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Chromium in soil and tea (*Camellia sinensis* L.) infusion: Does soil amendment with municipal solid waste compost make sense?

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Calcium chloride (PubChem CID: 6093260)
Diethylenetriaminepentaacetic acid (PubChem CID: 3053)
Hydrochloric acid (PubChem CID: 313)
Hydrofluoric acid (PubChem CID: 14917)
Hydrogen peroxide (PubChem CID: 784)
Hydroxylamine hydrochloride (PubChem CID: 443297)
Nitric acid (PubChem CID: 944)
Potassium dichromate (PubChem CID: 24502)
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A B S T R A C T

In this study, two clones (TV1 and TV23) of tea (*Camellia sinensis* L.) plants were allowed to grow in earthen pots in a greenhouse, treated with municipal solid waste compost (MSWC) to study the effect of MSWC to the fractionation of chromium (Cr) ion, plant biomass production, plant uptake of Cr, fate of Cr in soils through risk assessment code (RAC) and Cr in tea infusion. Increasing rate of MSWC applications increased both total Cr and DTPA-extractable Cr in soil. Fractionation studies have revealed that Cr in soils was mainly associated with the organic and Fe–Mn oxide and the contribution of residual fractions is nearly 87.7–96.1%. The biomass yields of the tea plants were increased with the increase in MSWC, indicating that nutrient uptake of the plants from MSWC was dependent on the root–MSWC interface. High accumulation of Cr in the root of tea plants and its subsequent lower movement towards aerial parts corroborated the hypothesis that the root of the tea plants acts as a buffer. In this experiment the transfer factor was <1, indicating that the tea plants did not have a significant phytoextraction potential. In the application of 10 t ha^{-1} MSWC, Cr was found to be in medium risk for both clones whereas the application of 8 t ha^{-1} MSWC showed medium risk with respect to the Cr for TV23 clone applying RAC. Therefore, MSWC amendment rate above 8 t ha^{-1} increased the total biomass of the tea plants but posed a threat on environmental prospect with respect to Cr. It was also found that only 2.5 μg L^{-1} to 4.8 μg L^{-1} Cr was present in tea infusion when infusion was prepared from tea leaves receiving different doses of MSWC. Furthermore, stepwise regression technique was applied to choose the most significant regression variables to express the variability in leaves, stem, main root and feeder root biomass. The one-way analysis of variance along with the Dunnett's multiple comparison method was used to determine the significant differences between the means of different treatments. On the application of hierarchical cluster analysis, treatments were grouped in two distinct homogeneous groups.

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1. Introduction

A significant interest towards the use of municipal solid waste compost (MSWC) application for sustainable crop production is getting popular due to the dearth of availability of conventionally used raw materials for compost preparation (Gigliotti, Businelli, & Giusquiani, 1996; Hargreaves, Adl, & Warman, 2008; Karak, Bhattacharyya, Paul, Das, & Saha, 2013; Lakhdar et al., 2010). On the other hand the growing interest on MSWC application is associated with an element of concern, which is the presence of considerable amount of heavy metals (HMs). These heavy metals (HMs) can critically degrade the soil in addition of being incorporated into food webs through the plant uptake (Carbonell, de Imperial, Torrijos, Delgado, & Rodriguez, 2011; Jordao, Nascentes, Cecon, Fontes, & Pereira, 2006; Kabata-Pendias and Pendias, 2001; Khan, 2001; Laborda, Górriz, Bolea, & Castillo, 2007; Shanker, Cervantes,

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Loza-Tavera, & Avudainayagam, 2005). Among several HMs, Cr is an environmentally toxic metal that is assumed to have a long atmospheric dwelling time and hence subject to the long-range of persistence (Unsal, Tuzen, & Soylak, 2014). It has an involvement in certain metabolic processes like metabolism of glucose (Ražić & Đogo, 2010). Lower bioavailability of Cr in soils amended by MSWC has also been documented by several studies, as it is mainly bounded to organic matter in soil (Becquer, Quantin, Sicot, & Boudot, 2003; López-Luna, González-Chávez, Esparza-García, & Rodríguez-Vázquez, 2009). In contrary, other studies also have shown several beneficial effects of MSWC towards better crop management, as it may be considered as a source of different major nutrients and micronutrients to plants (Hargreaves et al., 2008; Hicklenton, Rodd, & Warman, 2001; Zhou et al., 2013). Therefore, the advantages or disadvantages of using MSWC as fertilizer and soil amendment should be evaluated with the possible environmental and toxicological impacts together, due to the presence of potentially toxic elements such as heavy metals (HMs). In general, in the solid phase of MSWC amended soils, Cr occurs in various chemical forms, and its plant availability is largely dependent upon these chemical forms (Prudent, Domeizel, & Massiani, 1996). In order to assess the potential effects and contamination in soil amended with MSWC, if the total Cr concentration in soil is considered as a criterion, it would imply that all forms of the element have an equal impact on the environment and such an assumption is clearly untenable (McBride, 1995; Tessier, Campbell, & Bisson, 1979). On the other hand an increase in the total content of Cr in compost treated soil has not necessarily led to the corresponding increase in plant uptake (Giusquiani, Gigliotti, & Businelli, 1992; Sharma et al., 1995; Zheljazkov & Warman, 2003). The potency of Cr to contaminate the food chain in MSWC applied soil can be determined by its bioavailability in soil, which in turn will be determined by the sequential extraction of Cr in MSWC amended soils. Cr fractionation procedures through sequential extraction coupled with plant uptake bioassays hold promise for assessing metal bioavailability (Achiba et al., 2009; Namieśnik and Rabajczyk, 2012). Soils can be classified according to the element mobility based on the fractionation study on Cr through the risk assessment code (RAC). The RAC assesses the potential release of elements by the percentages of water-soluble, exchangeable and carbonate-bound fractions (exchangeable and carbonate-bound fractions being obtained following Tessier's sequential extraction scheme; Tessier et al., 1979) in soils (Singh, Mohan, Singh, & Malik, 2005). RAC is also a reliable indicator to ecosystem health (Singh et al., 2005). Besides RAC, Zayed, Gowthaman, and Teryy (1998) emphasized on the translocation factor (TF) and bio-concentration factor (BCF) of metals to plant (definitions of TF and BCF are given in Sections 2.13.1 and 2.13.2 respectively) where plant grows on heavy metal contaminated soils.

India is the second largest tea growing country in the world and the North-East Indian state Assam is characterized by all the favorable conditions for tea (*Camellia sinensis* L.) plantation (Bhuyan et al., 2013; Karak & Bhagat, 2010; Karak et al., in press). The total tea cultivation area in this state is ~510,492 ha and the total levels of production and exportation of tea in January 2009 were 21.57 million kg and 12.70 million kg respectively (Tea statistics of India, 2009). Tea is one of the major cash crops in India as well as one of the major sources of foreign currency from agricultural products. Tea plant normally takes 10–11 years to attain the optimum bearing stage and is considered as the most productive up to an age of 30–35 years. However, the plant continues to be productive up to the age of 70 years. Thus, tea plant is a perennial one and grows in an undisturbed soil with soil pH ranging from 4.5 to 5.5 (Karak, Abollino, Bhattacharyya, Das, & Paul, 2011; Karak & Bhagat, 2010; Karak et al., 2014a). Application of easily available composts (e.g. mature cow dung compost and vermicompost) is of common practice in tea cultivation for sustainable nutrient supply. However, a burgeoning demand of compost and lack of raw materials opens up the prospect of using municipal solid waste (MSW) as a raw material for compost preparation. Preparation and use of MSWC solve not only the problem of management of MSW but also the disposal of MSW, as it is one of the most formidable challenges for its management (Karak, Bhagat, & Bhattacharyya, 2012). However, the criticism on MSWC application in tea cultivation is usually challenged due to the presence of HMs in MSWC and its high salinity that can create phytotoxic effects in the plants (Hargreaves et al., 2008). Despite the above opinion, there is little evidence of phytotoxic effects or accumulation of metals in tea plant that may pose a risk to human health on the application of MSWC to the soil (Smith, 2009). To the best of our knowledge, positive effect of MSWC on various types of soil and plants has been frequently reported in the literature (Bini, Maleci, & Romanin, 2008; Cervantes et al., 2001; Fozia, Muhammad, Muhammad, & Zafar, 2008; Hargreaves et al., 2008; Ražić & Đogo, 2010; Warman, Burnham, & Eaton, 2009; Zupančič, Justin, Bukovec, & Šelih, 2009 and the references therein). However, reports on the application of MSWC for tea cultivation are scanty. Likewise, literature on Cr accumulation within tea plant and tea infusion amended with MSWC is less extensive, as it has received little attention from scientists, particularly in the field of tea cultivation (Shanker et al., 2005). Hence, the profound objectives of this study are to investigate the effect of MSWC and in particular to ascertain (1) the distribution of Cr in different soil fractions in the tea growing soils, (2) the effects of Cr on tea production, (3) Cr distribution in different parts of tea plants, (4) Cr in tea infusion and (5) finally, the risk assessment analysis.

2. Materials and methods

2.1. Soil sample used in this experiment

Sixteen soil samples (0–15 cm depth) from different locations were collected under standing tea plant areas from Tocklai Experimental firm at Borbheta (26°45′N, 94°13′E), Assam, India. Samples were put in the field for natural drying. All the samples were mixed uniformly to produce a representative sample. A small quantity from the mixture was further dried naturally under a shade, sieved through 2 mm mesh and then was kept in an air tight plastic jar at room temperature.

2.2. Compost preparation

MSW samples were collected by crap sampling method (Elango, Thinakaran, Panneerselvam, & Sivanesan, 2009) and were segregated manually. Samples were collected during the summer as well as during the winter season in the year 2010. Segregated MSW samples were chopped by a cutting mill to 1–2 cm particle size to increase the reacting surface and to obtain better aeration and moisture control (Karak, Bhattacharyya, Paul, Das, & Saha, 2013; Singh, Kalamdhad, Ali, & Kazmi, 2009). Composting was performed in both summer and winter seasons in concrete pits with three replicates, following the methodology described by Benito, Masaguer, and Moliner (2003). Samples (about 4 kg) were taken from five symmetrical locations of each of the concrete pits at the end of the composting process (56th day). Compost samples were air-dried and were then passed through 1 mm sieve for analysis.

2.3. Phytotoxicity assay of prepared compost

Germination index (GI) known as phytotoxicity test was determined to judge the suitability of the prepared compost for field application. The details of the methodologies used for this test will be found in our recently published work (Karak, Bhattacharyya, & Paul, 2014b). Phytotoxicity test was done on the basis of GI of wheat (*Triticum aestivum* L.; cv. PBW 3) and Indian mustard (*Brassica campestris* L.; cv. Pusa Jaikisan) seeds. Germination index (GI) was determined as follows:

$$\mathrm{RSG}(\%) = \frac{\text{number of seeds germinated in soil amended with compost}}{\text{number of seeds germinated in soil without compost}} \times 100$$

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