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Influence of sulfur amendments on heavy metals phytoextraction from agricultural contaminated soils: A meta-analysis☆

Sissou Zakari[a,b,c,1], Xiaojin Jiang[a,b,1], Xiai Zhu[a,b], Wenjie Liu[a,b,*], M. Gloriose B. Allakonon[c], Ashutosh Kumar Singh[a,b], Chunfeng Chen[a,b], Xin Zou[a,b], P.B. Irénikatché Akponikpè[c], Gbadamassi G.O. Dossa[a,b], Bin Yang[a,b]

[a] CAS Key Laboratory of Tropical Forest Ecology, Xishuangbanna Tropical Botanical Garden, Chinese Academy of Sciences, Menglun, Yunnan, 666303, China
[b] Center of Plant Ecology, Core Botanical Gardens, Chinese Academy of Sciences, Menglun, Yunnan, 666303, China
[c] Laboratory of Hydraulics and Environmental Modeling (HydroModE-Lab), Faculté d'Agronomie, Université de Parakou, 03 BP 351, Parakou, Benin

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ABSTRACT

Heavy metal pollution is becoming recurrent and threatens biota biosafety in many agricultural fields. Diverse solutions explore the application of amendments to enable remediation. Sulfur represents a nonmetallic chemical element that actively affects heavy metals phytoextraction, and promotes and alternatively mitigates soil functions. In this study, we conduct a meta-analysis to synthesize the current knowledge on the influence of sulfur amendments on plants heavy metals uptake from contaminated soil media. Random-effects model was used to summarize effect sizes from 524 data points extracted from 30 peer reviewed studies. The phytoextraction of cadmium, chromium and nickel were 1.6-, 3.3-, and 12.6-fold, respectively, higher when sulfur amendment was applied; while copper uptake was 0.3-fold lower. Irrespective of the sulfur type, heavy metal extraction increased with the raising sulfur stress. Individual organs showed significant differences of heavy metal uptake between sulfur applied and non-sulfur treatments, and combined organs did not. The heavy metals uptake in leaves and roots were higher in sulfur applied than non-sulfur applied treatments, while those in grain, husk, and stalks were lower. The heavy metals phytoextraction (response ratio) followed the order roots > leaves > stalk > grain > husk. Moreover, heavy metals uptake was 2-fold higher in the sulfur applied than the non-sulfur treatments under ideal (5.5–8) and alkaline conditions (8–14), and 0.2-fold lower under acidic pH (1–5.5). Cadmium, manganese and nickel, and chromium were the most extracted under sulfur application by *Vicia* sp.*, Sorghum* sp. *and Brassica* sp., respectively; while chromium, manganese, and iron were the most uptake without sulfur amendments by *Oryza* sp., *Zea* sp. and *Sorghum* sp., respectively. Our study highlights that the influence of sulfur on heavy metal phytoextraction depends on the single or combined effects of sulfur stress intensity, sulfur compounds, plant organ, plant type, and soil pH condition.

1. Introduction

Heavy metals, hereafter HMs, constitute metals or metalloids with relatively high atomic numbers, atomic weigh, or density. Most HMs, especially trace metals, play a dual role in the biochemical cycles within soil and water environments. Heavy metals show long-residence time and represent non-degradable environmental pollutants (Chen et al., 2015; Li et al., 2015), with toxic effects on most of the living organisms at higher or lower concentrations depending of the considered HMs. However, HMs can also act as essential micro-nutrients at low concentrations (Driscoll et al., 1994). HMs pollution originates from natural sources such as rocks and metalliferous minerals or from anthropogenic activities including agriculture, metallurgy (Yu et al., 2015), energy production, mining (Kapusta and Sobczyk, 2015), sewage sludge, and waste disposal (Farmer et al., 2002; Liu et al., 2018; Oves et al., 2016; Qu et al., 2016). Approximately, the European Union hosts 0.5 million

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* Corresponding author. CAS Key Laboratory of Tropical Forest Ecology, Xishuangbanna Tropical Botanical Garden, Chinese Academy of Sciences, Menglun, Yunnan, 666303, China.

E-mail address: lwj@xtbg.org.cn (W. Liu).

[1] These authors contribute equally.

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highly HMs polluted site with an extra 3.5 million potentially polluted ones in countries including Germany, England, Denmark, Spain, Italy, Netherlands, Finland, Sweden, France, Hungary, Slovakia, Austria, Greece, Poland, Ireland and Portugal (Perez and Young, 2012). Moreover, about 600,000 ha brown field sites in the US (Surriya et al., 2015), and one-sixth of the total farmland area in China (Convard et al., 2005; Liu et al., 2005) have been classified as polluted field with HMs. The environmental pollution with HMs can induce adverse effects on soil and its ecosystems (Pan and Yu, 2011; Wang et al., 2016), or on human health through food chain and produce deleterious effects on the biological systems (Adrees et al., 2015; Arora et al., 2008; Kara, 2005; Memon and Schröder, 2009; Shaheen et al., 2016). For instance, toxic metals can replace vital metals in enzymes and pigments manipulating their biochemical role (Malayeri et al., 2008). Hence, the environmental pollution due to HMs has gained more attention recently and has become a global concern which request special action and care (Morcillo et al., 2016) (see Table 1).

The immobilization of HMs from contaminated soils using different soil amendments represents one of the most effective way to remediate contaminated soils (Ahmad et al., 2012; Ok et al., 2011; Tica et al., 2011). Sulfur is a very reactive nonmetallic chemical element that can naturally occur in soil in inorganic (sulphides, sulphates, etc.) and organic fractions associated with nitrogen (N) and carbon (C). Sulfur is usually applied as soil amendment to (im) mobilize other chemical elements such as HMs, or as fertilizers and pesticides in agricultural soil (Zakari et al., 2020). Sulfur amendments are applicable and cost-effective soil amendments, which can promote and alternatively mitigate ecosystems functions. For instance, sulfur may raise the soil acidity, which activates soil calcium that in turn enhances soil pH for better crops nutrient absorption. Plants roots can also absorb anionic sulfate from soil and transport it to the shoot by the transpiration stream (Mugford et al., 2011). Sulfate then gets reduced and incorporated into organic metabolites in plant tissues for the synthesis glutathione, methionine and other metabolites (Leustek and Saito, 1999; Takahashi et al., 2011). However, a limited sulfur availability reduces nutrients (especially iron (Fe)) uptake and deficiency, leading to the modulation of sulfate uptake and assimilation (Zuchi et al., 2015). Indeed, when sulfur is applied as soil nutrient or as a pesticide (Jayasinghe et al., 1997; Zhang et al., 2008), it may affect the (im) mobilization and bioavailability of pollutants in the amended soils (Rochayati et al., 2011; Shaheen and Rinklebe, 2015; Shaheen et al., 2015), and may cause toxic effects to soils, plants or animals (Driscoll et al., 2003; Joniec et al., 2019; Ma et al., 2018; Weeks et al., 2002).

An excessive elemental sulfur amendment in the soil can lead to the destruction of the sorption complex, changes in soil micronutrients availability (Oku et al., 2012) and biological balance, and a drop in sulfur bioavailability (Hu et al., 2005; Joniec et al., 2019). Moreover, the repeated application of elemental sulfur in agricultural soil affects plants biodiversity (Bowman et al., 2008) and soil microbial activity (Wang et al., 2019; Zakari et al., 2020). In fact, elemental sulfur is thought to be important in the severity of soil pollution, and the concentration of sulfate, which is the second byproduct of elemental sulfur oxidation (Tourna et al., 2014; Zhao et al., 2015), is highly correlated with soil acidity (Kayser et al., 2000; Ngatunga et al., 2003). The soil acidification might enhance the solubility of HMs and contribute to metal mobility (Bashir et al., 2020; Kabil et al., 2014; Li et al., 2019b; Shaheen et al.,

Table 1
Summary of collected heavy metal types from searched articles, and characteristics of their contents, extraction plants and plants organs; and sulfur compounds used as amendments.

Heavy metal	Sulfur compound	Plant heavy metal content [a] (mg/kg)		Plants [b]	Plant organ [c]	Article
		Treatment	Control			
As	Elemental sulfur, Sulfate sulfur	0.55–6393.6	0.80–4250.4	*Brassica* sp., *Pteris* sp., *Oryza* sp.	Roots, Shoots and Leaves.	(Grifoni et al., 2015), (de Oliveira et al., 2014) and (Hu et al., 2007).
Cd	Elemental sulfur, Sulfur trioxide, Sulfate sulfur, gypsum	0.03–629.3	0.09–1135.3	*Sorghum* sp., *Brassica* sp., *Vicia* sp., *Zea* sp., *Nicotiana* sp., *Salix* sp., *Pisum* sp., *Oryza* sp., *Triticum* sp.	Shoots, Roots, Leaves, Stalks, Grain, Husk and Straws.	(Shaheen et al., 2017b), (Lu et al., 2019), (Wu et al., 2018), (Cui et al., 2004), (Dede et al., 2012), (Faessler et al., 2011), (Guo et al., 2019), (Iqbal et al., 2012), (Kroulikova et al., 2019), (Li et al., 2019a), (Mahar et al., 2016), (Shaheen et al., 2019b), (Fan et al., 2010), (Cui and Wang, 2006), (Qayyum et al., 2017), (Adhikari et al., 2018), (Liang et al., 2016), (Lou et al., 2017) and (Masood et al., 2012).
Cr	Elemental sulfur, Gypsum	0.03–179.0	0.025–144.0	*Sorghum* sp., *Brassica* sp., *Pisum* sp., *Zea* sp., *Sedum* sp., *Oryza* sp.	Shoots, Roots and Grain.	(Shaheen et al., 2017b), (Dede et al., 2012), (Wang et al., 2019), (Wu et al., 2019), (Cimrin et al., 2007) and (Shaheen et al., 2019b).
Cu	Elemental sulfur, Sulfate sulfur, Gypsum	1.08–677.4	2.22–317.5	*Sorghum* sp., *Oryza* sp., *Ricinus* sp., *Brassica* sp., *Pisum* sp., *Zea* sp.	Shoots, Roots, Stalks, Leaves, Husk and Grain.	(Shaheen et al., 2017b), (Sun et al., 2017), (Ren et al., 2017), (Dede et al., 2012), (Mahar et al., 2016), (Sun et al., 2016) and (Shaheen et al., 2019b).
Fe	Elemental sulfur	140.0–18292.0	329.7–4406.0	*Sorghum* sp., *Zea* sp.	Shoots and Roots.	(Shaheen et al., 2017b) and (Kroulikova et al., 2019).
Hg	Elemental sulfur, Sulfate sulfur	0.06–857.4	0.05–937.1	*Oryza* sp.	Shoots, Straws and Grain.	(Li et al., 2017) and (Li et al., 2019b).
Mn	Elemental sulfur	24.0–3201.0	17.8–821.0	*Sorghum* sp., *Nicotiana* sp., *Zea* sp.	Shoots, Roots and Leaves.	(Shaheen et al., 2017b), (Faessler et al., 2011) and (Kroulikova et al., 2019).
Ni	Elemental sulfur, Gypsum	2.41–31.3	0.35–12.0	*Sorghum* sp., *Brassica* sp., *Pisum* sp., *Zea*	Shoots and Roots.	(Shaheen et al., 2017b), (Dede et al., 2012) and (Shaheen et al., 2019b).
Pb	Elemental sulfur, Sulfate sulfur, Gypsum	1.69–2598.0	1.12–679.0	*Brassica* sp., *Pisum* sp., *Zea* sp.	Shoots and Roots.	(Cui et al., 2004), (Dede et al., 2012), (Kroulikova et al., 2019), (Mahar et al., 2016) and (Shaheen et al., 2019b).
Zn	Elemental sulfur, Gypsum	26.09–5407.0	23.5–6422.0	*Sorghum* sp., *Zea* sp., *Brassica* sp., *Nicotiana* sp., *Salix* sp., *Pisum* sp.	Shoots, Roots, Leaves and Stalks.	(Shaheen et al., 2017b), (Cui et al., 2004), (Dede et al., 2012), (Faessler et al., 2011), (Guo et al., 2019), (Iqbal et al., 2012), (Kroulikova et al., 2019) and (Shaheen et al., 2019b).

[a] Ranges of plants heavy metal uptakes in sulfur applied and non-sulfur applied (control) treatments.
[b] Plants genera used during the metal extraction experiments.
[c] Plants organs from which metal contents are obtained.

2019a), and alter soil through depletion of labile pools of nutrients cations (Ca and magnesium (Mg)), accumulation of sulfur and nitrogen, and the mobilization of elevated concentrations of inorganic monomeric aluminum to soil solutions in acid-sensitive areas (Driscoll et al., 2003). HMs (especially, Fe, cadmium (Cd) and nickel (Ni)) and sulfur speciation, therefore, change depending on soil redox conditions (Lin et al., 2010; Shaheen et al., 2017a). Acid soils have become a global problem (Boman et al., 2010; Dent and Pons, 1995); because both the acidity and heavy metals can be readily leached *via* runoff or drainage into adjacent surface waters, and metals in the acidic conditions become more soluble and available for plants absorption, affecting their phytoextraction (Amaral et al., 2012; Cook et al., 2000; Österholm and Åström, 2002; Sammut et al., 1996; Simpson et al., 2010).

In the present study, we used a systematic meta-analysis to summarize available knowledge on the contribution of sulfur compounds on plants heavy metals uptake from soils media. Meta-analysis represents 'a technique that statistically combines the results of quantitative studies to provide a more precise effect of the results' (Grant and Booth, 2009). This approach allows generalization of findings when these are consistent across local studies and thus, meta-analysis helps to advance scientific knowledge in a given field. We specifically focus on factors that have often been considered key determinants of heavy metal uptake by plants. These factors include sulfur type and its stress intensity, type of heavy metal, plants species and accumulating organs, and soil pH. A systematic and global quantification of the combined effects of sulfur amendment and these variables remains poorly examined. Thus, we sought to fill this gap by answering the following research questions: (1) How does sulfur amendment affect heavy metals uptake by plants from soils media? (2) Do sulfur types, sulfur level, soil pH differently influence heavy metals phytoextraction? (3) Are heavy metals content in plants organs different and safe for human and animals? We also discussed the relationships between heavy metals and plants (genera and organs), and the various mechanisms of heavy metal uptake by plants.

2. Methods

2.1. Data collection for meta-analysis

The articles of the present meta-analysis were selected through a literature search and retrieval, and a screening of identified studies to select those meeting selection criteria (Fig. 1). The articles were searched using the keywords "sulfur AND heavy metal AND soil", "sulfur AND heavy metal AND phytoextraction", and "sulfur AND heavy metal AND mobilization". Only articles published between 2000 and 2020 were retained for further steps. A total of 246 articles were identified from the Scopus and Web of Science databases. The tittles and abstract of these articles were first scrutinized, and we ended with only 37 scientific articles reporting data on heavy metals in agricultural soils. However, we also examined the references of the 37 selected articles to find more articles meeting our criteria, due to the little number of identified studies from the databases search. Finally, a total of 30 articles met our selection criteria and from those articles we compiled a total of 524 data points which were used for this meta-analysis (Fig. 1). The heavy metals with less than 5 data points were not considered for this meta-analysis.

2.2. Statistical analysis

2.2.1. Effect sizes and variances

Heavy metals contents in plants were used as response variables in the current meta-analysis. The effect size was estimated as log response ratio (hereafter, *RR*), which is the log ratio of the heavy metal concentration in experimental treatment concentration (X_t) over the heavy metal concentration in control (X_c) (Eq. (1)). The response ratio has the advantage to be applied for scale types variables (Borenstein et al., 2009), which is the case for the concentration of heavy metal expressed in mg kg^{-1}. Moreover, response ratio can help to estimate the proportional changes that can result from X_t using a random model.

$$RR = \log\left(\frac{X_t}{X_c}\right) \quad \text{(Eq. 1)}$$

where X_t is the mean value of plants heavy metal concentration in sulfur applied treatment and X_c is the mean value of plants heavy metal concentration in the non-sulfur (control) treatment.

A null effect size ($RR = 0$) suggests a similarity in the average outcome of plants heavy metals concentration from the sulfur applied and control treatments. A negative effect size ($RR < 0$), however, indicates a decline or reduction of plants heavy metals concentration in sulfur applied treatments due to sulfur application whereas positive ($RR > 0$) effect size indicates an increase of plant heavy metals concentration

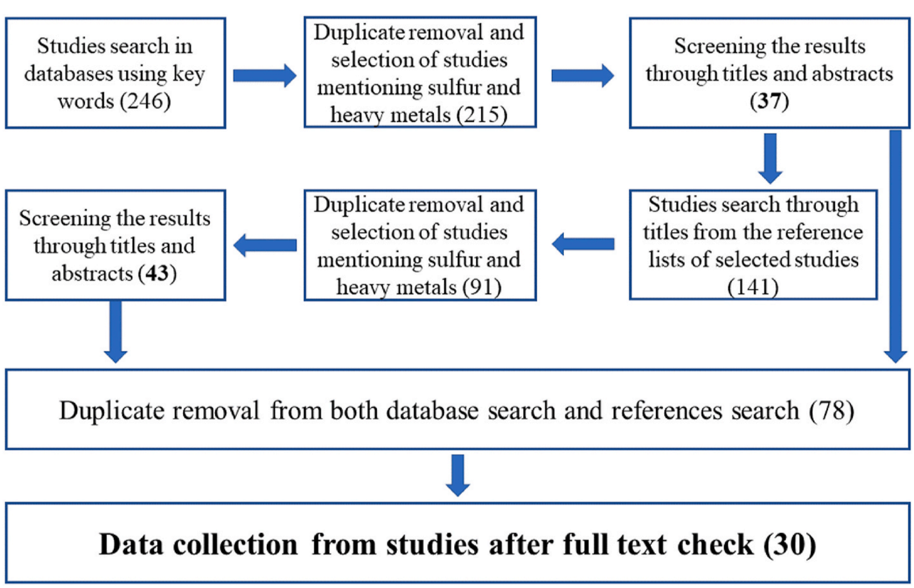

Fig. 1. Flowchart of literature search, screening, selection and internalization for data analysis. Two steps were used for the articles search: first from database resulting in 37 articles after abstract screening, and second from the 37 retained articles references which resulted in 43 new articles after abstract screening. A total of 78 papers were checked for full text and only 30 had data filling the requirements for the meta-analysis.

in sulfur applied treatments due to sulfur application. Finally, the sampling variances ($\sigma^2(RR)$) were estimated using the following formula as described in Benítez-López et al. (2017) (Eq. (2)).

$$\sigma^2(RR) = \frac{SD_t^2}{N_t\overline{X}_t^2} + \frac{SD_c^2}{N_c\overline{X}_c^2} \quad \text{(Eq. 2)}$$

where SD_t and SD_c represent the standard deviations of X_t and X_c, respectively; N_t and N_c are the size of experimental and control treatments replicates, respectively; and $\overline{X}_t$ and $\overline{X}_c$ are means of X_t and X_c, respectively.

2.2.2. Meta-analysis

The statistical analysis of this meta-analysis was computed using "metafor" package (Viechtbauer, 2010) in R Version 4.0.3 (R Foundation for Statistical Computing 2020). We used forest plots to synthetize the pooled studies outcomes (Borenstein et al., 2009), and forest plots sub-meta-analysis was performed for each factor level (sulfur type, sulfur level, plant organ etc.). In all cases, we presented each effect size with a diamond, which bounds is the confidence interval. The size of the diamond is proportional to the weight assigned to the factor level, and the width of the diamond reflects the precision of each estimate. The vertical line (at zero) is the line of no-effect, i.e. the effect size of sulfur applied treatments equals that of the non-sulfur treatments.

This meta-analysis used random-effects model to merge the studies effect sizes, to compute the pooled effect sizes, and to assess the variation of heavy metal uptake, under different metal types, plants organs, plants genera, soil pH etc. The random-effects model was selected because the variance between-studies was statistically significant. We quantified the heterogeneity of true effect sizes between studies using the *p-value* associated to the heterogeneity test (Q), and I^2 values. Assuming that all studies or group of studies share a common effect size, *p-value* less than α ($=0.05$) indicates heterogeneity effect sizes among studies or group of studies. I^2 value represents the proportion of the true variance, and was used to appreciate the homogeneity test that depends neither on the number of studies, nor on the scale of the effect size (Borenstein et al., 2009). I^2 value approximating zero indicates no heterogeneity between effect sizes.

2.2.3. Network analysis

Network analysis was performed to determine the co-occurrence network and the interaction relationships between heavy metals, plants genera and plants organs. The metals and plants organs were first concatenated, and a summary effect size was estimated for each concatenated moderator using random-effect model. The calculated summary effect sizes were used to conduct and visualize the network analysis using Gephi ™ (Version 0.9.2, 2017).

3. Results

3.1. Plants heavy metals phytoextraction under sulfur application

A total of 524 data points were collected from the 30 retained articles. The Q-statistic showed a substantial heterogeneity between data points of the collected articles ($Q > 0$, $p < 0.001$). However, the I^2-statistic exhibited a low heterogeneity of 23.46%, suggesting a fixed-effects model would be suitable to conclude on the summary effect size. The estimated summary response ratio was 0.036 ($p < 0.0001$). As a result, the uptake of heavy metals by plants was slightly higher under the sulfur applied treatments compared to the non-sulfur treatments (control). However, we used a random-effects model to evaluate the influence of various moderators on the response ratios because of differences in the methods used in different articles.

3.2. Effect of sulfur type, sulfur level and soil pH on heavy metals uptake

Elemental sulfur, sulfate sulfur, and gypsum were applied at rates ranging from 0.05 to 59.28 g kg^{-1}, 0.01–208.51 g kg^{-1}, and 2.00–8.00 g kg^{-1}, respectively. The application of elemental sulfur ($RR = 0.015$, $p < 0.001$) and sulfate sulfur ($RR = 0.089$, $p = 0.298$) resulted in slightly higher plants heavy metal uptake in the sulfur applied than the non-sulfur treatments, while mineral gypsum ($RR = -0.025$, $p = 0.881$) application gave an opposite trend (Fig. 2). However, the heavy metals extraction was not significantly different between sulfur applied and non-sulfur treatments for both sulfate sulfur and gypsum, while significant difference occurred between the two treatments ($p < 0.001$) for each level of sulfur stress and each pH ranges (Fig. 2). The response ratios increased with the increasing sulfur stress, with values of 0.037, 0.913, and 1.823 for low (0–10 g kg^{-1}), medium (10–50 g kg^{-1}) and high (>50 g kg^{-1}) sulfur levels, respectively; though no linear or nonlinear relationship was found between these two variables. Finally, significant differences occurred between plant heavy metals uptakes under sulfur applied and non-sulfur treatments at each pH range (Fig. 2), with higher heavy metals uptake in the sulfur applied-than the non-sulfur treatments under ideal (pH ranging from 5.5 to 8, $RR = 0.358$, $p < 0.001$) and alkaline (pH higher than 8, $RR = 0.281$, $p < 0.001$) conditions; and lower heavy metals uptake in sulfur applied than non-sulfur treatments under acidic pH (pH lower than 5.5, $RR = -0.623$, $p < 0.001$).

3.3. Effect of heavy metal, plant genera, and plant organ on heavy metals uptake

Eleven heavy metals were collected from the databases, however, only ten heavy metals (number of data point) including cadmium (132 data points), copper (77), zinc (59), chromium (57), lead (54), mercury (51), manganese (29), iron (25) arsenic (22), and nickel (17) were used to estimate the summary effect-sizes and to construct the forest plots (Fig. 3). The phytoextraction of nickel ($RR = 1.01$, $p = 0.001$), chromium ($RR = 0.52$, $p = 0.001$) and cadmium ($RR = 0.21$, $p = 0.027$) from soil were significantly higher in the sulfur applied treatment compared to the non-sulfur treatments ($RR = 0.28$, $p = 0.002$), and an opposite significant result occurred for copper ($RR = -0.15$, $p = 0.035$). The remaining heavy metals including arsenic, lead, zinc, mercury, manganese and iron exhibited non-significant plants uptakes between the sulfur applied and the non-sulfur treatments (Fig. 3A).

Overall, the heavy metal uptake by each plant genera was significantly different between sulfur applied and non-sulfur treatments (Fig. 3B). Two groups of plants genera arose according to the observed response ratios: (*i*) the groups of *Brasica* sp., *Nicotiana* sp., *Pisum* sp., *Pteris* sp., *Salix* sp., *Sedum* sp., *Sorghum* sp., and *Vicia* sp. where the heavy metal uptake was higher in the sulfur applied than the non-sulfur treatments; and (*ii*) the groups of *Oryza* sp., *Ricinus* sp., and *Tricicum* sp. where an opposite result occurred. However, *Pteris* sp. ($p = 0.078$) and *Zea* sp. ($p = 0.117$) showed moderate difference between sulfur applied and non-sulfur treatments, whereas the remaining genera exhibited significant differences (Fig. 3B). The heavy metal mainly accumulated in individual plants organs (roots, stalks, leaves, husk and grain), and combined organs (straws and shoots). Only roots ($RR = 0.25$, $p < 0.001$) and leaves ($RR = 0.21$, $p = 0.076$) showed significant higher heavy metal uptake in sulfur applied than non-sulfur treatments, while grain ($RR = -0.18$, $p = 0.022$), husk ($RR = -0.69$, $p = 0.004$) and stalk ($RR = -0.21$, $p = 0.128$) presented lower uptake (Fig. 3C, considering 0.07 as marginally significant). The metal extraction in combined organs was not significantly different between sulfur treatments. The response ratios were in the following order roots > leaves > stalk > grain > husk.

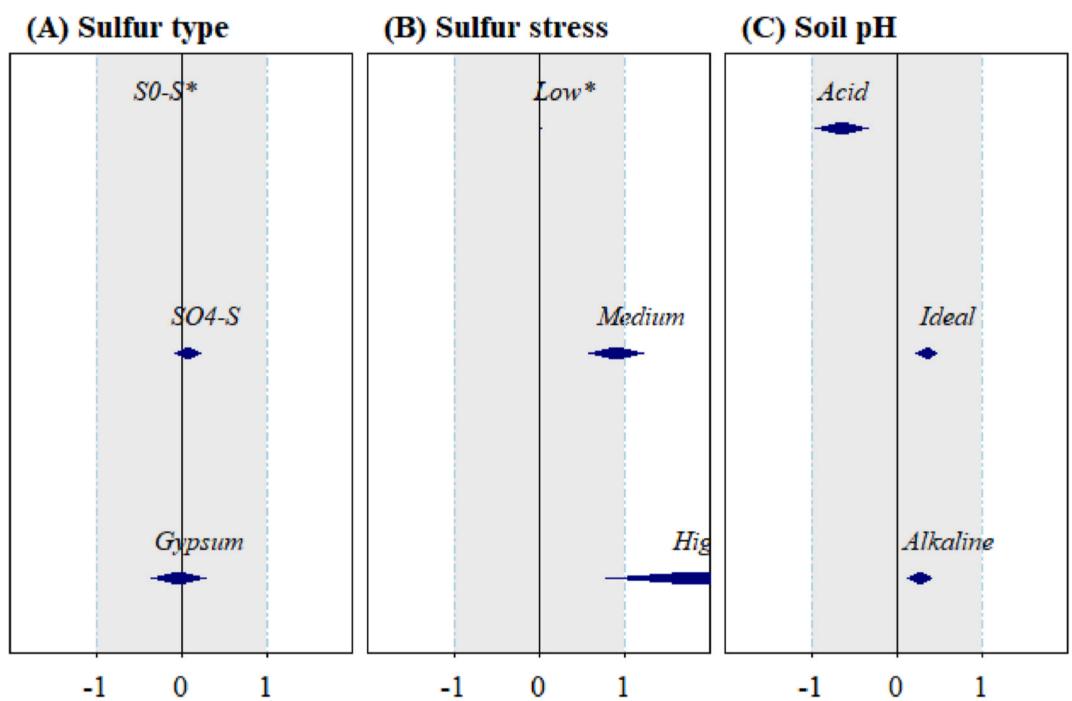

Fig. 2. Responses ratios, log(RR), of heavy metals uptakes under sulfur application as functions of sulfur types (a), sulfur stress (b), and soil pH (c). The diamond representing the effect-size of S0-S* and low* sulfur level did not appear in the graph because their response ratio and standard error are very close to zero. S0-S and SO4-S represent elemental sulfur and sulfate sulfur, respectively.

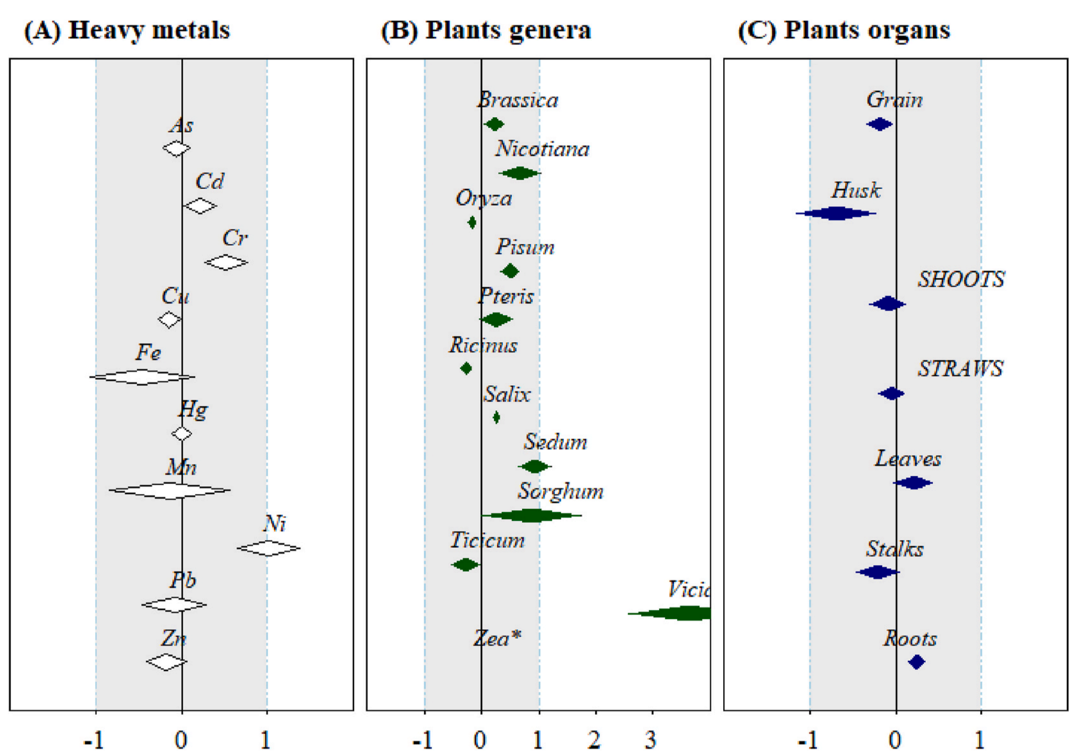

Fig. 3. Responses ratios, log(RR), of heavy metals uptakes under sulfur application as affected by various heavy metals (A), plants genera (B), and plants organs (C). The diamond representing the effect-size of *Zea** did not appear in the graph because of its response ratio and standard error, which are very close to zero. Combined organs are in uppercase and individual organs are in lowercase. STRAWS = Stalks + Leaves, and SHOOTS = Straws + Husks + Grains.

3.4. Network analysis of plants organs and plants genera on heavy metals uptake

Fifteen plants species corresponding to twelve plants genera and seven plants organs or combination of plants organs were found from the selected articles for the heavy metals extraction experiments. Twelve plant genera, seven plant organs and ten heavy metals fitted the selection criteria for random effect model (size > 5), and were therefore used to compute the network analysis. We used co-occurrence network analysis to determine and evaluate the relationships between heavy metals, plants genera and organs (genera-HMs, organ-HMs, and genera-organ); and to identify the occurring communities from each network analysis using the modularity function of Gephi (Fig. 4). The edges showing positive interactions (response ratio) genera-metal, organ-metal, and genera-organs occurred in 72.1%, 57.1% and 50% of cases, respectively, and those showing negative interactions occurred in 27.9%, 42.9%, and 50% of cases, respectively. Moreover, three communities (edges with same color) were found for each of the genera-metal and organ-metal networks, while four communities were identified for the genera-organ network (Fig. 4).

The results show that manganese, cadmium, chromium, and nickel were the most extracted under sulfur application by plant genera including *Vicia* sp., *Sorghum* sp., *and Brassica* sp. (heavier red edges) (Fig. 4a). Considering the negative response ratios, chromium, manganese and iron were more extracted in non-sulfur than sulfur applied treatment by plant genera such as *Oryza* sp., *Zea* sp., and *Sorghum* sp. (heavier blue edges). Besides, manganese, nickel, chromium, and iron mainly accumulated in leaves, roots and shoots under sulfur amendments, while manganese, iron, copper, and cadmium accumulated in shoots, husk, and grain under non-sulfur treatment compared to sulfur treatment (Fig. 4b). Moreover, *Vicia* sp. extracted heavy metals under sulfur application by magnitude beyond two in its roots and shoots; and *Zea* sp. and *Oryza* sp. inhibited heavy metals uptake under sulfur application in shoots and husk, respectively (Fig. 4C).

Overall, the communities for the genera-metal network consisted of (1) *Brassica* sp., *Oryza* sp., *Pisum* sp., *Sedum* sp., *Pteris* sp. which mainly extracted chromium, arsenic, lead, mercury; (2) *Sorghum* sp., *Zea* sp., *Ricinus* sp., and *Nicotiana* sp. which assimilated copper, zinc, nickel, iron, and manganese; and (3) *Triticum* sp., *Salix* sp., and *Vicia* sp. linked with cadmium (Fig. 4a). The communities for organ-metal network were composed of (i) cadmium, copper, and mercury which were extracted in grain, husk, straws, and stalks; (ii) magnesium and arsenic uptake in leaves; and (iii) lead, zinc, iron, nickel, and chromium which were uptake in roots and shoots (Fig. 4b). Finally, *Vicia* sp. and *Ricinus* sp. principally accumulated heavy metals in roots; *Pisum* sp., *Zea* sp., *Sorghum* sp., *Sedum* sp., and *Brassica* sp. in shoots; *Tricicum* sp. in grain and straws; and *Oryza* sp., *Nicotiana* sp., *Pteris* sp. and *Salix* sp. in stalks, husk and leaves (Fig. 4c).

3.5. Effect of soil pH on heavy metal uptake

The influence of sulfur application on heavy metals phytoextraction was evaluated under various soil pH. The soil pH was ranged in three groups: 1 to 5.5, 5.5 to 8, and 8 to 14 for acidic, ideal, and alkaline soil conditions, respectively. The results show that each heavy metal had a specific behavior regarding its phytoextraction under various pH (Fig. 5). Cadmium ($RR = -0.725$, $p < 0.001$) and zinc ($RR = -0.762$, $p < 0.001$) extractions by plants were lower in the sulfur applied compared to the non-sulfur applied treatments under acidic pH; but an opposite trend occurred for cadmium ($RR = 0.772$, $p < 0.001$) while no difference was observed for zinc ($RR = 0.093$, $p = 0.512$) extraction between the sulfur and non-sulfur treatments under ideal pH. Chromium, however, showed a similar trend under acidic ($RR = 0.646$, $p = 0.003$), ideal ($RR = 0.494$, $p = 0.003$) and alkaline ($RR = 0.587$, $p < 0.001$) conditions, with higher extraction in the sulfur applied treatment than the non-sulfur treatments. Finally, nickel ($RR = 1.009$, $p < 0.001$) and lead ($RR = 0.234$, $p = 0.056$) were more extracted in the sulfur applied treatment than the non-sulfur treatments under ideal condition.

4. Discussion

At global scale, awareness about environmental pollution caused by heavy metals is on rise. Heavy metals emanate from diverse sources including large metallurgy factories and diverse anthropogenic

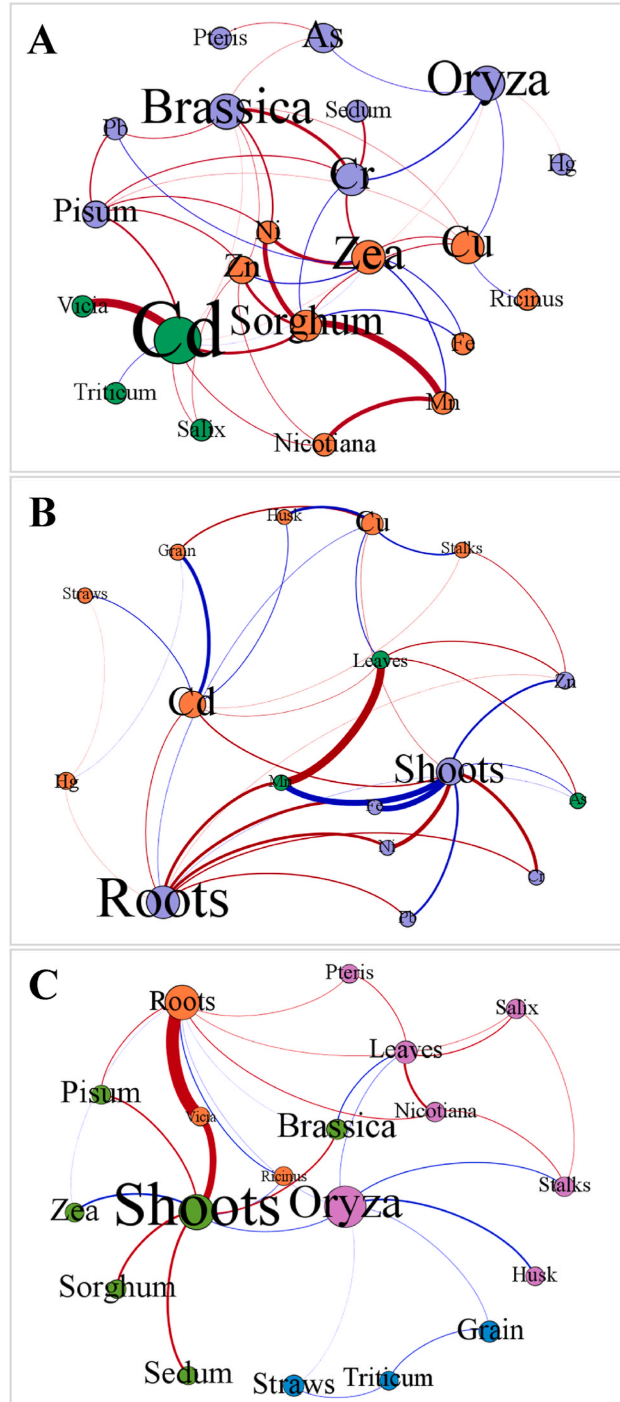

Fig. 4. Network analysis of the contribution of plants genera and plants organs on heavy metal (HMs) extraction (a and b), and the organs contribution in heavy metals uptakes by various plants genera (c). Nodes colors are communities obtained from the Gephi modular classes function; each community in the network is represented by the same node color. Wider nodes are the HMs or plant genera and organs that contribute the most in a community. Blue edge lines represent negative response ratios, i.e. HMs accumulation is lower in sulfur applied treatments than the non-sulfur applied treatments or sulfur addition inhibits HMs uptake in the treatment. Red edge lines are positive response ratios, i.e. HMs accumulation is higher in sulfur applied treatments than the no-sulfur applied treatments sulfur addition enhances HMs extraction in the treatment. The edges thickness represents the strength of the HMs extraction. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

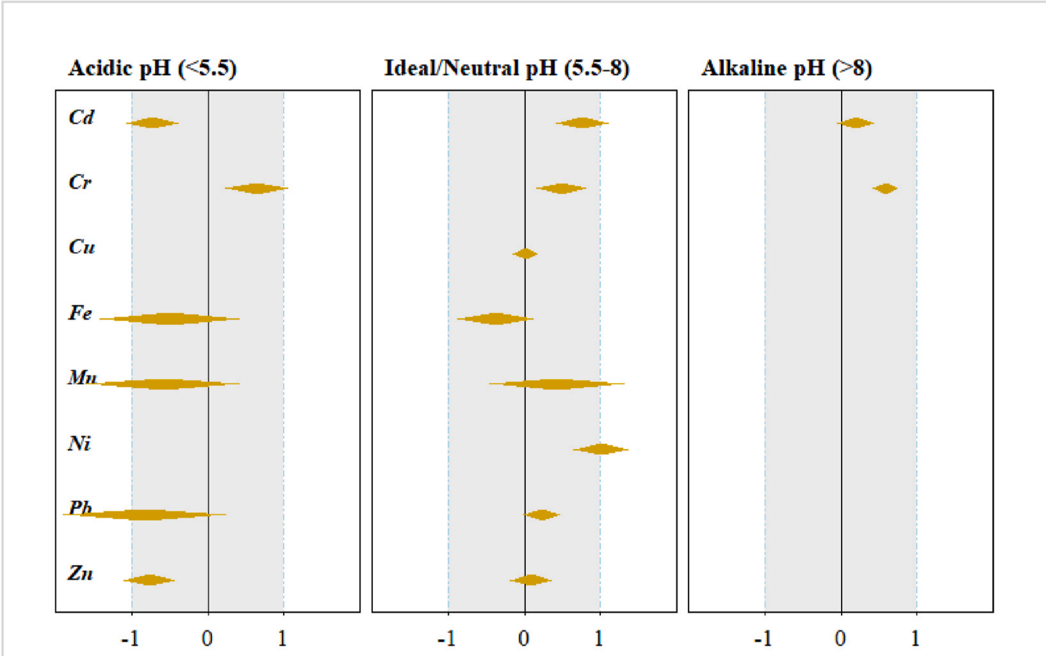

Fig. 5. Responses ratios, log(RR), of plants heavy metals uptakes under sulfur application as function of metal types and soil pH.

activities including agriculture practices. Both private and public sectors are merging efforts to tackle this issue as such pollution not only causes harm to the environment, but also to both plants and animals as well as human beings. One approach to remediate heavy metals from edaphic media is the use of sulfur amendments. Up to date, many local studies on diverse plants have examined the efficiency of plants and diverse of their organ in relation to heavy metals uptake. Here, we set out to evaluate how different plants uptake heavy metals and how these metals are accumulated in different plant organ under sulfur amendment as well as different edaphic pH levels through a meta-analysis. In general, several factors affect heavy metals phytoextraction, and the uptake rate of heavy metal is organ specific with different heavy metal behaving differently according to soil pH levels.

4.1. Factors affecting heavy metals phytoextraction

Phytoextraction of heavy metals varied according to factors such as sulfur level, sulfur type, and soil pH (Fig. 2) (Cui and Wang, 2006), and these factors differently affected plants heavy metal uptake between the sulfur applied and non-sulfur treatments, and between plants organs. Higher sulfur stress mainly leads to a higher heavy metal phytoextraction from soil (Guo et al., 2019; Iqbal et al., 2012; Kroulikova et al., 2019; Liang et al., 2016; Mahar et al., 2016; Qayyum et al., 2017; Wu et al., 2019); especially our results exhibited higher metal extraction in the sulfur applied than the non-sulfur treatments for elemental sulfur amendments, while no significant differences between the two treatments for sulfate sulfur and gypsum amendments (Fig. 2A). In fact, while sulfate sulfur and gypsum can be directly accessible to plants, elemental sulfur requires transformations such as microbial oxidation which can lead to both soil acidity and heavy metals solubility. For instance, when elemental sulfur is applied to the soil, chemolithotrophic bacteria (*Thiobacillus thioxidans*) of the family Thiobacteriaceae can oxidize elemental sulfur and thiosulphate to sulfuric acid under aerobic conditions (Van Ranst, 2006). The produced sulfuric acid during microbial sulfur oxidation decreases the rhizosphere soil pH (Cimrin et al., 2007; Cui et al., 2004; Dede et al., 2012; Li et al., 2019a; Mahar et al., 2016; Zhou et al., 2018), leading to the increase of metals solubility and bioavailability for plants uptake (Cimrin et al., 2007; Grifoni et al., 2015; Iqbal et al., 2012; Li et al., 2019b; Qayyum et al., 2017; Shaheen et al., 2019b; Wu et al., 2018; Zhou et al., 2018). Elemental sulfur addition can also reactivate the non-bioavailable metals to accelerate the phytoremediation processes (Li et al., 2019b). Indeed, the mobility and bioavailability of heavy metals is related to the biogeochemical cycle of sulfur in the soil (Sun et al., 2016), which highly affects the heavy metals phytoextraction from agricultural soil.

In the present meta-analysis, heavy metals uptake was 2.3-fold higher in the sulfur applied-than the non-sulfur treatments under ideal and alkaline conditions; and 0.2-fold lower in sulfur applied than non-sulfur treatments under acidic pH (Fig. 2C). Cadmium and zinc were less extracted in the sulfur applied treatments under acidic condition, chromium was more extracted under all pH conditions, and chromium and nickel were also more extracted from soil only under ideal pH condition (Fig. 5). The heavy metals extraction by plants under the ideal pH followed the order nickel > cadmium > chromium, suggesting nickel was more susceptible for plant extraction under sulfur application under ideal pH condition. Since most of the crops usually require ideal pH conditions for better growth, it is therefore worth knowing farming on agricultural fields may lead to bioaccumulation of nickel, cadmium and chromium in plants grown on contaminated soils. These heavy metals uptake by plants seem ineluctable in agricultural soils amended with elemental sulfur. Thus, farmers should be aware to grow plants under sulfur amendments to remediate the level of toxicity, but should test whether the produce from those soils are below the threshold level of heavy metal that safe for human consumption before selling such produce.

4.2. Metals phytoextraction

The phytoextraction of cadmium, chromium and nickel were significantly higher in the sulfur applied compared to the non-sulfur treatments, and an opposite result occurred for copper (Fig. 3A). The order of the extraction intensity (response ratio) was nickel > chromium > cadmium. Likewise, sulfur amendment enhances cadmium uptake in straws and grain of wheat (Qayyum et al., 2017), in shoots and roots of *Brassica* sp. (Liang et al., 2016), corn (Cimrin et al., 2007; Cui et al., 2004; Shaheen et al., 2019b; Wu et al., 2019), pea (Shaheen et al., 2019b), Cabbage (Mahar et al., 2016), mustard (Guo et al., 2019), tobacco (Faessler et al., 2011), in shoots of corn (Cui and Wang, 2006; Li et al., 2019a), and in rice grain (Fan et al., 2010). The high cadmium uptake has been linked to processes including iron plaque formation and plant enzyme biochemical activities. For instance, sulfur decreases the iron plaque formation on the roots surface of rice which raises cadmium accumulation in roots (Fan et al., 2010). More, sulfur supply can decrease maize malondialdehyde content, peroxidases and catalase activities compared to control treatments (Cui and Wang, 2006). Peroxidases are involved in several physiological and biochemical processes such as cell growth and expansion, differentiation and development, auxin catabolism, lignification, as well as abiotic and biotic stress responses; and catalase is an important enzymes involved in the removal of toxic peroxides (Cui and Wang, 2006). Sulfur application to cadmium-stressed plants alleviates cadmium-induced oxidative stress, and decreases cadmium translocation from Brassica roots to the shoots by enhancing phytochelatins biosynthesis (Liang et al., 2016). However, for Faessler et al. (2011) the high cadmium uptake in root and shoot are associated with up-regulation of the putative cadmium transporters and the genes involved in sulfur assimilation in root tissues (Faessler et al., 2011).

The results of the present meta-analysis showed that the application of sulfur in the soil overall increased cadmium extraction by 162% (RR = 0.21) in the sulfur applied treatments compared to the non-sulfur treatments. Moreover, chromium phytoextraction was 3.3-fold (RR = 0.52) higher in the sulfur applied compared to the non-sulfur treatments, and nickel phytoextraction was 12.6-fold higher (Fig. 3A). These results are supported by Shaheen et al. (2017a) and Li et al. (2011) who initially reported that sulfate sulfur addition in soil increases cadmium bioavailability in soil and raises its uptake in soybean and sorghum aboveground tissues. In contrast, an accumulation of cadmium in soil has been observed, followed by its low uptake in rice grain (Fan et al., 2010), maize (Adhikari et al., 2018), Brassica (Lou et al., 2017), and wheat (Qayyum et al., 2017) under sulfur application; while other research found no change in cadmium extraction between sulfur applied and non-sulfur applied treatments (Wu et al., 2019). In fact, sulfur compounds, especially sulfate sulfur, can react with cadmium and increase the soil $CdSO4^0$ content, which has faster diffusion in plants than Cd^{2+}, leading to higher accumulation of Cd in plants (Wu et al., 2018). Conversely, lower chromium uptake by plants under sulfur amendment can be attributed to the formation of iron plaques in soil (Li et al., 2017; Shaheen et al., 2017b; Zhang et al., 2019). In short, while cadmium phytoextraction is differently affected by sulfur types, sulfur amendments in overall lead to high absorption of cadmium in plants organs.

On the other hand, copper phytoextraction was overall 30% lower in the sulfur applied than the non-sulfur treatments (Fig. 3A). In fact, sulfur amendment decreases metal mobility in the rhizosphere soils (Li et al., 2017; Sun et al., 2016). Sun et al. (2016), Ren et al. (2017) and Zhihong et al. (2019) also concluded that sulfur lowers the bioavailability of copper in the soil, inducing the transformation of copper bioavailable fractions to copper bound to organic matter, or a decrease in the reducible copper fraction in the rhizosphere, and the increase of the oxidizable copper fraction. However, previous results found opposite trends where sulfur amendment increases the bioavailable/exchangeable fraction of copper in the rhizosphere soil, enhancing its phytoextraction in roots and shoots of Chinese cabbage (Mahar et al., 2016), in grain of rice (Sun et al., 2017), and in roots and shoots of *Brassica juncea* (Dede et al., 2012). The heavy metal phytoextraction from soil was influenced by soil condition (pH), metal bioavailability and plants physiology. Further research might target different soil metal as well as soil physico-chemical properties response to sulfur amendments.

4.3. Plants sensitiveness: genera and organs

More than 450–500 plants species have been identified as hyperaccumulators including Thalaspi and Arabidopsis and members from families such as Brassicaceae, Cyperaceae, Poaceae, Fabaceae, and several others. Factors such as metal type and traits of plants species also affected the plants heavy metal uptake under sulfur enriched environment. Plants can effectively extract heavy metals from soil, if the heavy metal is converted into its water soluble form (Sun et al., 2016). Studies on evolutionary trajectories suggested that the tolerance and metal accumulation are genetically different responses of plants against excess metal (Cappa and Pilon-Smits, 2014; Goolsby and Mason, 2015). In the present review, cadmium, manganese, nickel, and chromium were the most extracted under sulfur application by *Vicia* sp.*, Sorghum* sp. *and Brassica* sp., respectively (heavier red edges thickness); while chromium, manganese, and iron were the most uptake without sulfur amendments by *Oryza* sp., *Zea* sp. and *Sorghum* sp., respectively (heavier blue edges) (Fig. 3). Nonetheless, pure heavy metals alone present difficulties for phytoextraction. These heavy metals when combined with sulfur compounds are easily up taken by plants because the combination with other sulfur related compound increases heavy metals diffusion ability and these can easily be bioaccumulated in plants tissues. For instance, arsenic which cannot directly precipitate with sulfur uses iron as a mediator in the phytoextraction process (Amoakwah et al., 2014).

We demonstrated that heavy metals phytoextraction was different among plant organs and followed the order (of response ratio) roots > leaves > stalk > grain > husk. More, these individual organs showed significant differences between sulfur applied and non-sulfur applied treatment, and the combined organs did not. The metal uptake in leaves and roots was higher in the sulfur applied than the non-sulfur treatments, whereas opposite results occurred for grains, husks and stalks (Fig. 3C). The heavy metal uptake in grains, husks and stalks is known to increase with raising sulfur stress (Li et al., 2017; Xu et al., 2015; Xu et al., 2019). Moreover, previous studies showed that soil-available sulfur plays a critical role in inhibiting metal transfer, especially cadmium, in the soil-rice system (Zheng et al., 2019). Sulfur can be transformed into S^{2-} in reductive soil environments, thereby limiting grain heavy metal uptake and ensuring the grain quality (Hassan et al., 2005). As a result, roots from plants (e.g., *Vicia* sp.*, Pisum* sp.*, Pteris* sp. *and Nicotiana* sp.) growing in contaminated soils containing sulfur compounds can significantly accumulate high concentrations of heavy metal than those growing in uncontaminated soils. Moreover, plant parts, especially the husk from *Oryza* sp. growing in contaminated soils without sulfur compounds can significantly accumulate high concentrations of heavy metal than those growing in uncontaminated soils. These main findings should guide decision making during cleaning and reclamation of contaminated soils with phytoremediation.

4.4. Sensitivity analysis and future research needs

Substantial research and debate are still ongoing about the impact of various soil amendments on heavy metal phytoextraction/immobilization, but systematic meta-analysis on the impact of sulfur on plants heavy metal uptake remains rare. Here, the present meta-analysis laid some generalization, and recognized that responses to phytoextraction are heavy metal, plant species and the type of sulfur amendment specific. We showed that the answer to the question whether sulfur amendment could enhance heavy metals phytoextraction is not one answer fits it all. However, our results show that the direction of plants response to sulfur amendment highly depends on various factors such as type of amended sulfur, the heavy metal type and its stress intensity, plants organs and other experimental conditions such as soil pH. On this basis, we argue against quick generalization about the influence of sulfur application on HMs phytoextraction or immobilization in the soil without any baseline checking.

The results from the present meta-analysis remain of high importance as these represent knowledge to guide specific heavy metal phytoextraction in agricultural soil under sulfur amendment. These results are deemed useful to guide in selecting plant that can help speed up remediation as well as which organs food quality control service should target while testing agricultural produce safety for human consumption.

Nonetheless, many questions remain unanswered. Currently available studies on factors influencing heavy metals phytoextraction from sulfur amended agricultural soil are strongly oriented to the soil and plant heavy metals remediation processes. However, several different factors have been considered from the primary researches. We recognize that there might probably be multiple sources of heterogeneity in the data set, such as the experimental sites (e.g., soil type, moisture contents, water source, random sites etc.), amount of variation allowed in sampling sites (e.g., controlled for individual factors), or simple differences in measurement procedure.

To obtain more information on the importance of these factors, data accumulation effort should continue. In addition, in the future when the amount of data allows, other interesting questions such as plant genomic effect of heavy metals extraction, or what is the optimum pH to achieve higher phytoextraction rates could be answered. Moreover, combined sulfur and fertilizer amendment experiments should be explored to highlight their influence, since many agricultural soils are applying mineral or organic fertilizers. Apart from these overarching questions, the importance of plants organs and combined heavy metals needs to be tested more thoroughly in polluted agricultural soils. It would particularly be worth identifying and investigating the biochemical interactions between soil and plants organs, and linking these processes with plant genomes.

5. Conclusion

Generalizing the effect of sulfur on heavy metal phytoextraction seems unresolved due to the diverse range of factors involved in phytoextration processes. Nevertheless, our present study results reach important findings. First, cadmium, manganese and nickel, and chromium were the most extracted heavy metals under sulfur application by *Vicia* sp.*, Sorghum* sp.*, and Brassica* sp., respectively. Second, chromium, manganese, and iron were the most up taken under no sulfur amendments by *Oryza* sp., *Zea* sp. and *Sorghum* sp., respectively. The heavy metal phytoextraction increases with the rising sulfur stress, but sulfur type does not significantly affect the metal extraction between sulfur applied and non-sulfur applied treatments. However, factors such as metal type (Cd, Cr, Cu, and Ni), plant genera (*Brassica* sp., *Nicotiana* sp., *Oryza* sp., *Pisum* sp., *Ricinus* sp., *Salix* sp., *Sedum* sp., *Sorghom* sp., *Ticicum* sp., and *Vicia* sp.), plant individual organs (grain, husk, stalks, leaves and roots), and soil pH significantly influence the heavy metal uptake. *Vicia* sp. occurs as a hyperaccumulator under sulfur amendment. Sulfur application in soil may not affect grain quality and make it safe for human consumption. Nevertheless, people should be aware that some crops leaves highly accumulate more heavy metal beyond threshold levels under sulfur amendment and thus can become threats to human health through the food chain. Agricultural produce food quality control service should design tests that target those accumulating plant organs.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.117820.

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