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ECOTOXICOLOGICAL RISKS OF ROAD RUNOFF WATER SEDIMENTS ON AQUATIC ECOSYSTEMS

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Abstract

The management of road runoff water sediments is an environmental, economical and social important issue. In addition to the strengthening of regulations about waste management, the valorization of these heterogeneous materials, which are produced in high quantities on a national scale, is limited due to their physico-chemical characteristics and their polluting load (heavy metals and petroleum hydrocarbons).

The Laboratoire Central des Ponts et Chaussées (LCPC) of Nantes has developed a treatment process of such materials with the aim to valorize them in road building (road embankment, ...) (Pétavy, 2007). This process consists in separating the sediments into several valorizable fractions and discarding the finest particles onto which most of the metal and organic pollution is located.

In this study, raw and treated materials were characterized on a physico-chemical level and submitted to a leaching test reflecting the transfer of pollutants from the solid phase to the aqueous phase when the re-used materials are exposed to rainfall. Ecotoxicological single-species tests and microcosm assays were then carried out on the leachates to predict the potential effects of the leachates reaching a close aquatic lentic ecosystem.

The results obtained on 13 raw materials and 2 valorizable granulometric fractions for 3 of them, suggest absence of acute ecotoxicity of the leachates at dilution factor > 2 and absence of chronic ecotoxicity (21 day exposure) at dilution factor > 10 .

The analysis of metallic contents and bioassay results lead concordingly to propose a no chronic effect concentration close to 1% (v : v). On an ecotoxicological point of view, the granulometric fractioning does not seem to produce a valorizable fraction which would be systematically innocuous.

Keywords : road runoff water sediment ; heavy metal ; microcosm

Introduction

In urban or rural environments, the drainage of roads and motorways by rain water leads to more or less polluted effluents. These runoff waters must be collected and discharged into surface or groundwater systems. Due to the high volumes generated and the limited capacity of drainage networks, alternative techniques (retention/detention or infiltration basins, drainage ditches) have been developed. In addition to the quantitative (flow control) as well as qualitative (reduction of pollution) management of water that these techniques allow, they are economically beneficial and insert more easily in the landscape. However, the ditches and basins must be regularly cleaned in order to maintain their treatment performance and their hydraulic capacity. The maintenance of the works consists in extracting sediments that accumulate, leading to a total volume of 6 to 10 million tons of sediments per year in France, depending on the source (SETRA, 1995; ONR, 2000; Pétavy, 2007). The sweeping of roads, often considered as an efficient way of reducing urban pollution, also generates significant volumes of materials (1 million tons per year in France, Kempf, 2001). All these materials are polluted, due to the emission of pollutants (heavy metals, petroleum hydrocarbons, pesticides) by motor vehicles, road pavement and road equipment (crash barriers, road verges, ...) (Durand, 2003).

The common origin of particles produced by road sweeping and sediments accumulated in basins leads to manage them as a whole. Until now, these materials were landfilled or simply stored on soils in the vicinity of roads. But these classical ways of waste management are more and more non fitted to the strengthening of environmental regulations and the scarcity of natural primary materials. The re-use of these materials in civil engineering as building or earthwork spreading materials seems to be a relevant way of valorization. Due to their polluting load, treatment processes may be necessary prior to their re-use. Among techniques available, a method proposed by LCPC (Laboratoire Central des Ponts et Chaussées) is aimed at separating valorizable and non valorizable fractions by a granulometric sieving (Pétavy, 2007). This sieving removes the finest fractions which are also often the most polluted and the most difficult to use in civil engineering due to their geotechnical properties.

However, there are no data in the literature to support the assumption that these materials, with or without treatment, are re-usable in conditions acceptable for the environment. This study, carried out in the Laboratoire des Sciences de l'Environnement

(L.S.E.) of ENTPE, was aimed at assessing the risks for the environment of a scenario of re-use of road runoff water sediments (spreading on road dependency or use on embankment). The scenario considers the leachate produced by infiltration of rainwater through the re-used materials and the potential effects of leachate discharge into a lentic aquatic ecosystem. This ecosystem is represented by laboratory aquatic microcosms previously used by L.S.E. in various studies (Triffault-Bouchet *et al.*, 2004 ; Clément *et al.*, 2004; Triffault-Bouchet *et al.*, 2005). The microcosm assays were preceded by single-species tests aimed at giving a first idea of the acute ecotoxicity of leachates.

Materials and methods

The general approach consisted in producing a leachate from each studied sediment, either raw or treated, characterising the sediments and the leachates on a physico-chemical level, and assessing the ecotoxicity of leachates using single- and multi-species (microcosm) tests.

The study focussed on 13 sites, with a majority of urban retention ponds with high traffic roads (table 1). Three of these sediments (AhAh, Cheviré, Lille) were granulometrically sieved, leading to 4 fractions : the raw fraction, the 2-30 mm fraction, the 60 µm-2 mm fraction, and the <60 µm fraction.

Table 1. Origin of road sediments studied in this work (nd : data not available)

Sediment	Road	Location	Type	Work	Type	Vegetation	Volume (m ³)	Traffic (vh/day)
Wissous	nd	Paris region	urban	Underground basin	concrete	no	10000	/
AhAh	nd	Crosne (Essonne)	urban	Open basin	concrete	no	600	/
Cheviré	Pont de Cheviré	Nantes	Urban motorway	Open basin	Infiltration soil	no	1170	80000
Lille	nd	Lille centre	Urban street	Sweeping residues	/	no	/	/
RCE1	Rocade Est	Lyon region	Urban motorway	Open basin	PEHD liner + sand	yes	940	77091
RCE2	Rocade Est	Lyon region	Urban motorway	Open basin	PEHD liner + sand	yes	4670	77091
RCE3	Rocade Est	Lyon region	Urban motorway	Open basin	béton	no	nd	nd
RCE4	Rocade Est	Lyon region	Urban motorway	Open basin	PEHD liner + sand	yes	5510	83879
RCE8	Rocade Est	Lyon region	Urban motorway	Open basin	concrete	no	2360	83879
A1	A7/Bld Europe	Lyon region	Urban motorway	Open basin	PEHD liner + sand	yes	nd	nd
A2	A7/Subdi	Lyon region	Urban motorway	Underground basin	concrete	no	nd	nd
A7	A7/A450	Lyon region	Urban motorway baine	Open basin	PEHD liner + sand	yes	nd	nd
S	nd	Lyon region	Urban road	Open basin	PEHD liner + sand	yes	nd	nd

The leachates were obtained using the french protocol NF X 31-210 (AFNOR, 1992) which consists in a 24hr-mixing of 100 g sediment with a liquid-solid ratio of 10, and a centrifugation aiming at separating particles and water.

The sediments and their leachates were characterized according to the protocols displayed in table 2.

Table 2. Physico-chemical analyses carried out on road sediments and their leachates

Physico-chemical analyses	Sediment	Centrifuged leachate	0.22 µm-filtered leachate
pH	nd		pH-meter
Conductivity	nd		Conductimeter
% water	weight		nd
perte au feu	weight		nd
Heavy metals (Cd, Cr, Cu, Ni, Pb, Zn)	AAS, following mineralisation AFNOR 1998		AAS, AFNOR 1998
NH ₄ ⁺ , NO ₃ ⁻ , PO ₄ ³⁻	nd	nd	HACH colorimetry
SO ₄ ²⁻ , Cl ⁻	nd	nd	Ion Chromatography Dionex
Ca ²⁺ , Mg ²⁺ , Na ⁺ , K ⁺	nd	nd	Ion Chromatography Dionex

Daphnid and micro-algae tests were carried out on leachates following the protocols displayed in table 3.

Table 3. Single-species tests carried out on leachates

Bioassay	Algae	Daphnid	
Species	<i>Pseudokirchneriella subcapitata</i>	<i>Daphnia magna</i>	
End point	Growth inhibition	Immobilisation	Reproduction inhibition
Exposure time	72 h	48 h	21 days
Temperature	24 °C	20 °C	
Light	6000 lux continuously	Dark	2000 lux 16 h/day
Vessels	96-wells microplates	Glass tubes	polyethylene flask
Number of organisms	10 ⁴ cells per mL at t=0	10 daphnids/tube	1 daphnid per flask
Age of organisms at t = 0	/	< 24 h	
Replicates	5 /conc.	3 /conc.	10 /conc
Food	/	no	Algae : 3.10 ⁵ cells/mL 3 x/week
Tested concentrations	11.25, 22.5, 45.0, 90.0 %	6.25, 12.5, 25.0, 50.0, 100.0 %	12.5 %, 25 %, 50 %
Volume of solution	250 µL	10 mL	40 mL
Leachate treatment	filtration 0.22 µm	filtration 0.22 µm or not	

Microcosm tests were also carried out on all leachates according to the protocol developed by Clément & Cadier (1998) and applied in various studies (Clément *et al.*, 2004; Triffault-Bouchet *et al.*, 2004, 2005). Details on initial organism densities, monitoring of biological parameters are displayed in tables 4 and 5. Following a stabilisation phase of 5 days (microcosms with aeration in the dark, at 20°C), centrifuged leachates (not filtered) were introduced 1 day before organisms. The assays lasted 21 days, a duration which enabled to assess daphnid reproduction and chironomid emergence.

Table 4. Numbers of organisms in microcosms at t=0

Organisms	Number	Age
Daphnid <i>Daphnia magna</i>	10	< 24 h
Algae		
<i>Chlorella vulgaris</i>	10000 cells/mL	-
<i>Pseudokirchneriella subcapitata</i>	10000 cells/mL	-
Duckweed <i>Lemna minor</i>	12 fronds (6 two-frond colonies)	-
Amphipod <i>Hyalella azteca</i>	10	1 to 2 weeks
Chironomid larvae <i>Chironomus riparius</i>	25	2 to 4 days

Table 5. Biological monitoring in microcosms

Organism	Frequency	End point	Method
<i>Duckweeds</i>	twice per week	Growth	Fronds and colonies counting
	End	Growth	Fronds and colonies counting + dry mass
<i>Daphnids</i>	once or twice per week	Survival	Mother daphnids counting
		Reproduction	Sampling neonates, counting and discarding
<i>Chironomids (flying adults)</i>	Each day	Survival	Number of males Number of females
	End	Growth	Dry mass of males Dry mass of females
<i>Amphipods</i>	End	Survival	Number of living amphipods
		Growth	Dry mass

Results

Physico-chemical composition of sediments and leachates

All sediments were potentially toxic for aquatic organisms. As a matter of fact, their contents in heavy metals (table 6) often exceeded the thresholds for such materials (TEC and PEC, MacDonald *et al.*, 2000). The concentrations in heavy metals of leachates are displayed in table 7. These concentrations show that only a low fraction of sediment pollutants was mobilised by leaching. However, the 42 leachates showed, for at least one metal, concentrations > PNEC (respective values for Pb, Cd, Cr, Ni, Cu and

Zn : 5, 0.75, 4.7, 0.5, 1.6 et 8.6 µg/L, according to Ineris studies). Ni, Pb and Cu were mainly concerned, and this fact must be linked to their higher mobility. Leachates of fractions 60 µm-2 mm et 2-30 mm were generally less contaminated than leachates of raw sediments, but also showed values > PNEC for some metals.

Toxicity of leachates

The acute toxicity (48 h exposure) of leachates to *Daphnia magna* was low, no toxicity was observed for concentrations ≤ 25%. One leachate (Cheviré) was tested in the 21-day reproduction test and had no effect. Microalgae growth was only inhibited for concentrations ≥ 45% or 22.5%.

For most microcosm assays, only the concentration 10% was tested, which corresponds to concentrations below which no acute effect was expected, according to single-species tests results. The results for all leachates are summarised in table 8.

Table 6. Concentrations in heavy metals (mg/kg dry weight) of sediments (grey light : conc < TEC (MacDonald *et al.*, 2000) ; grey : conc > TEC and < PEC ; grey dark : conc > PEC)

	Cd	Cr	Cu	Ni	Pb	Zn
Wissous	4.5	348	324	499	323	1575
AhAh1 raw	4.26	68	139	61	244	631
AhAh1 2-30 mm	2.6	541	120	38	169	535
AhAh1 60 µm-2 mm	1.75	60	72	62	119	271
AhAh1 <60 µm	6.19	99	184	53	320	875
AhAh2 raw	0.8	29	72	50	69	268
AhAh2 2-30 mm	0.27	42	38	26	37	134
AhAh2 60 µm-2 mm	0.4	39	99	62	100	215
AhAh2 <60 µm	6.3	123	303	91	348	1152
Lille raw	0.9	202	97	45	106	356
Lille 2-30 mm	0.07	64	16	11	12	58
Lille 60 µm-2 mm	0.47	73	87	17	60	381
Lille < 60 µm	2	118	222	52	269	871
Cheviré raw	1.33	69	306	29	138	1180
Cheviré 2-30 mm	0.37	32	125	14	83	650
Cheviré 60 µm-2 mm	0.18	32	59	11	66	320
Cheviré <60 µm	2.48	120	518	46	279	2275
RCE1	0.33	28.7	24.6	10.6	21.9	80
RCE2	0.48	50	116.8	15.79	59.64	330
RCE3	0.49	49.8	140.6	14.4	70.7	410
RCE4	0.39	56.2	90.6	27.2	64.2	263
RCE8	1.45	92.7	457	35.99	212.6	1430
A1	0.28	27.43	75.48	14.88	28.18	200
A2	0.9	65.09	268	49.27	126.32	1120
A7	1.52	78.7	474.59	51.99	214.33	2040
S	0.22	33.97	69.66	11.76	28.46	210

Table 7. Heavy metal concentrations of leachates (NF : centrifuged leachate, analyses following mineralisation ; F : 0.22 µm-filtrated leachate, no mineralisation; < LD : below detection limit; grey : values > PNEC Ineris)

		Pb	Cd	Cr	Ni	Cu	Zn
Wissous	NF	1906	20.7	180.7	281.7	2127.5	10
	F	8.4	0.16	0.74	7.8	13.6	<LD
AhAh2 raw	NF	5.67	<LD	<LD	18.52	12.75	<LD
	F	<LD	<LD	<LD	17.34	8.56	<LD
AhAh2 2-30 mm	NF	1.26	<LD	<LD	4.16	2.27	<LD
	F	<LD	<LD	<LD	4.29	2.25	<LD
AhAh2 60µm-2mm	NF	4.13	<LD	<LD	4.41	7.16	<LD
	F	<LD	<LD	<LD	3.27	4.15	<LD
Lille raw	NF	183	1.43	3.7	30	103.25	0.43
	F	5	0	0.14	2.63	18.9	<LD
Lille 2-30 mm	NF	35.1	0.35	0.7	7.4	23.1	0.17
	F	1.22	<LD	<LD	0.9	6.67	<LD
Lille 60µm-2 mm	NF	36.1	0.7	0.9	8.8	37.9	0.2
	F	3.8	<LD	<LD	<LD	9.3	<LD
Cheviré raw	NF	105.7	0.98	1.63	18.3	289.7	0.85
	F	1.22	0.13	0.1	4.09	19.3	0.19
Cheviré 2-30 mm	NF	70	0.7	1.9	16.5	178.7	0.6
	F	<LD	0.48	<LD	3.04	18.4	0.15
Cheviré 60µm-2 mm	NF	25.9	<LD	0.19	1.2	23.77	0.15
	F	7.53	<LD	0.11	0.71	8.27	<LD
Cheviré <60 µm	NF	20.45	0.12	0.17	2.89	70.05	0.21
	F	<LD	0.1	<LD	1.57	23.98	0.21
RCE1	NF	21.1	2.03	13.32	9.77	23.3	<LD
	F	0.47	<LD	0.76	<LD	4.30	<LD
RCE2	NF	8.08	0.45	4.22	3.32	16.6	<LD
	F	0.66	0.07	0.39	0.52	4.70	<LD
RCE3	NF	11.5	0.83	4.52	2.28	46.3	<LD
	F	1.62	0.33	0.50	1.14	12.95	<LD
RCE4	NF	8.67	0.58	7.92	6.83	12.50	<LD
	F	<LD	0.32	2.23	1.14	6.49	0.13
RCE8	NF	47.30	1.48	21.50	15.70	307	0.45
	F	0.51	0.48	2.50	1.96	91.90	0.10
A1	NF	25.30	0.83	5.47	9.86	58.10	420
	F	2.47	<LD	0.79	6.78	6.19	140
A2	NF	97.10	0.99	15.8	28.31	189	980
	F	2.32	<LD	1.00	5.90	53.45	190
A7	NF	12.70	1.87	4.73	6.76	28.70	210
	F	<LD	<LD	1.30	4.12	1.41	80
S	NF	26.40	0.53	9.98	15.77	69.10	550
	F	0.86	<LD	1.88	54.58	6.25	210
S _{eau}	NF	193	4.58	69.50	53.74	964	2060
	F	<LD	<LD	1.03	7.28	1.14	190

Variations of pH, oxygen content and conductivity due to leachates were low and did not reflect any impairment. Algal growth was slightly impaired in some cases (RCE8 and Cheviré fraction 2-30 mm). Duckweed growth was generally not significantly reduced or was enhanced (Wissous, RCE4, RCE8), likely due to nutrient enrichment by leachates or to indirect effects resulting from competition between algae and duckweeds. The leachates, at tested concentrations, were not toxic for daphnids, as expected from single-species tests, except for Cheviré (fraction 2-30 mm) and Wissous which impaired growth or reproduction of cladocerans. Results were similar for the

amphipod *Hyalella azteca*. Chironomids survived and developed well in microcosms, with the exception of Wissous which reduced the weight of emerged adults. The comparison of observed effects and heavy metal nominal concentrations of the water column (table 9) suggests a causal relationship. As a matter of fact, Wissous and Chevire (fraction 2-30 mm) leachates, the most toxic, show several concentrations (for Cu, Ni, Zn) higher by a factor > 10 than threshold concentrations (PNEC). This is only an assumption, since these nominal concentrations are theoretical and must be related to a dissolved and a particulate fractions which have different meanings regarding exposure of organisms.

Table 8. Summary of results of microcosm assays (0 : no effect ; - : negative effect (inhibition, mortality, ...); + : positive effect (growth stimulation) ; nm : not measured)

Sediment	Leachate conc.	Duration (days)	pH	Conductivity	Algae Growth	Duckweeds growth (fronds)	final biomass
Wissous	10%	28	increases at 10 from D21 to D28	slight difference	?	+ 100%	
AhAh2 raw							
AhAh2 2-30 mm	10%	28	0	slight difference	0	+20%	+20 to +30%
AhAh2 60 µm-2 mm	12.5%		0	0	no data -	0	0
Cheviré 2-30 mm	25%	21	0	higher	according to	0	0
	50%		0	conductivity	pH	0	0
	12.5%		0	0	no data 0	0	0
Cheviré 60 µm-2 mm	25%		0	0	according to	0	0
	50%		0	0	pH	0	0
			0	0		0	0
RCE1			0		0	0	0
RCE2			0		0	0	0
RCE3			0		0	0	0
RCE4			0	Slightly higher	0	+ 70%	0
RCE8	10%	21	0	conductivity due to leachate	-	+ 100%	0
A1			0		0	0	0
A2			0		0	0	0
A7			0		0	0	0
S			0		0	0	0

Discussion and conclusion

The analysis of heavy metal contents of the 26 road sediments studied (raw materials and granulometric fractions for some of them) and their comparison with threshold values such as TEC or PEC (MacDonald *et al.*, 2000) indicates potentially dangerous materials, notwithstanding other contaminants (PAHs, PCB, pesticides, ...) which were not measured. The observations made on leachates produced from these sediments, according to the studied scenario (leaching of rainwater through the re-used materials

and transfer of the leachates to a close lentic ecosystem), show that, though only a low fraction of pollutants is leached, these effluents may be at risk for the ecosystem for dilution factor <100x.

Table 8 (continued). Summary of results of microcosm assays (0 : no effect ; - : negative effect (inhibition, mortality, ...); + : positive effect (growth stimulation) ; nm : not measured; EC50 : concentration that reduces by 50% the concerned end point)

Sediment	Daphnids			Amphipods			Chironomids	
	survival	growth	reproduction	survival	growth	survival	emergence	adult mass
Wissous	0	- (60%)	0 (but delay)	0	- (40%)	0	0	- (50 to 70%)
AhAh2 raw								
AhAh2 2-30 mm	0	0	0	0	0	0	0	0
AhAh2 60 µm-2 mm	0	0	0	0	0	0	0	0
Chevire 2-30 mm	0	0	EC50-19 d 35.6%	EC50-21 d 17.6%	EC50-21 d 14.6%	0	0	0
Chevire 60 µm-2 mm	0	0	0	0	0	0	0	0
RCE1 to RCE8	0	0	0	0	0	0	0	0
A1	0	0	0	0	nm	0	0	nm
A2	0	0	0	0	nm	0	0	nm
A7	0	0	0	0	0	0	0	nm
S	0	0	0	0	0	0	0	0

Table 9. Nominal contents (estimated from measured concentrations of centrifuged leachates) in heavy metals (particulate + dissolved) of microcosms water column (light grey : values > PNEC Ineris, dark grey : values > 10xPNEC)

Leachate/concentration (%)	Heavy metal contents of water column (µg/L)					
	Cd	Cr	Cu	Ni	Pb	Zn
Wissous 10%	2	18	213	28	190	1
AhAh2 raw 10%	<LD	<LD	1.28	1.85	0.57	<LD
AhAh2 2-30 mm 10%	<LD	<LD	0.23	0.42	0.13	<LD
AhAh2 60 µm-2 mm 10%	<LD	<LD	0.72	0.44	0.41	<LD
Chevire 2-30 mm 12.5%	0.09	0.24	22.34	2.06	8.75	75
Chevire 2-30 mm 25%	0.18	0.48	44.68	4.13	17.50	150
Chevire 2-30 mm 50%	0.35	0.95	89.35	8.25	35.00	300
Chevire 60 µm-2 mm 12.5%	<LD	0.03	3	0.15	3.25	18.75
Chevire 60 µm-2 mm 25%	<LD	0.05	6	0.30	6.5	37.5
Chevire 60 µm-2 mm 50%	<LD	0.10	12	0.60	13	75
RCE1 10%	0.20	1.33	2.33	0.98	2.11	/
RCE2 10%	0.05	0.42	1.66	0.33	0.81	/
RCE3 10%	0.08	0.45	4.63	0.23	1.15	/
RCE4 10%	0.06	0.79	1.25	0.68	0.87	/
RCE8 10%	0.15	2.15	30.7	1.57	4.73	0.05
A1 10%	0.08	0.55	5.81	0.99	2.53	42
A2 10%	0.10	1.58	18.9	2.83	9.71	98
A7 10%	0.19	0.47	2.87	0.68	1.27	21
S 10%	0.05	1.00	6.91	1.58	2.64	55

Ecotoxicological bioassays on leachates are helpful tools to improve this first appreciation, since they allow to predict effects on aquatic organisms living in such ecosystems. Microcosm assays are particularly relevant, as they are an attempt to better represent the complex environment (water column and sediment) and allow to measure effects on rather long exposure durations. The results of bioassays show that the leachates are generally not toxic or only slightly toxic at concentrations $\leq 10\%$. In the latter case, toxicity might be explained by heavy metals such as copper, nickel or zinc.

The analysis of physico-chemical composition of leachates by comparison with threshold values and the results of microcosm assays are convergent : in the first case, it seems that a dilution factor $\geq 100x$ is necessary to reach non dangerous metal concentrations, whereas the application of a uncertainty factor of 10 to microcosm assays results leads to propose a no chronic effect concentration (NOEC) of 1% (v : v) for such materials.

A secondary result of this study is that, on an ecotoxicological point of view, the granulometric fractioning does not seem to produce a valorizable fraction which would be systematically innocuous.

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