VALSOLINDUS, A DEMONSTRATION PROJECT OF SEDIMENT VALORISATION

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ABSTRACT

The VALSOLINDUS project aimed at validating the beneficial reuse in landscaping projects of dredged waterways sediments previously processed by mineral separation in the Solindus project. Experimental plots with increasing proportion of a processed sediment in an agricultural soil (0, 50 and 100% vol/vol) were set up and sown with ryegrass. The feasibility of the reuse of sediment was evaluated through (1) the assessment of habitat function of the re-constituted soils and (2) the assessment of environmental risks. For 18 months, plant growth and biomass on the plots were monitored and ecotoxicity tests on *Eisenia fetida*'s reproduction and on nitrifying bacteria were realized. Contaminant concentrations in re-constituted soils and heavy metal concentrations in water and plants were also analyzed. The experiment showed landscaping projects are a potential solution for sediment valorisation. Risks for grazers are low. Worms' reproduction decreased with sediment incorporation but no mortality was observed. No toxicity on nitrifying potential was observed either.

Keywords: Sediment, Field Experiment, Ecotoxicity, Lolium

INTRODUCTION

In Wallonia, dredged sediments management is regulated by two regional decrees from 1995 and 2001, which did not allow other operational solutions than dredging and landfilling to emerge so far. This resulted in important delay in dredging operations and in high costs related to sediment disposal. Moreover, the shortage of land for disposal is becoming an increasingly important problem. It is therefore necessary to demonstrate the feasibility of cost-effective valorisation solutions for sediments to favor sustainable regulation emergence and population acceptance of those practices [1]. In the Valsolindus project, the potential of landscaping management for sediments treated by the Solindus mineralurgical process has been assessed through a field experiment. Landscaping is a cheap and easy to implement solution, and it would allow the recycling of large sediments volumes in the frame of circular economy.

Here, we present the results from 4 field plots where ryegrass has been grown on an artificial sediment (dredged in the Scheldt river) incorporated into an agricultural soil. The specific objective of the study was to demonstrate the feasibility of the reuse of sediment in landscape management applications through (1) the assessment of habitat function of the re-constituted soils and (2) the assessment of environmental risks.

MATERIALS & METHODS

Experimental design

Four plots of 4 m² and 60 cm depth were settled on waterproof HDPE membrane on a site located in Châtelet, in central Belgium. The site is a former industrial mound.

Increasing proportions of sediment in an agricultural soil (0-50-100% vol/vol) were established. Labelling of the results is subsequently referred to as "T" and "T2" for the control plots (agricultural soil only), "S50" for 50% sediment incorporation in agricultural soil and "S100" for 100% sediment.

The agricultural soil was sampled in a potato collection center and used as control soil for the experiment after verification of compliance with the Walloon reference values for an agricultural use of the Soil Decree of 2008.

In order to get enough volume for the field experiment, an artificial sediment was re-constituted from different fractions (15-63 µm and 63-250 µm) of two sediment batches dredged in the Scheldt river. Its heavy metal and PAHs concentrations were below the maximum acceptable values (TMA) of the Walloon government agreement of November 30th 1995, classifying it as "non-polluted" sediment.

Tornado 2 (Barenbrug), a mixed variety of Italian and hybrid ryegrass (80% *Lolium multiflorum* + 20% *Lolium hybridum*), was sown at the start of the experiment, in May 2014. Neither irrigation nor weeding was performed on the plots. During the winter, excess water was pumped into 1m³ tanks.

Three months after sowing (July 2014), plant biomass was harvested and dry biomass production was compared and analysed for inorganic element concentrations. Plant and insect colonization was also recorded. Composite soil samples were collected to analyze soil heavy metal and organic component concentrations as well as agronomic parameters.

The same assessment was made after one year of cultivation (August 2015).

Soil, water and ryegrass analyses

Soil analysis

Three soil samples were collected from the 0-10 cm top layer in each experimental plot at the start of the experiment, prior to sowing. For following soil sampling (at harvesting in summer 2014 and summer 2015), composite samples were taken.

Soil concentrations of Cd, Cr, Cu, Ni, Pb and Zn were determined by ICP-OES (ISO 22036, 2008), Hg by CVAFS (ISO 16772, 2004), following aqua-regia digestion (ISO 12914, 2012). PAHs concentrations were quantified using UPLC (ISO 13859, 2014). Hydrocarbon Index was also determined (ISO 16703, 2004) after ASE extraction. Dry matter content was analysed according to ISO 11465 (1993).

For the evaluation of the nutrient status in soil, composite samples were taken at 0-10 cm depth. Tot-N was analysed according to ISO 13878 (1998) and tot-C according to ISO 10694 (1995). Available phosphorus, potassium and sodium were analysed with an ammonium acetate EDTA solution (pH 4,65) according to CMA/2/IV/6 (Flemish regulation, 1994). CEC was also measured (ISO 23470, 2007).

Ryegrass analyses and BCFs

In July 2014 and August 2015, ryegrass was harvested at 5 cm above the ground on a 1m² area in the center of each plot. Ryegrass was let to dry in ambient air during 10 days. Dry and milled ryegrass samples were digested by aqua-regia in a closed microwave-assisted system. Metals (Cu, Ni and Zn) were quantified by ICP-OES (ISO 22036, 2008).

Water analyses

Exceeding water in the plots was pumped once a week and stocked in 1m³ water tanks. Before water elimination through the Solindus treatment platform, trace metals concentrations and turbidity in water were controlled for compliance with Solindus environmental permit. Cd, Cr, Cu, Ni, Pb and Zn concentrations were determined by ICP-OES (ISO 22036, 2008), Hg by CVAFS (ISO 16772, 2004), following aqua-regia digestion (ISO12914, 2012).

Plant and insects colonization

Plant and insect colonization was assessed throughout the summer and recorded the day of harvesting in summer 2014 and summer 2015.

Ecotoxicity tests

Composite soil samples for ecotoxicity testing were taken at 0-20 cm depth at the same time than soil samples (prior to cultivation, at harvesting in summer 2014 and in summer 2015).

Nitrification potential

Toxicity on nitrification potential was studied using a simple 6h incubation test (ISO 15685, 2012). This test consists of blocking nitrate formation by sodium chlorate addition. The resulting nitrites accumulation is then measured over a 6h period to assess potential activity of nitrifying micro-organisms. Nitrites were analysed by segmented flow analysis (ISO 14256-2, 2005).

NH_4^+ oxidation rate (ng NO_2-N/g $_{\text{dry weight}}$.h) is calculated by the difference between NO_2-N concentrations at 2 and 6h. Tested soils are considered as toxic if NH_4^+ oxidation in the mixture is inferior to 90% of the average oxidation activity of both soils (tested and control) taken separately , following the equation:

$$\bar{A}_m + s_m < 0{,}9.\frac{(\bar{A}_c + \bar{A}_p)}{2} \qquad (1)$$

where $\bar{A}_m$ is the average NH_4^+ oxidation activity in the mixture ; s_m is the standard deviation of the NH_4^+ oxidation activity in the mixture ; $\bar{A}_c$ and $\bar{A}_p$ are the average NH_4^+ oxidation activities in the reference soil and the tested soil respectively.

The reference soil was sampled in an agricultural soil in Ath.

Toxicity on Eisenia fetida*'s reproduction*

Worm *Eisenia fetida*'s reproduction toxicity test was implemented following the standard procedure (ISO 11268-2, 2012). Prior each test, worms were acclimated for 3 days in artificial soil, considered as the control soil. 40 worms (10 worms in 4 replicates) were put in glass beakers filled with 500 g $_{\text{dry weight}}$ of samples to be tested, and of artificial soil with increasing boric acid concentrations for the calibration curve. The worms were nourished twice a week during 4 weeks. pH and moisture were controlled. After 4 weeks, adult worms were removed. Cocoons were counted and replaced in the beakers for another 4 weeks. pH and moisture were controlled but no more food was added. At the end of the 8th week, juvenile worms were counted placing the beakers in water bath at 50°C to make the worms reach the surface.

The studied endpoints are worms' mortality and fecundity.

Risks of environmental contamination

The translocation of metals from tested substrate to plants was evaluated using bioconcentration factors (BCFs) using the following equation [1]:

$$BCF = C_{biomass}/C_{soil} \quad (2)$$

RESULTS & DISCUSSION

Chemical analyses

Chemical compositions of field plots at the beginning of the experiment and after one year of cultivation are reported in Table 1 and Table 2, respectively. Results before cultivation refer to triplicates and are presented as mean and standard deviation. Results after one year of cultivation apply to composite samples. Dry weight of soil samples in Table 2 was measured in triplicates from a single sample.

Tested substrates contaminant concentrations show exceedance of Belgian threshold values (VS) for a recreational use of the Soil Decree of 2008 for Zn, Benzo(b)fluoranthene, Benzo(a)pyrene and Indeno(1,2,3-c,d)pyrene in S100 and for Zn in S50.

Cation Exchange Capacity (CEC), organic nitrogen (N_{org}) and potassium (K) are found to be low in the sediment as compared to the agricultural soil (T and T2), which is likely to have an adverse effect on plant growth. pH values are similar amongst plots and over time.

Tested substrates chemical composition doesn't seem to evolve significantly over time.

Risk for the environment

Metal concentrations in plants and BCFs

Metal concentrations in ryegrass are presented in Table 3. Cu and Zn concentrations are in the range of normal concentrations in grasses [4] whereas Ni is found in higher concentrations even in the control plots (T and T2).

BCFs were well below the unit for Cu, Ni and Zn for all plots (Table 4), showing no accumulation of these metals in ryegrass and limited risk to grazers [1].

Metal concentrations in water

Table 5 gives metal concentrations analysed in excess water before emptying the tanks in January and in March 2015.

Only Zn concentrations are beyond quantification limit. However Zn concentrations decrease rapidly with time (from 47 to 7 µg/l between January and March 2015). Zn being a mobile element [2], [3] and the most abundant inorganic element in sediments, it is not surprising to detect it in excess water.

Table 1 Soil concentration for plots T, T2, S50 and S100 at the beginning of the experiment (May 2014)

Before cultivation					
Items	Units	Plot T	Plot T2	Plot S50	Plot S100
Cd	mg/kg_{dw}	<0,500	<0,500	1,45 ± 0,05	2,65 ± 0,67
Cr	mg/kg_{dw}	23,1 ± 6,1	25,0 ± 0,23	23,8 ± 2,4	19,4 ± 2,8
Cu	mg/kg_{dw}	12,5 ± 0,50	11,6 ± 0,26	15,0 ± 0,55	15,2 ± 2,85
Hg	mg/kg_{dw}	0,05 ± 0,01	0,05 ± 0,00	0,13 ± 0,01	0,30 ± 0,09
Ni	mg/kg_{dw}	17,6 ± 0,66	16,6 ± 0,05	12,1 ± 1,03	8,39 ± 1,40
Pb	mg/kg_{dw}	31,33 ± 5,35	34,4 ± 0,22	94,39 ± 5,57	137,87 ± 30,35
Zn	mg/kg_{dw}	89,7 ± 5,76	106 ± 0,51	280 ± 10,3	461 ± 122
Σ16 PAHs	mg/kg_{dw}	1,6 ± 0,4	2,4 ± 0,1	5,1 ± 0,6	6,9 ± 0,1
HC index	mg/kg_{dw}	<20	30 ± 11	49 ± 4	113 ± 45
dw	%	85,9 ± 1,9	83,41 ± 0,06	86,58 ± 1,8	81,60 ± 0,50
pH-KCl	-	7,2	7,5	7,5	7,5
C_{org}	%	1,29	1,15	1,63	1,61
N_{org}	‰ N	1,09	1,15	0,66	0,34
P	mg/kg_{dw}	11,3	9,86	12,9	14,5
K	mg/kg_{dw}	20,7	26,3	10,3	4,17
CEC	meq/100g	15,6	10,8	7,61	4,30

Table 2 Soil concentration for plots T, T2, S50 and S100 after 1 year of cultivation (August 2015)

Items	Units	Plot T	Plot T2	Plot S50	Plot S100
Cd	mg/kg_{dw}	<0,500	<0,500	1,11	2,54
Cr	mg/kg_{dw}	23,3	25,4	22,7	15,0
Cu	mg/kg_{dw}	11,7	11,7	12,8	18,7
Hg	mg/kg_{dw}	0,055	0,055	0,110	0,253
Ni	mg/kg_{dw}	17,2	16,9	15,1	7,85
Pb	mg/kg_{dw}	28,4	34,7	53,6	159
Zn	mg/kg_{dw}	85,8	108	165	469
Σ16 PAHs	mg/kg_{dw}	1,32	2,36	4,20	6,30
HC index	mg/kg_{dw}	13	18	36	44
dw	%	88,06 ± 0,09	83,09 ± 0,13	87,72 ± 0,13	86,54 ± 0,55
pH-KCl	-	7,6	7,6	7,7	7,8
C_{org}	%	1,41	1,11	1,53	1,80
N_{org}	‰ N	1,16	1,08	0,75	0,32
P	mg/kg_{dw}	9,25	10,70	4,63	4,48
K	mg/kg_{dw}	19,10	19,20	11,70	4,26
CEC	meq/100g	13,18	11,87	8,23	4,31

Table 3 Trace Element Concentrations in plant biomass after 1 year of cultivation

Element	Unit	T	T2	S50	S100	Normal concentrations in grasses [4]
Cu	mg/kg_{dw}	4,52	3,87	9,93	4,12	7,4-15
Ni	mg/kg_{dw}	2,83	1,73	3,56	5,16	0,9-1,3
Zn	mg/kg_{dw}	<50	<50	64	107	15-80

Table 4 Biomass Concentration Factors (BCFs) for Cu, Ni and Zn after 1 year of cultivation

Element	T	T2	S50	S100
Cu	0,36	0,33	0,66	0,27
Ni	0,16	0,10	0,29	0,62
Zn	-	-	0,23	0,23

Habitat function

Plant biomass yield

Plant biomass yield was assessed by dry matter harvested in a 1m² area (Table 6). After one year of cultivation, dry biomass on sediment plots S50 and S100 is inferior to control plots. It was also observed that ryegrass showed a higher density and a greener colour on control plots than on S50 and S100 plots.

This could be explained partly by nitrogen deficiency in the sediment. Amendment addition should be considered in landscaping projects in order to balance sediment nutriments (N, P, K).

Table 5 Metal Concentrations in water cisterns in January 2015 and March 2015

Element	01/2015 (µg/l)	03/2015 (µg/l)
Cd	<2,5	<2,5
Cr	<5	<5
Cu	<5	<5
Hg	<0,01	<0,0025
Ni	<5	<5
Pb	<5	<5
Zn	47 ± 12	7 ± 2

Table 6 Plant biomass yield

(g_{dw}/m^2)	T	T2	S50	S100
July 2014	110	-	130	100
August 2015	299	408	179	85

Flora and insect colonization

Flora colonization of the experimental plots was found to be low. Higher presence of orthoptera and locusts was observed in S50 and S100 plots. Those organisms were almost absent in control plots (T and T2) whereas the latter were distinguished by abundance of gastropods. However this is probably biased by the low biodiversity of the site, a former industrial field, which is not representative of Walloon soils.

Nitrification potential

Results of nitrification potential tests show no toxicity for any of the four plots at any stage of the experiment. Nitrification potential of the mixture is not lower than 90% of the average potential of the reference soil and the soil tested taken separately.

Nitrification activity in sediment plots (S50 and S100) is lower than in the control plot (T). This observation could be explained by the low nitrogen concentrations in sediment plots. However, when in contact with the reference soil and its microbial flora, nitrification is not inhibited.

Toxicity on Eisenia fetida*'s reproduction*

One endpoint is mortality. Tested concentrations in the artificial soil were not high enough to note adult mortality. No mortality was observed in tested substrates either.

The second studied endpoint is fecundity (number of juveniles produced). Results of the test for the samples taken at the beginning of the experiment (May 2014) are presented in Fig. 1 and Fig. 2. The same conclusions can be drawn from the samples taken subsequently.

In artificial soil, number of juveniles decreases with increasing boric acid concentration and reaches zero around 300 mg boric acid/kg_{dw}. Juvenile production decreases with sediment incorporation, virtually reaching zero in S100.

Juvenile production in the control plot T is lower than in the artificial soil (0 mg boric acid/kg_{dw}). This allow us to think that physical features have great influence on earthworms, as it has been observed in several former studies [5], [6]. During the experiment, it was observed that adult worms tried to escape the recipients. Sediments present fine particulates and a high retention capacity, possibly asphyxiating worms. Humidity between 40 and 60% of water retention capacity is recommended for the test. Here it was settled at 45%.

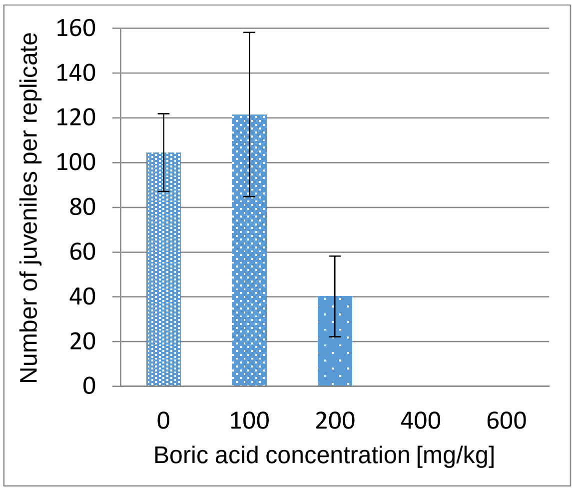

Fig. 1 Number of juveniles in controls (May 2014)

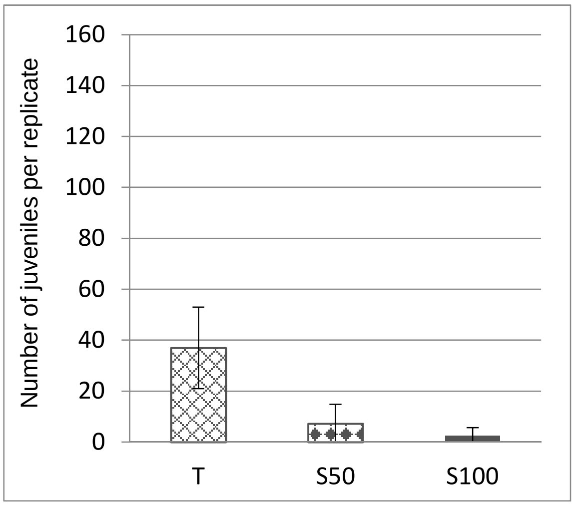

Fig. 2 Number of juveniles in field plots (May 2014)

CONCLUSION

The Valsolindus field experiment demonstrated landscaping projects are a potential solution for sediment beneficial reuse. Chemical composition of the substrate samples doesn't seem to evolve significantly over time. Assessed risks for the environment by transfer through grass were low. There was no toxicity on nitrifying potential. Even if worms' reproduction decreased with increasing sediment incorporation, no mortality was observed. Plant growth and biomass yield decreased with increasing sediment proportions probably due to sediment nutriments (N, K) deficiency. Amendment should be considered to ensure a balanced N, P, K nutriments equilibrium when valorizing sediments on soils.

In conclusion, landscaping allows beneficial reuse of important volume of sediments that would otherwise be disposed of in confined landfills, an expensive and not sustainable solution.

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REFERENCES

[1] Enell A *et al*., "Risk management and regeneration of brownfields using bioenergy crops", J. Soils Sediments, Vol. 16 (3), Mar. 2016, pp 987-1000.

[2] Singh SP *et al*., "Trace Metal Leachability of Land-Disposed Dredged Sediments", J. Environ. Qual. Vol 29 (4), 2000, p 1124-1132.

[3] Néel C *et al*., "Mobility and bioavailability of Cr, Cu, Ni, Pb and Zn in a basaltic grassland: Comparison of selective extractions with quantitative approaches at different scales", Appl Geochemistry, Vol 22(4), 2007, pp 724-735.

[4] Kabata-Pendias A, Pendias K, "Trace Elements in Soils and Plants", Boca Raton: CRC Press, 2011, chap. 15-16-17.

[5] Kumpiene J *et al.*, "Selecting chemical and ecotoxicological test batteries for risk assessment of trace element-contaminated soils (phyto)managed by gentle remediation options (GRO)", Sci. Total Environ., Vol. 496, Aug. 2014, pp. 510-522.

[6] Demuynck S *et al*., "Effects of field metal-contaminated soils submitted to phyto-stabilisation and fly-aided phytostabilisation on the avoidance behavior of the earthworm *Eisenia fetida*", Ecotoxicol. Environ. Saf., Vol. 107, 2014, pp. 170-177.