suggested that the direction and intensity of the prevailing winds may contribute to this variability by affecting metal dispersion, since they noticed some differences in the soil contents on either side of the road (Alsbou and Al-Khashman, 2018; Jaradat and Momani, 1999).

Inter-site comparisons are somewhat scarcer for PAHs, and their conclusions are non-concordant as well. Kim *et al*. (2019) evidenced a positive correlation between PAHs concentrations in roadside soil and traffic density, however the values fell within a narrow range ($\sum^{16}$ PAHs between 0.3-1.8 mg/kg)

compared to other studies – *cf*. the data synthesized by Marusenko *et al*. (2011). The influence of traffic density was also supported by the results of Kumar and Kothiyal (2012), and to a lesser extent Marusenko *et al*. (2011); in the latter case, the most influent factor was found to be soil organic matter. Conversely, Agarwal (2009) noticed a lack of correlation between total PAH concentrations and the soil's organic carbon, and stated that the variations of concentrations could be explained in terms of vehicular composition (two-wheelers, cars, heavy-duty vehicles), as well as immediate surroundings of the road that modify atmospheric transport mechanisms. Similar assumptions were made by Garcia-Flores *et al*. (2016) to explain their data. Dierkes and Geiger (1999) observed a relationship between PAHs and the age of the road shoulders; in their study, the effects of traffic density appeared to be less marked, but it should be mentioned that all samples were not taken at the same distance from the road.

The above-mentioned divergences could be due to different levels of representativeness of the samples collected from one study to another (*e.g.*, different sampling depths or distances from the road, with single or composite samples). In any case, given the absence of recurring correlation in the available literature, soil contamination does not appear to be systematically predictable. This literature overview emphasizes the interest of combining a fine assessment of the site-specific distribution of metals and PAHs in road shoulders with a large panel of study sites. This may (i) confirm the recurring patterns of concentrations within each site, (ii) help identify accurate descriptors of soil contamination among site-specific characteristics, and (iii) provide insight into its consequences in terms of soil management. These are the objectives of the present work, for which the French department of *Seine-et-Marne*, in the Paris region, has been retained as a study area, targeting 40 shoulders receiving direct runoff from departmental roads with different characteristics.

2 MATERIAL AND METHODS

2.1 Study sites

Seine-et-Marne is a department of ~6,000 km^2. It is highly urbanized in its western edge which intersects the Paris region, and more rural on the eastern part (Fig. 2). It contains around 4,500 kilometers of roadways managed by the county council. The 40 study sites were spread across all the department's area to encompass most of its soil types and environments: scattered dwellings, open countryside, forest. They were chosen to guarantee a uniform bivariate distribution of two road factors which were deemed relevant and were systematically available from the department's database: the annual average daily traffic (AADT) and the percentage of semi-trucks, which in our panel respectively varied from 900 to 25,000 veh/day and from 2 to 26%. The site characteristics are presented as *Supplementary material*, Table S1.

So as to specifically target the traffic-related sources of contamination, none of the study sites had a metallic guardrail – except two of them which were retained for comparison purposes – nor had them road

signs or street lights along the sampled sections. The latter were chosen in straight and relatively flat areas, which are most likely to induce a diffuse inflow of runoff on the shoulder and limit the influence of the driving style; in particular, the absence of local depression on the road (which could have created a "concentration effect" for runoff) was systematically checked before undertaking soil samplings.

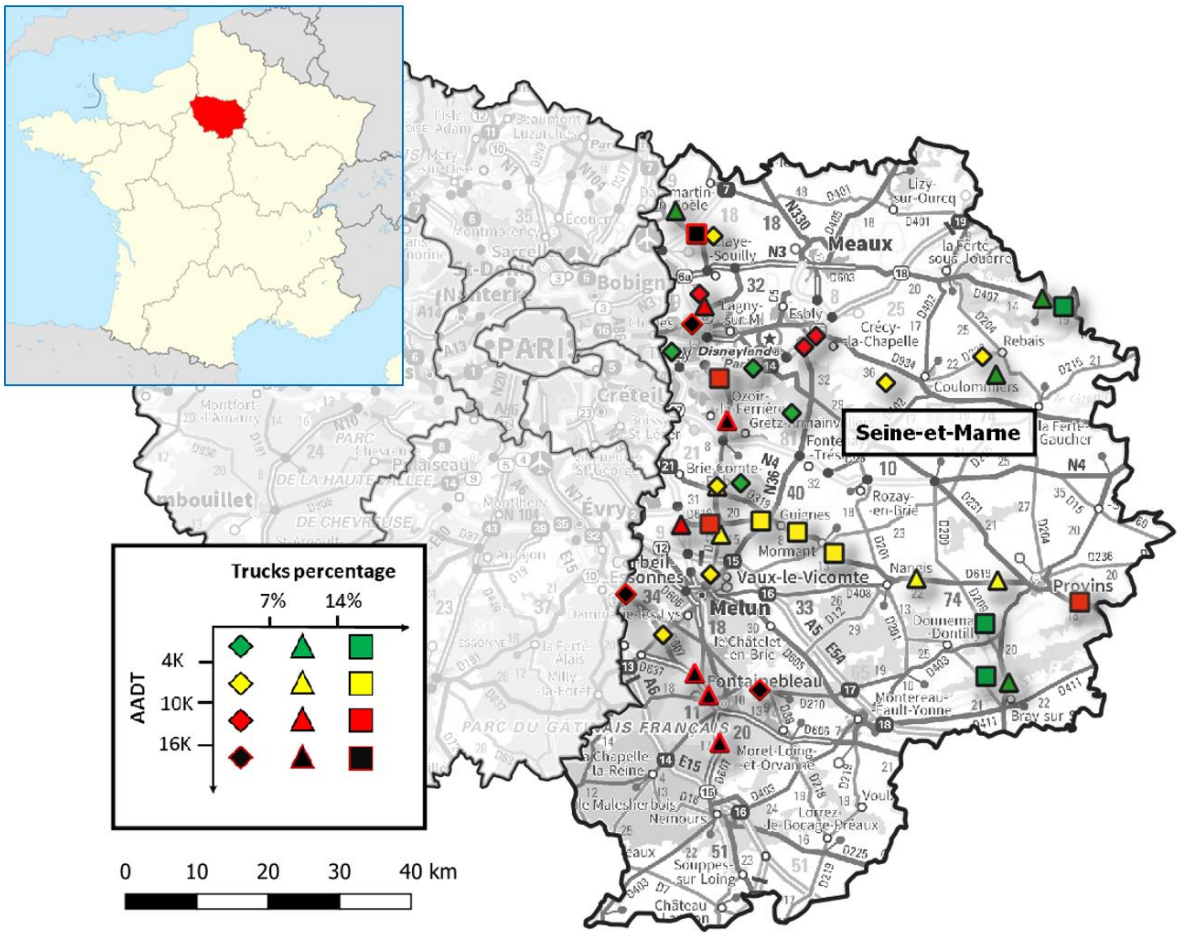


Fig. 2 – Location of the study sites in the Paris region. The colors and shapes indicate respectively the classes of traffic and trucks percentage on the adjacent road.

The width of the road shoulder varied from 1.5 to 3 m – generally the roads with low traffic had narrower shoulders. The road sub-base extended under the shoulder to a certain distance, which varied from a few tens of centimeters to > 1 m depending on the traffic characteristics. During road construction, this sub-base was covered with topsoil from the site to create a flat shoulder; then the growth of spontaneous vegetation, combined with the accumulation of sediment brought by road runoff, induced a pedogenesis at the soil surface. All study sites were uniformly covered with grass (similar to the example shown in Fig. 1), thus vegetation is not expected to contribute to inter-site variability in soil contamination. The soil depth depended on the geometry of the sub-base: it was often ~2 cm close to the road, and increased with increasing distance. The date of the latest maintenance (*i.e.*, soil scraping) was not always known with precision, but the operation time of each retained site was in any case higher than five years, and almost certainly lower that ten years (usual frequency of such operations).

In order to set some meteorological context, the average annual precipitation depth in the Paris region is ~640 mm/year, and the rainfall distribution is dominated by small events: during the period 1993-2008, 60% of the daily rainfall depths were $\leq$ 10 mm, and 80% were $\leq$ 17 mm. A 1-year frequency storm corresponds to 18 mm in 1 h, and 33 mm in 24 h; a 10-year frequency storm corresponds to 32 mm in 1 h, and 61 mm in 24 h (source: *Météo France*).

2.2 Sampling methodology

Field investigations were undertaken between August and December 2018. The sampling methodology was designed to capture the transversal trend of contaminant concentrations, *i.e.*, their distribution with respect to the distance from the road. Soil samples were collected at four to five distances depending on the width of each shoulder (0, 0.3, 0.7, 1.2, and 1.8-3 m whenever possible) with stainless steel sample rings. The sampling depths corresponded to the upper 2 cm, and the underlying layer (2-5 cm) whenever the sub-base was deep enough.

Former assessments evidenced a potential variability of soil concentrations at a given distance from the road (Tedoldi *et al.*, 2017a). In order to mitigate these effects, a mean sample was composited for each investigated distance from 6 different subsamples collected over a 25-m length (Fig. 3); it was then quartered on the field to achieve a 100-g representative sample. The sampling material was cleaned and rinsed twice with ultrapure water between two distances from the road. This procedure led to a maximum of 10 samples in each site (4-5 distances × 1-2 depths).

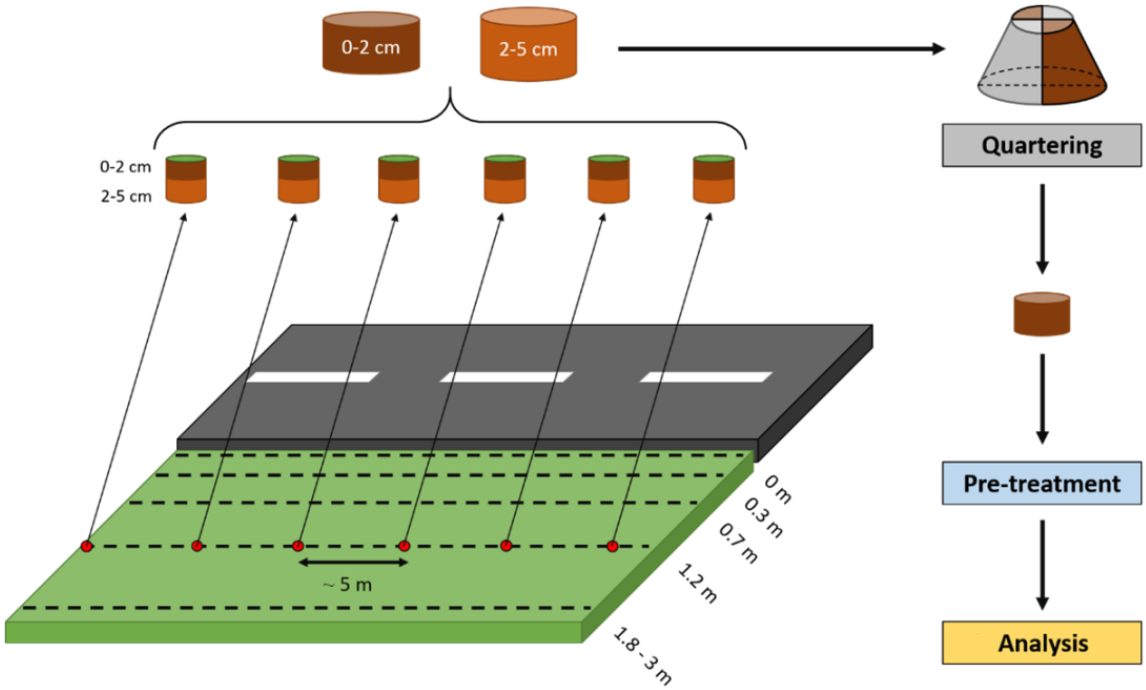


Fig. 3 – Schematic representation of the methodology for soil sampling and analysis (in the case of metals).

All 10 samples could not be analyzed for PAHs due to cost constraints: it was therefore chosen to retain (i) a composite sample formed by merging the subsamples collected at 0 and 0.3 m, and (ii) a sample collected on the opposite side of the shoulder.

2.3 Laboratory analyses

2.3.1 Metals

The soil samples intended for metal analysis were pre-treated according to the international standard ISO 11464 (2006), *i.e.* oven-dried at 40 °C for 7 days, ground with a pestle, then passed through a 2-mm nylon sieve. Elemental analysis was achieved *via* X-ray fluorescence (XRF) spectrometry (*Thermo Scientific*, portable Niton™ analyzer XL3t in a standard soil mode, 60 s per beam). Among the analytical range of the apparatus, copper, lead and zinc were retained as tracers of traffic-derived contamination, because of their well-documented relevance in urban and highway stormwater runoff (Huber *et al.*, 2016). 5 to 6 homogenized subsamples were analyzed independently, thus enabling the repeatability to be checked, then each sample was assigned its average metal concentrations. All soil concentrations (in $mg.kg^{-1}$) presented in the subsequent developments are based on dry mass. The limits of quantification are sample-dependent, as they vary according to the signal received by the analyzer, but they were in any case $< 30\ mg.kg^{-1}$ whatever the metal.

Portable XRF being a non-conventional practice for the elemental analysis of solid samples, a comparison with a reference approach (ICP-based analyses following acid digestion) was carried out on soil samples from three different sites with contrasting characteristics and contamination levels. The results are presented as *Supplementary material*. For copper and zinc, the data points were distributed on the first bisector (Fig. S1a and c), whereas lead displayed a slight offset (Fig. S1b), indicating that XRF analysis consistently underestimates total Pb contents by $\sim 15\ mg.kg^{-1}$. Overall, this comparison showed that combining an advanced pre-treatment method (sample drying, grinding and sieving at 2 mm) and an averaging process of ≥ 5 measurements enabled a satisfying reliability of the XRF-determined concentrations. This good accuracy was confirmed by a comparison with NIST soil standards (*i.e.*, materials with well-characterized composition): relative errors ranged from -12 to 5% for Cu, -7 to 16% for Zn, and -35 to -13% for Pb (negative values indicate an underestimation of the certified contents).

2.3.2 PAHs

The two samples from each site intended for PAH analysis were sent to an analytical laboratory. The analyzed compounds were the 16 PAHs classified as priority pollutants by the US-EPA: naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(ah)anthracene, indeno(123-cd)pyrene, and benzo(ghi)perylene. After fine grinding of the soil samples, PAHs were extracted with a mixture of acetone and dichloromethane, followed by GC/MS

analysis. The complete method is referenced in the European standard EN 15527 (2008). The limit of quantification was 10 µg.kg^{-1} for each individual compound except for naphthalene (30 µg.kg^{-1}). Additional details regarding the analytical uncertainties are provided as *Supplementary material*.

2.3.3 Soil organic matter

Given the importance of soil organic matter (SOM) in the physico-chemical retention of dissolved metals and hydrophobic compounds such as PAHs (Turer and Maynard, 2003), the volatile matter (VM) content – often taken as a proxy for SOM – was determined in two surface samples from each site: (i) the closest sample to the road (composed of a mixture of soil and sediment), and (ii) the sample collected at 1.2 m (most likely composed of soil only). 10-g subsamples, previously oven-dried, were calcined at 550 °C for 6 hours, then the volatile matter content was calculated from mass difference.

2.4 Estimation of the reference contents in soils

Characterizing the site-specific contamination requires evaluating the reference contents in the soils of the study area. The latter term is preferred to "*background* contents", as all soils in urban or semi-rural regions undergo diffuse contamination, *e.g.*, due to long-range atmospheric transport. For metals, usual content ranges were estimated from a national database of trace metals in agricultural topsoils called "BDETM": over the period 2000-2010, ~2300 soil samples were collected within the *Seine-et-Marne* district and analyzed for eight metals (Baize *et al*., 2007). The 1st and 9th deciles of the measurements were respectively 8-20 mg.kg^{-1} for copper, 13-28 mg.kg^{-1} for lead, and 38-68 mg.kg^{-1} for zinc. For each metal, the 9th decile was presently retained as a conservative estimate of the reference content, *i.e.*, the exceeding accumulation could be reasonably attributed to site-specific sources of contamination.

As to PAHs, data are available at the national level *via* a measurement network called "RMQS – *Réseau de Mesures de la Qualité des Sols*", which conducts long-term monitoring of soil quality based on a regular sampling grid with 2200 points over the entire metropolitan territory (Jolivet *et al*., 2006). The 1st and 9th deciles of $\sum^{16}$ PAHs were respectively 0.13-0.40 mg.kg^{-1}.

2.5 Data availability

The present dataset has been added to an online data repository (Charafeddine *et al*., 2020).

3 RESULTS AND DISCUSSION

3.1 Typical patterns of soil contamination in the road shoulders

3.1.1 Metals

Copper, lead and zinc were consistently above the analyzer's limits of quantification. Across all sites, Cu and Zn appeared to be strongly correlated, with Spearman's $\rho = 0.93$ ($p \ll 0.01$); on average, Zn concentrations were 3 times higher than Cu concentrations, as previously observed by Horstmeyer *et al*. (2016) in various roadside swales. This usual Zn/Cu ratio enabled the detection of anomalous behaviors due to additional sources of metals: for instance, in the two sites equipped with a galvanized guardrail, a steep albeit localized increase in Zn concentrations (> 900 $mg.kg^{-1}$) was visible under the guardrail: this resulted in a Zn/Cu ratio > 6 at this point, while the rest of the shoulders did not appear to be impacted. In one site that was adjacent to a cultivated field, Cu concentrations exceeded that of Zn, as a result of which agricultural contamination of copper was suspected (*e.g.*, by copper sulfate).

Within each shoulder, Cu and Zn displayed significant accumulation close to the road, followed by an overall decrease in concentrations with increasing distance; the median ratio between the concentrations found at 0 and 1.2 m was 2:1 (Fig. 4a-b). At the highest distance from the road, their concentrations were approaching the usual content ranges in agricultural soils of *Seine-et-Marne*. Generally, no significant differences were observable between the surface sample (0-2 cm) and the underlying one (2-5 cm), whatever the distance from the road (Fig. S2). These patterns were however not systematically replicated by Pb, the distributions of which showed a relative uniformity across the entire width of the road shoulders, without the systematic buildup close to the road that characterized Cu and Zn behavior (Fig. 4c). These concentrations corresponded to a moderate contamination relative to usual agricultural contents.

The reason for the bounded accumulation of copper and zinc lies in their main transport processes, and notably (i) the deposition of particles at the soil surface, and (ii) the non-uniform nature of spreading and infiltration processes within road shoulders. The latter is especially true for the low-intensity, long-duration rainfall events which prevail in the Paris region and induce a localized infiltration (Tedoldi *et al*., 2017a). The spatial variability of copper and zinc in the surface soil constitutes a demonstration that runoff water from the road rarely reaches the other end of the permeable shoulder, which was confirmed by other approaches such as hydrological modelling (Flanagan *et al*., 2016) or direct observations of the water pathways (García-Serrana *et al*., 2017). Metal patterns provide an overall, time-integrated signature of the cumulative infiltration fluxes and sedimentation processes at the soil surface, most of which occur within 1 to 1.5 m from the road edge.

Conversely, the even distribution of lead, as well as its moderate concentrations in most sites, is probably a consequence of the diminishing sources of this metal in road environments. The review carried out by Huber *et al*. (2016) highlighted a clear historical trend in Pb concentrations in road runoff, with a net

decrease during the 1990s and 2000s resulting from the elimination of leaded gasoline, and substitution of Pb in tires and brake linings, lubricating oil and grease. The studies on roadside soil contamination published prior to the 2000s did show the same distribution for lead as for other metals (Dierkes and Geiger, 1999; Jaradat and Momani, 1999; Olajire and Ayodele, 1997; Yassoglou et al., 1987). Contrariwise, for newer sites such as those in this study, lead probably accumulates in amounts insufficient to generate a significant spatial trend.

Within the soil matrix, particulate contaminants undergo mechanical filtration, while the reactive transport of dissolved metals induces (i) a gradual increase in surface concentrations due to sorption onto the upper horizon, and (ii) a downward migration of the "front" between contaminated and uncontaminated soil. These two processes lead to a *local equilibrium state* in which the soil's sorption capacities are exhausted up to a certain depth. Dissolved metals are then transported downwards and retained onto the underlying horizon. This vision is consistent with the fact that differences in metal levels between the upper and lower samples were often negligible.

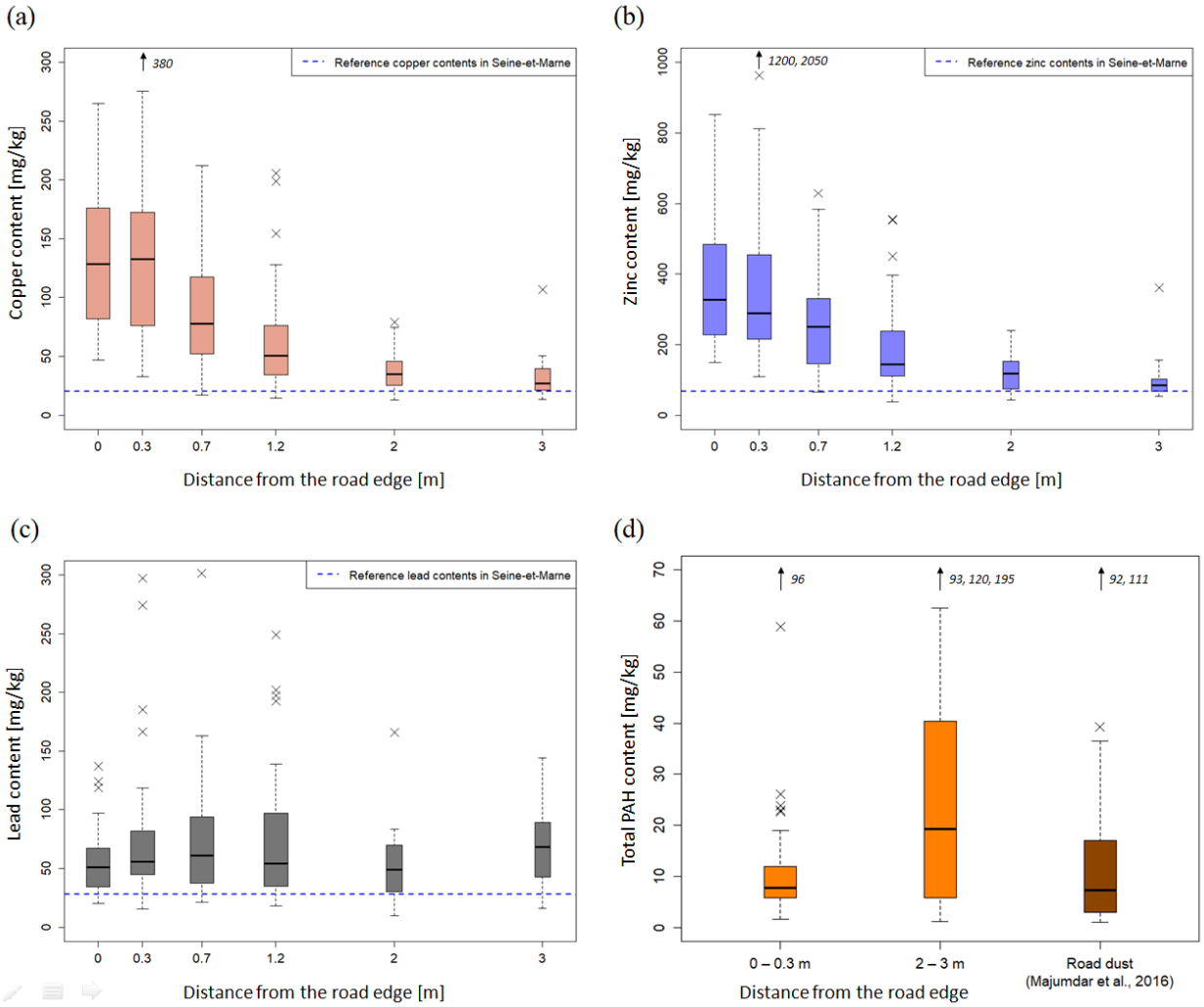


Fig. 4 – Concentrations of (a) copper, (b) zinc, (c) lead, and (d) $\sum^{16}$ PAHs across all sites, as a function of the distance from the road. The bottom and top of each box correspond to the first and third quartiles, and the thick line represents the median value (N = 40).

3.1.2 PAHs

PAH concentrations in road shoulders were significantly higher than reference values: across all sites, the minimum values of $\sum^{16}$ PAHs measured close and far from the road were respectively 1.6 and 1.1 mg.kg^{-1}, whilst the 9th decile of usual contents in agricultural topsoils amounted to only 0.4 mg.kg^{-1}, indicating a strong influence of traffic-based emissions. The distribution of PAHs among the 16 analyzed compounds appeared to be fairly homogeneous from one site to another, with limited differences between the two sampling locations (Fig. 5). Fluoranthene, pyrene and benzo(b)fluoranthene were the three predominant PAHs, together contributing > 35% of $\sum^{16}$ PAHs. These profiles were somewhat dissimilar to what was previously found in the soil of various infiltration devices for urban runoff management (Tedoldi *et al*., 2017b), where phenanthrene and chrysene concentrations were proportionally higher than in the present roadside soils (resp. 10 and 12% of $\sum^{16}$ PAHs), and benzo(b)fluoranthene and benzo(a)pyrene concentrations were lower (resp. 8 and 6%).

It has been widely documented that "diagnostic ratios" between certain PAH concentrations enable the identification of the main pollution emission sources (Tobiszewski and Namiesnik, 2012). Let us consider for instance the ratio K_1 = BaA/[BaA + CHR], where BaA and CHR respectively refer to benzo(a)anthracene and chrysene concentrations in soil. $K_1 < 0.2$ indicates petrogenic sources of PAHs, $0.2 < K_1 < 0.35$ uncovers pyrogenic sources dominated by coal or wood combustion, while $K_1 > 0.35$ corresponds to vehicular emissions. Unsurprisingly, the soil samples from all study sites belonged to the latter category ($K_1 \geq 0.4$). Applying other ratios, *i.e.*, K_2 = ANT/[ANT + PHE], K_3 = IcdP/[IcdP + BghiP], and K_4 = BaP/BghiP (where ANT, PHE, IcdP, BghiP, and BaP respectively refer to anthracene, phenanthrene, indeno(123-cd)pyrene, benzo(ghi)perylene, and benzo(a)pyrene concentrations), led to the same conclusion based on the threshold values reported by Tobiszewski and Namiesnik (2012). However, for the most polluted sites (> 90 mg.kg^{-1}, *cf*. Fig. 4d), these ratios were approaching the threshold values between coal/wood and traffic-based emissions, as a reason of which "historical" contamination of the soil was also suspected.

In contrast to the spatial patterns of metals, PAH concentrations were often lower in the samples collected near the road than at the other end of the shoulder (Fig. 4d). This dissimilarity, which was also acknowledged in most reviewed studies (*cf*. Introduction), probably indicates that the major transport and dispersion processes are different. While most metal fluxes appear to be associated with wet weather (runoff and spray), with a short-range influence, as previously discussed, atmospheric transport plays a greater role for traffic-emitted PAHs (Agarwal, 2009): air turbulence caused by passing vehicles likely results in less deposition in the immediate vicinity of the road.

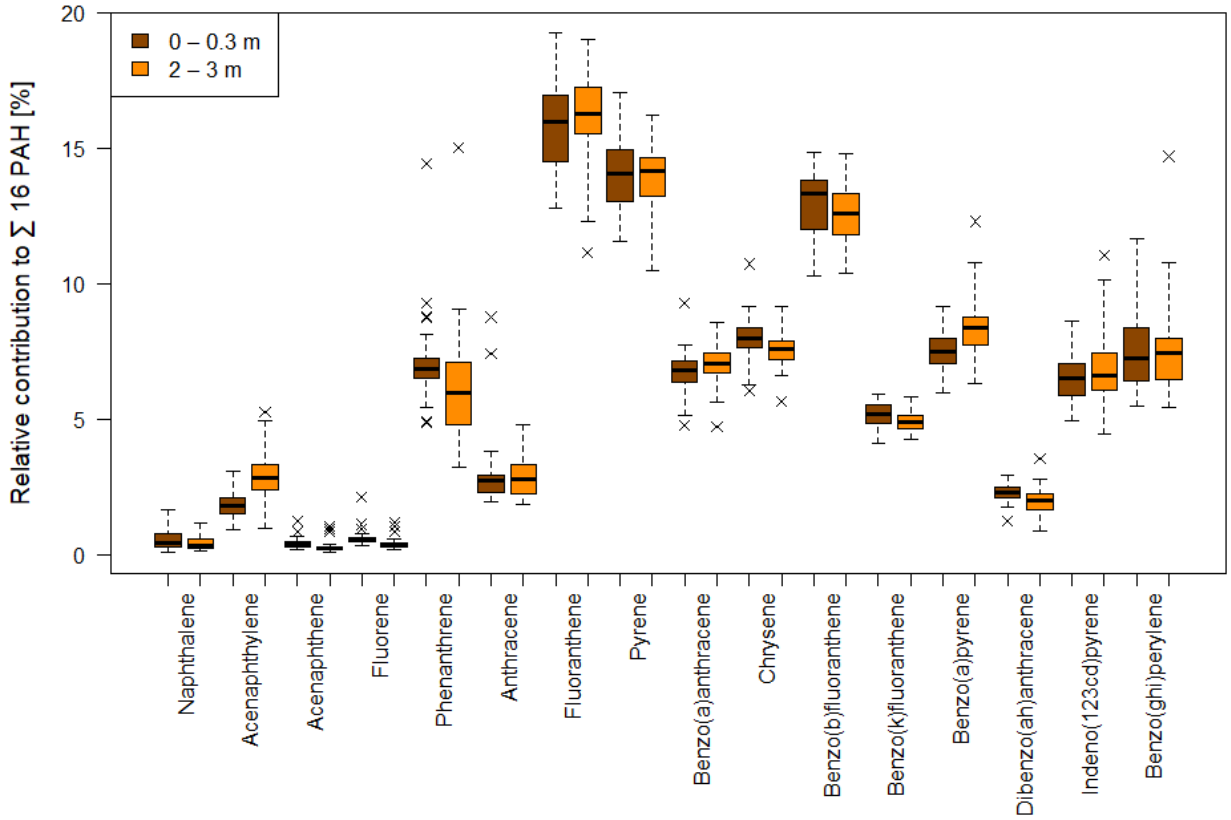


Fig. 5 – Relative contribution of each individual PAH to their total concentration, across all sites.

3.2 Inter-site variability of the contamination levels

3.2.1 Influence of the traffic characteristics on metal concentrations

Beyond the recurring structure of metal distribution within each study site, Fig. 4 emphasizes the important inter-site dispersion which affects the values measured at a given distance. For example, close to the road edge, copper and zinc concentrations ranged respectively from 45 to 270 mg.kg^{-1}, and from 150 to 850 mg.kg^{-1}. So as to further explore the causes of such variability, inter-site comparisons were carried out. The data from each site were synthesized into three indicators to be conceivably explained by different influencing factors: (i) the mean concentrations of copper and zinc in the first 30 cm from the road, (ii) the mean concentrations in the middle of the shoulder (70-120 cm), and (iii) the values measured on the opposite side. They are hereinbelow plotted against AADT in the case of zinc, with color gradients representing the percentage of semi-trucks (Fig. 6). The same graph is provided as *Supplementary material* for copper (Fig. S3). The samples with "anomalous" Zn/Cu ratios, reflecting additional sources of contamination (*cf.* paragraph 3.1.1), were not accounted for in the present approach.

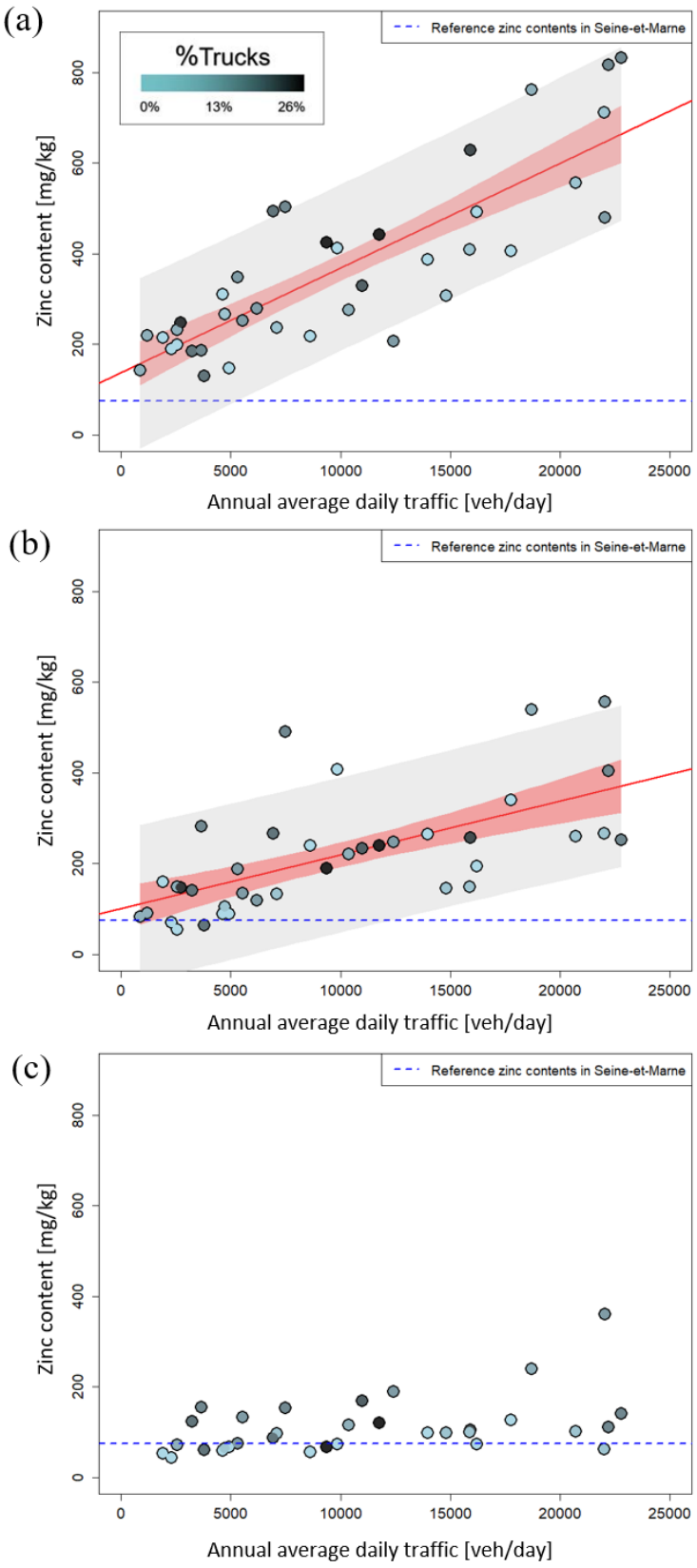


Fig. 6 – Relationship between the traffic characteristics (AADT on the horizontal axis and %Trucks represented as a color gradient) and the mean Zn concentrations (a) in the first 30 cm, (b) between 70 and 120 cm from the road, and (c) on the opposite side of each shoulder. The red and grey envelopes correspond respectively to the 95% confidence and prediction intervals of the regression.

Despite a vertical dispersion of the data points, concentrations from the most contaminated zone (indicator i) displayed a clear increasing trend over the entire range of traffic (Fig. 6a); interestingly, the influence of AADT on soil contamination was still visible in the center of the shoulder (indicator ii) – albeit less markedly and with more deviating behaviors (Fig. 6b). These trends reflect the effect of increasing traffic-related emission sources – which in road environments have been shown to be essentially brake and tire wear for copper and zinc, respectively (Huber *et al*., 2016) – on pollutant loads and build-up in roadside soil. AADT alone explained respectively 81% and 69% of the variability of Cu and Zn levels *via* the following linear models:

$$\mathrm{Cu}_{\,0-30\ \mathrm{cm}} = (49 \pm 16) + (7.7 \pm 1.3) \cdot \frac{\mathrm{AADT}}{1000} \quad [\mathrm{mg.kg^{-1}}] \tag{1a}$$

$$\mathrm{Zn}_{\,0-30\ \mathrm{cm}} = (141 \pm 61) + (22.6 \pm 5.1) \cdot \frac{\mathrm{AADT}}{1000} \quad [\mathrm{mg.kg^{-1}}] \tag{1b}$$

The constant term of each model is close but still slightly higher than the estimated reference contents of the soils in *Seine-et-Marne* (*cf.* paragraph 2.4). The residuals were tested for normality (Shapiro-Wilk test) and absence of autocorrelation (Durbin-Watson test); homoscedasticity was established in the case of copper, but not zinc (Breusch-Pagan test) at a 0.05 level.

Conversely, the contamination levels at 1.8-3 m from the road appeared to fluctuate independently from the traffic characteristics, in a relatively narrow range close to the usual agricultural contents – except for a few points (Fig. 6c). Such observation confirmed that this area of the shoulder was only marginally affected by runoff-derived contamination.

In order to evaluate the influence of truck traffic on the residual variability of metal concentrations, a multiple regression was carried out, including both AADT and %Trucks as predictors. The quality of the model was slightly improved for Zn (the adjusted R^2 increased to 0.73, and the residuals could be considered homoscedastic) but not for Cu. This was not deemed enough to justify the use of a two-variable model in the following developments. The variability unexplained by the traffic characteristics may therefore be interpreted in terms of (i) differences in traffic conditions from one site to another (*e.g.*, frequency of traffic jams), (ii) differences in road environments (open countryside, forest, proximity of dwellings), (iii) differences in operation time, and/or (iv) differences in soil characteristics – resulting in variable retention properties of dissolved metals – which are discussed in the following paragraph.

3.2.2 Influence of the soil organic matter

Among the different soil properties, SOM is frequently reported as the most important characteristic governing the sorption of dissolved metals, as it participates both in chemisorption and physisorption processes (*i.e.*, chemical reactions with surface functional groups, and charge-driven mechanisms, respectively (Tedoldi *et al*., 2016)). As presented in the Material and Methods section, the volatile matter (VM) content was presently retained as a proxy for SOM, which is a common approach in soil studies. In

the samples collected close to the road, VM ranged from 4 to 14% depending on the site; the lowest value corresponded to a calcareous shoulder, while most values fell between 7 and 11%. Fig. 7 displays the Zn concentration versus the VM content of these samples, with colors representing the AADT on the adjacent road.

Organic matter appears to control the maximum concentration of metals that can be sorbed onto the soil, *i.e.*, the upper envelope of the data points, below which Zn concentrations spread according to the traffic density, as previously discussed. This envelope forms approximately a straight line for VM contents ≤ 11%, the equation of which was obtained from a linear regression after isolating the upper points:

$$\mathrm{Zn}_{\mathrm{max}} = (80 \pm 12) \cdot \mathrm{VM} + (-90 \pm 115) \quad [\mathrm{mg.kg^{-1}}] \qquad (2)$$

The same behavior is observable for copper (see *Supplementary material*, Fig. S4).

Although the present result is of great interest from a scientific perspective, including the VM content in a predictive model of soil contamination is not operationally functional, as the latter, just like metal contents, is unknown *a priori*. While the design of Best Management Practices (and especially swales or bioretention systems) is subject to precise guidelines in several countries, which set an acceptable range for the organic matter content of the soil/filter media, this is usually not the case for road shoulders in France. As a result, VM would have to be determined from *in-situ* sampling due to its potential variability. Thus, in the subsequent part, where the practical implications are looked upon, only the above-mentioned traffic-based regression model will be focused on.

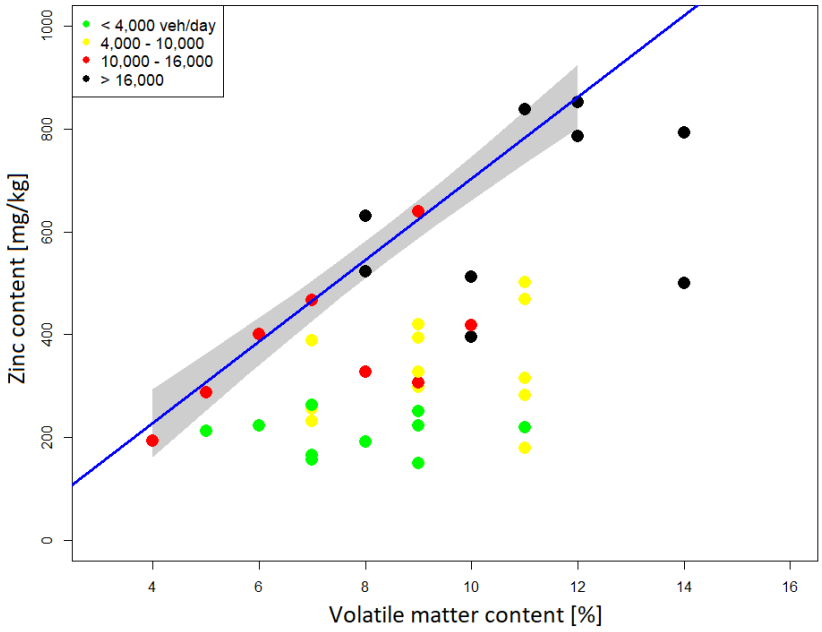


Fig. 7 – Relationship between volatile matter and zinc concentrations in the samples collected close to the road. The upper envelope of the data points (defined as the highest Zn concentration for each value of the volatile matter content) was modeled *via* a linear regression in the range 4% ≤ VM ≤ 12% (blue line, along with its 95% confidence interval).

3.2.3 Case of PAHs

PAH concentrations did not exhibit the same dependence on road traffic as copper and zinc. As a reminder, not all examined studies mentioned traffic density as a significant factor influencing PAH contamination either (Kim *et al*., 2019; Kumar and Kothiyal, 2012; Marusenko *et al*., 2011). Another remarkable difference with metals was that the contamination levels across all study sites displayed a noticeably lower variability at 0-0.3 m than at 2-3 m (Fig. 4d). So as to interpret this observation, it should be kept in mind that the samples collected close to the pavement were composed of a mixture of soil and sediment, the latter originating from road dust mobilization by runoff. Interestingly, a literature review conducted by Majumdar *et al*. (2016) showed that PAH concentrations in road dust from worldwide traffic areas were distributed within a very narrow range of values: $\sum^{16}$ PAHs frequently fell between 1 and 35 $mg.kg^{-1}$, as reported on Fig. 4d. This relative stability of road dust composition is consistent with the low dispersion and the range of the measurements near the roadway.

At the other end of the shoulder, not only did the values span a wider range, but they were also relatively high compared to the available data about roadside soil contamination. For instance, the third quartile of the present measurements at 2-3 m (40 $mg.kg^{-1}$) corresponds to the highest value reported by Marusenko *et al*. (2011) from a literature survey gathering 23 different studies. Nevertheless, such contamination levels were also measured by Mikkelsen *et al*. (1996) in two old infiltration systems for road runoff ($\sum^{16}$ PAHs between 31 and 220 $mg.kg^{-1}$). A conceivable assumption for this behavior is that this zone may not have been scraped during all previous maintenance operations, depending on the length of the equipment used: if so, this would entail different accumulation times from one site to another at 2-3 m. While this is not of major importance for metals, which have been proven to be mostly transported by short-range, wet weather-related processes (and therefore accumulate close to the roadway), it may lead to larger differences for PAHs if the prevalence of mid- to long-range atmospheric transport is ascertained. Under such assumptions, PAH concentrations measured far from the road would result from a continuous integration of the deposits at the soil surface over a larger period of time. It may also reinforce the influence of the road environment on the inter-site variability of PAH concentrations – inasmuch as the presence of trees or other obstacles affects atmospheric dispersion, as discussed by Agarwal (2009) – making the derivation of predictive relationships more complex.

3.3 Practical implications

3.3.1 Optimization of maintenance practices

The unequally polluted soil which is scraped during usual maintenance operations of road shoulders is ordinarily managed as a whole, inducing a “dilution” of the contamination. Due to the recurrent patterns of soil contaminants, it may be practically interesting to consider a differentiated management for the different parts of the vegetated strip. This concern may be rephrased as follows: is it possible to

systematically isolate a certain proportion of the pollutants accumulated in each shoulder, by excavating it over a controlled width? For PAHs, the spatial resolution of the data was not deemed sufficient to carry out such appraisal; however, for metals, this proportion was calculated as a function of the distance d from the road, according to Eq. 3.

$$F(d) = \frac{\int_0^d \rho\, C_{\mathrm{acc}}(x)\, dx}{\int_0^D \rho\, C_{\mathrm{acc}}(x)\, dx} \tag{3}$$

where D is the entire width of the road shoulder (1.5 to 3 m depending on the site), ρ is the soil's bulk density (which is assumed to be uniform in the sampled layer, and thus no longer intervenes in this equation), and $C_{\mathrm{acc}}(x)$ is the concentration of accumulated metal at a distance x from the road (averaged over the soil depth). The latter term was conservatively estimated as:

$$C_{\mathrm{acc}}(x) = \max(C(x) - C_{\mathrm{ref}}\,, 0) \tag{4}$$

where C_{ref} is the upper bound of usual agricultural contents in *Seine-et-Marne* (*cf.* paragraph 2.4), and $C(x)$ results from linear interpolation of the concentrations measured at 0, 0.3, 0.7, 1.2, and 1.8-3 m.

The results are displayed on Fig. 8 in the case of zinc. As can be seen of this graph, scraping 1.5 m of the shoulder starting from the road will likely remove between 75 and 100% of the accumulated metal. In nine cases out of ten, a 80% removal can be achieved by targeting ≤ 1.8 m. This evaluation is of particular interest near heavily trafficked roads, where metal contamination levels in the soil can be locally high, thus emphasizing the benefits of treating this pollution separately *via* a differentiated management of the scraped materials.

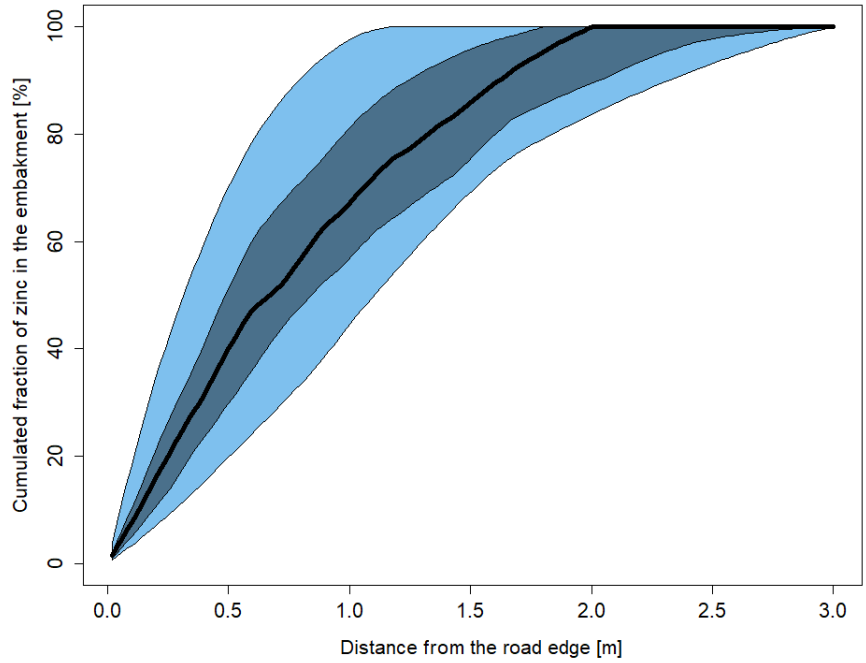


Fig. 8 – Fraction of zinc accumulated in the shoulder as a function of the distance from the road. The lines and envelopes represent statistics established from the 40 study sites: median accumulation (solid line), 1st and 3rd quartiles (dark envelope), 1st and 9th deciles (light envelope).

3.3.2 Potential for reuse of the excavated soils

So as to evaluate the suitability of partially contaminated soils for certain uses (*e.g.*, residential, agricultural or commercial), a conventional approach consists of comparing contaminant concentrations to threshold values. In case of exceedance, it is considered that such uses are to be prohibited and/or specific actions are needed: for this reason, these values are often referred to as "intervention thresholds" or "soil cleanup standards". Despite the apparent simplicity of this procedure, Provoost *et al*. (2006) underlined the fact that thresholds can differ significantly from one country to another – where they exist: for instance, in France, the only regulatory guidelines about soil contamination are aimed to restrict the spreading of sewage sludge in agricultural fields deemed "too polluted". In previous research works addressing soil contamination in stormwater Best Management Practices, authors referred to the Dutch, Swiss, Austrian, Swedish, or Danish standards, in which intervention thresholds vary from 100 to 1000 $mg.kg^{-1}$ for copper, and from 500 to 2500 $mg.kg^{-1}$ for zinc (Tedoldi *et al*., 2016). For PAHs, it is common for these thresholds to be additionally compound-dependent, as several PAHs have been proven to present carcinogenic hazards and are therefore subject to stricter regulations.

It is beyond the scope of this paper to discuss which thresholds should be preferably retained and which ones have a lesser relevance to quantify the impacts of soil pollution; instead, a different strategy may be proposed for metals. Considering the established relationships between soil concentrations and vehicular density (paragraph 3.2.1), each existing threshold can be associated a "compatible" range of traffic, such that all roads below the "cutoff" AADT would be expected to generate lower concentrations than the pollution threshold. A conservative approach can be adopted by considering the upper limit of the prediction interval for the data points at 0-30 cm (Fig. 6a): this corresponds to the highest possible concentrations on the road shoulder for a given AADT. As an illustration, let us consider the intervention thresholds adopted in the Netherlands (NMHSPE, 2000) and in Sweden (Swedish EPA, 1997), which are relatively close (respectively 190 and 200 $mg.kg^{-1}$ for copper, and 720 and 700 $mg.kg^{-1}$ for zinc). Considering the aforementioned regression analysis (see Fig. S3 for copper), roads with daily traffic below 17,000 veh/day are not expected to generate metal contamination exceeding these thresholds.

Conversely, the state of the art as well as the present work reveal our lesser understanding of the characteristics of PAH contamination, and more generally organic micropollutants of emerging concern such as phthalates, alkylphenols and their derivatives. These substances have been shown to be ubiquitous in road runoff (Flanagan *et al*., 2018), but investigations on roadside soils are still scarce (Hongjun *et al*., 2013; Lü *et al*., 2018). Further research is needed to address their emissions from road traffic, retention and persistence in soil, so as to move towards a more integrated approach to environmental hazards and possible remediation techniques.

3.3.3 Operational perspectives: road shoulders as treatment devices for contaminated runoff

Although road shoulders are not initially designed and maintained for this purpose, they offer interesting perspectives for the mitigation of runoff-generated contaminant fluxes to the environment, since the soil

acts as a filter for both dissolved and particulate substances. Hence, it may be valuable to consider these infrastructures as multi-functional *nature-based solutions*, the characteristics of which may be enhanced towards an effective treatment of road runoff. The fact that soil organic matter controls the maximum amount of metals that can be retained from contaminated water (paragraph 3.2.2) emphasizes the interest of avoiding low-organic materials such as calcareous soils when implementing road shoulders, and bringing some organic-rich material to maximize contaminant retention in the presence of sensitive issues (*e.g.*, groundwater used as a source of drinking water). Similarly, the depth of this filter material should be such that subsequent scraping operations would not expose deeper soil horizons – with potentially lower sorption capacities and poorer biological activity – otherwise the post-maintenance retention efficiency would be significantly lessened.

4 CONCLUSIONS

In the present study, soil contamination in road shoulders was investigated through an extensive sampling campaign on 40 study sites, with a dual focus on intra- and inter-site variability of concentrations. The achieved results demonstrated:

i. a good reproducibility of the "typical" distribution of metal concentrations within each site: for copper and zinc, this consisted of a significant accumulation close to the road, and a continuous decrease with increasing distance, so that the first 1.5 m of the shoulder systematically contained $\geq 75\%$ and up to 100% of the accumulated metals. Lead displayed more uniform distributions, closer to the usual content range for agricultural soils from the study area.
ii. contrasting patterns for PAHs, the concentrations of which fell within a relatively narrow range of values close to the road, whereas highest buildup occurred at the other end of the shoulders.
iii. a satisfying predictability of copper and zinc levels from the annual average daily traffic of the nearby road, which explained respectively 81% and 69% of the inter-site variability.
iv. the existence of a "ceiling" concentration of metals in soil, controlled by its organic matter content, which limits the amounts that can be intercepted from road runoff.

Unlike metals, which are mostly carried by short-range, wet weather-related processes, the dispersion mechanisms by which PAHs are transported from the road to the adjacent shoulders remain unclear. The higher concentrations found further away from the road could either indicate the prevalence of longer-range mechanisms, mainly governed by atmospheric transport, or be due to longer accumulation times.

Overall, these results demonstrate that road shoulders enable the mitigation of runoff-generated contaminant fluxes to the environment, and may therefore be considered as treatment devices and optimized as such. Additionally, the existence of explicit relationships between traffic density and metal concentrations makes it possible to route the scraped soil towards the most suitable valorization or treatment option according to the site characteristics. Further research could refine the understanding of

PAH behavior and include other emerging organic micropollutants in these considerations, to achieve an integrated vision of the hazards posed by roadside soil contamination.

Interestingly, most studies which focused on road runoff hardly evidenced a correlation between traffic indicators and metal concentrations in water, owing to their significant inter-event variability. However, their long-term accumulation in soil acts like an averaging process which enables this trend to be reconstructed. Considering the soil's propensity to act as a "passive sampler" of contaminants – insofar as the latter are not or only slightly subject to dissipation processes – studying soil contamination in stormwater Best Management Practices may offer promising perspectives for the further characterization and modelling of pollutant fluxes.

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