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Field evaluation of industrial non-food crops for phytomanaging a metal-contaminated dredged sediment

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Abstract

Phytomanagement is a concept fit for a bio-based circular economy that combines phytotechnologies and biomass production for non-food purposes. Here, ten annual and perennial industrial non-food crops (*Sorghum* Biomass 133, *Sorghum* Santa Fe red, *Linum usitatissimum* L., *Eucalyptus* sp., *Salix* Inger, *Salix* Tordis, *Beta vulgaris* L., *Phacelia tanacetifolia* Benth., *Malva sylvestris* L., and *Chenopodium album* L.) were studied under field conditions for phytomanaging a (Cd, Cu, Pb, and Zn) metal-contaminated dredged sediment in the North of France. The crops were selected according to their relevance to pedoclimatic and futur climatic conditions, and one or more non-food end-products were proposed for each plant part collected, such as biogas, bioethanol, compost, natural dye, ecocatalyst, and fiber. Based on the soil-plant transfer of metals, eight out of the crops cultivated on field plots exhibited an excluder behavior (Bioconcentration Factor, BCF < 1), a trait suitable for phytostabilization. However, these crops did not change the metal mobilities in the dredged sediment. The BCF < 1 was not sufficient to characterize the excluder behavior of crops as this factor depended on the total dredged sediment contaminant. Therefore, a BCF group ranking method was proposed accounting for metal phytotoxicity levels or yield decrease as a complementary way to discuss the crop behavior. The feasibility of the biomass-processing chains were discussed based on these results and according to a survey of available legislation in standard and scientific literature.

Keywords: Bioconcentration Factor, phytoremediation, Cd, Zn, Cu, Pb, metal exposure

1) Introduction

In the European Union (EU), the Green Deal plan set in 2019 aimed at a sustainable neutral carbon economy by 2050 (European Commission communication of 11th December 2019). Soil as a resource and the remediation of degraded soil were recognized as significant points for the success of the European Green Deal since soil provides ecosystem services and highly contributes to carbon storage, bio-based circular economy, and human health (European Commission communication 17th November 2021). In 2018, in Europe, around 650,000 sites were reported to have polluting activity, and 125,000 need to be remediated (Pérez and Rodríguez-Eugenio 2018).

The management of contaminated sites with phytotechnologies was thought of as an *in situ* alternative to physical and chemical soil clean-up (soil excavation, soil washing, etc.), which have high environmental and economic costs, decreasing their suitability for large areas (Robinson et al. 2007; Robinson et al. 2009; Puschenreiter et al. 2009, Bert et al. 2009; Kidd et al. 2015; Ali et al. 2013; Gong et al. 2018). When metals are the contaminants, phytotechnologies can be applied as a partial clean-up technology in a phytoextraction option or as a partial sequestration technology in a phytostabilization option (Moreira et al. 2021). Usually, metal phytoextraction consists of the growth of either accumulator (Bioconcentration Factor, BCF : ratio between [metal] in plant parts and total [metal] in soil- $BCF > 1$, Translocation Factor, TF: ratio between [metal] in shoots and [metal] in roots- $TF > 1$) or hyperaccumulator ($BCF > 10$, $TF > 1$) plant species with high metal concentrations in the aboveground biomass, which result in the progressive clean-up of the topsoil (mainly the phytoavailable metal fraction) by the repeated export of the harvested biomass (Meers et al 2010; Tangahu et al. 2011; Conesa et al. 2012; Chalot et al. 2012; Delplanque et al. 2013; Tang et al. 2019; Grignet et al. 2020; Moreira et al. 2021). Phytostabilization, on the other hand, consists of the cultivation of metal-excluder plants ($BCF < 1$, $TF < 1$), lowering the metal transfer and pollutant linkages in the environment as the interaction of root systems changes the bioavailable metal fraction to a less-available one, reducing the metal mobility in the soil (Robinson et al. 2009; Garbisu et al. 2020).

Contrary to widely used remediation techniques, phytotechnologies are adapted to large areas and produce plant biomass, having similar practices to agriculture inducing related cost (Cundy et al. 2016; Kidd et al. 2015; Kumpiene et al. 2014). To alleviate costs and to allow the needed long-term management of the site, phytomanagement therefore has additional aims of producing an income on the site by processing the biomass in environmentally and economically sustainable local chains while maintaining/enhancing soil ecosystem services

(Robinson et al. 2009; Meers et al. 2010; Burges et al. 2018; Moreira et al. 2021). Due to plant species diversity and their characteristics, the biomass obtained during the site phytomanagement could be used to produce energy (Cheng et al. 2016; Delplanque et al. 2013; Meers et al. 2010; Bert et al. 2017 a; Pogrzeba et al. 2019; Perlein et al. 2021 a) or as a raw material for bio-based products (Deyris et al. 2018; Grignet et al. 2020; Ziegler-Devin et al. 2019; Asad et al. 2017; Zhejzakov et al. 2008; De Vos et al. 2022; Perlein et al. 2021 b, c).

However, phytomanagement has some limits, and studies are still in progress to address them and enable the uptake of this remediation solution by site managers/owners (Burges et al. 2018; Moreira et al. 2021). Whereas pot experiment results are difficult to extrapolate to field conditions, field experiments are still rare (Moreira et al. 2021). In addition, plant relevance for phytomanagement cannot be generalized and should be studied for each site accounting for its pedoclimatic and agronomical conditions, type and level of soil contaminants, and possibilities for local biomass processing (Zine et al. 2020). Therefore, plant selection for phytomanaging a specific site should be based on a holistic approach with on-field results to confirm its potential and/or identify its limits. Figure 1 proposes a base of four main factors and sub-sections to be studied for selecting a plant species for a phytomanagement solution. The approach shows the importance of field results to support crop selection (Fig. 1).

Based on Fig. 1 and a previous field experiment performed on a metal-contaminated sediment landfill site, this work aimed to enlarge *in situ* scientific knowledge by assessing a set of 10 annual and perennial industrial non-food crops (*Linum usitatissimum* L., *Eucalyptus* sp., *Beta vulgaris* sub sp. *vulgaris* L., *Phacelia tanacetifolia* Benth., *Malva sylvestris* L., *Chenopodium album* L., two cultivars of *Sorghum bicolor* L. (Biomass 133 and Santa Fe red), and two cultivars of *Salix* sp. (Inger and Tordis) for the site phytomanagement (Table 1). The tested crops were selected for their potential non-food processing (biogas, bioethanol, ecocatalyst, essential oil, natural dye, and fiber production) and relevance to pedoclimatic conditions. By taking into account the potential heterogeneity of the site contamination, the first objective was to study the relevance of the chosen crops for phytotechnologies according to their metal behavior (metal concentrations in the aboveground part, BCF), tolerance (yield and vegetation cover), and the crop effect on the extractable metal concentrations and metal mobility in the topsoil. Based on the BCF results, we proposed a BCF group ranking method to classify more precisely the crop metal behavior. Regarding the metal concentrations in the plant parts of the crops, suitable processing options were proposed according to a survey of available legislation in the standard and scientific literature.

2) Material and Methods

2.1) Site description and experiments set up

The 1 ha experimental site located at Fresnes-sur-Escaut (Hauts-de-France, France; 50°25'41.1" N 3°35'01.9" E, Fig. 2) is a former dredged sediment deposit site characterized by a metal contamination (Table 2). From 1978 until 1988, the site received dredged metal-contaminated canal sediment. Previous studies (Perlein et al. 2021 a,b) already evidenced that extractable and total concentrations of Cd, Cu, Pb, and Zn in the dredged sediment (Table 2) were the main traits characterizing its contamination, so a focus was given to these elements in this study. To study the soil-plant metal transfer under field conditions, two micro-plots experiments were set up in 2019 and 2020, one with seven crops (*B. vulgaris* cv. Dickens, *L. usitatissimum*, *P. tanacetifolia*, *M. sylvestris*, *S. bicolor* cv. Biomass 133, *S. bicolor* cv. Santa Fe red, and *Eucalyptus* sp. clone 645) on three areas and one with willows (Inger and Tordis) cultivated on site since 2012 as a very short rotation coppice (VSRC) (Fig. 2, Table 1). The climatic conditions on the site were characterized by similar rainfall of around 627 mm in 2019 and 2020, and mean temperatures of 12°C in 2019 and 12.6°C in 2020 (Supplemental Fig. 1). Infoclimat.fr indicated that the annual mean temperature calculated for 29 years (1981-2010) was 1.2-1.8°C above normal and the rainfall was 16% below normal.

2.1.1) Micro-plot experiment with annual crops

In April 2019, three areas of 225 m² each (Fig. 2: area 1, area 2, and area 3) were defined according to previous studies (Perlein et al. 2021a,b). The three areas, which showed different dredged-sediment metal levels (Table 2), were surrounded by a 2 m high fence buried 50 cm below ground to protect crops from herbivory during cultivation. The annual crops (*B. vulgaris*, *L. usitatissimum*, *P. tanacetifolia*, *M. sylvestris*, *Sorghum* Biomass 133, and *Sorghum* Santa Fe red) were selected according to their potential processing in the non-food sector and potential growth capacity in the Hauts-de-France region (Table 1). The sowing and planting period (Table 1) and the experimental design (sowing density, space between seedling ligne and seeds, space between tree; Supplemental Fig. 2) were derived according to agricultural recommendations to be as close as possible to classic cultivation conditions. During cultivation, mechanical weed control was performed to limit the impact of weeds on plant growth.

In 2019, the first sowing was performed. For each area, 25 m² were sown with *B. vulgaris* (Sugarbeet, cv. Dickens from Deleplanque, <https://www.deleplanque.fr>) (Table 1). In spring 2020, in each area, five new plots of 25 m² were cultivated according to a sowing design (Table 1, Supplemental Fig. 2) with either *L. usitatissimum*

(kindly provided by a local farmer), *P. tanacetifolia* (seed origin Graines de Semences, <https://www.graines-semences.com>), *M. sylvestris* (seed origin Graines de Semences, <https://www.graines-semences.com>), *Sorghum* Santa Fe red (seed origin Kokopelli, <https://kokopelli-semences.fr>) or *Sorghum* Biomass 133 (seed origin Semence de Provence, <https://www.semencesdeprovence.com>). Different plant parts were collected (Table 1) and analyzed for their metal concentrations according to the local processing chain options. In addition, control plants (*L. usitatissimum*, *P. tanacetifolia*, *Sorghum* Biomass 133, *Sorghum* Santa Fe red, and *M. sylvestris*) were cultivated on 1 m² plots in a garden near the experimental site (50°25'43.5 N, 3°33'16.5 E), and similar plant parts were collected to obtain common metal concentrations.

2.1.2) Micro-plot experiment with perennial crops

Two willow cultivars (Inger: *Salix triandra* x *S. viminalis* and Tordis: *Salix schwerinii* x *S. viminalis*) were initially planted in June 2012 on the site (Fig. 2). For that, 11,110 cuttings (Inger: 5566 and Tordis: 5544) of willows were planted following a planting design that consisted of 21 lines of double ranks of cutting (0.70 m) with lines spaced by 1.5 m. After seven years of growth, in January 2019, the willows were harvested, allowing the plants' regrowth. After this harvest, four areas, two by cultivar (Inger A, Inger B, Tordis A, Tordis B), were defined on the site with 12 trees per area (Fig. 2). In these areas, leaf metal concentrations were monitored for three years (2019, 2020 and 2021) in July to study their changes with elapsed time and, consequently, the potential processing of willow leaves in the industrial non-food sector according to these leaf metal concentrations (Table 1).

In April 2019, 9 trees in clods (30 cm height) of *Eucalyptus* sp. clone 645 (*E. gundal*: *E. gunnii* x *E. dalrympleana*, kindly provided by the Technologic Institute FCBA, Champs-sur-Marne, France, Melun et al. 2016) were planted on 25 m² plots on the three areas (Fig. 2) according to the planting design (Supplemental Fig. 2). The FCBA selected this clone to be tolerant to the lowest temperatures in the South of France (Melun et al. 2016).

2.2) Sediment sampling and analysis

On the three areas defined in the micro-plot experiment with the cultivated annual crops and for each plot, two dredged-sediment sampling campaigns were done, one before sowing and the second at harvest. A hand auger allowed the dredged-sediment sampling of the first 20 cm of depth. The sampling before sowing consisted of the collection of five random points per plot. At the harvest, ten random points were collected per plot. Each dredged-sediment sample was dried until constant weight in a forced-air oven (250 g of fresh dredged-sediment,

40 °C). The dry dredged-sediment (DS) samples were mechanically ground (Retsch BB51, Retsch GmbH, Germany) and sieved to < 2 mm. On the prepared DS, an ammonium nitrate extraction of metal was performed following the NF ISO 19730:2008 (E), in which 25 mL NH₄NO₃ (1 M) was mixed with 10 g of DS and shaken for two hours. The mixture was filtered (0.45 µm, Millipore, USA) to obtain the leachate containing the extractable metal fraction and acidified until a pH of 2 (HNO₃, 69%). This measure was not performed in the case of *C. album*, this species being a colonist and not a cultivated annual crop.

As described in Perlein et al. (2021 a), for each area, a dredged-sediment composite was created by mixing 50 g of fresh dredged-sediment per sample collected before sowing or planting. On these composites, agronomic parameters were determined. After a preparation step (drying at 40 °C; agate ball mill grinding at 250 µm, Laval lab Pulverisette 6, Laval lab, Canada), acid digestion was performed in triplicate (0.2 g of DS, 6 mL of hydrofluoric acid [HF] 48%) in a microwave digester (Mars 2 Xpress CEM, CEM, USA). These solutions were completed with boric acid (5.5%) to neutralize the HF and with Milli-Q water until 50 mL and were filtered at 0.45 µm (hydrophilic Teflon). The metal concentrations in digestates were analyzed either by inductive-coupled plasma optical emission spectrometry (ICP-OES, Agilent 5100, Agilent Technologies inc., USA) or inductive-coupled plasma mass spectrometry (ICP-MS, Agilent 7500, Agilent Technologies inc., USA) depending on the concentration range in the dredged-sediment sample. One standard reference material was used for analytical quality control (NIST SRM 2710, Montana soil, National Institute of Standards & Technology, USA). For all of digestion sets, recoveries were between 80% and 93% for Cu, 95% and 98% for Pb, and 96% and 104% for Zn in the reference sediment. For Cd, no recoveries could be calculated, as the results obtained from ICP-OES analysis were below the limit of quantification (LoQ).

The 1M NH₄NO₃-extractable and total metal concentrations in the dredged-sediment were used to calculate the % metal mobility, with % mobility = [extractable metal concentration in dredged-sediment / total metal concentration in dredged-sediment] * 100.

2.3) Plant sampling and analysis

At harvest on the micro-plot set up with annual crops, the bottom of aboveground part of the plants was cut with a hand pruner or directly pulled out from the sediment. Ten plants were randomly sampled simultaneously to the sediments per area and plot. After harvest, all of the fresh biomass (FW) was weighed with a portable hand scale (OHAUS defender 3000 inox, OHAUS, Switzerland) to obtain the yield per area. Leaves of *Eucalyptus* sp. were randomly sampled from trees in July 2020 (4 on areas 1 and 2; 3 on area 3) and 2021 (3 on area 1; 4 on area 2; 2

on area 3) with the same methodology. A composite per tree was made with leaves collected at about 1 m height. The number of collected samples differed between years due to tree mortality. As *Chenopodium album* L. naturally colonized the micro-plots and was most abundant on the one sown with *L. usitatissimum*, three aboveground parts of this species were sampled per area (Table 1). In addition from each area (Fig. 1: A and B), the leaves of 12 trees of each Tordis and Inger cultivars were randomly collected at about 1.3 m height. From these samples, 12 composites per cultivar were made and transported to the laboratory for further analyses. The plant parts were washed with tap and deionized water and were dried until constant weight (40 °C). Each dry sample was ground with a blender to obtain a powder. Microwave digestion was performed with 0.5 g of DW and 10 mL of HNO₃ (69%) for each sample. The obtained solution was completed until 50 mL with Milli-Q water and filtered (0.45 µm, hydrophilic Teflon). The metal concentrations in the eluates were measured by ICP-OES or ICP-MS, as previously described. One standard reference material was used for analytical quality control (Branches and leaves of Bush “NCS DC 73349”, NCS Testing Technology, China, <http://www.ncsstandard.com>, accessed on 5 June 2018). For all of the digestion sets, recoveries of the reference material were between 96% and 99% for Cu, 103% and 105% for Pb, and 103% for Zn in the branches and leaves. The reference value for Cd was lower than the analytical LoQ.

Using the metal concentrations in the plant parts and the total concentrations in the dredged sediment (Table 2), the Bioconcentration Factor (BCF) was calculated as follows:

$BCF_{tot} = \text{metal concentration in plant parts} / \text{total metal concentration in dredged-sediment}.$

Five rank groups were defined for BCF below 1 ([0;0.01[; [0.01,0.1[; [0.1;0.25[; [0.25;0.75[; [0.75;1]), allowing for each crop and metal to calculate a repartition of BCF in the different groups such as follow :

$BCF \text{ ranking repartition (\%)} = [\text{number of BCF in the rank group} / \text{total number of BCF calculated}] * 100.$

The comparison of the metal concentrations in the crops cultivated under either contaminated or uncontaminated conditions was performed by the calculation of an enrichment factor (EF) as follows:

$EF = \text{metal concentration in crop grown on contaminated condition} / \text{metal concentration in crop grown in uncontaminated condition}.$

2.4) Statistical analysis

The statistical analysis of the values was performed with the software R 4.0.01 (6 June 2020) and R studio. Graphics were made with the Microsoft office license 2019 (Excel and Power Point). A Shapiro test was used to

verify data normality. The Bartlett test (parametric) or the Fligner–Killeen test (non-parametric) were applied to determine variance homogeneity. The means of the different parameters (plant part metal concentration, BCF tot) between areas were compared with a one-way ANOVA test when applicability conditions were respected; in the other case, a Kruskal–Wallis test was used. A two-way ANOVA was used to compare the means of the parameters (1 M NHNO₃-extractable metal concentration, metal concentration in plant parts, metal mobility in the dredged-sediment) between two variables (area and plant parts, area and cultivars, or area and time (sowing, harvest)). At an $\alpha=5\%$ risk or below, a post hoc test (Tukey Honestly Significant Difference (HSD) test, or pairwise *t*-test) was used. A Pearson correlation test was used to study correlations between variables (1 M NH₄NO₃ extractable metal concentration in sediment and metal concentration in plant parts; metal concentrations between plant parts).

3) Results and discussion

3.1) Annual crops

3.1.1) Plant yields

Plant yields measured on the plot highly varied according to the area. Yields peaked in area 2 (Table 3, Fig. 2), which was the least contaminated area according to total dredged-sediment metals (Table 2). This was especially true for the crops sown in 2020. Our previous studies also evidenced a yield difference according to the area but concluded that the pedo-agronomic parameters could not explain this area effect for *S. bicolor* and *M. sylvestris* (Perlein et al. 2021 a,b). The yield for *Sorghum* biomass 133 and *M. sylvestris* in area 2 was equal to the expected agronomic yield (Table 3), which correspond to the yield reported in Perlein et al. (2021a,b). The *Sorghum* Santa Fe red cultivar was bought as a free of right seed so, to our knowledge, no reference yield value is available in the scientific literature. A yield of 280 kg ha⁻¹ FW was reported for *P. tanacetifolia* (*Cap filières*), which was from 10 to 30 times lesser than the yield calculated on our metal-contaminated site (Table 3), suggesting the high tolerance of this crop to site condition, especially metal contamination. Metal concentrations in *Sorghum Santa fe red* and *P. tanacetifolia* below upper critical thresholds could explain this result. In the case of *Sorghum Biomass 133* and *M. sylvestris*, where shoot Cd or Zn concentrations were higher than upper critical thresholds (Table 4), a high tolerance to these metals could explain this result.

The sugarbeet yields below its agronomic values indicated that the dredged-sediment contamination could have decreased the yield for this plant species. Singh and Agrawal (2007) reported that high metal (Cd, Cu, Cr, Ni, Pb, and Zn) concentrations in *B. vulgaris* decreased its yield (by around 20%), when grown in a metal-

contaminated sewage sludge mixed with soil, due to the disturbance of physiological functions such as decreasing photosynthetic rate and increasing lipid peroxidation and proline or protein contents. The leaf Zn concentration of *B. vulgaris* exceeded the Zn upper critical threshold (Table 4) and phytotoxicity values (Table 5) and supported the assumption of the Zn impact on the sugarbeet yield.

In the case of *L. usitatissimum*, it was not feasible to obtain a viable yield in areas 1 and 3 due to the parallel growth of *C. album* (Fig. 2). In area 2, the yield was fairly below the expected value of 6.9 t ha⁻¹ (Table 3). It was reported that the most impacting factor on *L. usitatissimum* yield might be the presence of a colonist species (*Lin et Chanvre Bio*. <https://linetchanvrebio.org/index.php/fiche-technique/> (26/01/2022)) which could lead to a poor development of the flax root system. Thus, soils with a high plant colonist seed bank could limit the cultivation of *L. usitatissimum*. In addition, several studies reported that Cd and Zn contamination could reduce the growth of *L. usitatissimum* leading to lower yield and decreasing competitive fitness against colonist plant species (Lebrun et al. 2021; Amna et al. 2015; Hosman et al. 2017). However, the most explaining factor of the very low yield might be the competition with colonist as all metal concentrations in *L. usitatissimum* aboveground parts were lower than their respective metal upper critical thresholds (Table 4).

Similarly to *L. usitatissimum*, a more vigorous establishment of native plant species was observed simultaneously with the cultivation of other crops in 2020 in areas 1 and 3 compared to area 2. This might be due to a synergic effect of total dredged-sediment metals and highest weed competition leading to a decrease of the fitness and the energy allocation in plants grown in areas 1 and 3.

Although *C. album* impacted the growing of crop species, its implantation and vigor on our site were parameters indicating that this plant species could be of interest in the context of metal-contaminated site if its biomass could be processed. Our results confirmed *C. album* as a colonist species in our site conditions (Tózsér et al. 2019; Sharma et al. 2020; Gupta and Sinha et al. 2007; Río-Celestino et al. 2006; Walker et al. 2004).

3.1.2) Metal concentrations in plant parts and transfer in the crops of the micro-plot experiments

As expected, Zn and Cu, which are essential elements for plants (Kabata-Pendias 2011, Jogawat et al. 2021), were measured in higher concentrations in sampled plant parts than Pb and Cd (Table 4). All evaluated crops showed similar metal ranking pattern (Zn > Cu > Pb > Cd) both in contaminated and uncontaminated conditions (Table 5).

The comparison of the metal concentrations in the crops cultivated under either contaminated (Table 4) or uncontaminated (Table 5) conditions was performed to refine the concentration deviation from uncontaminated condition values (Table 6). The calculated EF for Cd, Pb, and Zn were mostly in the blocks of 1.3-5, 5-10, and >10 (Table 6), showing that Cd, Pb, and Zn were slightly to strongly accumulated in the crops under contaminated conditions. For Zn and Cd, most crops had 1.3 to 5 times higher concentrations in collected plant parts when grown on the contaminated condition than in the uncontaminated one, indicating that most crops have a similar Cd and Zn enrichment pattern. The Pb EF values were equally distributed in the three blocks (Table 6) showing a heterogeneous enrichment between crops. The Cu concentrations in sampled plant parts could be considered physiological for all crops in the contaminated site as they were lower or equal to the uncontaminated condition concentrations (Table 4, Table 5, Table 6).

Higher Cd, Pb, and Zn concentrations in leaves of *B. vulgaris* than in roots ($p < 0.05$) were evidenced for Cd in one area, Pb in two areas, and Zn in the three areas (Table 4) whereas Cu concentrations in leaves and roots were similar for the three areas. In other study (Singh and Agrawal 2007), Zn concentrations in sugarbeet's leaves were 7 to 10 times lesser than ours, whereas Cd, Cu, and Pb concentrations were higher than or in the same range as ours. The authors also reported a reverse organ effect, with Cd, Pb, and Zn concentrations higher in roots than in leaves (Singh and Agrawal 2007). The experimental design performed in pots with other metal soil concentrations could explain these differences.

To our knowledge, *Sorghum* cultivar Santa Fe was never studied in a metal-contaminated field. In accordance to Perlein et al. (2021a), Cd ($p < 0.05$) was present in a higher concentration in aboveground part of "Biomass 133" than in the "Santa Fe red" cultivar. Compared to other cultivars of *Sorghum* cultivated under contaminated conditions, our Pb concentrations in both cultivars were particularly low (37-42 $\mu\text{g g}^{-1}$ DW in Zhuang et al. 2009), whereas the Cu concentrations were similar (Angelova et al. 2011). On the contrary, the Cd and Zn concentrations in aboveground parts (Table 4) were both highly above the concentrations reported in Marchiol et al. 2007 (Cd: 0.2 $\mu\text{g g}^{-1}$ DW) and Epelde et al. 2009 (Zn: 50 $\mu\text{g g}^{-1}$ DW), which suggests that metal transfer in aboveground parts mainly depends on cultivar than other factor such as total and extractable (Cd: $r : 0.36$ and 0.38 for Biomass 133 and Santa fe red respectively; $p < 0.05$) metals in dredged-sediment.

Saleem et al. (2020) reported aboveground metal concentrations in *L. usitatissimum* (9-49, 31-814, 110-332, 116-255 $\mu\text{g g}^{-1}$ of DW for Cd, Cu, Pb, and Zn, respectively) grown under spiked contaminated pot conditions

highly above those measured in our study (Table 4). This difference highlighted the need to perform field experiments to measure real concentrations.

Furthermore, differences in metal concentration in crops (Table 4) were found depending on cultivation area, sampled plant part, and cultivar. In 2/3 of the cases, the cultivation area did not change metal concentrations in crops ($p>0.05$) (Table 4). In the other cases, the change in metal concentrations in crops between areas was not supported by the correlation of 1M NH_4NO_3 -extractable metal concentrations in dredged-sediments and metal in plant parts in each area, suggesting that the cultivation area was not the main factor leading to changes in metal concentration in crops in our site conditions.

Amongst the annual crop studied, Zn concentration in *B. vulgaris* (leaves) and *M. sylvestris* (aboveground part) and shoot Cu concentration in *C. album* exceeded the phytotoxicity values (Table 5) or the upper critical thresholds for these elements (Table 4). With the exception of *C. album*, all the Cd concentrations in collected plant parts were above the Cd phytotoxicity value (Table 5) or in the range of the Cd upper critical threshold (Table 4). Despite this, no visible toxicity symptoms was evidenced on the plant shoots. Contrary to *C. album* for Cu, all other annual crops presented an excluder behavior with a $\text{BCF}_{\text{tot}} < 1$ (Supplemental Table 1) although crop Cd, Pb, Cu, and Zn concentrations on the contaminated field were above uncontaminated one (Tables 5 and 6).

To better understand the potential of living organisms to uptake and accumulate pollutants and the subsequent pollutant exposure, a ranking approach of BCF_{tot} was proposed for aquatic media (Donnachie et al. 2014) and crops (Christou et al. 2019). Similar to what has been done for either accumulator ($\text{BCF}_{\text{tot}} > 1$) or hyperaccumulator ($\text{BCF}_{\text{tot}} > 10$) species, few studies proposed to enlarge the ranking of excluder plant species. Tang et al. (2019) defined species as excluders when $\text{BCF}_{\text{tot}} < 0.1$ to improve the accuracy of the definition. Based on these approaches, an extension of excluder groups according to site condition and metal could be of interest to maximize the cultivation success (implementation, biomass yield) and minimize the metal trophic chain exposure. Because each metal has different physiological functions and/or phytotoxicity, the BCF_{tot} assessment should be defined metal by metal. When BCF_{tot} is < 1 this indicates an excluder plant behavior, whereas growth, yield, and plant metabolism could be impeded due to, amongst factors, plant metals concentration(s) above phytotoxicity value(s) (Table 5) and/or value(s) leading to a yield decrease (upper critical threshold, Table 4). To precise the metal behavior of the excluder crops studied on our field, we proposed a

BCF_{tot} ranking with five groups from the most to the least excluder, as follows: [0;0.01[, [0.01;0.1[, [0.1;0.25[, [0.25;0.75[, [0.75;1]. This approach was not a way to understand the physiological mechanisms.

An analytical matrix was created to divide the BCF_{tot} into two groups according to the physiological role of the four metals, i.e., essential or non-essential element (Zn/Cu or Cd/Pb, respectively) (Tables 7 and 8). These matrices allowed the visual comparison of the BCF_{tot} between crops and elements. Regardless of the element, 73% of the BCF_{tot} were included in the [0;0.01[and [0.01;0.1[groups (Tables 7 and 8). For Pb, all BCF_{tot} were below 0.1, showing similar crop behavior towards this metal (Table 7). 96% and 63% of the BCF_{tot} were below 0.1 in the case of Zn and Cu, respectively (Table 8). The high percentage of Pb and Zn BCF_{tot} under 0.1 could be explained by the high total Pb and Zn concentrations in the dredged-sediment, indicating that these elements did not result in aerial crop transfer in the site conditions. For Cd, 67% of the BCF_{tot} were above 0.1, with 37% in the [0.1;0.25[group and 30% in the [0.25;0.75[group (Table 7). The Cd BCF_{tot} repartition in the rank was the most heterogeneous between crops and plant parts, suggesting different Cd uptake regulation mechanisms (e.g., metal transporter, root exudates) (Chen et al. 2017; Montiel- Rozas et al., 2016; Sterckeman and Thomine, 2020)). In *B. vulgaris* (Yolcu et al. 2022) and *L. usitatissimum* (Khan et al. 2020), several genes from NRAMP and HMA families were assumed to play a role in Cd uptake and translocation. To our knowledge, no information is available about Cd transporters in the other studied crops. Table 7 evidenced the crop behavior difference between the two non-essential elements and suggested that Cd should be monitored, especially for *M. sylvestris* and *Sorghum* Biomass 133. Interestingly, Table 8 demonstrates the same behavior towards the transfer of Cu and Zn for all crops, except for *C. album* and *M. sylvestris*. Copper in *M. sylvestris* would have higher physiological importance than in the other plant species.

Consequently, the plants with Cu, Pb, and Zn BCF_{tot} values within the [0;0.01[and [0.01;0.1[ranges can be qualified as strongly excluders and excluders, respectively, with *a priori* no deleterious health consequences to the plants. For Cd, except *C. album*, the other crops presented concentrations above this toxicity threshold or inside the upper critical threshold, although some had BCF_{tot} values in the [0.01;0.1[range. Accordingly, the Cd BCF_{tot} in the [0;0.01[range could refer to an excluder behavior without any impact on the plant. When the plant's BCF_{tot} value is above the excluder group and shows metal concentrations above phytotoxicity values or upper critical thresholds, long-term monitoring could be performed to ensure that no phytotoxicity sign will appear as well as aerial metal transfer will not increase.

3.1.3) Impact of cultivation on the extractable metal dredged-sediment fraction

The mean values of extractable metal concentrations were compared according to the crops and the area. Those measured at the sampling periods (at the start and harvest, Table 9) exceeded the concentrations mentioned in the standard ISO 19730:2008 (E) corresponding to the frequently retrieved concentrations after a soil extraction with 1M NH₄NO₃ (Cd: 0.005 µg g⁻¹ DS, Cu: 0.25 µg g⁻¹ DS, Pb 0.02 µg g⁻¹ DS, and Zn: 0.25 µg g⁻¹ DS). In addition, for each metal, the percentage of mobility was calculated (Supplemental Table 2). During the whole experiment, the percentage of mobility in the dredged-sediment was in the following order: Pb < Zn < Cu ≤ Cd (Supplemental Table 2).

In most cases (80%), extractable metal concentrations and mobilities did not vary between the sowing and the harvest ($p < 0.05$, Table 9 and Supplemental Table 2). Extractable Cd and Zn concentrations, to a lesser extent, increased or decreased ($p < 0.05$). In contrast, extractable Pb and Cu concentrations in the dredged-sediment did not change, like Pb mobilities. Changes depended on crops. The cultivation of *S. Biomass 133*, *S. Santa Fe red*, *L. usitatissimum*, *P. tanacetifolia*, and *M. sylvestris* decreased the extractable dredged sediment Cd and Cd mobility in area 1, increased them in area 2, and increased extractable dredged-sediment Zn and its mobility on the areas 2 and 3. In contrast, sugarbeet cultivation did not change the extractable metal fractions nor their mobility in the dredged-sediment.

Overall, the crops have a small impact on the metal mobility and extractable metal fractions, even when a significant difference was found, as it was not evidenced on all areas or was antagonistic in different areas. As already indicated in Perlein et al. (2021 a,b), the cultivation of the crop alone could not reduce metal mobilities in this dredged-sediment. The addition of a soil amendment, allowing to either complex or precipitate, on a long-term basis, available Cd, and to a lesser extent available Zn in this dredged-sediment, could homogenize and reduce metal extractability and mobility on the whole experimental site. The use of biochar or arbuscular mycorrhizal fungi when crops are mycorrhizable could fill this role without impacting pedoagronomic parameters, such as pH, which in our site, promotes a less available Cd form (Phanthavongsa 2018; Van Poucke et al. 2018; Perlein et al. 2021c).

3.2) Perennial crops - *Eucalyptus* sp. and willow (*S. viminalis* hybrid) leaves

For three years, after a first harvest in January 2019, foliar metal concentrations for both willow cultivars were monitored on the site to study changes in ionome in the regrowths. In addition, BCF for willow leaves

(Supplemental Table 3) were calculated using the mean values of total metal concentrations in the dredged-sediment of the whole VSRC experiment (Table 2, Phanthavongsa 2018).

The foliar Cd and Zn concentrations (Fig. 3) were extremely high. The Cd BCF values oscillated around 1 (Supplemental Table 3) with a maximum of 1.55, and the Zn BCF values varied between 0.14 and 0.6, below 1 due to the high total dredged-sediment Zn. Such high concentrations confirmed the Cd and Zn accumulator phenotype of both willow cultivars (Meers et al. 2005; Meers et al. 2007; Van Slycken et al. 2013; Grignet et al. 2020).

The sampling area had a limited impact on the foliar Zn and Cd concentrations and BCF of these elements for Inger and Tordis willows (Fig. 3, Supplemental Table 3). For Zn, the area effect only concerned the Tordis cultivar in 2019 ($p < 0.05$), and for Cd, this effect was significant ($p < 0.05$) for the Inger cultivar in 2020 and 2021. In addition, for both metals, a cultivar effect was evidenced ($p < 0.05$) each year with Tordis > Inger. However, the difference intensity between these cultivars varied according to the years and metals considered. In line with several studies performed on many *Salix* cultivars, foliar Cd and Zn concentrations were strongly correlated ($p < 0.01$, $r = 0.88$; Dos Santos Utmazian and Wenzel 2007; Wieshammer et al. 2007; McBride et al. 2016; Grignet et al. 2020). As suggested for many *Populus* cultivars, similar pathways could be used for Cd and Zn storage or uptake in leaves (Pottier et al. 2015). It is admitted that Cd, a non-essential element, could interact with essential element, such as Zn or Mn by competing with many transporters of ZIP and HMA families (Sterckeman and Thomine 2020). All foliar concentrations of Cu and Pb for both willow cultivars (Fig. 3) could be considered as common values (Cu: 15-20 $\mu\text{g g}^{-1}$ DW; Pb: 0.2-3.8 $\mu\text{g g}^{-1}$ DW; Kabata-Pendias 2011) with the Pb and Cu BCF values below 0.001 and 0.1 respectively (Supplemental Table 3). For foliar Cu and Pb concentrations, a moderate correlation ($p < 0.01$, $r = 0.38$) explained the similar patterns for these metals (Fig. 3). In 2020, a small effect of the area and cultivar was observed ($p < 0.05$) with a higher foliar Pb and Cu concentrations for Inger B. In addition, a cultivar effect ($p < 0.05$) was evidenced in 2021 for foliar Cu concentrations with Tordis < Inger. The Pb and Cu BCF values confirmed these results (Supplemental Table 3).

Phanthavongsa (2018) reported concentrations in leaves of Inger (in $\mu\text{g g}^{-1}$ DW, Cd: 5, Cu: 10, Pb: roughly 1, and Zn: 1000-1500) and Tordis (Cd: 5, Cu: 10, Pb: around 1, and Zn: 1300-2000) before the first cut of the trees and, for both cultivars. Our study reported similar concentrations (Fig. 3). Moreover, the four foliar metal concentrations varied only slightly for both cultivars during the three years (Fig. 3). This is in accordance with Van Slycken et al. (2013), who showed for several willow cultivars that foliar Cd and Zn concentrations did not

change during four years. Overall, our study supports the assumption that the cut of the trees did not change the metal concentration in willow leaves during regrowth.

Similarly to willow, at a smaller scale, the foliar metal concentrations of *Eucalyptus* sp. was followed for two years. Contrary to willow cultivars, such concentrations could be considered common as they were below values leading to phytotoxicity, and the BCF values were below 0.2 (Table 4 and Supplemental Table 1). In year 2, i.e., one year after planting, a higher foliar Cd concentration in area 1 and a lower foliar Pb concentration in area 3 were observed (Table 4). Foliar Cd, Pb, and Zn concentrations differed significantly depending on the area ($p < 0.05$), with year 2 showing higher values than year 3, i.e., two years after planting. For Cd, this was evidenced in areas 1 and 2; for Pb, in area 3; and for Zn, in areas 1 and 3. In addition, a reduction tendency of the foliar concentrations for the four metals during growth was observed. Mughini et al. (2013) reported similar foliar concentrations for several *Eucalyptus* cultivars grown under contaminated conditions as compared to our data after a 3-year growth period. Other pot experiments with biosolid highlighted an accumulator behavior of *Eucalyptus* sp. and reported higher concentrations and/or BCF in leaves (Mok et al. 2013).

The low BCF values of *Eucalyptus* sp. and the fact that the metal concentrations in *Eucalyptus* leaves were highly below those of both *Salix* cultivars suggested that *Eucalyptus* sp. exhibited an excluding behavior in our site condition. Even lower metal concentrations could be expected in bark (Mughini et al. 2013; Evangelou et al. 2015).

This study was impacted by the mortality of *Eucalyptus* sp. on the site, leading to a decrease in the number of trees between years 1, 2, and 3 (§2.3). Several studies reported the tolerance of *Eucalyptus* sp. to high metal concentrations in soils, indicating that this would not be the main factor explaining such mortality rate (66% in 2021) during our study (Shukla et al. 2011; Marchiol et al. 2013; Mok et al. 2013). The low water access of the growing root system of *Eucalyptus* sp. due to low rainfall and high temperatures in July and August 2019 (Supplemental Fig. 1) might explain it. Between 2020-2021, the overall climatic conditions of the site (Supplemental Fig. 1), such as the negative temperature in winter might also explained it. In the foreseeable future, if the climatic change would lead to global warming in the North of France and, by extension the North of Europe, *Eucalyptus* sp. could become an effective option for phytostabilization. However, in the climatic change context, other parameters must be assessed such as wildfire risk and local biodiversity disturbance.

3.3) Scenario of phytomanagement option for each crop

As indicated in Fig. 1, the soil phytomanagement consists of the combination of several parameters that could be resumed as the adequation of the phytotechnology with the economic valorization of the biomass produced to allow long-term management of the site (Robinson et al. 2009). Therefore, each crop studied was assessed as a phytomanagement option by considering the following main factors: plant behavior on the metal contaminated soil, agricultural parameters, plant characteristics for non-food processing, and regulation (Fig. 1, Table 1). As no processing test of the biomass was performed, the following discussion was based on a survey of the scientific literature, standard, and regulation.

3.3.1) Annual crops

Based on BCF calculation, all the annual crops exhibited an excluder behavior with the exception of *C. album* with Cu. As a result, in our field conditions, annual crops fit with exclusion as the metal transfer in the harvested parts was limited. In addition, plant cover rate, biomass yield, adapted machinery for large-scale application, weed control, and cultural practices are key factors to ensure the success of phytomanagement (Fig. 1, Kidd et al. 2015).

For each crop, the last point to evaluate within the framework of phytomanagement is the compliance of the biomass with potential local biomass-processing chains (Tables 1 and 5).

Sorghum Biomass 133 and *P. tanacetifolia* were selected for anaerobic digestion (AD) to produce biogas (Table 1). A relevant biomethane potential (BMP) and biogas yield were evidenced for *Sorghum* Biomass 133, which allowed similar energy production to forage maize in a cogeneration scenario (Perlein et al. 2021a). The BMP of *P. tanacetifolia* was 274 L CH₄ Kg⁻¹ organic dry matter indicating that the biogas production with this plant species could be relevant (Herrmann et al. 2016). If the digestate was applied as a fertilizer, metal concentrations in the digestate should respect the threshold set in the Regulation (EU) 2019/1009 (Table 5). In this case, the shoot Cd concentration in the *Sorghum* Biomass 133 should not exceed 1.3 µg g⁻¹ of DW, or a pretreatment to remove Cd in the digestate should be performed. *Phacelia tanacetifolia* respected the metal threshold of the Regulation (EU) 2019/1009 (Table 5) for all the metals, allowing the direct usage of the digestate obtained during AD as a fertilizer product. The direct fertilization of contaminated soils by the grinding of the *P. tanacetifolia* biomass could also be relevant. However, the influence of mixing such biomass with metal-contaminated soils on the metal mobility should be studied as the organic matter could interact with the available metal fraction, or the final low pH after ensiling could affect metal mobility leading to a higher metal exposure of the following crop (5.2 pH unit, Herrmann et al. 2016).

Two crops were selected based on their tinctorial characteristics: *M. sylvestris* and the *S. bicolor* cultivar Santa Fe red to produce natural dye from flowers and leaf sheaths, respectively (Table 1, Akogou et al. 2018; Perlein et al. 2021b). The metal transfer from the flowers of *M. sylvestris* to the dye extract and the dyed textile was limited, which respected metal thresholds set in the Eco Passport by OEKO-Tex or OEKO-Tex standard 100 (Table 5, Perlein et al. 2021b). The Cd concentration in the *M. sylvestris* flowers was quite similar (Table 4) to the one already reported (Perlein et al. 2021b), which suggested that this Cd concentration should not be a limitation for this biomass processing option. Contrary to Akogou et al. (2018), who studied another tinctorial *Sorghum* cultivar, we did not observe a red color on the foliage on the harvested biomass. Further investigation is necessary to check the relevance of this cultivar for the dyeing value chain, including metal fate in the process and natural dye color (Perlein et al. 2021b; Hou et al. 2017). The residues of the dye production should be valorized in a complementary processing chain as they compose the major part of the plant biomass. Alternative non-food processing chains were proposed for *M. sylvestris*: essential oil (EO) production, water extract containing high-value compounds and, Hg and Cu adsorbent in water (Ramavandi et al. 2016; Ramavandi et al. 2018; Delfine et al. 2017; Kostic et al. 2019). As reported for other plant species, a limited metal transfer from *M. sylvestris* to EO or water extract can be expected (Angelova et al. 2016; Zheljazkov et al. 2008; Perlein et al. 2021 b; Perlein et al. 2021c; Elallem et al. 2020). The biomass from both crops could also be used to produce compost or energy via AD with regard to BMP and metal concentrations. Zinc biofortification for animal feeding could be envisaged if *S. Santa Fe red* is edible by animals and shoot Cd concentration (Table 4) respects the directive 2002/32/EC (Table 5) (Clemens et al. 2017, Anderson et al. 2012, Vamerali et al. 2014, Wang et al. 2021).

Linum usitatissimum could be used to produce fiber for the textile sector (Table 1). Cadmium, Pb, and Zn concentrations in the hemp fibers were similar to or lower than the concentrations in its total aboveground part (De Vos et al. 2022). As for hemp, it could be assumed that the metal concentrations in *L. usitatissimum* fibers be equal to the concentrations of the whole aboveground part. After the harvest, the biomass should be laid on the soil to allow the fiber separation from the biomass. In the context of metal-contaminated site, the contamination of the fibers during the retting could occur. If the biomass retting on the contaminated site can lead to exceed the thresholds of the OEKO-Tex standard 100, one alternative way is to perform the retting in water with a suitable process to avoid metal transfer from the fiber to the water and the environment. After fiber production, the residues (shives) should be processed to produce energy as proposed in De Vos et al. (2022) for hemp.

Beta vulgaris is a sugar crop allowing biofuel production by the alcoholic fermentation of roots (Table 1). Sugarbeet containing 16 percent of sugar in roots could produce 103.5 L of ethanol per ton of fresh matter (Shapouri et al. 2006; Panella and Kaffka 2010). To our knowledge, metal thresholds in bioethanol do not exist. Anaerobic digestion would be suitable for this plant part given that the roots show a high BMP (374.9 L kg⁻¹ ODM; Herrmann et al. 2016) and metal concentrations are below regulation (Table 5, Panella and Kaffka 2010). The leaves could be valorized in complementary processing chains such as compost if foliar Cd concentrations do not limit this option (Regulation (EU) 2019/1009, Table 5). The use of the existing agricultural machinery to wash *B. vulgaris* roots potentially covered by metal-contaminated dredged-sediment could be sufficient to limit metal transfer in further processes.

3.3.2) Perennial crops

The Inger and Tordis willow cultivars exhibited a Zn/Cd accumulator behavior with a high concentration in the leaves, indicating that these cultivars could better fit with phytoextraction. These high concentrations prevent the use of this biomass in compost production or mulching (Regulation (EU) 2019/1009, Table 5). The high Zn concentration at leaf fall would allow the production of Zn-ecocatalyst (Deyris et al. 2018; Grignet et al. 2020). To ensure the viability of the overall chain, wood processing in the energy sector would be an option if the metal thresholds for fuels stated in the French regulation for combustion (Cd: 5; Cu: 30; Pb: 50; Zn: 200 µg g⁻¹ DW; Decree of the 3 August 2018) are respected. Based on the measurement performed in Inger and Tordis willow wood at harvest in January 2019 (unpublished data; concentrations (µg g⁻¹ DW) for Inger and Tordis, respectively: Cd: 2.96 (±1.24) - 3.68 (±0.52); Cu: 10.95 (±4.25) - 11.42 (±4.69); Pb: 1.38 (±0.84) - 1.20 (±0.71); and Zn: 581.37 (±242.13) - 635.18 (±88.63)), only the Zn concentrations in the wood of both willow cultivars exceeded the Zn threshold. Thus, to use willows in combustion, the installation of a filtration system at the chimney could be relevant to avoid any metal dispersion in the environment (Bert et al. 2017b).

Leaves of *Eucalyptus* sp. have chemical activity, allowing EO production as an alternative to synthetic pesticides (antibacterial, antifungal, insecticidal, herbicidal; Batish et al. 2008). *Eucalyptus* sp. presented low metal concentrations in leaves and exhibited an excluder behavior, a relevant characteristic for the safe production of EO (Table 4, Zheljzkov et al. 2008, Perlein et al. 2021c). As wood composes the major part of the aboveground biomass, for instance the wood processing in the energy sector should be studied (Pereira and Costa 2017). To our knowledge, no data are available on metal concentrations in this clone wood.

4) Conclusion

The potential of 10 annual and perennial crops for phytomanaging a dredged sediment was investigated in field conditions (North of France) according to their relevance for phytotechnologies and their non-food biomass processing. Most crops were relevant for the site phytomanagement, except *L. usitatissimum* and *Eucalyptus* sp. due to competition with colonist species and pedoclimatic conditions. As the eight metal-tolerant plant species well developed in the dredged-sediment of the site, it could be expected that they will perform similarly or better on less metal-contaminated soils. Nevertheless, metal-tolerant plant species could be challenged by ruderal plant species, or other adapted plant species with other traits, leading to competition which could impede their maintenance or yield. The field experiment evidenced that, with the exception of willow cultivars (Inger and Tordis), all the crops exhibited an excluder behavior ($BCF < 1$) for Zn and Cd, a suitable characteristic for phytostabilization. In this study, we proposed a new way to define and assess the plant excluder behavior on site. The BCF group ranking approach could be considered as a macroscale tool allowing a preliminary step of crop assessment. The methodology selects the most suitable crops for one metal-contaminated site by comparing several crops according to their BCF_{tot} , phytotoxicity values and upper critical thresholds. This approach could help the site manager/owner to select the proper action for phytomanagement according to future land use and their management objectives. BCF_{tot} ranking recommendations were site-specific and should not be generalized as total metal concentrations and other soil characteristics differ from one site to another. Nevertheless, the proposed methodology could be applied to different typology of site. We confirmed that the definition of excluders species as $BCF_{tot} < 1$ was not accurate enough and that the new groups ranking should provide a better estimate of crop exposure to metal excess. The reduction of the metal mobility in the soil was not achieved by the cultivation of the annual crops. For Cd especially, which remained highly mobile in our field experiment, the addition of a suitable amendment which did not change pedoagronomic parameters (e.g., biochar, arbuscular mycorrhizal fungi) to decrease Cd mobility could be studied. Finally, for all crops, one or several non-food biomass processing options could be proposed. This selection was facilitated by the excluder behavior of the crops, as all plant parts could be processed without any metal constraint or after a pretreatment to remove Cd. Biomass processing tests are however needed to conclude on the feasibility of the phytomanagement options for all the studied crops, i.e., different scale assays, metal impact on the process, and plant adequation to the process.

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Figure captions:

Fig. 1. Conceptual model to assess plant selection suitable for a phytomanagement option based on metal plant behavior and other factors necessary for a relevant biomass valorization option at field level. (*Tang et al. 2019)

Fig. 2. Aerial view of the site with the experiments' areas (left) and pictures of the different studied plants (right)

Fig. 3. Foliar metal concentration in Inger and Tordis willow cultivars over the 3-year growth period.

Supplemental Fig. 1. Monthly climatic parameters (rainfall, temperature, and maximal and minimal temperatures) on the site during the 3-year experiment (Info.climat.fr: meteorological station of Lille-Lesquin).

Supplemental Fig. 2. Sowing and planting design of the annual crops and *Eucalyptus* adapted from agricultural or field practices on 25 m² plot on the three areas.

Table 1. Plant species tested on the field experiment, date of planting and harvest, plant parts collected and selected traits for non-food biomass processing.

Species	Sowing / planting date	Harvest date	Collected plant parts	Selected trait for processing	Potential valorization
<i>Beta vulgaris</i> sub sp. <i>vulgaris</i> L.	05/16/2019	09/17/2019	Leaf, root	High sugar content	Bioethanol production
<i>Eucalyptus</i> sp.	06/19/2019	July 2020- July 2021	Leaf	Aromatic plant	Essential Oil
<i>Malva sylvestris</i> L.	04/16/2020	07/16/2020	Aboveground part, flower	Tinctorial plant	Dye production
<i>Linum usitatissimum</i> L.	04/16/2020	08/20/2020	Aboveground part	Fiber crop	Fiber production
<i>Phacelia tanacetifolia</i> Benth.	04/16/2020	07/16/2020	Aboveground part	High biomethane potential	Biogas production / Green fertilizer
<i>Sorghum bicolor</i> Biomass 133	05/19/2020	09/16/2020	Aboveground part	High biomethane potential	Biogas production
<i>Sorghum bicolor</i> Santa fe red	05/19/2020	09/16/2020	Aboveground part	Tinctorial plant*	Dye production
<i>Chenopodium album</i> L.	Natural colonizer	09/16/2020	Aboveground part	TE tolerance	Composting
<i>Salix Inger and Tordis</i>	June 2012 First coppice harvest in January 2019	July 2019 July 2020 July 2021	Leaf	Zn-accumulator	Zn-ecocatalyst production

* information noticed on the purchase site.

Table 2. Total metal concentration in the dry dredged-sediment (DS) (mg kg⁻¹ DS) at the three areas, and at the area with the very short rotation coppice (VSRC) experiment and metal threshold values (S1) for sediment management.

Parameters		Area 1	Area 2	Area 3	VSRC*	S1
Total metal (mg kg⁻¹ DS)	Zn	6685 ± 509	6084 ± 132	8980 ± 340	6089	300
	Pb	774 ± 18	592 ± 12	1043 ± 12	956	100
	Cd	6.3 ± 0.14	5 ± 0.08	9 ± 0.13	8.1	2
	Cu	87 ± 2.3	76 ± 1.5	101 ± 1.3	110	100

*Phanthavongsa (2018)

Table 3. Crop yields measured on each area at the harvest (kg of FW 25m²) and extrapolated at the hectare (t of FW ha⁻¹) and reference value on either contaminated or uncontaminated site.

	Area 1		Area 2		Area 3		Reference values
	kg 25 m ⁻²	t ha ⁻¹	kg 25 m ⁻²	t ha ⁻¹	kg 25 m ⁻²	t ha ⁻¹	t ha ⁻¹
<i>B. vulgaris</i> (whole plant)	82.99	33.2	136.46	54.58	101.36	40.54	85-90
<i>M. sylvestris</i> (aboveground part)	4.92	1.97	32.44	12.98	0.6	0.24	10.5-15.5 (FW) ⁺
<i>P. tanacetifolia</i> (aboveground part)	9.2	3.68	26.72	10.69	9.2	3.68	0.280 (FW)
<i>L. usitatissimum</i> (aboveground part)	-	-	5.76	2.3	-	-	6.0
<i>S. Santa Fe red</i> (aboveground part)	5.28	2.11	62.6	25.04	12.52	5.01	-
<i>S. Biomass 133</i> (aboveground part)	21.22	8.49	131.7	52.68	14.82	5.93	54.97 (FW) [']

⁺ Perlein et al. 2021 a; ['] Perlein et al. 2021 b

Table 4. Mean metal concentrations and standard deviation ($\mu\text{g g}^{-1}$ of DW, $\pm\text{SD}$) in the collected plant part of the annual crops and *Eucalyptus* on the different areas (area 1, area 2, and area 3), and upper critical threshold values for metal concentration in plant parts. For each crop and metal, significant differences between conditions (according to area of cultivation, plant parts, cultivars, year) are indicated by different letters at the level of $\alpha = 0.05$.

		Upper critical threshold values*	<i>B. vulgaris</i>		<i>S. Bicolor</i>		<i>L. usitatissimum</i>	<i>P. tanacetifolia</i>	<i>M. sylvestris</i>		<i>C. album</i>	<i>Eucalyptus (Leaves)</i>	
			Leaves	Root	Biomass 133	Santa fe red			Aerial part	Flower		Years 2	Years 3
Cd	Area 1	3-10	1.53 (± 0.48) ^a	0.57 (± 0.11) ^b	3.12 (± 0.93) ^a	1.42 (± 0.43) ^b	1.20 (± 0.26) ^a	0.69 (± 0.21) ^a	3.04 (± 1.07) ^a	1.23 (± 0.35) ^c	0.23 (± 0.03) ^a	1.25 (± 0.41) ^a	0.26 (± 0.16) ^{b,c}
	Area 2		0.83 (± 0.73) ^b	0.56 (± 0.16) ^b	2.91 (± 1.36) ^{a,c}	1.50 (± 0.68) ^b	2.14 (± 0.67) ^b	0.51 (± 0.18) ^{a,b}	2.30 (± 0.82) ^a	0.77 (± 0.21) ^c	0.39 (± 0.02) ^b	0.63 (± 0.02) ^b	0.13 (± 0.06) ^c
	Area 3		1.07 (± 0.40) ^{a,b}	0.62 (± 0.40) ^b	3.95 (± 0.72) ^a	1.87 (± 0.65) ^{b,c}	1.44 (± 0.28) ^a	0.49 (± 0.11) ^b	3.84 (± 1.33) ^b	1.12 (± 0.23) ^c	0.38 (± 0.01) ^b	0.57 ± 0.20) ^{b,c}	0.058 (± 0.02) ^{b,c}
Cu	Area 1	20-30	8.87 (± 0.91) ^a	6.69 (± 1.52) ^a	7.98 (± 1.96) ^a	7.06 (± 1.39) ^a	7.71 (± 1.13) ^a	7.33 (± 0.90) ^a	10.49 (± 3.12) ^a	13.74 (± 7.46) ^{a,b}	65.52 (± 97.53) ^a	12.58 (± 2.04) ^a	8.68 (± 1.49) ^a
	Area 2		5.54 (± 4.88) ^a	6.39 (± 1.15) ^a	7.65 (± 2.52) ^a	5.77 (± 1.35) ^a	6.67 (± 1.73) ^a	7.25 (± 1.80) ^a	9.49 (± 1.48) ^a	8.90 (± 3.08) ^a	105.97 (± 91.73) ^a	9.15 (± 1.64) ^a	7.63 (± 3.99) ^a
	Area 3		8.11 (± 1.77) ^a	6.77 (± 0.68) ^a	6.08 (± 1.98) ^a	5.97 (± 1.44) ^a	7.05 (± 1.34) ^a	5.77 (± 0.84) ^b	16.39 (± 8.22) ^b	8.06 (± 1.40) ^a	183.46 (± 272.94) ^a	8.46 (± 1.31) ^a	8.14 (± 0.44) ^a
Pb	Area 1	19-35	2.87 (± 1.49) ^{a,b,d}	0.64 (± 0.26) ^c	1.13 (± 0.31) ^{a,b}	1.42 (± 1.19) ^b	2.90 (± 2.86) ^a	5.19 (± 3.20) ^a	5.26 (± 2.32) ^a	3.57 (± 1.63) ^a	3.41 (± 3.10) ^a	0.69 (± 0.27) ^a	0.27 (± 0.20) ^a
	Area 2		1.67 (± 1.47) ^{b,c}	1.10 (± 0.42) ^c	0.58 (± 0.23) ^a	0.57 (± 0.44) ^a	2.45 (± 1.57) ^a	2.25 (± 1.62) ^b	7.02 (± 5.65) ^a	7.98 (± 8.66) ^a	3.99 (± 3.09) ^a	1.94 (± 0.99) ^a	0.17 (± 0.05) ^a
	Area 3		3.65 (± 2.67) ^a	1.10 (± 0.45) ^{c,d}	0.88 (± 0.28) ^{a,b}	0.55 (± 0.13) ^a	1.01 (± 0.47) ^a	1.93 (± 1.45) ^b	7.71 (± 4.53) ^a	5.61 (± 5.03) ^a	1.55 (± 0.11) ^a	4.52 (± 1.92) ^b	0.17 (± 0.13) ^a
Zn	Area 1	200-300	705.12 (± 146.54) ^a	187.59 (± 37.84) ^c	210.94 (± 71.19) ^{a,b}	225.97 (± 57.05) ^{a,b}	146.93 (± 29.42) ^a	185.12 (± 33.19) ^a	331.67 (± 61.62) ^a	163.06 (± 24.10) ^b	247.83 (± 91.78) ^a	121.32 (± 10.67) ^a	43.54 (± 11.67) ^b
	Area 2		575.67 (± 149.51) ^b	150.23 (± 39.27) ^c	176.98 (± 51.56) ^a	210.90 (± 36.45) ^{a,b}	114.81 (± 20.30) ^b	173.21 (± 52.88) ^a	314.70 (± 127.96) ^{a,c}	190.91 (± 101.91) ^b	349.26 (± 62.13) ^a	94.00 (± 14.42) ^{a,c}	57.89 (± 14.86) ^{b,c}
	Area 3		541.14 (± 85.18) ^b	150.53 (± 20.55) ^c	208.04 (± 36.23) ^{a,b}	256.29 (± 53.25) ^b	99.78 (± 17.52) ^b	122.09 (± 30.91) ^b	388.77 (± 120.02) ^a	211.80 (± 48.65) ^{b,c}	391.30 (± 208.19) ^a	103.17 (± 27.10) ^a	46.76 (± 5.68) ^b

* Macnicol and Beckett 1985, Anna-Maj balsberg Pahlsson 1988

Table 5. Metal concentrations measured under uncontaminated field conditions or from the literature ($\mu\text{g g}^{-1}$ DW) for plant parts of annual crops and maximum permitted concentrations for plant biomass processing.

	Cd	Cu	Pb	Zn
<i>B. vulgaris (root)</i>	0.3**	17.6*; 9**	0.1**	59.5*; 12**
<i>B. vulgaris (leaves)</i>	0.4**	15.6*; 10**	0.1**	45.6*; 20**
<i>S. Santa Fe red</i>	0.29	4.68	0.32	55.97
<i>S. Biomass 133</i>	0.16	7.22	0.62	47.84
<i>L.usitatissimum</i>	0.55 (± 0.22)	11.19 (± 6.26)	0.41 (± 0.16)	32.38 (± 7.55)
<i>P. tanacetifolia</i>	0.14 (± 0.03)	6.80 (± 1.24)	0.92 (± 0.49)	26.81 (± 5.33)
<i>M. sylvestris (aerial part)</i>	0.25 (± 0.07)	14.09 (± 3.31)	0.42 (± 0.09)	84.10 (± 28.57)
<i>M. sylvestris (flower)</i>	0.44	7.80	0.22	46.64
<i>C. album</i>	0.08 (± 0.01)	33.73 (± 28.32)	1.26 (± 0.87)	97.24 (± 13.94)
Phytotoxicity limit value²	0.05 - 0.4	15 - 20	0.2 - 3.8	400
Threshold values fertilizing product⁺	2	300	120	800
Threshold values Animal feed (12% DW)⁺⁺	1	-	10	-
OEKO-Tex standard 100⁺⁺⁺	0.1 (extractable) – 40 (total)	50 (extractable)	1 (extractable) – 90 (total)	750 (extractable)
Eco Passport Threshold Value[‘]	20	250	90	1500







- ² Kabata-Pendias 2011, * S. Chibowski 2000; ** Singh and Agrawal 2012; ⁺ Regulation (EU) 2019/1009; ⁺⁺ Directive 2002/32/EC, ⁺⁺⁺ OEKO-Tex Standard 100, [‘] Eco Passport by OEKO-Tex

Table 6. Metal enrichment (EF = [metal concentration in crop grown in contaminated condition] / [metal concentration in crop grown in uncontaminated condition]) and matrix of crops grown in contaminated conditions compared to control (uncontaminated condition).

crops	area	Cd		Cu		Pb		Zn	
		Leaves	Root	Leaves	Root	Leaves	Root	Leaves	Root
<i>B. vulgaris</i>	part								
	1	>	>	<	<	>>>	>	>>>	>
	2	>	>	<	<	>>>	>>	>>	>
	3	>	>	<	<	>>>	>>	>>	>
<i>S. Biomass 133</i>	1	>>>		=		>		>	
	2	>>>		=		=		>	
	3	>>>		<		>		>	
<i>S. santa fe red</i>	1	>		=		>		>	
	2	>		=		>		>	
	3	>>		=		>		>	
<i>L. usitatissimum</i>	1	>		<		>>		>	
	2	>		<		>>		>	
	3	>		<		>		>	
<i>P. tanacetifolia</i>	1	>		=		>>		>>	
	2	>		=		>		>>	
	3	>		=		>		>	
<i>M. sylvestris</i>	part	AP*	Flower	AP	Flower	AP	Flower	AP	Flower
	1	>>	>	=	>	>>>	>>>	>	>
	2	>>>	>	<	=	>>>	>>>	>	>
	3	>>>	>	<	=	>>>	>>>	>	>
<i>C. album</i>	1	>		>		>		>	
	2	>		>		>		>	
	3	>		>>		=		>	





















< : EF inferior to concentration measured on control or uncontaminated conditions; = :EF equal or ~1; > : EF 1.3-5 times higher; >> : EF 5-10 times higher; >>> : EF above 10 times higher. * AP: aboveground part

Table 7. Cd and Pb BCF tot group ranking matrix according to collected plant part (aboveground part, leaves, root and flower) of annual crops on the area 1, area 2, area 3 and BCF repartition in the group.

metal	Crops	area	part		metal	Crops	area	part				
			Root	Leaves				Root	Leaves			
Cd	<i>B. vulgaris</i>	1			Pb	<i>B. vulgaris</i>	1			[0;0.01[	
		2					2					
		3					3					
	<i>S. Santa fe red</i>	1				<i>S. Santa fe red</i>	1				[0.01;0.1[
		2					2					
		3					3					
	<i>S. Biomass 133</i>	1				<i>S. Biomass 133</i>	1				[0.1;0.25[
		2					2					
		3					3					
	<i>L. usitatissimum</i>	1				<i>L. usitatissimum</i>	1				[0.25;0.75[
		2					2					
		3					3					
	<i>P. tanacetifolia</i>	1				<i>P. tanacetifolia</i>	1				[0.75;1[
		2					2					
		3					3					
	<i>M. sylvestris</i>			AP*		Flower	<i>M. sylvestris</i>		AP	Flower	>1	
		1						1				
		2						2				
		3						3				
	<i>C. album</i>	1				<i>C. album</i>	1					
		2					2					
3				3								
BCF repartition		0	33	37	30	0						
BCF repartition		93	7	0	0	0						

*AP: aboveground part

Table 8. Cu and Zn BCF tot group ranking matrix according to collect plant parts (aboveground part, leaves, root and flower) of annual crops on the area 1, area 2, area 3 and BCF repartition in the group.

metal	Crops	area	Part		metal	Crops	area	Part				
			Root	Leaves				Root	Leaves			
Cu	<i>B. vulgaris</i>	1			<i>B. vulgaris</i>	1			[0;0.01[		
		2				2			[0.01;0.1[		
		3				3			[0.1;0.25[		
	<i>S. Santa fe red</i>	1			<i>S. Santa fe red</i>	1			[0.25;0.75[		
		2				2			[0.75;1[		
		3				3			>1			
	<i>S. Biomass 133</i>	1			<i>S. Biomass 133</i>	1			[0.25;0.75[		
		2				2			[0.75;1[		
		3				3			>1			
	<i>L. usitatissimum</i>	1			<i>L. usitatissimum</i>	1			[0.25;0.75[		
		2				2			[0.75;1[		
		3				3			>1			
	<i>P. tanacetifolia</i>	1			<i>P. tanacetifolia</i>	1			[0.25;0.75[		
		2				2			[0.75;1[		
		3				3			>1			
	<i>M. sylvestris</i>			AP*	Flower	<i>M. sylvestris</i>		AP	Flower			
		1					1				[0.25;0.75[
		2					2				[0.75;1[
	<i>C. album</i>	1				<i>C. album</i>	1			[0.25;0.75[	
		2					2			[0.75;1[	
		3					3			>1		
BCF repartition		0	63	26	0	0						
BCF repartition		0	96	4	0	0						

*AP: aboveground part

Table 9. 1M NH₄NO₃-extractable metal concentrations at the start and the harvest time for each area of plot sown with annual crops. Significant differences between conditions are indicated by different letters at the level of $\alpha = 0.05$.

		Cd		Cu		Pb		Zn	
		At the start	At harvest	At the start	At harvest	At the start	At harvest	At the start	At harvest
<i>B. vulgaris</i>	Area 1	0.04 (±0.01) ^a	0.04 (±0.01) ^a	0.57 (±0.30) ^a	0.58 (±0.11) ^a	0.04 (±0.01) ^a	0.05 (±0.01) ^a	1.44 (±0.69) ^a	1.10 (±0.12) ^a
	Area 2	0.03 (±0.01) ^a	0.03 (±0.01) ^a	0.64 (±0.22) ^a	0.59 (±0.11) ^a	0.04 (±0.01) ^a	0.04 (±0.01) ^a	1.04 (±0.14) ^a	1.13 (±0.19) ^a
	Area 3	0.04 (±0.01) ^a	0.07 (±0.09) ^a	0.64 (±0.11) ^a	1.34 (±1.73) ^a	0.04 (±0.01) ^a	0.08 (±0.07) ^a	0.88 (±0.22) ^a	2.22 (±3.41) ^a
<i>S. Biomass 133</i>	Area 1	0.05 (±0.01) ^{a,c}	0.03 (±0.00) ^b	0.8 (±0.61) ^a	0.46 (±0.06) ^{a,b}	0.05 (±0.03) ^a	0.09 (±0.06) ^a	6.42 (±0.75) ^a	7.68 (±1.04) ^a
	Area 2	0.02 (±0.01) ^b	0.03 (±0.01) ^b	0.36 (±0.08) ^{a,b}	0.40 (±0.06) ^b	0.04 (±0.02) ^a	0.08 (±0.07) ^a	6.77 (±0.51) ^a	10.84 (±2.74) ^b
	Area 3	0.03 (±0.01) ^a	0.04 (±0.01) ^c	0.76 (±0.40) ^{a,b}	0.47 (±0.08) ^{a,b}	0.05 (±0.03) ^a	0.08 (±0.06) ^a	6.31 (±1.02) ^a	10.27 (±1.72) ^b
<i>S. Santa Fe red</i>	Area 1	0.04 (±0.01) ^a	0.02 (±0.01) ^b	0.78 (±0.80) ^a	0.45 (±0.09) ^a	0.05 (±0.04) ^a	0.08 (±0.06) ^a	6.4 (±1.10) ^a	6.4 (±0.83) ^a
	Area 2	0.03 (±0.00) ^{b,c}	0.03 (±0.01) ^b	0.34 (±0.04) ^a	0.42 (±0.05) ^a	0.05 (±0.03) ^a	0.05 (±0.01) ^a	6.85 (±0.62) ^{a,b}	8.3 (±0.68) ^b
	Area 3	0.05 (±0.01) ^a	0.04 (±0.01) ^{a,c}	0.47 (±0.09) ^a	0.5 (±0.13) ^a	0.07 (±0.07) ^a	0.06 (±0.03) ^a	5.76 (±0.92) ^a	8.7 (±1.65) ^b
<i>L. usitatissimum</i>	Area 1	0.04 (±0.00) ^{a,d}	0.02 (±0.00) ^b	0.37 (±0.06) ^a	0.59 (±0.18) ^{a,b}	0.03 (±0.01) ^a	0.09 (±0.04) ^a	5.12 (±0.17) ^{a,b}	5.25 (±0.57) ^a
	Area 2	0.03 (±0.01) ^{a,b}	0.03 (±0.00) ^{a,b}	0.88 (±0.38) ^b	0.4 (±0.06) ^{a,b}	0.09 (±0.04) ^a	0.12 (±0.12) ^a	8.02 (±0.94) ^{b,c}	8.27 (±0.78) ^c
	Area 3	0.07 (±0.01) ^c	0.05 (±0.01) ^d	0.55 (±0.14) ^{a,b}	0.59 (±0.26) ^{a,b}	0.13 (±0.16) ^a	0.07 (±0.06) ^a	8.4 (±1.49) ^c	10.31 (±2.87) ^c
<i>P. tanacetifolia</i>	Area 1	0.05 (±0.01) ^a	0.03 (±0.00) ^b	0.45 (±0.10) ^a	0.68 (±0.42) ^a	0.04 (±0.01) ^a	0.03 (±0.02) ^a	5.44 (±0.23) ^a	8.69 (±2.98) ^{a,c}
	Area 2	0.03 (±0.00) ^b	0.04 (±0.01) ^a	0.74 (±0.11) ^a	1.52 (±2.78) ^a	0.07 (±0.05) ^a	0.07 (±0.09) ^a	8.1 (±0.17) ^{a,c}	12.7 (±2.40) ^b
	Area 3	0.06 (±0.01) ^c	0.06 (±0.01) ^c	1.63 (±2.01) ^a	0.86 (±0.44) ^a	0.05 (±0.02) ^a	0.04 (±0.02) ^a	7.88 (±3.55) ^{a,c}	11.15 (±1.57) ^{b,c}
<i>M. sylvestris</i>	Area 1	0.05 (±0.01) ^a	0.04 (±0.01) ^a	0.40 (±0.05) ^a	0.82 (±0.84) ^a	0.03 (±0.01) ^a	0.06 (±0.04) ^a	5.82 (±0.55) ^a	7.06 (±1.46) ^a
	Area 2	0.03 (±0.00) ^b	0.05 (±0.01) ^a	0.63 (±0.38) ^a	0.63 (±0.13) ^a	0.04 (±0.01) ^a	0.03 (±0.01) ^a	7.11 (±0.74) ^{a,b}	9.61 (±1.60) ^b
	Area 3	0.08 (±0.01) ^c	0.06 (±0.01) ^c	0.78 (±0.54) ^a	1.3 (±1.13) ^a	0.06 (±0.03) ^a	0.05 (±0.08) ^a	8.14 (±1.70) ^{a,b}	12.65 (±2.06) ^c

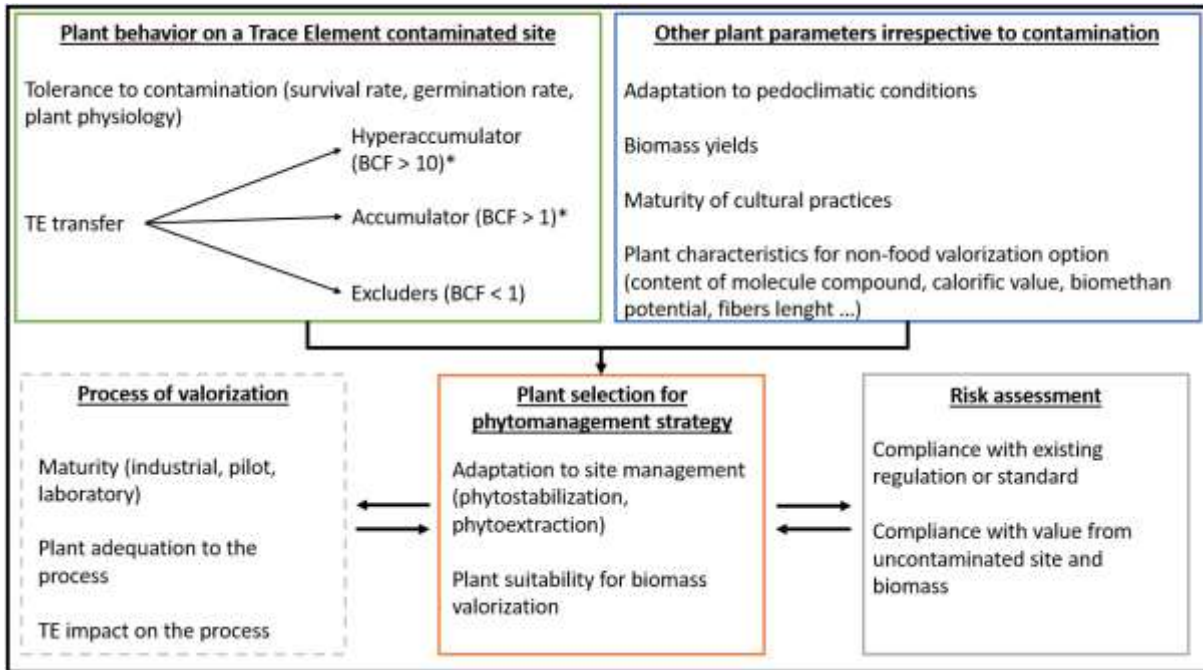


Fig. 1.

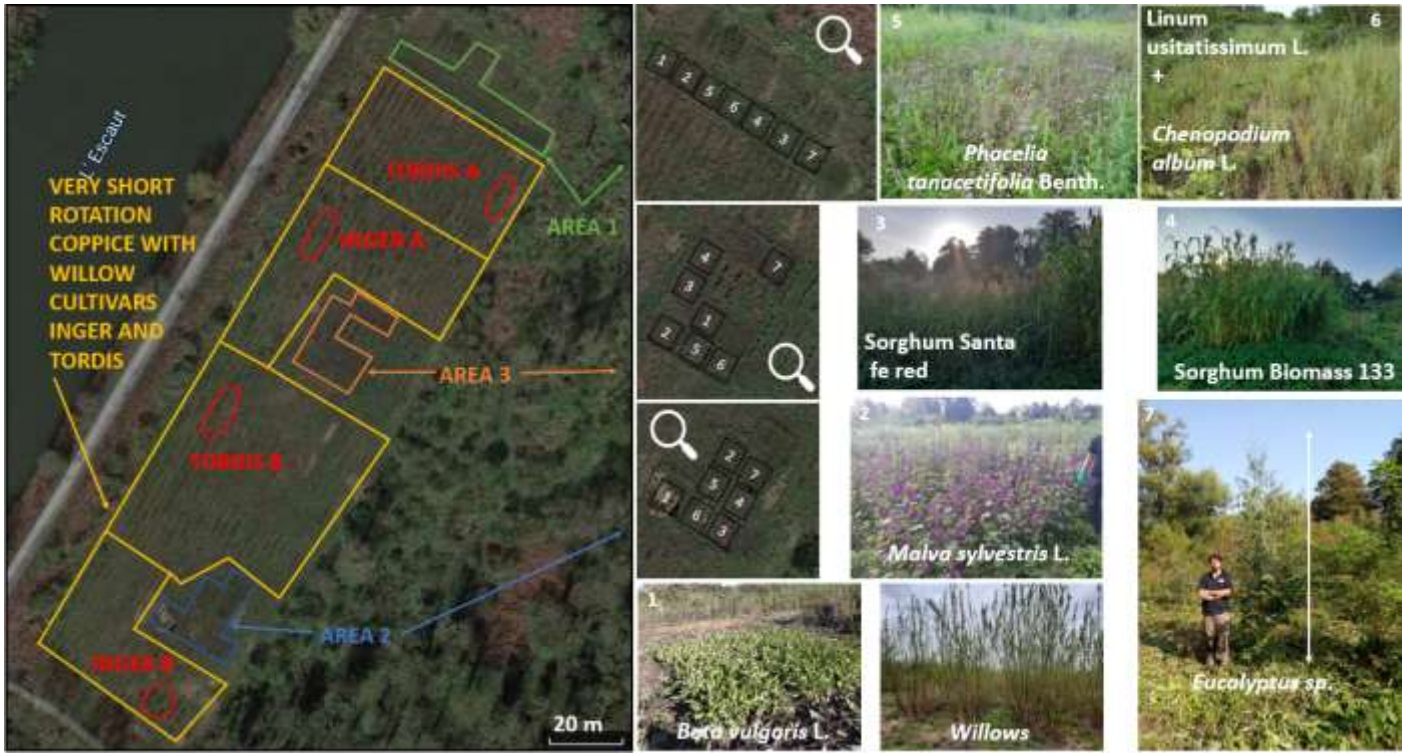


Fig. 2.

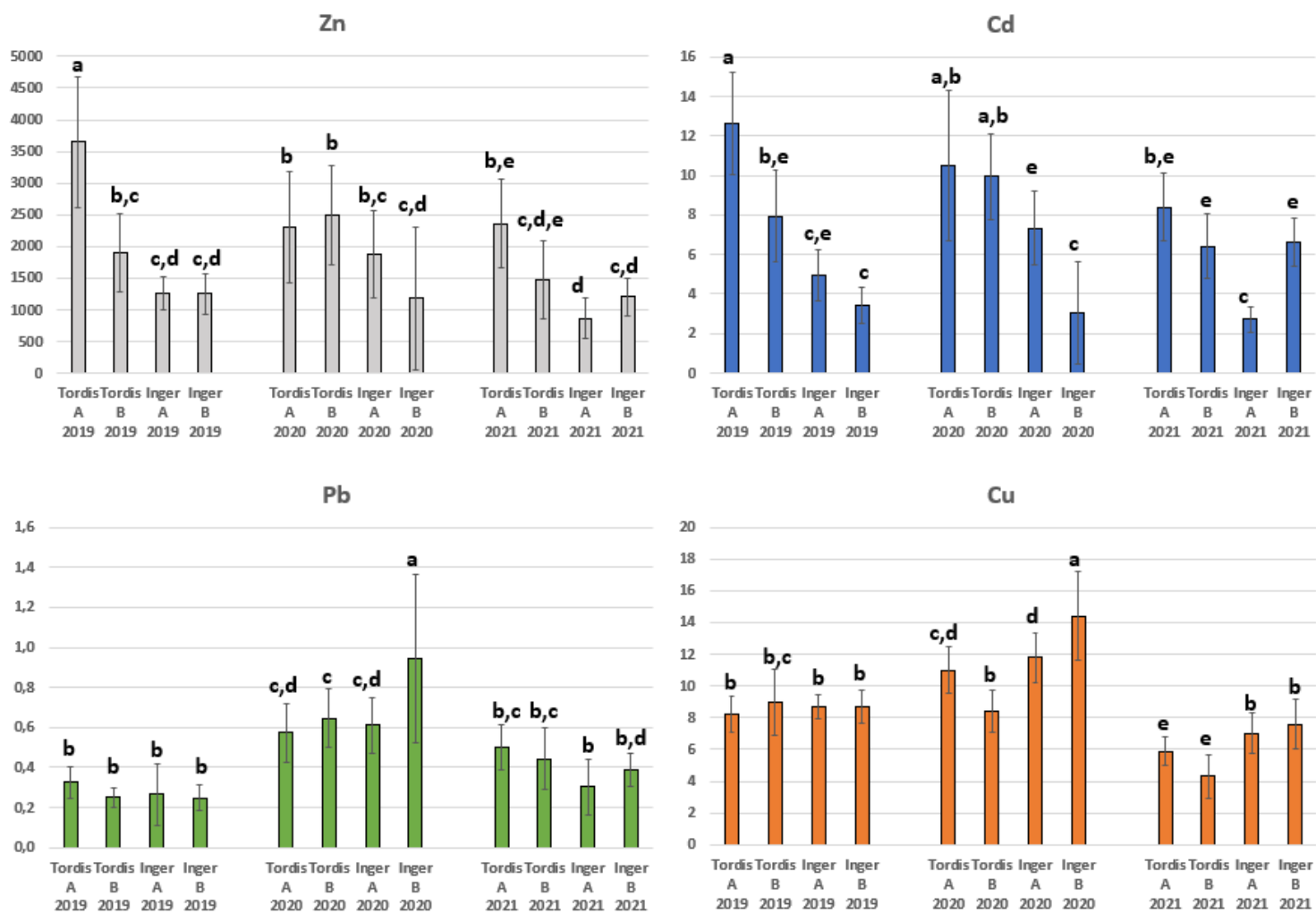


Fig. 3.

Supplemental Table 1. Mean BCF tot and standard deviation ($\mu\text{g g}^{-1}$ of DW, $\pm\text{SD}$) in the collected plant part of the annual crops and Eucalyptus on the different areas (area 1, area 2, area 3). Significant differences between conditions (area of cultivation, plant parts, cultivars, year) are indicated by different letters at the level of $\alpha = 0.05$.

BCF _{tot}		<i>B. vulgaris</i>		<i>S. Bicolor</i>		<i>L. usitatissimum</i>	<i>P. tanacetifolia</i>	<i>M. sylvestris</i>		<i>C. album</i>	<i>Eucalyptus</i> (leaves)	
		Leaves	Root	Biomass 133	Santa fe red			Aerial part	Flower		Years 2	Years 3
Cd	Area 1	0.245 (± 0.076) ^b	0.091 (± 0.018) ^a	0.498 (± 0.148) ^a	0.227 (± 0.068) ^b	0.191 (± 0.041) ^a	0.110 (± 0.033) ^a	0.485 (± 0.170) ^a	0.196 (± 0.056) ^b	0.037 (± 0.005) ^a	0.199 (± 0.065) ^a	0.041 (± 0.025) ^{c,d}
	Area 2	0.158 (± 0.139) ^{a,b}	0.108 (± 0.031) ^a	0.557 (± 0.261) ^a	0.288 (± 0.13) ^{b,c}	0.409 (± 0.128) ^b	0.098 (± 0.035) ^a	0.441 (± 0.156) ^a	0.147 (± 0.039) ^b	0.076 (± 0.004) ^b	0.121 (± 0.004) ^{b,d}	0.025 (± 0.011) ^c
	Area 3	0.121 (± 0.045) ^a	0.070 (± 0.015) ^a	0.447 (± 0.082) ^{a,c}	0.212 (± 0.074) ^b	0.163 (± 0.031) ^a	0.055 (± 0.013) ^b	0.434 (± 0.150) ^a	0.127 (± 0.026) ^b	0.043 (± 0.001) ^a	0.065 (± 0.022) ^{b,c}	0.007 (± 0.002) ^c
Cu	Area 1	0.102 (± 0.010) ^a	0.077 (± 0.017) ^a	0.092 (± 0.022) ^a	0.081 (± 0.016) ^{a,b}	0.088 (± 0.013) ^a	0.084 (± 0.010) ^a	0.120 (± 0.036) ^{a,b,c}	0.158 (± 0.086) ^b	0.752 (± 1.119) ^a	0.144 (± 0.023) ^a	0.100 (± 0.017) ^a
	Area 2	0.083 (± 0.064) ^a	0.084 (± 0.015) ^a	0.100 (± 0.033) ^a	0.076 (± 0.018) ^{a,b}	0.087 (± 0.023) ^{a,b}	0.095 (± 0.024) ^a	0.124 (± 0.019) ^{a,b,c}	0.117 (± 0.040) ^{a,b,c}	1.389 (± 1.202) ^a	0.120 (± 0.022) ^a	0.100 (± 0.052) ^a
	Area 3	0.080 (± 0.017) ^a	0.067 (± 0.007) ^a	0.060 (± 0.020) ^b	0.059 (± 0.014) ^b	0.070 (± 0.013) ^b	0.057 (± 0.008) ^b	0.162 (± 0.081) ^{a,b,c}	0.080 (± 0.014) ^c	1.812 (± 2.696) ^a	0.084 (± 0.013) ^a	0.080 (± 0.004) ^a
Pb	Area 1	0.004 (± 0.002) ^a	0.001 (± 0.000) ^b	0.001 (± 0.000) ^{a,b}	0.002 (± 0.002) ^a	0.004 (± 0.004) ^a	0.007 (± 0.004) ^a	0.007 (± 0.003) ^a	0.005 (± 0.002) ^a	0.004 (± 0.004) ^a	0.001 (± 0.000) ^{a,c}	0.000 (± 0.000) ^c
	Area 2	0.003 (± 0.002) ^{a,b,c}	0.002 (± 0.001) ^{a,b,c}	0.001 (± 0.000) ^{a,b}	0.001 (± 0.001) ^{a,b}	0.004 (± 0.003) ^a	0.004 (± 0.003) ^{a,b}	0.012 (± 0.010) ^a	0.013 (± 0.015) ^a	0.007 (± 0.005) ^a	0.003 (± 0.002) ^{a,b}	0.000 (± 0.000) ^c
	Area 3	0.004 (± 0.003) ^{a,b}	0.001 (± 0.000) ^c	0.001 (± 0.000) ^{a,b}	0.001 (± 0.000) ^b	0.001 (± 0.000) ^b	0.002 (± 0.001) ^b	0.007 (± 0.004) ^a	0.005 (± 0.005) ^a	0.001 (± 0.000) ^a	0.004 (± 0.002) ^b	0.000 (± 0.000) ^c
Zn	Area 1	0.105 (± 0.022) ^a	0.028 (± 0.006) ^c	0.032 (± 0.011) ^{a,b}	0.034 (± 0.009) ^b	0.022 (± 0.004) ^a	0.028 (± 0.005) ^a	0.050 (± 0.009) ^a	0.024 (± 0.004) ^b	0.037 (± 0.014) ^a	0.018 (± 0.002) ^a	0.007 (± 0.002) ^c
	Area 2	0.095 (± 0.025) ^a	0.025 (± 0.006) ^c	0.029 (± 0.008) ^{a,b}	0.035 (± 0.006) ^b	0.019 (± 0.003) ^a	0.028 (± 0.009) ^a	0.052 (± 0.021) ^a	0.031 (± 0.017) ^{b,c}	0.057 (± 0.010) ^a	0.015 (± 0.002) ^{a,b}	0.010 (± 0.002) ^c
	Area 3	0.060 (± 0.009) ^b	0.017 (± 0.002) ^c	0.023 (± 0.004) ^a	0.029 (± 0.006) ^{a,b}	0.011 (± 0.002) ^b	0.014 (± 0.003) ^b	0.043 (± 0.013) ^{a,c}	0.024 (± 0.005) ^b	0.044 (± 0.023) ^a	0.011 (± 0.003) ^{b,c}	0.005 (± 0.001) ^c

Supplemental Table 2. Percent of Cd, Cu, Pb, Zn mobility in the sediment on the three areas at both sampling time (at the start and at harvest). Significant differences between conditions are indicated by different letters at the level of $\alpha = 0.05$.

Mobility %		Cd		Cu		Pb		Zn	
		At the start	At harvest	At the start	At harvest	At the start	At harvest	At the start	At harvest
<i>B. vulgaris</i>	Area 1	0.586 (±0.211) ^a	0.574 (±0.071) ^a	0.655 (±0.344) ^a	0.669 (±0.122) ^a	0.005 (±0.002) ^a	0.006 (±0.002) ^a	0.022 (±0.010) ^a	0.016 (±0.002) ^a
	Area 2	0.481 (±0.083) ^a	0.553 (±0.089) ^a	0.832 (±0.293) ^a	0.770 (±0.150) ^a	0.007 (±0.002) ^a	0.007 (±0.001) ^a	0.017 (±0.002) ^a	0.019 (±0.003) ^a
	Area 3	0.441 (±0.030) ^a	0.829 (±1.130) ^a	0.636 (±0.113) ^a	1.325 (±1.710) ^a	0.003 (±0.001) ^a	0.008 (±0.007) ^a	0.010 (±0.002) ^a	0.025 (±0.038) ^a
<i>S. Biomass 133</i>	Area 1	0.772 (±0.098) ^a	0.330 (±0.041) ^d	0.919 (±0.696) ^a	0.455 (±0.059) ^b	0.007 (±0.004) ^a	0.009 (±0.005) ^a	0.096 (±0.011) ^a	0.086 (±0.012) ^a
	Area 2	0.477 (±0.106) ^b	0.568 (±0.147) ^{b,c}	0.467 (±0.108) ^{a,b}	0.520 (±0.079) ^{a,b}	0.008 (±0.003) ^a	0.014 (±0.012) ^a	0.111 (±0.008) ^{a,c}	0.178 (±0.045) ^b
	Area 3	0.652 (±0.134) ^{a,b}	0.674 (±0.127) ^{a,c}	0.754 (±0.399) ^{a,b}	0.541 (±0.089) ^{a,b}	0.005 (±0.003) ^a	0.011 (±0.008) ^a	0.070 (±0.011) ^a	0.154 (±0.026) ^{b,c}
<i>S. Santa fe red</i>	Area 1	0.706 (±0.115) ^a	0.267 (±0.059) ^b	0.897 (±0.920) ^a	0.445 (±0.087) ^a	0.006 (±0.006) ^a	0.008 (±0.006) ^a	0.096 (±0.016) ^{a,c}	0.071 (±0.009) ^{b,c}
	Area 2	0.557 (±0.043) ^a	0.559 (±0.095) ^a	0.444 (±0.049) ^a	0.555 (±0.062) ^a	0.008 (±0.005) ^a	0.008 (±0.002) ^a	0.113 (±0.010) ^{a,d}	0.136 (±0.011) ^d
	Area 3	0.552 (±0.117) ^a	0.613 (±0.116) ^a	0.463 (±0.088) ^a	0.579 (±0.150) ^a	0.007 (±0.006) ^a	0.008 (±0.003) ^a	0.064 (±0.010) ^b	0.130 (±0.025) ^d
<i>L. usitatissimum</i>	Area 1	0.616 (±0.049) ^{a,d,e}	0.275 (±0.027) ^c	0.427 (±0.066) ^a	0.585 (±0.181) ^a	0.004 (±0.001) ^a	0.009 (±0.004) ^a	0.077 (±0.003) ^a	0.059 (±0.006) ^a
	Area 2	0.545 (±0.096) ^{a,d}	0.507 (±0.057) ^d	1.155 (±0.501) ^b	0.524 (±0.074) ^a	0.015 (±0.006) ^a	0.020 (±0.020) ^a	0.132 (±0.015) ^{b,c}	0.136 (±0.013) ^c
	Area 3	0.817 (±0.157) ^{b,e}	0.722 (±0.102) ^e	0.543 (±0.138) ^a	0.680 (±0.294) ^a	0.013 (±0.015) ^a	0.009 (±0.008) ^a	0.093 (±0.017) ^{a,b}	0.154 (±0.043) ^c
<i>P. tanacetifolia</i>	Area 1	0.721 (±0.127) ^{a,d,e}	0.452 (±0.039) ^c	0.514 (±0.119) ^a	0.782 (±0.483) ^a	0.005 (±0.002) ^a	0.004 (±0.003) ^a	0.081 (±0.003) ^{a,b,c}	0.130 (±0.045) ^b
	Area 2	0.516 (±0.053) ^{b,c}	0.783 (±0.104) ^d	0.969 (±0.147) ^a	1.995 (±3.637) ^a	0.012 (±0.009) ^a	0.011 (±0.016) ^a	0.133 (±0.003) ^a	0.209 (±0.039) ^c
	Area 3	0.673 (±0.143) ^{a,b,d,e}	0.661 (±0.058) ^e	1.611 (±1.939) ^a	0.848 (±0.433) ^a	0.005 (±0.002) ^a	0.004 (±0.002) ^a	0.088 (±0.040) ^{a,b}	0.124 (±0.018) ^b
<i>M. sylvestris</i>	Area 1	0.774 (±0.103) ^{a,b,d}	0.602 (±0.097) ^c	0.455 (±0.059) ^a	0.939 (±0.961) ^a	0.004 (±0.001) ^a	0.007 (±0.006) ^a	0.087 (±0.008) ^a	0.106 (±0.022) ^a
	Area 2	0.606 (±0.056) ^{a,c,e}	0.871 (±0.126) ^d	0.826 (±0.499) ^a	0.820 (±0.174) ^a	0.006 (±0.002) ^a	0.005 (±0.002) ^a	0.117 (±0.012) ^a	0.158 (±0.026) ^b
	Area 3	0.845 (±0.103) ^{b,d,e}	0.719 (±0.088) ^{c,e}	0.769 (±0.529) ^a	1.284 (±1.119) ^a	0.006 (±0.003) ^a	0.005 (±0.007) ^a	0.091 (±0.019) ^a	0.141 (±0.023) ^b

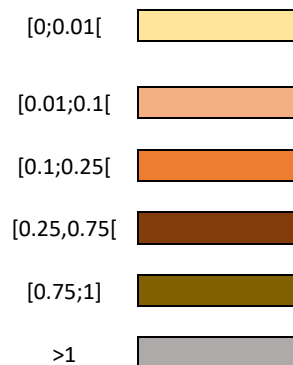
Supplemental Table 3. Bio Concentration Factor (BCF) tot of Cd, Cu, Pb and Zn in the leaves of the willow' cultivars Tordis and Inger on the area A and B in 2019, 2020 and, 2021. Significant differences between conditions are indicated by different letters at the level of $\alpha = 0.05$.

	Cd	Cu	Pb	Zn
Tordis A 2019	1.55 (± 0.32) ^{a,d}	0.07 (± 0.01) ^{a,d}	0.0003 (± 0.0001) ^{a,e}	0.60 (± 0.17) ^a
Tordis B 2019	0.98 (± 0.28) ^{b,d,f}	0.08 (± 0.02) ^{a,e}	0.0003 (± 0.0001) ^a	0.31 (± 0.10) ^{b,c,e}
Inger A 2019	0.61 (± 0.16) ^{c,e}	0.08 (± 0.01) ^{a,d}	0.0003 (± 0.0002) ^a	0.21 (± 0.04) ^{b,d,e}
Inger B 2019	0.42 (± 0.11) ^c	0.08 (± 0.01) ^{a,e}	0.0003 (± 0.0001) ^a	0.21 (± 0.05) ^{b,d}
Tordis A 2020	1.3 (± 0.47) ^d	0.1 (± 0.01) ^f	0.0006 (± 0.0002) ^{b,f}	0.38 (± 0.14) ^{c,f}
Tordis B 2020	1.23 (± 0.27) ^d	0.08 (± 0.01) ^{a,e,g}	0.0007 (± 0.0002) ^b	0.41 (± 0.13) ^{c,g}
Inger A 2020	0.91 (± 0.23) ^e	0.11 (± 0.01) ^f	0.0006 (± 0.0001) ^c	0.31 (± 0.11) ^{b,c}
Inger B 2020	0.38 (± 0.32) ^c	0.13 (± 0.03) ^g	0.001 (± 0.0004) ^d	0.19 (± 0.18) ^{b,d}
Tordis A 2021	1.04 (± 0.21) ^{b,f}	0.05 (± 0.01) ^{b,c}	0.0005 (± 0.0001) ^{b,e}	0.39 (± 0.11) ^{f,g}
Tordis B 2021	0.79 (± 0.20) ^{b,f}	0.04 (± 0.01) ^c	0.0005 (± 0.0002) ^{a,b,c}	0.24 (± 0.10) ^{e,f}
Inger A 2021	0.33 (± 0.08) ^c	0.06 (± 0.01) ^{b,d}	0.0003 (± 0.0001) ^a	0.14 (± 0.05) ^{d,e}
Inger B 2021	0.81 (± 0.15) ^{b,e,f}	0.07 (± 0.01) ^{b,d,e}	0.0004 (± 0.0001) ^{a,c,f}	0.20 (± 0.05) ^{b,d,e}

Supplemental Table 4. Cd and Pb Bio Concentration Factor (BCF) groups ranking matrice for the Eucalyptus sp. (year 2 and year 3) and the Willow cultivars Tordis and Inger (2019, 2020, 2021) on the sampling areas and BCF repartition in the different groups.

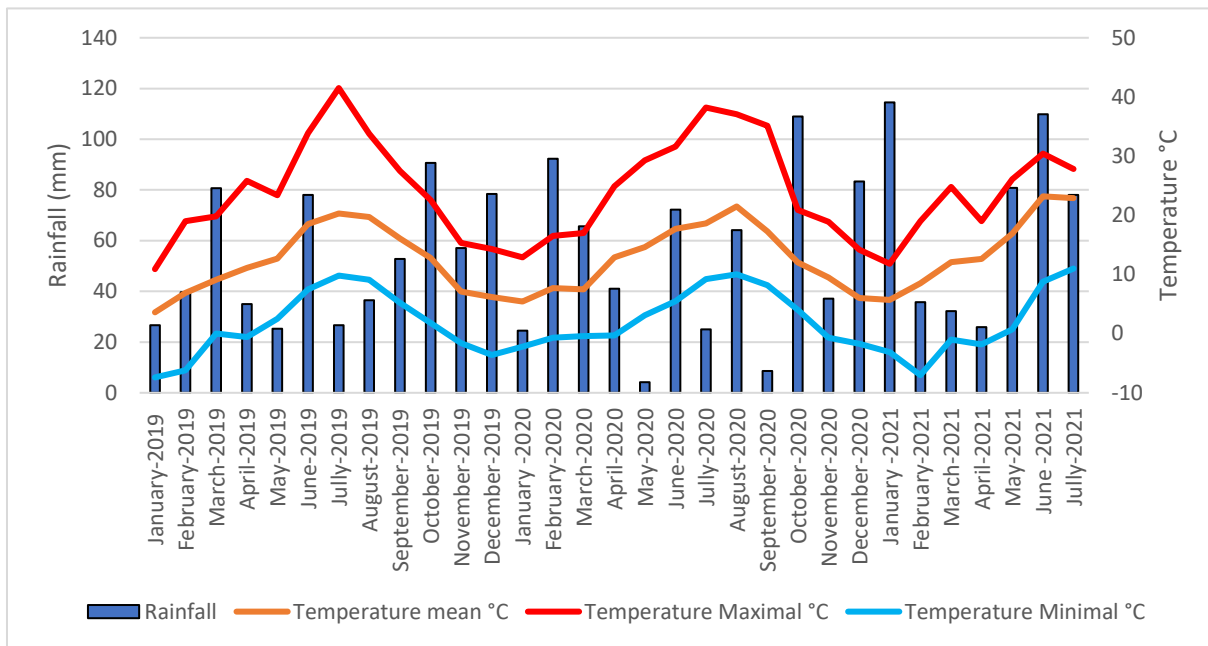
I	Crops	Area / Year					
		Year	Year 2	Year 3			
Cd	<i>Eucalyptus sp.</i>	1					
		2					
		3					
	<i>Tordis</i>	Area	A	B			
		2019					
		2020					
		2021					
	<i>Inger</i>	Area	A	B			
		2019					
		2020					
		2021					
	BCF repartition		6	17	11	22	22

	Crops	Area / Year					
		Year	Year 2	Year 3			
Pb	<i>Eucalyptus sp.</i>	1					
		2					
		3					
	<i>Tordis</i>	Area	A	B			
		2019					
		2020					
		2021					
	<i>Inger</i>	Area	A	B			
		2019					
		2020					
		2021					
	BCF repartition		100	0	0	0	0



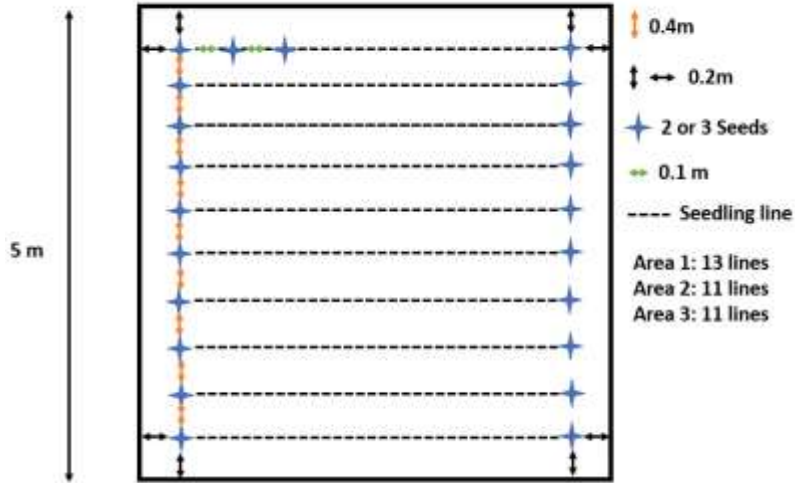
	Crops	Area / Year					
		Year	Year 2	Year 3			
Cu	<i>Eucalyptus sp.</i>	1					
		2					
		3					
	<i>Tordis</i>	Area	A	B			
		2019					
		2020					
		2021					
	<i>Inger</i>	Area	A	B			
		2019					
		2020					
		2021					
	BCF repartition		5	67	28	0	0

	Crops	Area / Year					
		Year	Year 2	Year 3			
Zn	<i>Eucalyptus sp.</i>	1					
		2					
		3					
	<i>Tordis</i>	Area	A	B			
		2019					
		2020					
		2021					
	<i>Inger</i>	Area	A	B			
		2019					
		2020					
		2021					
	BCF repartition		10	20	34	34	0

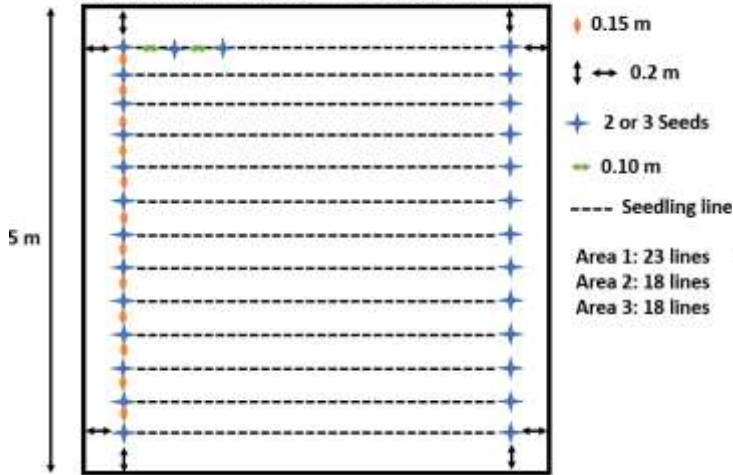


Supplemental Fig. 1.

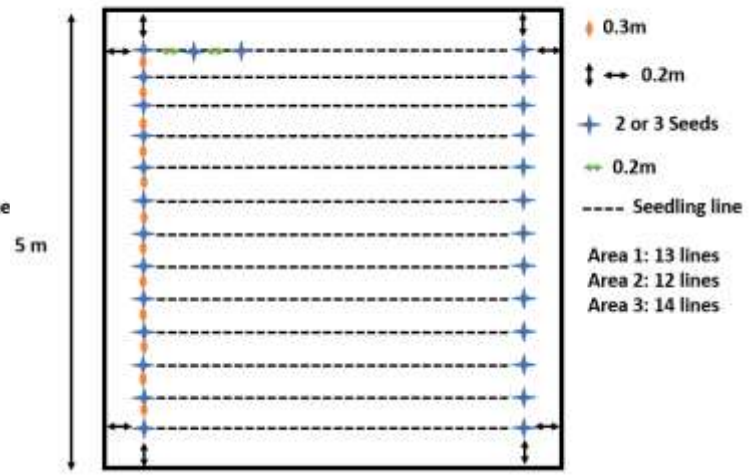
Sorgho sowing design by 25 m² plot



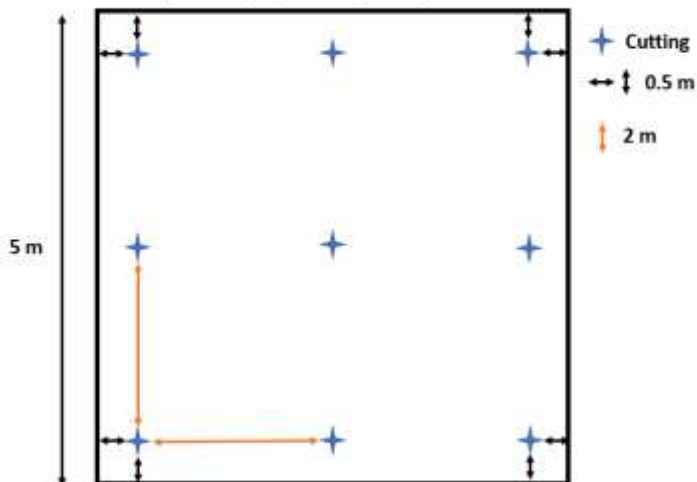
Linum usitatissimum and Phacelia tanacetifolia sowing design by 25 m² plot



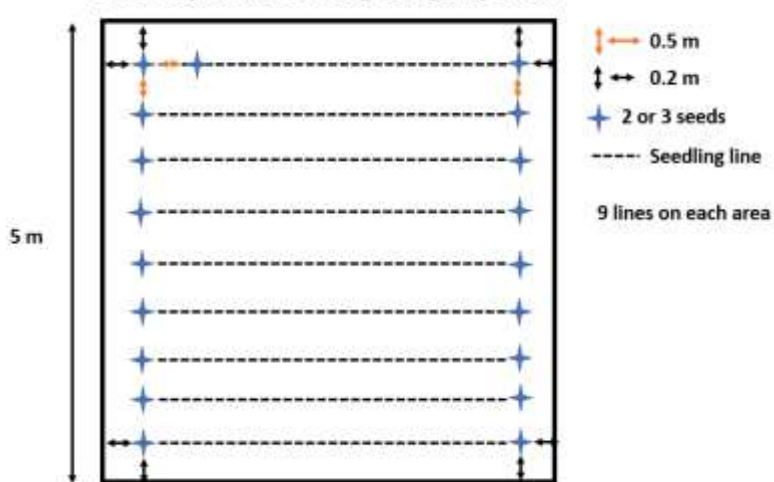
Beta vulgaris sowing design by 25 m² plot



Eucalyptus sp. planting design on 25 m²



Malva sylvestris sowing design by 25 m²



Supplemental Fig. 2.