Ecological Risk Assessment of Heavy Metals in the Fortified Organic Fertilizers produced from Ganga Sewage Sludge (SS) in Haridwar

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Introduction

Rapid urbanization and increasing population generate huge volumes of discharge which are beyond the self-purifying capacity of the ecosystem (Abdoli 2022). These non-farm waste streams may be municipal or industrial in origin, and often presents an environmental and health challenge (Mokhtari et al. 2017). However, in a circular economy, these waste materials are transformed into an opportunity by recycling and reusing(Velasco-Muñoz et al. 2021). The residual, semi-solid material produced as a by-product in sewage treatment plants is known as sewage sludge (SS) (Mokhtari et al. 2017). Management of this SS is a challenge both in economic and environmental terms. The various methods for disposal of SS include incineration, sanitary burial and composting (Abdoli 2022). Burning and burial of SS on land and oceans create health and environmental hazards and hence, are legally restricted (Smith 2009; Tchounwou et al. 2012). Due to high organic carbon, nitrogen (N) and phosphorous (P) content, the use of SS as a fertilizer is encouraged as it recycles nutrients and contributes to circularization in agriculture (Camargo et al. 2016; Velasco-Muñoz et al. 2021). However, the presence of various contaminants and pathogenic organisms in SS can pose a high risk to public health, and hence, its use for agricultural purposes requires special guidance

Organic agriculture is a system of agriculture that avoids chemical inputs (FAO). SS offers a good organic source of NPK and other micronutrients (Shafieepour et al. 2011). It acts as a cheap fertilizer for use in barren areas for improving plant growth (Shafieepour et al. 2011). It is especially useful in arid and semi-arid regions as a cheap source of water, organic matter and nutrients (Tabatabaei et al. 2020). Improvements in growth and yield parameters of various foodgrains have been observed by various researchers, including rice (Latare et al. 2014), wheat (Fathololomi and Asghari 2015; Fathololomi et al. 2015; Shahbazi F et al. 2018), corn (Karimpour et al. 2010; Saadat K. et al. 2012; Hoshyar and Baghaie 2017), and barley (Chorom and Aghaei Foroushani 2007). Further, vegetables, like tomato (*Lycopersicon esculentum* L.) (Hossain et al. 2015) and eggplant (*Solanum melongena*) (Kumar and Chopra 2016) also showed significant improvement in health due to the application of SS as a fertilizer. However, the effect of SS on the above plants was influenced by many factors, such as the origin of SS (urban or industrial), rate of SS application, and the plant type (Abdoli 2022).

Heavy and toxic metal contamination of SS-based compost

The direct usage of SS for agricultural uses is discouraged due to the presence of various pathogens and toxic metals (Camargo et al. 2016). Among the various contaminants, heavy metals have gained significant attention due to their peculiar characteristics, including high reactivity, lipophilic nature, non-biodegradability, and toxicity (Duruibe et al. 2007). Heavy metal composition of municipal waste, and in turn, SS, depends on various factors, including the source of the wastes, composting process and geographical location (He et al. 1992; Krogmann 1999). Electronic wastes contribute almost 70 percent of heavy metals to landfill waste (Wuana and Okieimen 2011). Small and medium enterprises located around urban centres often dispose of their waste along with municipal wastes (Esakku et al. 2003).

Heavy metals contamination of SS is a major hindrance in their use for agricultural purposes. Earlier studies found heavy metals to be over the specified limits in municipal solid wastes (Merian 1991; Cebula et al. 1995; Ciba et al. 1999). Many heavy metals were found to be present in municipal wastes, including lead, nickel, zinc, cobalt, cadmium, and manganese (Ciba et al. 1999). Most heavy metals exert a detrimental effect on the health of living organisms if present beyond certain concentrations. If such contaminated compost is used as manure for food production, the heavy and toxic metals may enter the food chain and become a major health hazard (Lopes et al. 2011). Hence, it is necessary to monitor the levels of these heavy and toxic metals in the compost produced using SS from such STPs. In this context, the present study is a preliminary investigation of the heavy and toxic metal content present in SS and products derived from SS compost.

Methodology

Study area description

The SS samples were collected from the Jagjeetpur sewage treatment plant (STP) located in Haridwar, Uttarakhand, INDIA. The STP uses the Sequencing Batch Reactor (SBR) process and has an installed capacity of 68 million litres per day (MLD). The STP was built under the Namami Gange program of the Government of INDIA and is responsible for the treatment of domestic sewage from urban areas of Haridwar.

Sample collection and analysis

The SS samples were collected from the dumping ground of the STP on 8^{th} and 15^{th} February 2022 in bulk in sealed packets and brought to the laboratory for further analysis. For products derived from SS-compost, we used two products manufactured by Patanjali Organic Research Institute (PORI) using the sludge collected from Jagjeetpur STP, i.e., Poshak (CHOS/PJPA/0322/450) and Jaivik Khad (CHOS/PJKA/0322/448). Poshak is a mycorrhiza-based granular biofertilizer, while Jaivik Khad is 100 percent organic manure with balanced levels of all the essential nutrients. Both the products are patented and are usually prepared using in-house compost. However, for this study, the Poshak and Jaivik Khad were prepared using the compost derived from STP sludge. The samples were analyzed for the presence of heavy and toxic metal contaminants according to the guidelines of the Fertilizer Control Order (FCO), 2009 (SWMR 2016). Samples were collected and analyzed in triplicates. The results are represented as average.

Results and Discussion

The SS from Jagjeetpur STP was found to be rich in organic matter, along with various micro-and macro-nutrients. . In the present study, a total of seven heavy and toxic metals were analyzed in the samples and compared with the tolerance limits as stated in FCO, 2009 (SWMR 2016). The SS collected from the Jagjeetpur STP contained all the analyzed metals within the permissible limits, except mercury (Hg), which was approximately 9-times more than the permissible limit (Table 1). Processed products contained decreased levels of Hg compared to the unprocessed SS, but still, it was around 4-times of the allowed limit in Poshak, while it was below the tolerance limit in Khad. The levels of lead (Pb) in Khad were similar to unprocessed sewage, but it doubled in Poshak. Similarly, the level of chromium (Cr) decreased slightly in Khad but doubled in Poshak. The processing of SS does not affect the levels of As in both the products. The level of Ni remained similar in both unprocessed SS and processed products. Copper (Cu) increased almost 50 percent in Poshak compared to SS, while it almost tripled in Khad. On the other hand, zinc (Zn) levels remained similar in Khad but tripled in Poshak. Despite the increase in the levels of various heavy metals in processed products, the levels were still below the permissible limits. An increase in some of the heavy metals, which are also macronutrients (Cr, Ni, Cu, Zn), is good for the soil.

The variation in the quantity of the various heavy metals is directly related to the input waste quality of the STP, which is prone to temporal fluctuations (SWMR 2016). Being close to an industrial area, the input waste streams of the STP is bound to contain numerous heavy metals. The fact that most of the heavy and toxic metals (except Hg) are within the permissible limit indicates the proper functioning of the STP and proper treatment of effluents by the various industrial units in the area. The high levels of various metals found in the processed products are expected since the removal of water concentrates the SS and its components. However, being within limits ensures that they are safe for use in organic farming. Further, the long-term effects of the application of these SS-compost based products is unlikely to have any major effects since any residual heavy and toxic metals often migrate deeper into the soil (Sun et al. 2017).

Mercury is a major pollutant in industrial areas released due to coal combustion and is associated with kidney damage (Wuana and Okieimen 2011). The pH of the soil determines the stability of the Hg in the SS. Mercurous and mercuric forms are more stable under oxidizing conditions. Under reducing conditions, organic or inorganic Hg may be converted to elemental form, which is further alkylated by biotic and abiotic processes. This alkylated form of Hg is water-soluble and volatile in air, making it the most toxic form (Smith et al. 1995). Sorption of Hg in soils, sediments and humic materials is the best remedial solution, which increases with an increase in pH (Wuana and Okieimen 2011). Under anaerobic conditions, both organic and inorganic forms of mercury may be alkylated by sulfur-reducing bacteria or may be converted to elemental form by demethylation or reduction of Hg(II). While acidic pH favours the methylation of Hg, higher pH leads to precipitation of HgS (Smith et al. 1995). Hence, the best solution to tackle higher Hg contamination in SS would be aerobic composting at higher pH to make them less toxic.

Chromium contamination is due to discharge from industries involved in electroplating industries (Smith et al. 1995). It is associated with allergic dermatitis in humans (Scragg 2006). Cr(VI) is the most common contaminating and toxic form. It can be reduced to Cr(III) under anaerobic conditions by soil organic matter. Mobility of Cr depends on the sorption capabilities of the soil, including mineral oxide content, amount of clay and organic matter. Cr(VI) is very mobile, which can be decreased by adsorption to oxide minerals and clays at pH below 5. In addition, solubility can be reduced by the formation of $Cr(OH)_3$ at pH above 5 (Chrostowski et al. 1991). Hence, Cr content in SS-derived compost can be controlled by ensuring high organic matter and mineral oxides.

Lead (Pb) is naturally present in surface soils ranging from 10 to 67 mg/Kg. It has a wide range of uses in industries, including storage batteries (Wuana and Okieimen 2011). Pb enters the body through inhalation and ingestion, leading to Pb poisoning or even death (Charkiewicz and Backstrand 2020). Lead sulfide (PbS) is formed under reducing soil conditions, while anaerobic conditions lead to the formation of volatile organo-lead due to microbial alkylation (Sevak et al. 2021). At higher pH, insoluble Pb forms are formed (McBride et al. 2005; Wuana and Okieimen 2011). Hence, Pb pollution can be controlled by compost formation under aerobic conditions at higher pH.

Naturally, nickel occurs at very low levels. It is essential in small quantities and is toxic above tolerable limits, responsible Nickel contamination occurs mostly through industries, and is removed from the environment through adsorption by sediments or soil particles, which makes it immobile. In acidic soils, Ni is more mobile and may leach into groundwater. It is not known to accumulate in plants or animals, and hence, doesn't pose a health risk via biomagnification through the food chain (Wuana and Okieimen 2011). Similarly, copper and zinc are also essential micronutrients. Both are necessary for human and plant health. While Cu does not bioaccumulate through the food chain, Zn may accumulate in some fishes and lead to biomagnification through the food chain (Colaço et al. 2006; Wuana and Okieimen 2011). In addition, Zn contamination in the soil can also affect the activity of various microorganisms and earthworms (Cheng and Wong 2002; Colaço et al. 2006; Wuana and Okieimen 2011).

Table 1. Heavy and toxic metal analysis of unprocessed SS and products based on SS-compost.

Heavy metals (mg/Kg)	Limits*	Sludge	Poshak	Khad
Mercury (Hg)	0.15	1.39	<0.1	0.66
Lead (Pb)	100	18.01	43.68	15.03
Arsenic (As)	10	3.00	4.00	3.41
Chromium (Cr)	50	14.15	32.55	11.61
Nickel (Ni)	50	12.42	14.63	11.88
Copper (Cu)	300	40.29	68.76	119.17
Zinc (Zn)	1000	230.3	774.98	228.16

*Tolerance limits as per Fertilizer Control Order (FCO), 2009.

Conclusion

The disposal of SS is a major problem and composting for agricultural uses is the most recommended method due to its high organic content. The input quality of waste streams influences the characteristics of the output SS, which in turn influences the quality of the products derived from it. Since the Jagjeetpur STP caters to urban areas near an industrial area, it is bound to be contaminated with heavy and toxic metals to some extent. The proper regulation and operation of the STP ensure that the level of heavy and toxic metals is within permissible limits. Although the level of Hg was well above the tolerable limit, it may be a temporal situation or a permanent problem in this geographical area. The products derived from the compost of SS had heavy and toxic metals within permissible limits. For a few metals, the levels increased in the processed products which may be due to the concentration of the sludge after water removal. However, the composting process renders most of the metals immobile or insoluble, rendering them unavailable for uptake by plants or animals. While the unprocessed SS is not recommended for agricultural purposes due to the presence of pathogenic and toxic metals, processed SS-derived product is suitable for organic farming due to permissible levels of toxic metals and the absence of pathogenic organisms.

References

Abdoli M (2022) Sustainable Use of Sewage Sludge in Soil Fertility and Crop Production. Sustain Manag Util Sew Sludge 297–333. https://doi.org/10.1007/978-3-030-85226-9_15

Barati Rashvanlou R, Farzadkia M, Marousi M (2018) Selection of Suitable Guidelines for the Application of Sewage Sludge in Agricultural Using Hierarchical Analysis System (AHP). J Environ Heal Enginering 5:299–308. https://doi.org/10.29252/JEHE.5.3.299

Camargo FP, Sérgio Tonello P, dos Santos ACA, Duarte ICS (2016) Removal of Toxic Metals from Sewage Sludge Through Chemical, Physical, and Biological Treatments—a Review. Water, Air, Soil Pollut 2016 22712 227:1–11. https://doi.org/10.1007/S11270-016-3141-3

Cebula J, Bodzek M, Loska K (1995) Contaminated Soil. Kluwer Academic Publishers, Dordrecht

Charkiewicz AE, Backstrand JR (2020) Lead Toxicity and Pollution in Poland. Int J Environ Res Public Heal 2020, Vol 17, Page 4385 17:4385. https://doi.org/10.3390/IJERPH17124385

Cheng J, Wong MH (2002) Effects of earthworms on Zn fractionation in soils. Biol Fertil Soils 2002 361 36:72–78. https://doi.org/10.1007/S00374-002-0507-Z

Chorom M, Aghaei Foroushani M (2007) Effects of amended sewage sludge application on yield and heavy metal uptake of barley: a case study of Ahvaz sewage treatment plant. J Water Wastewater 18:53–63

Chrostowski P, Durda JL, Edelmann KG (1991) The use of natural processes for the control of chromium migration. Remediation 2:341–351

Ciba J, Korolewicz T, Turek M (1999) The occurrence of metals in composted municipal wastes and their removal. Water Air Soil Pollut 111:159–170

Colaço A, Bustamante P, Fouquet Y, et al (2006) Bioaccumulation of Hg, Cu, and Zn in the Azores triple junction hydrothermal vent fields food web. Chemosphere 65:2260–2267. https://doi.org/10.1016/J.CHEMOSPHERE.2006.05.034

Duruibe JO, Ogwuegbu MOC, Egwurugwu JN (2007) Heavy metal pollution and human biotoxic effects. Int J Phys Sci 2:112–118

Esakku S, Palanivelu K, Joseph K (2003) Assessment of Heavy Metals in a Municipal Solid Waste Dumpsite. In: Proceedings of Workshop on Sustainable Landfill Management. pp 139–145

FAO Organic Agriculture: What is organic agriculture? In: FAO. https://www.fao.org/organicag/oa-faq/oa-faq1/en. Accessed 31 Mar 2022

Fathololomi S, Asghari S, Goli Kalanpal E (2015) Effects Of Municipal Sewage Sludge On The Concentration Of Macronutrients In Soil And Plant And Some Agronomic Traits Of Wheat. Electron J Soil Manag Sustain Prod 5:49–70

Fathololomi S, Asghari SH (2015) Effects of Ardabil Municipal Sewage Sludge on Some Physical and Hydraulic Properties of a Coarse-Textured Soil under Wheat Cultivation. Water Soil 24:169–183

He XT, Traina SJ, Logan TJ (1992) Chemical properties of municipal solid waste compost. J Env Qual 21:318–329

Hoshyar P, Baghaie AH (2017) Effect of Arak municipal sewage sludge application on corn Cd uptake in a loamy soil. Iran J Soil Res 31:303–314

Hossain MK, Strezov V, Nelson PF (2015) Comparative assessment of the effect of wastewater sludge biochar on growth, yield and metal bioaccumulation of cherry tomato. Pedosphere 25:680–685

Karimpour M, Afyuni M, Sari AE (2010) Effect of sewage sludge on mercury concentration in soil and corn. J Sci Technol Agric Nat Resour 14:115–123

Krogmann U (1999) Effect of season and population density on source separated waste composts. Waste Manag Res 17:109–123

Kumar V, Chopra AK (2016) Agronomical performance of high yielding cultivar of eggplant (Solanum melongena L.) grown in sewage sludge amended soil. Res Agric 1:1

Latare AM, Kumar O, Singh SK, Gupta A (2014) Direct and residual effect of sewage sludge on yield, heavy metals content and soil fertility under rice–wheat system. Ecol Eng 69:17–24. https://doi.org/10.1016/J.ECOLENG.2014.03.066

Lopes C, Herva M, Franco-Uría A, Roca E (2011) Inventory of heavy metal content in organic waste applied as fertilizer in agriculture: evaluating the risk of transfer into the food chain. Environ Sci Pollut Res 2011 186 18:918–939. https://doi.org/10.1007/S11356-011-0444-1

McBride M, Sauvé S, Hendershot W (2005) Solubility control of Cu, Zn, Cd and Pb in contaminated soils. Eur J Soil Sci 48:337–346. https://doi.org/10.1111/J.1365-2389.1997.TB00554.X

Merian E (1991) Metals and their compounds in the Environment. VCH, New York

Mokhtari M, Vaziri AS, Zareyi T, Jalili M (2017) An Investigation on Feasibility of Composting of Yazd WWT Dewatered Sludge with Windrow Method Using Different Treatments - Iranian Journal of Health and Environment. Iran J Heal Environ 9:493–502

Rahmani HR, Moayyeri M, Kouhanestani ZM, et al (2014) Studying some of the qualitative properties and concentration of heavy metals in dried sewage sludge from Shahin-Shahr WWTP in Isfahan. J Environ Sci

Technol 16:55–66

Saadat K., Barani Motlagh M., Dordipour E, Ghasemnezhad A (2012) Influence Of Sewage Sludge On Some Soil Properties, Yield And Concentration Of Lead And Cadmium In Roots And Shoots Of Maize. Electron J Soil Manag Sustain Prod 2:27–48

Scragg A (2006) Environmental Biotechnology, 2nd Editio. Oxford University Press (OUP), Oxford ,UK

Sevak PI, Pushkar BK, Kapadne PN (2021) Lead pollution and bacterial bioremediation: a review. Environ Chem Lett 2021 196 19:4463–4488. https://doi.org/10.1007/S10311-021-01296-7

Shafieepour S, Ayati B, Ganjidoust H (2011) Reuse of Sewage Sludge for Agricultural Soil Improvement (Case Study: Kish Island). J Water Wastewater 22:85–93

Shahbazi F, Ghasemi S, Sodaeizadeh H, et al (2018) The Effects of Sewage Sludge on Some Physical and Chemical Properties of Soil and Wheat Yield. Water Soil Sci 28:193–206

Smith LA, Means JL, Chen A (1995) Remedial Options for Metals-Contaminated Sites. Lewis Publishers, Boca Raton, Fla, USA

Smith SR (2009) A critical review of the bioavailability and impacts of heavy metals in municipal solid waste composts compared to sewage sludge. Environ Int 35:142–156. https://doi.org/10.1016/J.ENVINT.2008.06.009

Sun N, Shang HP, Ru SH, Su DC (2017) [Effects of Continuous Application of Sewage Sludge Compost on Heavy Metals Accumulation and Mobility Characteristics in Soil Profile and on Heavy Metals Uptake of Wheat]. Huan jing ke xue= Huanjing kexue 38:815–824. https://doi.org/10.13227/J.HJKX.201608031

SWMR (2016) The Solid Waste Management Rules, Ministry of Environment, Forest and Climate Change. INDIA

Tabatabaei SH, Nourmahnad N, Kermani SG, et al (2020) Urban wastewater reuse in agriculture for irrigation in arid and semi-arid regions - A review. Int J Recycl Org Waste Agric 9:193–220. https://doi.org/10.30486/IJROWA.2020.671672

Tchounwou PB, Yedjou CG, Patlolla AK, Sutton DJ (2012) Heavy Metals Toxicity and the Environment. EXS 101:133. https://doi.org/10.1007/978-3-7643-8340-4_6

Velasco-Muñoz JF, Mendoza JMF, Aznar-Sánchez JA, Gallego-Schmid A (2021) Circular economy implementation in the agricultural sector: Definition, strategies and indicators. Resour Conserv Recycl 170:105618. https://doi.org/10.1016/J.RESCONREC.2021.105618

Wuana RA, Okieimen FE (2011) Heavy Metals in Contaminated Soils: A Review of Sources, Chemistry, Risks and Best Available Strategies for Remediation. ISRN Ecol 2011:1–20. https://doi.org/10.5402/2011/402647