Steel Mill Wastes Application in Soil: Dynamics of Potentially Toxic Elements in Rice and Health Risk Perspectives

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Research Article

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Abstract

Potentially toxic elements (PTE's) are of great concern in steel mill wastes. Therefore, in other to use them as potential fertilizers in soil, risk assessments are needed. Three steel mill wastes were tested as possible amendments for soils at six different doses (0; 0,5; 1; 2; 4; 8; 16 t ha^{-1}): Phosphate mud (PM), Metallurgical press residue (MPR) and Filter press mud (FPM) during rice cultivation in a pot experiment in a Haplic Gleisol. Analysis on rice tissues, including roots, shoots, husk and grains were conducted and contents of Cu, Cd, Ni, Zn, Mn and Pb were assessed. Translocation and bioaccumulation factors were calculated for each element. In general, PTE's are more accumulated in roots and greater contents of Zn and Mn were found, while the lowest ones were found for Pb, probably due to its lack of functional roles during plants development. Higher translocation was observed for Mn, which is associated to the redox conditions of rice cultivation and the high mobility of this element under this condition. Application of steel mill wastes can increase PTE's bioavailability and translocation factors, especially PM, but all of the wastes reveal a high hazard index.

1. Introduction

According to the World Steel Association (2019), 1,808 million tons of crude steel were produced in the world in 2018, 34.9 of them produced by Brazil and approximately 100% of the wastes produced by them could be reused, for instance, as fertilizers. However, potentially toxic elements (PTE's) can compose these wastes (Wendling et al. 2013; Guerrini et al. 2017), what could impair their utilization as fertilizers in soils (Das et al., 2007). Contamination in agricultural soils by PTE's is a worrisome problem (Cao et al. 2020; Rinklebe et al. 2019) and could affect human health in several ways.

When in soils, PTE's can show different behaviors depending on soil properties and element characteristics (Antoniadis et al. 2008; Shaheen et al. 2017; Zhao et al. 2020). Sorption process in soils are of high relevance because they avoid movement of toxic elements through soil profile and consequent contamination of subsurface water (Gul et al. 2019). Among the PTE's in steel mill wastes, Cd, Ni, Pb, Zn, Mn and Cu stand out due to the many harmful effects they can pose in in living beings, when present in high concentrations (Yu et al. 2017).

The increasing contents of PTE's in soils are responsible for greater antioxidant enzymes production as part of the defense mechanism of plants (Cândido et al. 2020; Guo et al. 2019). Accumulating PTE's in roots and consequently decreasing roots-shoots translocation can increase plants' resistance to harmful elements (Soleimani et al. 2020). The translocation capacity varies with plants species and with the PTE's (Bonanno et al. 2018). Plants with greater translocation factor can jeopardize food security when cultivated in soils contaminated with PTE's (Khan et al. 2017; Antoniadis et al. 2017), especially when translocation is toward the consumable plant' part such as grain or leafy vegetables (Sihlahla et al. 2019).

Some PTE's, such as Pb and Cd, did not show any beneficial role in plants or animals (Zulfiqar et al. 2019) and in order to estimate risks of oral exposure, health hazards are calculated using indexes that consider a reference dose (RfD) as a dose of daily oral exposure for the human population that does not cause deleterious effects throughout life (Peng et al. 2018; Chen et al. 2018; Halder et al. 2019; Shahriar et al. 2020). On the other hand, some PTE's can work as micronutrients for plants, such as Zn and Mn. Micronutrients deficiency is a problem of concern, especially for Zn, Mn and Cu (Rashid 2005; Nadeem and Farooq 2019).

Rice is an important staple food in many countries and better understanding the dynamics of PTE's in paddy soils is of paramount relevance. This study aims to evaluate the potential hazard of adding three different steel mill wastes as fertilizers, taking advantage of their micronutrients contents. Health risks index (HRI) and hazard index (HI) based on reference doses were also used as an auxiliary tool to evaluate food safety.

2 Material And Methods

2.1 Soil characterization

A greenhouse study was performed in the city of Lavras, Minas Gerais, Brazil. The surface layer (0 – 0.2m) of an Haplic Gleisol was collected and used for rice cultivation in pots (7 dm^3). Samples were air dried and physically and chemically characterized according to Teixeira et al. (2017) and PTE's were analyzed following the methodology preconized by U.S. Environmental Protection Agency (USEPA) 3051A (USEPA 2007) (Table 1).

2.2 Steel mill wastes characterization

The steel mill wastes chosen to be used in this study were: Metallurgical press residue (FPM), Filter press mud (FPM) and Phosphate mud (PM), collected in the city of Juiz de Fora, Minas Gerais, Brazil, where a metallurgical industry is installed. The available contents of macro and micronutrients in these wastes were analyzed following Mehlich-1 extraction and they were also analyzed regarding PTE's following USEPA 3051A (USEPA 2007) (Table 2). The elements analyzed were: P, S, Mg, K, Ca, Cr, Mn, Fe, Ni, Cu, Zn, Cd and Pb, all of them determined by spectrophotometry of atomic absorption with air-acetylene flame, except for phosphorus that was determined by colorimetry and K that was analyzed by flame photometry (Malavolta et al. 1989).

2.3 Experiment conduction

The experiment was set in a completely randomized design with a factorial scheme (3×7) in triplicates, consisting of three steel mill wastes combined at seven wastes doses (0; 0.5; 1; 2; 4; 8; 16 t ha^{-1}) which corresponds to the following doses per pot: 0; 1; 2; 4; 8; 16 and 32 g pot^{-1}.

Soil samples and metallurgical wastes were weighed, mixed homogeneously in plastic bags and incubated for 60 days. Then, soil samples (7 dm^3) were fertilized with N, K, P, Mg, S, B, Cu, Zn and Mo (450; 450; 200; 30; 50; 0.5; 1.5; 5 and 0.2 mg kg^{-1} respectively) following recommendations of Malavolta (1981) and using analytical grade reagents. Iron and Mn were not added due to the natural content of them in soil and their low demand by plants. Micronutrients and S were added at once during cultivation while N and K were split out in topdressing applications. The first nutritional supplementation occurred 32 days after installing the crop. From then on, N and K were supplied weekly. Cultivation was performed in pots with 7 dm^3 capacity.

Rice crop (*Oryza sativa* L.) cultivar Curinga was chosen due to the interest in studying PTE's behavior under flooding and because rice is an important staple food all over the world (Laborte et al. 2012). Twelve seeds were cultivated per pot but only five seedlings were carried on until the end of five months. Most of rice crops are conducted in floodplain soils, and in order to approach such condition, a 5 cm water blade was maintained until the end of the experiment.

2.4 PTE's in rice parts

Plant material was cleansed with distilled water in order to remove any soil particle and avoiding contamination, especially roots. After drying the plant material in an oven with forced air circulation at 65 ºC up to constant mass, the shoots, husk, grain and roots dry matter of rice was determined. Then, the material was ground in a Wiley mill and digested with nitroperchloric method in a digester block. About 1 g of plant material was added to each tube in the digester, together with 6 ml of $HClO_4$ + HNO_3 solution in a 1:2 ratio. Samples were allowed to pre-digestion for approximately 4 hours and then, they were gradually heated until temperature reached 190ºC. When samples became colorless and a 2 mL aliquot was left, digestion was completed. After cooling down, the extracts were diluted with distilled water to 16 mL and filtered through Whatman 40 filter papers (Scott 1978). PTE's concentration (Mn, Ni, Cu, Zn, Cd and Pb) was then determined in an atomic absorption spectrophotometer (AAS).

2.5 Bioaccumulation Factor (BAF) and Translocation Factor (TF)

PTE's concentrations (Ni, Cd, Pb, Cu, Zn and Mn) in the plant and in the soil were calculated based on dry weight. The bioaccumulation factor (BAF) is an index that shows the plant's ability to accumulate an element according to the concentration of that element in the soil. Thus, BAF was calculated according to the following equations (Galal and Shehatab 2015; Usman et al. 2013; Usman and Mohamed 2009):

BAF (grain) = Mgrain / Msoil (1)

BAF (husk) = Mhusk / Msoil (2)

BAF (shoot) = Mshoot / Msoil (3)

BAF (roots) = Mroot / MCsoil (4)

Where Mgrain, Mhusk, Mshoot and Mroot are the concentrations of Ni, Cd, Pb, Cu, Zn and Mn in the grains, in the husk, in shoot and root, respectively, and Msoil is the concentration in the soil.

The translocation factor (TF) evaluates the relative displacement of Ni, Cd, Pb, Cu, Zn and Mn from plants' roots towards other plants' parts such as shoots, grains and husk. The TF is calculated using the following equations (Azzia et al. 2017):

$$TF\ (grain) = Mgrain / Mroot \quad (5)$$

$$TF\ (husk) = Mhusk / Mroot \quad (6)$$

$$TF\ (shoot) = Mshoot / Mroot \quad (7)$$

2.6 Health Risk Assessment

2.6.1. Health Risk Index (HRI)

Health risk index is calculated to verify exposure risks of PTE's through food intake. It reveals the risk of consuming contaminated foodstuffs. HRI is the ratio between the exposure and the reference oral dose (RfD) (USEPA, US Environmental Protection Agency 2002). It is generally used in EPA non-cancer health assessments. The RfD values used in this study were 0.02, 0.04, 0.004, 0.001, 0.03, 0.30 mg kg^{-1} for Ni, Cu, Pb, Cd, Mn and Zn, respectively. (USEPA, 2010; WHO, 1993; Food and Nutrition Board, 2004). Therefore, the health risk index was calculated as in the following equation:

$$HRI = (DI) \times (Cmetal) / RfD \times B \quad (8)$$

Where DI is the daily rice intake rate (kg per day), Cmetal is the metal concentration in the grain (mg kg^{-1}), RfD is the oral reference dose for the metal (mg kg^{-1} per weight per day) and B is the human body mass (kg). The DI used was 34 kg $year^{-1}$ for adults and for children (0 to 6 years) the consumption was established as 1/3 of the rate for adults (IBGE 2004, 2006; WHO 1993). The average B value was 70 kg for adults and 19.5 kg for children (IBGE 2004, 2006; Guerra et al. 2012; WHO 1993).

HRI greater than 1 for any metal in rice grains is considered unsafe for human health and serious health risks are possible. However, HRI < 1 means that oral exposure to PTE's is considered safe. (Storelli 2008).

2.6.2 Hazard Index (HI)

The potential health risk from exposure of several metals in rice grains (HI) was calculated according to the EPA guides for health risk assessment (Ogunkunle et al. 2016) using the following equation:

$$HI = \Sigma HRI = HRICd + HRINi + HRIPb + HRICu + HRIMn + HRIZn \quad (9)$$

For the Health Risk Index (HRI) and Harzard Index (HI) calculations, the three metallurgical wastes (Metallurgical press residue; FPM: Filter press mud; PM: Phosphate mud) were considered at 8 t ha^{-1}

dose.

2.7 Quality Control and Quality Assurance

The qualitative detection limit for each analytical method (MDL) was calculated after determining Ni, Cd, Zn, Pb, Mn, Cu in seven blank samples and using the equation below (APHA 2012): MDL = (x + t × s)×d, whereas, x is the average content of the substance in seven blank samples, t is the Student value at 0.01 probability and n-1 degrees of freedom (for n+7, α = 0.01 and t = 3,14), s is the standard deviation of these seven samples, d is the diluton eventually applied. The MDL for Zn, Cu, Cd, Mn, Ni and Pb were 2.9, 2.35, 7.7, 11.6, 1.36 e 0.72 μg $kg^{-1,}$ respectively. All glassware used was rinsed several times with 10% HNO_3 in order to avoid contamination. An internal sample was used to ensure quality control, and determination was only run if recovery was higher than 90%.

2.8 Statistical Analysis

Statistical analysis was performed by conducting a variance analysis (F test, P <0.05). The effect of steel-mill wastes application and their doses on the soil regarding the contents of Zn, Cu, Cd, Mn, Ni and Pb in husks, grains, shoots and roots were evaluated with the Tukey test (P <0.05).

For bioaccumulation factor (BAF) and translocation factor (TF), only the effect of steel-mill wastes was evaluated through the mean of all doses (n = 18) by applying the Tukey test (P <0.05).

3 Results And Discussion

Rice crops have developed normally until the end of the experiment, and the presence of steel mill wastes did not promote significat visual changes in plants. However, physiological studies must be conducted in order to deeply study their effects in rice crops, and even in other crops of interest.

3.1. PTE’s in plant parts

PTE’s concentrations in rice plant parts cultivated in soils treated with residues presented similar pattern of distribution (Figs. 1 and 2). Higher concentration of metals studied were found in root, except for Mn, that presented higher concentrations in the husk and shoots and Zn which revealed similar contents in shoots and roots. Both elements are the ones at greater contents compared to other PTE’s. This result is expected, since these two elements are micronutrients and their use in plants’ upper parts is greater than other PTE’s. Copper is also a micronutrient, but its contents is lower than Zn and Mn in rice parts, which could be attributed to the natural lower contents of Cu in the wastes. Lead contents in rice were the lowest ones compared to other PTE’s, even without application of steel mill wastes, probably due to its toxic effect and its natural low contents of Pb in soils (Zulfiqar et al. 2019; Cândido et al. 2020).

Cadmium contents in shoots and roots were not affected by wastes application, but differences were observed in husk and grains with the application of PM and MPR, respectively, both at doses greater than

4 t ha^{-1} (Fig. 1). The FPM is the waste with natural greater contents of Cd, but no mobilization of Cd was observed. The natural contents of Cd in FPM is within the range of background contents in soils (5.96 and 2.48 mg kg^{-1}) which promoted a safe value for Cd in rice according to the CAC (Codex Alimentarius Comission 2004) and according to the Chinese Standard, respectively (Luo et al. 2020). However, the Brazilian limit for Cd in rice is 0.1 mg kg^{-1} (ANVISA 2013) and some residues have exceeded this limit in grains, especially the MPR applied at doses above 4 t ha^{-1}. In MPR, the increase in Cd contents in grains is worrisome, contents reached up to 1.5 mg kg^{-1} while the commom content is betwwen 0.3 and 0.6 (Li et al. 2017).

Pb concentrations in grains and roots were not affected by application of MPR doses (Fig. 1). In husk, application of MPR had actually decreased Pb contents, when compared to control. FPM doses influenced the Pb concentration in all plant compartments and it is the waste with greater natural contents of Pb. In grains and shoots, there was an increase in Pb contents after application of 0.5 t ha^{-1} of FPM. However, Pb contents decreases to contents similar to the control treatments after 2 and 8 t ha^{-1} application doses respectively, in grains and shoots, suggesting a possible barrier against Pb absorption at greater contents due to a toxic effect. It is well known that Pb accumulation occurs in the roots'cells, due to the blockage of Casparian Strips, inside the endoderm, and possible retention by the negative charge of the cell walls in roots (Pourrut et al. 2011). Other grains such as barley and wheat reveals background concentration of Pb between 0.2 and 0.5 mg kg^{-1} which is in our range of variation even with the application of wastes (Kabata-Pendias, 2015). In Brazil, the Health Ministry establishes a maximum concentration of Pb in rice of 0.2 mg kg^{-1} (ANVISA 2013).

FPM, MPR and PM application at different doses affected the concentration of Mn only in husk, and in the case of PM, also in grains (Fig. 1). At the 4, 8, and 16 t ha^{-1} PM application, Mn concentration in grains were reduced in relation to control, suggesting a possible defense mechanism that avoids Mn accumulation in rice at greater wastes doses. For rice, the commom variation for Mn contents in grains is 13.2–27.1 mg kg^{-1} according to Kabata-Pendias (2015). However, greater contents of Mn in rice grains were already reported by Kumar et al. (2017) in a range between 61–70 mg kg^{-1} which is best related to our results. The Brazilian Food Composition Table (TACO 2011) recommends 10.3 mg kg^{-1} as normal Mn concentrations in rice. In our study, the contents of Mn in grains is greater than other elements due to its essentiality on plants functions. Also, the reducing conditions during cultivation could be responsible for a higher mobility of Mn and therefore, high contents of Mn in rice parts (Halder et al. 2019).

Regarding Cu content in the plant parts, applied doses of MPR affected the Cu concentration in grains, husk and shoot. In the grains, the concentration of Cu increased with application of up to 2 t ha^{-1} of MPR, with subsequent decrease in Cu contents with rising doses (Fig. 2). Besides, in grains the application of 2 t ha^{-1} of MPR promoted higher Cu concentration than the application of the same rates of FPM and PM and it is good to highlight that MPR is the waste with greater contents of Cu. For FPM and PM, Cu concentration in all the plants compartiments were affected by doses, except grains. In the husk, shoot, and roots, a tendency of increased concentration with rising doses of FPM and PM is

observed (Fig. 2), which is benefital up to some extent due to the essentiality of Cu. Our results approach other studies also made with Cu contents in rice grains that reported 12.71 mg kg^{-1} of Cu in rice grains (Sun et al. 2017).

In rice root and shoot, Zn contents increased only with PM application, which is probably associated with the elevated Zn content in this residue (Table 2). However, none of residues were able to increase the Zn concentration in rice grains (Fig. 2). Therefore, some aspect could be responsible for blocking Zn transportation to upper parts, such as grains. A considerable variation on Zn contents in husk was also noticed with FPM application, however, a random behaviour was observed.

At the highest dose (16 t ha^{-1}), plants treated with MPR showed the highest Ni concentration compared with other residues, followed by PM and FPM, which is in accordance with the increasing natural Ni contents in these wastes (Fig. 2). No differences in Ni contents in the roots were observed with application of different residues at increasing doses (Fig. 2). In shoots, the doses of FPM and MPR affected the Ni concentrations. In case of FPM, no clear pattern was observed, but for MPR, rising concentrations were found with increased of applied doses. Increased concentrations of Ni in grains were found with the application of 4, 8, and 16 t ha^{-1} of MPR. The application of steel mill wastes at different doses increased Ni contents in grains more than fifteen times compared to normal Ni contents in rice, which according to Sommella et al. (2013) varied from 0.08 and 0.35 mg kg^{-1}.

These types of wastes were considered promising soil amendments, improving urban soil quality (Guerrini et al. 2017). The reason that justifies the application of PM in rice crops is the increase promoted by it in micronutrientes contents such as Cu, Zn and Mn. Application of wastes seemed to favor accumulation of these micronutrientes in different plant parts, however, increasing contents in grains seems to be a minor process. This is in agrement with a review made by Ali et al. (2020) who studied accumulation of PTE's in rice paddy soils, and shows that translocation to grains is little. Also, it is good to highlight that even not having the speciation of these elements or a deeper study on the physiological process of elements absorption, this study is the first step towards stell mill wastes utilization in soils considering their risk assessment.

3.2. Bioaccumulation factor and translocation factor

The BAF represents the real activity of potentially toxic elements when agricultural crops are cultivated in contaminated soil (Table 3). Assessing the element content in plant part is the most accurate way to evaluate its availability, even extraction solutions having been used with the same purpose. Roots were the organs most responsible for accumulation of PTE's, except for Mn, which revealed greater bioaccumulation in shoots, being even greater after wastes application. Manganese is the element with greater BAF and even being a miucronutrient, it can be responsible for toxic effects on plants (Faria et al. 2020) and humans (Williams et al. 2012), when the intake is greater than the suggested. Therefore, in a soil with high levels of Mn, or high available levels of Mn, bioaccumulation in rice would be worrisome.

Table 3
Mean values (n = 18) of the bioaccumulation factor (BAF) of Ni, Cd, Pb, Cu, Zn and Mn in grains, husk and shoots in rice plants grown in soil treated with steel mill wastes: MPR: Metallurgical press residue; FPM: Filter press mud; PM: Phosphate mud and control treatment

Waste	**Grains**	**Husk**	**Shoots**	**Roots**	**Grains**	**Husk**	**Shoots**	**Roots**
	Ni				**Cu**			
MPR	0.70a	1.10a	0.65b	1.85a	0.43ab	0.27a	0.50ab	0.97a
FPM	0.33a	1.71a	0.46ab	2.24a	0.40a	0.33ab	0.60b	1.33ab
PM	0.61a	1.08a	0.52b	1.82a	0.43ab	0.42b	1.21c	1.80b
Control	0.00a	0.46a	0.15a	2.50a	0.63b	0.20a	0.27a	1.17a
MSD	0.82	1.32	0.35	1.18	0.21	0.14	0.29	0.51
	Cd				Zn			
MPR	2.32a	1.26a	2.39a	3.21a	0.72a	0.99a	1.74ab	1.51a
FPM	0.00	1.16a	0.97a	2.09a	0.69a	0.93a	2.08b	3.11bc
PM	0.20a	2.07a	1.48a	2.61a	0.44a	0.68a	2.77b	4.48c
Control	0.10a	0.76a	1.06a	2.60a	0.76a	0.96a	0.90a	1.56ab
MSD	2.83	1.55	2.85	2.86	0.33	0.42	1.14	1.57
	Pb				Mn			
MPR	0.08a	0.28ab	0.86a	1.63a	0.62b	6.15b	9.91b	3.91a
FPM	0.05a	0.16a	0.24a	2.18a	0.63b	6.72b	10.03b	3.90a
PM	0.07a	0.43b	0.37a	1.28a	0.46ab	6.60b	12.5b	3.50a
Control	0	0.13a	0.32a	1.20a	0.30a	1.23a	5.89a	2.16a
MSD	0.17	0.26	0.41	1.15	0.26	2.41	2.91	1.76
Lower case letters compare bioaccumulation factor (BAF) in rice plants’parts in each steel mill waste and control treatment. MSD: minimum significant difference.								

Nickel revealed greater bioaccumulation in roots with or without wastes application, but the application of wastes increase bioaccumulation in shoots. For copper, the waste PM increases bioaccumulation in husk, shoots and roots compared to the control and this was unexpected because this waste has the lowest content of Cu among the wastes. Zn was affected by wastes application only in shoots and roots being FPM and PM increasing accumulation in roots in a greater extent compared to the control. Lead was only affected in husk, increasing bioaccumulation when PM was applied. Cadmium was not affected at all by the three wastes. Therefore, steel mill wastes can increase elements bioavailability,

especially PM, and this increase can be associated to changes in metal especiation (Islam et al. 2015) or soil pH (Ali et al. 2020) due to different inputs of other elements and the element.

Table 4 shows the translocation factor for each element in each plant'part. In general, most of the elements show a low translocation capacity, regardless of waste application. However, Mn have the greatest capacity to be translocated, showing TF > 1 except in grains, followed by Zn. Zinc was translocated at significant amounts for shoots after MPR application and Mn was translocated at significant amounts for husk after applying all wastes. Control treatments show a high translocation factor for Zn and Mn in shoots, meaning that this process naturally occurs in rice. Translocation of Mn to rice shoots was already described (Lidon et al. 2004) and the fact that Mn is more mobile under reducing conditions can explain its high acumulation in rice cultivated under such conditions (Halder et al. 2019). Considering the fact that Zn and Mn are also micronutrients for plants, higher concentrations of these elements is expected (Bonnano et al. 2018).

Table 4
Mean values (n = 18) of the translocation factor (TF) of Ni, Cd, Pb, Cu, Zn and Mn in grains, husk and shoots in rice plants grown in soil treated with steel mill wastes: MPR: Metallurgical press residue; FPM: Filter press mud; PM: Phosphate mud and control treatment

Waste	Grains	Husk	Shoots	Grains	Husk	Shoots
	Ni			**Cu**		
MPR	0.42a	0.67ab	0.40b	0.48b	0.27b	0.53b
FPM	0.16a	0.79b	0.24ab	0.32ab	0.25b	0.45b
PM	0.33a	0.66ab	0.27b	0.22a	0.24b	0.71c
Control	0.00a	0.18a	0.06a	0.50b	0.18a	0.24a
MSD	0.44	0.54	0.19	0.20	0.05	0.17
	Cd			Zn		
MPR	0.51a	0.34a	0.53a	0.48b	0.69b	1.17b
FPM	0.07a	0.44a	0.54a	0.23a	0.31a	0.69a
PM	0.09a	0.51a	0.57a	0.11a	0.16a	0.63a
Control	0.03a	0.32a	0.42a	0.50b	0.62b	0.59a
MSD	0.52	0.37	0.40	0.12	0.15	0.33
	Pb			Mn		
MPR	0.05a	0.16ab	0.51b	0.19a	1.72b	2.85a
FPM	0.027a	0.08a	0.15a	0.15a	1.79b	2.64a
PM	0.06a	0.37b	0.37ab	0.11a	2.04b	3.58a
Control	0.00a	0.10a	0.27ab	0.13a	0.58a	2.80a
MSD	0.13	0.22	0.24	0.10	0.87	2.70

Lower case letters compare translocation factor (TF) in rice plants’parts in each steel mill waste and control treatment. MSD: minimum significant difference.

Some translocation capacity was noticed in shoots and husk for the elements Ni, Cu and Pb when compared to control treatments, but concentrations in roots are still greater than any plant’ part. The presence of Ni and Pb in rice grain can be considered a potential route for exposure (Halder et al. 2019), however, in general Pb is indeed very little mobilized to shoots (Memoli et al. 2017). The capacity of not translocating PTE’s from roots can be considered a defense mechanism avoiding toxic symptons in aboveground organs (Bonnano et al. 2017, 2018).

Cadmium translocation was not affected at all by wastes application. Rice has become one of the main sources of Cd intake worldwide (Li et al. 2017) which would suggest great translocation to plants and grains. However, Cd translocation depends on many factors such as root morphology, roots anatomy and cultivation conditions (Huang et al. 2018).

3.3. Health risk index and Harzard index

Table 5 shows that rice crops being cultivated in soils after steel mill application are worrisome concerning some PTE's. Considering only adults, Mn would be a problem in rice crops after applying MPR and FPM in soils (HRI > 1), but not with PM. This could be due to the lower content of this element in this latter waste, and therefore, less bioavailable Mn is in soil. Cadmium concentration can be hazardous after applying MPR in soils but not with other wastes.

Table 5
– Values of Health Risk Index (HRI) and Hazard Index (HI).

Waste		Ni	Cd	Pb	Cu	Zn	Mn	HI
MPR	Adults	0.92	1.81	0.0002	0.27	0.19	2.18	5.36
FPM		0.19	0.01	0.0002	0.31	0.20	2.13	2.84
PM		0.56	0.44	0.0002	0.33	0.15	0.90	2.38
MPR	Children	1.10	2.17	0.0003	0.32	0.22	2.60	6.42
FPM		0.22	0.01	0.0003	0.37	0.24	2.55	3.39
PM		0.67	0.53	0.0003	0.39	0.19	1.07	2.85

Metallurgical press residue; FPM: Filter press mud; PM: Phosphate mud

The lowest HRI is attributed to Pb and it ranks the second place in the substance priority list of ATSDR (ATSDR 2019a). This could be a consequence of the low mobility of Pb, which reduces the risk of rice intake. The risks involved with Pb oral exposure are related to neurological, renal, cardiovascular, immunological and reproductive effects (ATSDR 2019b). According to Table 4, this element has low absorption of Pb from soil, even in the control treatment. It is expected, therefore, that Pb is not so available in soil. Considering the reducing conditions of the soil in our study, some papers discuss that continuous flooding of soils can decrease Pb availability to rice (Ye et al. 2018), converting the acid-soluble fraction of Pb to the reducible fraction.

Since children are more sensitive to PTE's (Chen et al. 2018), it can be seen a pattern of increasing HRI when compared to adults values. For children, Ni concentration would be also risky after application of MPR, likewise Cd. Manganese would be risky for all three wastes. Therefore, one can state that none of the steel mill wastes can be used as fertilizers, since all of them can pose health risks, especially for children.

Health risks index in rice was already reported by many authors and highlighted the importance of Cd (Chen et al. 2018; Shahriar et al. 2020), Pb and Ni (Fakhri et al. 2018; Halder et al. 2019) as PTE's in this route of exposure. Some countries, such as Bangladesh, that has a greater rice comsuption rate (Shahriar et al. 2020) must manage rice crops with greater caution, trying to reduce availability of PTE's and protect population health, especially in paddy soil, where reducing conditions prevail.

The sum of individual elements and their health risks index show that MPR would be the most risky waste to be used in soils, while PM would be the least one. However, application of industrial wastes in soils must therefore be done with caution because all of them can be considered hazardous. Even some benefitial effect having been observed, this study already alerts for the risks associated with steel mill wastes application in soils.

4. Conclusion

Phosphate mud revealed to be the most promising waste to be used as fertlizer. However, PTE's absorbed by rice after steel mill waste application is a potential risk for consumers. Application of steel mill wastes, even being encouraged as an option to fertilize soils due to micronutrients content, is not recommended. This reinforces the importance of health risk assessment when evaluating the feasibility of wastes reuse. Other studies with these wastes, with other crops, in other environmental conditions are highly encouraged.

Declarations

Ethics approval

Not applicable.

Consent to participate

Not applicable.

Consent to publish

Not applicable.

Availability of data and materials

The datasets used and/or analysed during the current study are available from the corresponding author on reasonable request.

Competing Interests

The authors declare that they have no competing interests.

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Authors Contributions

GSC conducted the experiment, analyzed, interpreted data and helped writing the manuscript. JRO interpreted data, organized figures and tables, helped writing and revising the manuscript. ICFV helped interpreting data and writing and revising the manuscript. MJ helped interpreting data, writing the manuscript and revising it. MLTS helped interpreting data, writing the manuscript and revising it. MTPJ organized figures and tables, helped writing and revising the manuscript. FRDL helped writing and revising the manuscript. JJM supervised the research and helped searching funding for the project.

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Tables

Table 1. Soil characterization for Haplic Gleisol (0 – 0.2m)

Soil attributes	Values	Soil attributes	Values
pH (H_2O)	6.4	Cu (mg dm^{-3}) (1)	4
P (mg dm^{-3}) (1)	1.7	Cu (mg dm^{-3}) (6)	13
K (mg dm^{-3}) (1)	55	Fe (mg dm^{-3}) (1)	678
Ca^{2+} ($cmol_c$ dm^{-3}) (2)	2.6	Mn (mg dm^{-3}) (1)	52
Mg^{2+}($cmol_c$ dm^{-3}) (2)	0.4	Mn (mg dm^{-3}) (6)	79
Al^{3+} ($cmol_c$ dm^{-3}) (2)	0	Zn (mg dm^{-3}) (1)	7
H+Al ($cmol_c$ dm^{-3}) (3)	1.7	Zn (mg dm^{-3}) (6)	8
CEC ($cmol_c$ dm^{-3})	3.1	Ni (mg dm^{-3}) (6)	14
CEC at pH 7($cmol_c$ dm^{-3})	4.8	Pb (mg dm^{-3}) (6)	17
Base saturation (%)	65	Cd (mg dm^{-3}) (6)	0.21
Aluminum saturation (%)	0	Cr (mg dm^{-3}) (6)	53
O. M. (g kg^{-1}) (4)	21		
Sand (g kg^{-1}) (5)	530		
Silt (g kg^{-1}) (5)	110		
Clay (g kg^{-1}) (5)	360		

(1) Mehlich-1. (2) KCl 1 mol L^{-1}. (3) CaOAC 0.5 mol L^{-1}. (4) Walkley-Black (DeFilippo and Ribeiro, 1997). (5) Teixeira et al. (2017). (6) USEPA 3051A (USEPA, 2007).

Table 2. Steel mill wastes chemical characterization: MPR: Metallurgical press residue; FPM: Filter press mud; PM: Phosphate mud

Waste	pH	P	K	Ca	Mg	S	Na	Zn
				——mg kg^{-1}——				
FPM	7.4	2.1	735	249	37.6	270	693	10,725
PM	2.4	18.7	25	21	1.2	2.4	3.7	52.5
MPR	7.1	75	11	2.5	2.4	390	138	31
Waste	Ni	Cr	Pb	Cd	Mn	Fe	Cu	
				——mg kg^{-1}——				
FPM	151	57	6,072	4.3	1.6	296.0	459	
PM	214	8	19	0.2	330	12.9	10	
MPR	307	566	10	0.9	3,104	477.0	677	

Table 3. Mean values (n = 18) of the bioaccumulation factor (BAF) of Ni, Cd, Pb, Cu, Zn and Mn in grains, husk and shoots in rice plants grown in soil treated with steel mill wastes: MPR: Metallurgical press residue; FPM: Filter press mud; PM: Phosphate mud and control treatment

Waste	Grains	Husk	Shoots	Roots	Grains	Husk	Shoots	Roots
	Ni				Cu			
MPR	0.70a	1.10a	0.65b	1.85a	0.43ab	0.27a	0.50ab	0.97a
FPM	0.33a	1.71a	0.46ab	2.24a	0.40a	0.33ab	0.60b	1.33ab
PM	0.61a	1.08a	0.52b	1.82a	0.43ab	0.42b	1.21c	1.80b
Control	0.00a	0.46a	0.15a	2.50a	0.63b	0.20a	0.27a	1.17a
MSD	0.82	1.32	0.35	1.18	0.21	0.14	0.29	0.51
	Cd				Zn			
MPR	2.32a	1.26a	2.39a	3.21a	0.72a	0.99a	1.74ab	1.51a
FPM	0.00	1.16a	0.97a	2.09a	0.69a	0.93a	2.08b	3.11bc
PM	0.20a	2.07a	1.48a	2.61a	0.44a	0.68a	2.77b	4.48c
Control	0.10a	0.76a	1.06a	2.60a	0.76a	0.96a	0.90a	1.56ab
MSD	2.83	1.55	2.85	2.86	0.33	0.42	1.14	1.57
	Pb				Mn			
MPR	0.08a	0.28ab	0.86a	1.63a	0.62b	6.15b	9.91b	3.91a
FPM	0.05a	0.16a	0.24a	2.18a	0.63b	6.72b	10.03b	3.90a
PM	0.07a	0.43b	0.37a	1.28a	0.46ab	6.60b	12.5b	3.50a
Control	0	0.13a	0.32a	1.20a	0.30a	1.23a	5.89a	2.16a
MSD	0.17	0.26	0.41	1.15	0.26	2.41	2.91	1.76

Lower case letters compare bioaccumulation factor (BAF) in rice plants’parts in each steel mill waste and control treatment. MSD: minimum significant difference.

Table 4. Mean values (n = 18) of the translocation factor (TF) of Ni, Cd, Pb, Cu, Zn and Mn in grains, husk and shoots in rice plants grown in soil treated with steel mill wastes: MPR: Metallurgical press residue; FPM: Filter press mud; PM: Phosphate mud and control treatment

Waste	Grains	Husk	Shoots	Grains	Husk	Shoots
	Ni			Cu		
MPR	0.42a	0.67ab	0.40b	0.48b	0.27b	0.53b
FPM	0.16a	0.79b	0.24ab	0.32ab	0.25b	0.45b
PM	0.33a	0.66ab	0.27b	0.22a	0.24b	0.71c
Control	0.00a	0.18a	0.06a	0.50b	0.18a	0.24a
MSD	0.44	0.54	0.19	0.20	0.05	0.17
	Cd			Zn		
MPR	0.51a	0.34a	0.53a	0.48b	0.69b	1.17b
FPM	0.07a	0.44a	0.54a	0.23a	0.31a	0.69a
PM	0.09a	0.51a	0.57a	0.11a	0.16a	0.63a
Control	0.03a	0.32a	0.42a	0.50b	0.62b	0.59a
MSD	0.52	0.37	0.40	0.12	0.15	0.33
	Pb			Mn		
MPR	0.05a	0.16ab	0.51b	0.19a	1.72b	2.85a
FPM	0.027a	0.08a	0.15a	0.15a	1.79b	2.64a
PM	0.06a	0.37b	0.37ab	0.11a	2.04b	3.58a
Control	0.00a	0.10a	0.27ab	0.13a	0.58a	2.80a
MSD	0.13	0.22	0.24	0.10	0.87	2.70

Lower case letters compare translocation factor (TF) in rice plants’parts in each steel mill waste and control treatment. MSD: minimum significant difference.

Table 5 – Values of Health Risk Index (HRI) and Hazard Index (HI).

Waste		Ni	Cd	Pb	Cu	Zn	Mn	HI
MPR	Adults	0.92	1.81	0.0002	0.27	0.19	2.18	5.36
FPM		0.19	0.01	0.0002	0.31	0.20	2.13	2.84
PM		0.56	0.44	0.0002	0.33	0.15	0.90	2.38
MPR	Children	1.10	2.17	0.0003	0.32	0.22	2.60	6.42
FPM		0.22	0.01	0.0003	0.37	0.24	2.55	3.39
PM		0.67	0.53	0.0003	0.39	0.19	1.07	2.85

Metallurgical press residue; FPM: Filter press mud; PM: Phosphate mud

Figures

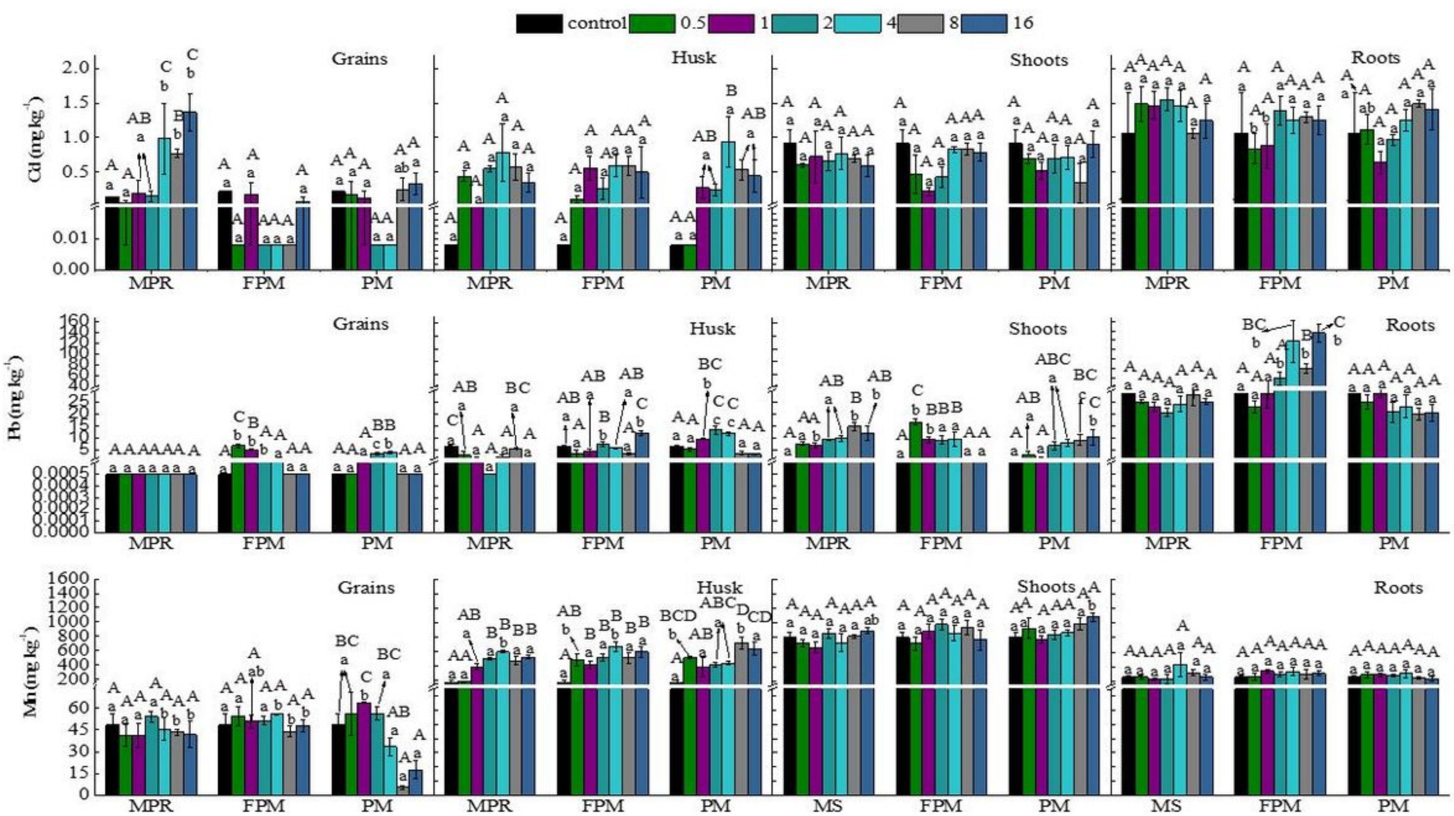

Figure 1

Cd, Pb and Mn contents in rice: grains, husk, shoots and roots considering six concentrations of steel mill wastes: MPR: Metallurgical press residue; FPM: Filter press mud; PM: Phosphate mud. Upper case letters

compare results between doses and lower letters compare results between wastes, according to Tukey test (P < 0.05). Error bars represent error (n = 3).

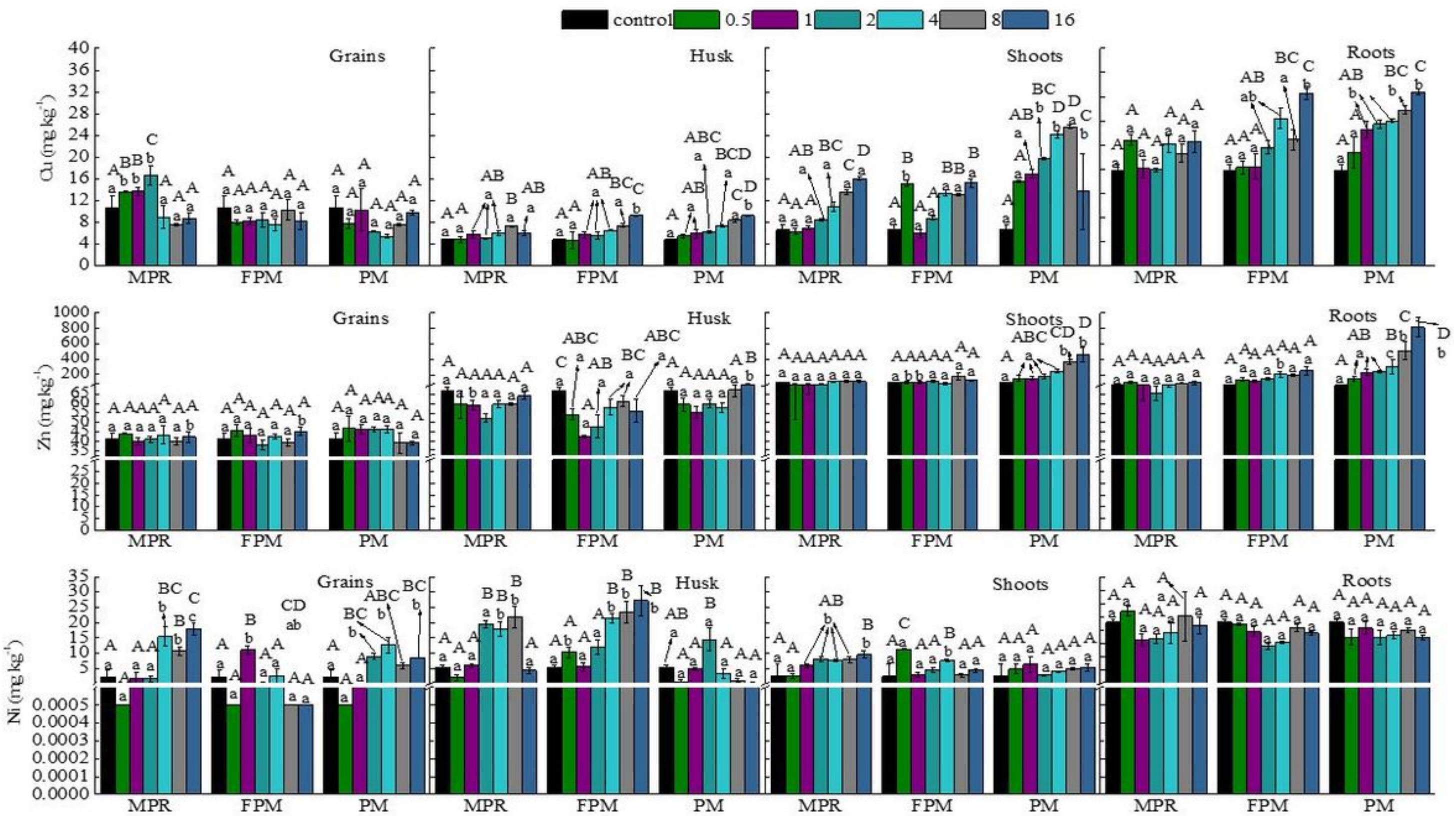

Figure 2

Cu, Zn and Ni contents in rice: grains, husk, shoots and roots considering six concentrations of steel mill wastes: MPR: Metallurgical press residue; FPM: Filter press mud; PM: Phosphate mud. Upper case letters compare results between doses and lower letters compare results between wastes, according to Tukey test (P < 0.05). Error bars represent error (n = 3).