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## Influence of the vegetative cover on the fate of trace metals in retention systems simulating roadside infiltration swales

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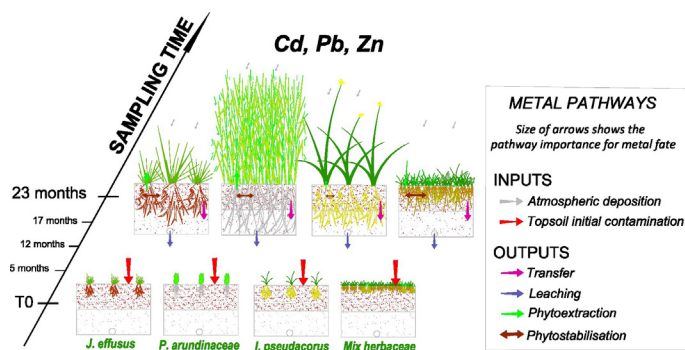
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### HIGHLIGHTS

- Mesocosms were designed to understand Cd, Pb, Zn pathways in urban roadside swales.
- Macrophytes and grass were studied for their influence on metals fate in mesocosms.
- Transfer of the initial stock of metals to water outflow was very low over two years.
- Metal mephytoextraction/phytoextraction were efficient using *P. arundinacea* or grass.
- Metal standing stocks must be considered instead of concentrations in plant tissues.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Large-scale outdoor mesocosms were designed and co-contaminated with metals (Cd, Pb, Zn) and organic compounds to better understand the complex functioning of urban roadside swale environments. Infiltration systems were planted with macrophytes (*P. arundinacea*, *J. effusus* and *I. pseudacorus*) or grassed, and natural or spiked target metals were monitored over two years. In the non-spiked mesocosms, atmospheric metal inputs were slightly higher than outputs, leading to low metal accumulation in topsoils and to very low outflow water contamination (<0.7% of the initial metal stock). In the spiked infiltration systems that simulated point pollution through water inflow, transfer of the initial stock of metals to the deeper soil layers was quite low and outflow water contamination was very low (<0.6% of the initial stock). The main metal output from these systems occurred in the first days of their installation because of the high metal solubility in water and insufficient plant cover at that time. The infiltration systems stabilized after a few weeks, probably because of stronger sorption to soil aggregates, and because of plant root development. Mephytoextraction in plant roots was more efficient in mesocosms planted with *P. arundinacea* and grass. Metal phytoextraction in plant aerial parts was also better for grass and *P. arundinacea*, when considering metal standing stocks instead of their concentration in plants. *J. effusus* was a good metal accumulator, but its low aboveground biomass development was less favorable to metal removal through harvesting.

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## 1. Introduction

The urban population, which accounted for 54% of the global world population in 2014, still keeps growing, leading to a sharp increase in impervious surfaces. Among the consequences on the environment, water quality alteration can be noted. In recently urbanized areas, new facilities are currently being developed to manage stormwater runoff, combining the necessity to better infiltrate and store total runoff volumes, but also to remediate pollutants through a variety of processes (Davis et al., 2012; Bressy et al., 2014; Kabir et al., 2014). Among the different types of bioretention facilities, swales have been historically designed to collect runoff from roadways (Dietz, 2007; Göbel et al., 2008; Stagge et al., 2012; Tromp et al., 2012). Swales are shallow drainage channels that receive road runoff laterally onto gentle side slopes and promote sedimentation and filtration through soil.

Metals and organic pollutants are often found together in urban runoff because they are discharged by the same or neighboring sources. In urban areas, sources of metals include traffic emission (vehicle exhausts, tire wear or weathered street surface particles, fluid leaks, corroded metal, etc.) and industrial emissions (power or chemical plants, steel industry, etc.) (Kabir et al., 2014; Jiang et al., 2015). Unlike organic pollutants, trace metals cannot be degraded and can, therefore, accumulate in the environment and in organisms. In the human body they can lead to toxic reactions even at very low concentrations.

Particle-phase pollutants accumulate on impervious surfaces through dry atmospheric deposition and dissolved contaminants accumulate through wet atmospheric deposition. Consequently, metal inputs reaching bioretention facilities can originate from runoff (mobilizing deposited contaminants from the road), but also from direct rainfall onto the soil surface (Leroy et al., 2016). Metals associated with particles are generally mostly trapped in topsoil aggregates, bonding to soil organic matter or soil clays (Li and Davis, 2008; Horstmeyer et al., 2016). Thus, topsoil quality in retention facilities can be degraded by metal accumulation. Unfortunately, immobilized metals can be released thereafter through chemical and biochemical dissolution reactions in the infiltration water. These dissolved metals can be partly assimilated by plants and microorganisms (Charlesworth et al., 2012; Horstmeyer et al., 2016). Finally, only a small portion of the metals entering the system leach to the deep soil along with the seepage of water (Kabir et al., 2014).

Plants can be incorporated into swales: they can favor the deposition of suspended solids with their sorbed metals because they contribute to reducing flow-rates; they can precipitate and accumulate metals on the surface of roots through iron plaques attached around them (phytostabilization) (Rahman et al., 2011); the roots can excrete protons, organic acids and chelating molecules to favor metal solubilization; microorganisms associated with the roots can also help in reducing metal toxicity through speciation modifications, increasing their uptake by plant roots, with the subsequent transport to the aerial parts (phytoextraction) (Upadhyay et al., 2016). A great variety of macrophytes can be used in humid zone systems such as roadside swales. They should be tolerant to flooding conditions, high organic and inorganic loadings, and adapted to local weather conditions and diseases (Gomes et al., 2014).

Swales are therefore complex systems with multiple reaction pathways at the water/soil-plant interface, and more knowledge is needed to better understand the mechanisms involved in the potential accumulation of metals in swale soils or the possible mass transfer of metals to infiltrated water. In order to better understand metal contaminants inputs, transfer and outputs, large-scale outdoor mesocosms were set up to simulate roadside swales, grassed or planted with three macrophytes (*Juncus effusus*, *Phalaris arundinaceae*, *Iris pseudacorus*). After their initial co-contamination with Cd, Pb, Zn and polycyclic aromatic hydrocarbons (PAHs) in May 2012, the evolution of metal contamination in the various compartments of the systems (surface soil 0–15 cm, deeper soil 15–30 cm, aboveground and belowground parts of plants, leachate)

was monitored for two years. The fate of PAHs has been described in a separate study (Leroy et al., 2015).

## 2. Experimental procedures

### 2.1. Chemicals and materials

Zn(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O (>98% purity), Cd(NO<sub>3</sub>)<sub>2</sub>·4H<sub>2</sub>O (>99.9% purity) and Pb(NO<sub>3</sub>)<sub>2</sub> (>99% purity) were purchased from Sigma-Aldrich (St Quentin-Fallavier, France), similarly to phenanthrene (98% purity), pyrene (98% purity) and benzo[*a*]pyrene (>96% purity). They were used to spike the soils of half of the mesocosms. Nitric acid (65%) and hydrochloric acid (37%), used for digestion, were obtained from Fisher Scientific (Illkirch, France).

Approximately 2.5 tons of a soil typical of the Normandy region (France) was collected (15–50 cm) from a suburban area of Rouen to fill outdoor mesocosms. This soil (pH = 7.8) was composed of 19.1% clay, 68.8% silt, 12.1% sand, contained an organic carbon content of 1.68%, and background levels of Cd (1.45 ± 0.02 mg kg<sup>-1</sup>), Pb (19.74 ± 0.63 mg kg<sup>-1</sup>), Zn (99.53 ± 1.28 mg kg<sup>-1</sup>) and total PAHs (0.43 ± 0.06 mg kg<sup>-1</sup>). Two-month old juvenile plants of *Juncus effusus*, *Phalaris arundinaceae*, *Iris pseudacorus* were obtained from Alisma (Taurignan-Castet, France) and a mix of grass seeds (25% *Festuca arundinacea*, 25% *Festuca rubra* and 50% *Lolium perenne*) from Gondian (Eure, France). These plants were therefore grown in the mesocosms.

### 2.2. Experimental setup and sampling

Four experimental mesocosms simulating grassed or vegetated swales were installed outdoors in Rouen, Normandy, North-West France (49°25'N; 1°4'E). The climate is temperate oceanic with a mean annual rainfall of 820 mm and a mean annual temperature of +10 °C. Fig. S1 (supporting information) gives the average monthly rainfall for the city of Rouen for 2012 and 2013.

Four large polypropylene tanks covered by a geomembrane were divided into two parts: a contaminated part (1.5 m × 0.9 m × 0.4 m) and a control part (1.2 m × 0.9 m × 0.4 m), and were respectively filled with 430 ± 2 kg dry weight (DW) and 350 ± 2 kg DW of a 30-cm thick silty-loam soil (Fig. 1). A drainage outlet was installed on the bottom of each tank, surrounded by a layer of gravel to avoid clogging of the pipe (Leroy et al., 2015).

Young plants of *J. effusus* (mesocosm M<sub>1</sub>), *P. arundinaceae* (mesocosm M<sub>2</sub>), *I. pseudacorus* (mesocosm M<sub>3</sub>) were planted in May 2012 (11 plants m<sup>-2</sup>) and acclimatized for 30 days. Mesocosm M<sub>4</sub> was sown at the same time using a grass mix. Then, parts of the mesocosms were spiked with a mix of PAHs and metals uniformly spread on the top of the soil to contaminate the surface, just like roadside swales. Three PAHs were introduced at a mass concentration of 10 mg kg<sup>-1</sup> each (approximately two orders of magnitude above the PAH background level) (details of the spiking process are given in Leroy et al., 2015). To make sure that the contamination was homogeneous over the complete mesocosm, the contaminated parts were divided into 24 cells. Then, 330 mL of deionized water containing Zn, Pb, and Cd nitrates (76.1 g L<sup>-1</sup>, 8.9 g L<sup>-1</sup> and 0.3 g L<sup>-1</sup> respectively) were spread on each cell. Cd, Pb and Zn were spiked to reach concentrations of 2, 100 and 300 mg kg<sup>-1</sup>, respectively, which are the limits given in the Council Directive 86/278/EEC to regulate the use of contaminated sewage sludge in soils for agriculture. These contamination levels correspond to soil concentrations reached if contaminants diffuse uniformly from the surface to the whole soil volume.

Two weeks after spiking, soil contamination was measured in each mesocosm (artificially contaminated parts and non contaminated parts) to obtain the initial contamination level (t<sub>0</sub> = June 2012). Ten soil samples from each mesocosm, obtained using a 2.5-cm inside diameter core sampler, were mixed to get a composite sample at 0–15 cm and 15–30 cm soil depths. Soil sampling was performed 5, 12, 17 and

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