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HEALTH RISK DUE TO ENRICHMENT OF CADMIUM, CHROMIUM, COPPER, NICKEL AND LEAD IN SOME COMMON MEDICINAL PLANTS USED IN AYURVEDIC AND HERBAL FORMULATIONS

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ABSTRACT

Medicinal plants are widely used in Ayurvedic and herbal treatment systems for their easy cultivation, wide availability and low cost. Heavy metals from agri- soil are bio-accumulated in the roots and above ground parts of the plants in varying concentrations. Several studies reported that medicinal plants enriched with one or more heavy metals beyond their safe limits, when used as raw materials in Ayurvedic and herbal formulations may induce health risk to the consumers. In view of this backdrop, present study was carried out to assess the enrichment levels of Cd, Cr, Cu, Ni and Pb in ten medicinal plants grown on garden soil. For comparison purpose, five medicinal plants collected from waste dump sites were also investigated. The consumer health risk in terms of metal enrichment factor (EF) and metal pollution index (MPI) were also evaluated. The levels of tested metals in all the 50 samples of ten medicinal plants collected from walled garden were below WHO acceptable limits; however, all 25 plant samples collected from waste dump sites recorded much higher levels of these metals. The EFs of tested metals in ten medicinal plants were Cd (0.26-0.58), Cr (0.37- 0.51), Cu (0.36-0.53), Ni (0.36-9.52) and Pb (0.44- 0.62). The MPI values due to tested metals computed for ten medicinal plants collected from walled garden were < 0.50, however, all the MPI for five plants collected from waste dump sites were < 1.0 and the values varied as: *Vasaka* (1.21) > *Parijat* (1.19) > *Papaya* (1.11) > Giloy (1.07) > *Curry leaf plant* (1.07mg/Kg).

KEY WORDS: Medicinal plants, Heavy metals, Enrichment Factors, Metal pollution index.

INTRODUCTION

Medicinal plants have contributed immensely to human health care and have registered notable contribution in the economy of the developed and developing countries, because of its being a potential field (CCR, 2018). This traditional system of herbal usage in management of disease has recorded a comparatively higher success rate than their synthetic counterpart (Patwardhan *et al*. 2004). Herbal based medicines are directly from nature and therefore much admired over allopathic medicines globally (Kulhari *et al*., 2013). Indian traditional medicines that commonly assumed to be safe and more affordable and avoid the adverse effects of synthetic counterparts (Kala *et al*., 2006). Ayurvedic and herbal formulations are composed of single or more medicinal herbs with a known combination, which are considered much safer way of treatment and also for strengthening immune system (Shankar *et al.,* 2017). Entire plant or its roots, leaves, bark, flowers, seeds etc along with some minerals such as sulphur, arsenic, lead, copper sulfate and metal bhasms (ashes) are used for treatment of depression, cardiovascular problems, etc. The effectiveness of the herbal treatment depends on the active principles of the medicinal plants present in formulation such as essential oils, vitamins, glycosides etc. Moreover, medicinal plants also served a source for the discovery of noval phyto-constituents with therapeutic properties for the pharmaceutical industries (Annan *et al*., 2010; Sarpong *et el.,* 2017). In the modern medicinal system, R & D Scientists derive intuition based ideas from phytochemicals that are derived from huge folk of varieties of wild or cultivated medicinal plants during the development, recognition and synthesis of '*lead compounds*' which finally may lead to the development of a evidence –based new Allopathic drug (Singh *et al*., 2011; Shah *et al*., 2013). As per WHO estimate, nearly 70–80% of the world population still primarily relies on nonconventional medications, mostly derived from herbal plants (WHO, 2002, 2004). The market of Ayurveda and Herbal medicines is one of the fast growing markets at global scenario and will continue to grow for long (Joshi *et al*., 2004). To maintain such a vibrant growth opportunity and to safeguard end users safety, the selection of high quality herbal raw materials and clean manufacturing protocol is to be ensured (Sdhu *et al.,* 2015).

Minerals are key factors for human health and body function and metals such as Fe, Co, Cr, Cu, Mo, Mn, Ni, Se, Zn etc. in very low concentration are necessary for human body. However, excessive levels beyond their safe limits can be damaging (Singh, 2005). Whereas mercury, arsenic, lead and cadmium are non-essential metals that can be toxic even in trace amounts (Singh *et al.*, 2011). Due to unique tendency of bio-accumulation heavy metals from agri- soil are deposited in the roots and above ground parts of the medicinal plants beyond the recommended safe limits recommended for human (Bakar and Bhattacharya, 2012, Barthwal *et al.*, 2008). Now, the accumulation of toxic metals in medicinal plants has become a major concern in traditional medicine. Prolonged exposure to As, Hg, Cd, Cr, Cu, Pb, Ni etc can cause deleterious health effects in humans and may disrupt metabolic functions of vital organs such as the heart, brain, kidneys, bone, liver, etc. (Singh *et al.,* 2011). Herbal raw materials are also contaminated during collection, drying, preservation, transportation and storing stages. Usually, selection of substandard raw materials, escaping of proper scientific identification, gradation and drying protocols of raw materials are other faults of the herbal product industry. In order to ensure optimum therapeutic benefits of herbal products, it is therefore important to monitor the quality of the raw materials with respect to their heavy metal contents (Sadhu *et al.*, 2015).

In view of the concern about heavy metal contamination of herbal raw materials, this study was undertaken to quantify the accumulated levels of heavy metals, Cd, Cr, Cu, Ni and Pb in ten therapeutically important medicinal plants commonly used as raw materials in various Ayruvedic and herbal products, to assess the consumer health risk in terms of the metal enrichment factors (EF) and metal pollution index (MPI). Medicinal plants selected for this study were, *Ocimum sanctum L*, *Mentha arvensis*, *Withania sonmifera, Andrographis Paniculata, Boerhavia diffusa*, *Tinospora cordifolia, Carica papaya*, *Adhatoda vasica, Nyctanthes arbor-tristis, and Murraya koenigii*. These plants were collected from the walled garden where the reach of waste water and street runoff is restricted. The garden soil samples were also analysed for levels of tested metals. For comparison purpose, samples of *Tinospora cordifolia, Carica papaya, Adhatoda vasica,* Nyctanthes *arbor-tristis, and Murraya koenigi were* also collected from waste dump sites. The metal enrichment factors (EF) and metal pollution index (MPI) were evaluated.

MATERIALS AND METHODS

Selection of plant species

For this study ten therapeutically important medicinal herbs and plants were selected in order to evaluate their metal enrichment potentials for Cd, Cr, Cu, Ni and Pb. The vernacular and scientific names, family and important medicinal uses of the plants are presented in the Table 1.

Sampling and pre-treatment of soil samples

Garden soil samples were collected during June 2017 from five different locations at nearly 10 cm depth of the walled garden. The samples were air dried in sunlight at room temperature (39–40 °C) for 4-5 days then oven- dried (80 ^{0}C) for 24 hours, finally grinded, thoroughly mixed and stored separately in labeled bottles for heavy metals analysis.

Sampling and pre-treatment of medicinal plants samples

For heavy metal enrichment study, 50 samples, five each of the matured 10 medicinal plants were collected randomly from five locations of the walled garden during August to October 2017. Here, only rain water from open sky and water supplied by government agency were used for irrigation and no street run off reaches there. Additionally, for comparison purpose, five samples each of *Giloy, Papaya*, *Basaka, Parijat* and *Curry leaf plants,* totaling 25 were also collected from waste dump sites of Nalapani Rao area, Dehradun, India. Samples were washed thoroughly under the tap water 2-3 times to

Table 1. Description and Applications of Medicinal plants selected for study

S.N.	Vernacular Name	Scientific Name, Family	Important Medicinal Applications
1.	Tulsi	*Ocimum sanctum L* Lamiaceae	Has adaptogen, boosts immune, lowers stress, anxiety, nausea, vomiting, eczema, insect bite.
2.	Mint (Pudina)	*Mentha arvensis*, Lamiaceae	Rich in antioxidants, phyto-nutrients, helps in acidity, boost digestion, manages blood sugar.
3.	Aswagandha	*Withania sonmifera* Solanaceae	Balances Cholesterol, helps kidney, liver, blood sugar level, reduce appetite and fever.
4	Kalmegh (Kirata)	*Andrographis Paniculata,* Acanthaceae	Root is diuretic, anti-helminthic, antioxidant, anti-depressant, anti-diabetic, immune booster.
5.	Punarnava	*Boerhavia diffusa* Nyctaginaceae	Antipyretic, anti-inflammatory, antibacterial, antioxidant, anti-parasitic, hepato-protective.
6.	Giloy (Amrita)	*Tinospora cordifolia*, Menispermaceae	Miracle herb, supports kidney, heart, RBC formation, helps in kidney stones and uric acid.
7.	Papaya	*Carica papaya, Caricaceae*	Immunity booster, anti-ageing, anxiety high antioxidants, detoxifier, cures dengue, malaria.
8.	Vasaka (Basinga)	*Adhatoda vasica* (Acanthaceae)	Anti-ageing supports kidney, heart, liver, diabetes; leaf extracts boosts platelet.
9.	Parijat (Har singar)	*Nyctanthes arbor-tristis*, Oleaceae	Used against arthritis, sciatica, cough, anxiety, fever, malaria, diarrhea, diabetes, worms.
10.	Curry leaf plant	*Murraya koenigii,* Rutaceae	Rich in aroma, antioxidants, maintains sugar level, immune booster and anti-inflammatory.

remove the adhered soil and dirt and finally cleaned with distilled water and air dried at room temperature (30- 32 ^{0}C) by placing over clean filter paper sheets in dust free area for 4- 5 days. All the plant samples were oven -dried (70 ^{0}C) for 48 hours, grinded and stored separately in air tight labeled bottles for analysis of metals under study.

Digestion of soil and medicinal plant samples

To determine the accumulated metals in medicinal plants, grinded samples (1.0g) were subjected to HNO_3 – $HClO_4$ (5:1, v/v) digestion at nearly 80 ^{0}C for several hours following the standard methods (APHA, 2005) to get a transparent light colored liquid. The liquid was then filtered in 100 mL volumetric flasks and filled up to the mark with double distilled water.

For estimation of metal concentration in garden soil samples, 1.0 g of the finely grinded soil samples were digested with HNO_3 –HCl–$HClO_4$ mixture (5:1:1) for several hours to get transparent extract which was filtered and diluted to a volume of 100 mL with double distilled water (Jones J. Benton Jr, 2001).

Estimation of metals in soil and plant samples

The accumulated concentration (mg/kg, dw) of cadmium, chromium, copper, nickel and lead in the samples of soil and medicinal plants were determined on Atomic Absorption Spectrometer, Analyst 200 (Perkin Elmer), using air-acetylene flame, in accordance to standard methods (APHA, 2005; Jones J. Benton Jr, 2001). The standard stock solutions of metal ions used were from *Sigma Aldrich*, which were diluted to required concentrations for preparing working standards. Other reagents, chemicals, and solvents used were of analytical grade (Kulshrestha *et al*, 2012). Double distilled water was used for all purposes. All the estimations were run in triplicate.

RESULTS AND DISCUSSION

The observed concentrations of Cd, Cr, Cu, Ni and Pb in garden soil samples as mg/Kg, d.w. of soil and the accumulated metals as mg/ Kg, dw of selected medicinal plant samples collected from walled garden are presented in Table 2. The levels of metals in medicinal plant samples collected from waste dump site are given in parenthesis. The metal concentrations recorded in soil and vegetable samples under study were compared with the recommended safe limits.

Heavy Metals in Garden Soil

Metals present in soil interact differently in soil solutions and can accumulate in soil system. The bioavailability and toxicity of metals in soil are significantly influenced by pH condition (Badawy *et al*., 2002; Wang *et al*., 2006). Due to their long-term interactions, accumulation of heavy metals in soil poses a threat to animals and plants (Chang *et al.*, 2014). The observed order of Cd, Cr, Cu, Ni and Pb levels in garden soil under study was: Pb (1.87±0.103) > Cr (0.76±0.034) > Cu (0.47±0.020) > Ni (0.44±0.023) > Cd (0.31±0.016 mg/Kg). All these values were below safe limits set for these metals in soil samples (Awasthi, 2001, WHO/FAO, 2007).

Heavy Metals in Medicinal Plants

Because of the widespread presence of heavy metals in the environment, their residues also reach and are assimilated into medicinal plants (Sarma *et al.*, 2011). The cell wall of plant root system absorbs and translocates heavy metals with varying concentration in the roots, stem and other aerial parts of the plant through xylem, whereas foliar transport involves the phloem vascular system (Prabhat *et al*., 2019). Water loss due to transpiration as a result of evaporation of water through leaf stomata, serves as a pump to absorb micronutrients and contaminants including heavy metals into the plant shoots (Chang *et al.*, 2014). The accumulation of heavy metals in plants depends on the degree of uptake of these elements by plants from the soil (Sadhu *et al.*, 2015). The heavy metals accumulated in various parts of the medicinal plants may enter consumer system via intake of herbal products. Therefore, heavy metal contaminations of herbal products are of great concern and for optimal therapeutic benefits of herbal products, regular monitoring of heavy metals contents in herbal raw materials is essential (Sadhu *et al.*, 2015). Due to the concern associated with bio-enrichment of heavy metals, WHO has developed a series of technical guidelines for medicinal plants used as raw materials for production of herbal products (FAO/WHO 1984, 1998, 2007, 2011).

The observed results (Table 2 and Fig. 1, 2) showed that the studied metals were present in varied concentrations in the tested medicinal plants. The levels of Cd, Cr, Cu, Ni and Pb in all the medicinal plant samples collected from walled garden were below WHO acceptable limit (WHO, 2005,1998) and varied from 00.08±0.010 to 0.21±0.016, 0.26 ±0.016 to 0.39±0.016, 0.17±0.014 to 0.25±0.018, 0.16±0.010 to 0.23±0.018 and 0.83±0.034 to 1.17±0.034 mg/Kg, respectively. However, the levels of these metals in medicinal plants collected from waste dump sites varied from 0.43±0.031 to 0.66±0.038, 1.41±0.043 to 1.72±0.049, 0.81±0.033 to 0.98± 0.039, 0.78 ±0.033 to 1.06±0.051 and 2.46±0.051 to 2.81±0.061 mg/Kg, respectively.

The wide variations recorded in accumulated

Table 2. Concentration (mg/Kg, dw) of metals in garden soil and medicinal plants collected from walled garden and from waste dump sites

Metals →	Cadmium	Chromium	Copper	Nickel	Lead
Soil (n = 5) →	0.31±0.016	0.76±0.034	0.47±0.023	0.44±0.023	1.87±0.103
Safe limit, soil [a]	3.0-6.0	–	135-270	75-150	250-500
Plants (n =5) ↓					
Tulsi	0.11±0.010	0.28±0.013	0.17±0.014	0.17±0.014	0.96±0.028
Mint	0.09± 0.008	0.30±0.016	0.19±0.016	0.18±0. 016	1.06±0.025
Aswagandha	0.10±0.014	0.29±0.016	0.21±0.016	0.19±0.014	0.83±0.034
Kalmegh	0.08±0.010	0.26±0.016	0.20±0.014	0.16±0.010	0.98±0.034
Punarnava	0.10±0.010	0.31±0.014	0.21±0.016	0.17±0.014	1.02±0.032
Giloy	0.12±0.010	0.33±0.018	0.18±0.016	0.15±0.014	0.87±0.024
	(0.43±0.031)[b]	(1.52±0.049)[b]	(0.98±0.039)[b]	(0.84±0.038)[b]	(2.64±0.061)[b]
Papaya	0.11±0.010	0.32±0.016	0.20±0.014	0.19±0.014	0.91±0.010
	(0.51±0.033)[b]	(1.41±0.043)[b]	(0.86±0.035)[b]	(0.98±0.039)[b]	(2.76±0.066)[b]
Vasaka	0.18±0.016	0.36±0.016	0.25±0.018	0.23±0.018	1.17±0.034
	(0.66±0.038)[b]	(1.58±0.045)[b]	(0.87±0.038)[b]	(1.06±0.051)[b]	(2.67±0.056)[b]
Parijat	0.21±0.016	0.39±0.016	0.21±0.016	0.21±0.016	0.94±0.035
	(0.58±0.036)[b]	(1.72±0.049)[b]	(0.95±0.033)[b]	(0.89±0.035)[b]	(2.81±0.061)[b]
Curry leaf Plant	0.12±0.014	0.31±0.016	0.19±0.014	0.18±0.014	1.16±0.035
	(0.50±0.028)[b]	(1.48±0.044)[b]	(0.81±0.033)[b]	(0.78±0.033)[b]	(2.46±0.051)[b]
Acceptable limits (medicinal herbs)	0.3 mg/kg (WHO, 2005)	1.5 mg/kg (WHO, 1998)	10 mg/kg (WHO, 2005)	1.5 mg/kg (WHO, 2005)	10 mg/·kg (WHO, 2006)

[a] (Awasthi, 2001, WHO/FAO, 2007).
[b] Values in parentheses are metal levels in plant samples collected from waste dump sites.

metal levels in the studied medicinal plants may be due to differences in the metal uptake and translocation potentials of plants (Chang *et al.* 2014). WHO acceptable range of Cd in herbal raw materials is 0.3 mg/kg (WHO, 2005). The high level of cadmium exerts adverse effect on liver, kidney and immune system (Dghaim *et al.*, 2012). Cadmium in all the 50 samples collected from walled garden were found below acceptable limit (WHO, 200) the lowest Cd level (0.08±0.010 mg/Kg) was found in *Kalmegh* and the highest (0.21±0.016 mg/Kg) was in *Parijat*. While, Cd levels in all the samples collected from waste dump sites were much higher than WHO limit and the order was: *Vasaka* (0.66±0.038) > *Parijat* (0.58±0.036)>*Papaya* (0.51±0.033) >*Curry leaf plant* (0.50±0.028) > *Giloy* (0.43±0.031mg/Kg).

Trivalent chromium is essential nutrient needed in very small amount by human body (Dghaim *et al.*, 2012) while hexavalent chromium is toxic to human and may cause even cancer (Kabata and Pendias, 1993). However, its elevated levels in herbal raw materials and other crops may be hazardous to the consumers (Bakar and Bhattacherjy, 2012). The acceptable limit of Cr set by WHO was 1.5 ppm for herbal raw materials (WHO, 2007). Out of the 75 tested samples, only 20 % samples (collected from waste dump sites) recorded higher levels of Cr than the WHO acceptable limit. However, all the 25 samples collected from the waste dump sites were much higher (1.41±0.043 to 1.72±0.049 mg/Kg), than in samples collected from walled garden (0.26±0.016 to 0.39±0.016 mg/Kg). Copper is important constituent of several enzymes and an essential trace element for metabolic processes and needed for normal growth and development.

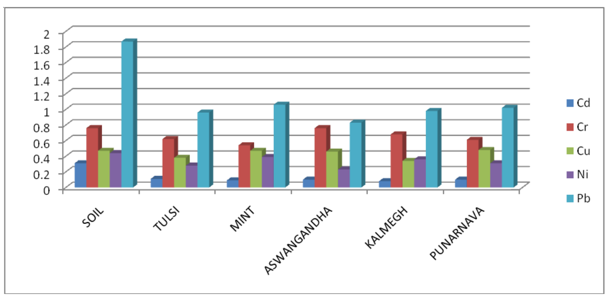

Fig. 1. Levels of Cd, Cr, Cu, Ni and Pb in Garden Soil and Medicinal Plants collected from walled garden

But its high intake may result anemia, arthritis, depression, hypertension, diabetes, dermatitis, kidney and liver problems (Kabata and Pendias, 1993, Ulla *et al*., 2012). The level of Cu in all 25 samples collected from waste dump sites were much higher (0.81±0.033 to 0.98±0.039 mg/Kg) than in samples collected from walled garden (0.17±0.014 to 0.25±0.018 mg/Kg). Nickel is necessary for formation of red blood cells and the regulation of lipid contents in tissues, yet at high level, Ni

Table 3. Enrichment factors (EFs) of Cd, Cr, Cu and Pb in Medicinal Plants collected from walled garden.

Medicinal plants	Cd	Cr	Cu	Ni	Pb
Tulsi	0. 35	0.37	0. 36	0.39	0.51
Mint	0.29	0.39	0.40	0.41	0.57
Aswagandha	0.32	0.38	0. 45	0.43	0.44
Kalmegh	0.26	0.34	0.42	0.36	0.52
Punarnava	0.32	0.41	0.45	0.39	0.54
Giloy	0.39	0.43	0.38	0.34	0.46
Papaya	0.35	0.42	0.42	0.43	0.49
Vasaka	0.58	0.47	0.53	0.52	0.62
Parijat	0.51	0.51	0.45	0.48	0.50
Curry leaf Plant	0.39	0.41	0.40	0.41	0.62

Table 4. Metal pollution index (MPI) due to Cd, Cr, Cu, Ni and Pb in Medicinal Plants.

S. N.	Medicinal Plants (Collected from walled Garden)	MPI (mg/Kg)	SN	Medicinal Plants (Collected from)	MPI (mg/Kg)	
					Walled Garden	Waste dump Sites
1.	Tulsi	0.24	6	Giloy	0.25	1.07
2.	Mint	0.25	7	Papaya	0.26	1.11
3.	Aswagandha	0.25	8	Vasaka	0.34	1.21
4.	Kalmegh	0.23	9	Parijat	0.32	1.19
5	Punarnava	0.26	10	Curry leaf Plant	0.27	1.03

adversely affects vision, lungs, heart and liver (Bakar and Bhattacherjy, 2012, McGrath and Smith, 1990). The WHO permissible limit of nickel in medicinal plant is 1.5mg/kg (WHO, 2005). The recorded Ni levels (Table 2, Fig. 2) in all 75 samples were much below the permissible limits. However, in 25 samples of five medicinal plants collected from dump sites, Ni levels were much higher (0.78 ±0.033 to 1.06±0.051 mg/Kg) than all 50 samples collected from walled garden that varied from 0.16±0.010 to 0.23±0.018 mg/Kg. Lead is a non essential heavy metal and when deposited in the bones and teeth, makes these brittle and weak (Shah *et al*., 2013; Bakar and Bhattacherjy, 2012). Lead causes hazardous effect on the nervous system, kidney, heart, brain, immune system etc (Dghaim *et al.*, 2012). The FAO/WHO maximum permissible limit set for lead in medicinal herbs was 10 mg/Kg (WHO 2007; Dghaim *et al.*, 2012). The recorded levels of Pb in all the 75 samples were below WHO permissible limit, however Pb levels in all 25 plant samples collected from waste dump sites were much higher (2.46±0.051 to 2.81±0.061 mg/ Kg) than in 50 samples collected from walled garden (0.83±0.034 to 1.17±0.034 mg/Kg). The levels of heavy metals varied from site to site, plant to plant and also in different parts of each plant, depending upon the chemical composition of soil and absorption potential of plants (Shah *et al*., 2013). The results of this study are comparable with earlier findings (Barthwal *et al.*, 2008; Dghaim *et al.*, 2012; Bakar and Bhattacherjy, 2012; Sarpong Kofi *et al.*, 2017).

Metal Enrichment Factor

The degree of metal enrichment in plants can be described with *enrichment factor* (EF), which is defined as the ratio of particular element content in a plant to that in soil (Chang *et al.* 2014). The metal uptake by plants is normally depends on bio-availability of the concerned metal ions in the soil-

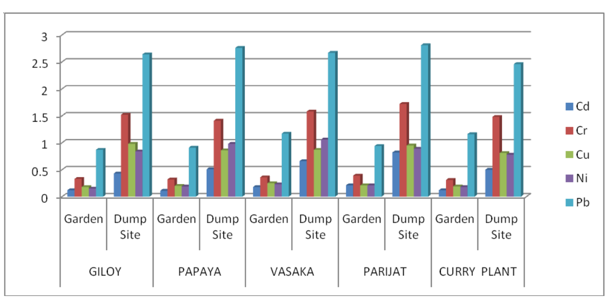

Fig. 2. Levels of Cd, Cr, Cu, Ni and Pb in Medicinal Plants collected from walled garden and waste dump sites

water system, soil pH, organic matter, plant species, plant age etc. (Sanghamitra *et al.*, 2012). Although the enrichment factor (EF) was initially proposed to quantify the soil–plant transfer of heavy metals, other terms, such as Bio-concentration factor (BCF) and the plant uptake factor, are also widely used (Rai *et al.*, 2019). Bio-accumulation of heavy metals in roots and above ground parts of the plants varies greatly that generally depends on the plant species, levels of metals in soil and characteristics of the study area (Barthwal *et al.*, 2008). The accumulation and magnification of heavy metals in human organs through consumption of herbal remedies can cause hazardous impacts on health (Kulhari *et al.*, 2013). The enrichment factor was evaluated the ratio of heavy metal concentration in vegetables (mg/kg) and in the soil (mg/kg) by using following relation (1):

EF = [Metals] plants D [Metals] soil (Cui *et al*. 2004, Yoon *et al*., 2006) .. (1)

The computed values of bio-enrichment levels of different metals in tested medicinal plants are presented in Table 3. The wide variations in heavy metals enrichment factors among studied medicinal plants might be due to the variation in the labels, bioavailability of metals in the soil and metal uptake potential of the plant (Cui *et al.* 2004). The overall order of enrichment factor (Table.3) was, Pb > Cu > Ni > Cr > Cd. However the individual enrichment order of Cd in studied plants was: *Vasaka* (0.58) > *Parijat* (0.51) > *Gilow* (0.39) > *Curry leaf Plant* (0.39) >*Tulsi* (0.35) > *Papaya* (0.35) >*Aswagandha* and *Punarnava* (0.32) > *Mint*(0.29).

The metal enrichment order of Cr was: *Parijat* (0.51) >*Vasaka* (0.47) >*Gilow* (0.43)> *Papaya* (0.42) *Punarnava* and *Curry leaf plant* (0.41) >*Mint* (0.39) > *Aswagandha* (0.38) > *Tulsi* (0.37) > *Kalmegh* (0.34). The order of EFs for Cu was: *Vasaka* (0.53) > *Aswagandha, Punarnava* and *Parijat* (0.45) > *Kalmegh* and *Papaya* (0.42) > *Mint and Curry leaf plant* (0.40) > *Giloy* (0.38) > *Tulsi* (0.36). The metal enrichment order of Ni was: *Vasaka* (0.52) > *Parijat* (0.48) > *Aswagandha and Papaya* (0.43) > *Mint* and *Curry leaf plant* (0.41) >Tulsi and *Punarnava* (0.39) > Kalmegh (0.36) > Giloy (0.34). The highest enrichment of Pb was in *Vasaka* and *Curry leaf plant* (0.62) , which was followed by *Mint* (0.57), *Punarnava* (0.54), *Kalmegh* (0.52), *Tulsi* (0.51), *Parijat* (0.50), *Papaya* (0.49), *Giloy* (0.46) and *Aswagandha* (0.44). The TF value beyond 0.50 indicates the elevated level of metal in the concerned medicinal plant being used as herbal raw material

(Zhang and Liu, 2002; Hussain *et al.,* 2019).

Metal Pollution Index (MPI)

Metal pollution index (MPI) signifies the overall level of metals in edible parts of a vegetable and calculated as the geometric mean of concentration of tested metals accumulated in edible parts of vegetable (Usero *et al.* 1997). The values of MPI, as mg/kg were obtained by putting the metal concentrations found in vegetables, in equation (2) mentioned earlier and the results are presented in the Table 4.

MPI (mg/ Kg) = $(Cf_1 \times Cf_2 \times Cf_3 \times Cf_4 \ldots\ldots Cf_n)^{1/n}$ …………………(2), where, Cf_n is the concentration of nth heavy metal in edible parts of vegetable (medicinal plant).

MPI values signify the overall metal levels in a vegetable and the possible quantum of risk to the consumers (Usero *et al*., 1997). The MPI score up to 1is termed medium, 1 to 2- moderate, 3- strong, while >3 strong, >4 very strong metal pollution (Rashed, 2010). All the MPI values due to enrichment of Cd, Cr, Cu, Ni and Pb in 10 medicinal plants collected from walled garden were < 0.50, however, all the MPI values computed for five plants collected from waste dump sites were > 1.0 and varied as: *Vasaka* (1.21) > *Parijat* (1.19) > *Papaya* (1.11) > Giloy (1.07) > Curry leaf Plant (1.07mg/Kg). All these MPI values fall under the '*moderate heavy metal pollution*' (Rashed *et al.,* 2010). Uptake of heavy metals by plants and further accumulation along the food chain is a potential threat to animal and human health (Luo *et al*., 2021). Thus, in the context of heavy metal contamination the medicinal herbs used as the raw materials need intense chemical examination before processing for further pharmaceutical purposes or for local human consumption (Sharma *et al*., 2009; Shah *et al*., 2013). WHO has emphasized the need to ensure the quality of medicinal plants as raw materials and developed a series of technical guidelines relating to the quality control of herbal medicines and guidelines on good agricultural and collection practices (GACP) for medicinal plants (WHO, 2003; AHPA, 2006).

CONCLUSION

In India the Ayurvedic and herbal medicines based sector is one of the most vibrant and fast growing fields that has unlimited direct or indirect employment generation potential. However, during cultivation, identification, collection, gradation, drying, transportation and processing stages of medicinal plants used as raw materials, quality control protocols are not followed. This study assessed enrichment levels Cd, Cr, Cu Ni and Pb in 50 samples of ten medicinal plants collected from walled garden and 25 samples of five medicinal plants were also collected from waste dump sites and their health risk in terms of MPI. The findings of this study revealed that all the samples of five medicinal plants collected from waste dump sites accumulated elevated levels of tested heavy metals and all the MPI values due to Cd, Cr, Cu, Ni and Pb for these plants were >1.0 which is '*moderate category*' of heavy metal pollution. The MPI values varied as: *Vasaka* (1.21) > *Parijat* (1.19) > *Papaya* (1.11) > Giloy (1.07) > Curry leaf Plant (1.07mg/Kg). On the other hand, the MPI values were < 0.50 (*safe category*) for all the samples of ten medicinal plants collected from walled garden. These results suggested that in order to get optimum benefits of Ayurvedic and

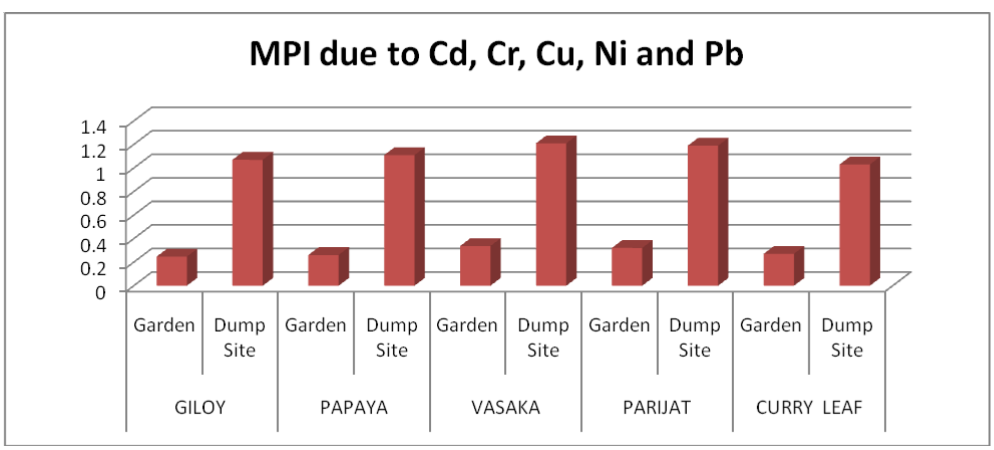

Fig. 3. Metal Pollution Index due to presence of Cd, Cr, Cu, Ni and Pb in medicinal plants collected from walled garden and waste dump sites.

herbal products, WHO technical guidelines relating to the quality control and on good agricultural and collection practices (GACP) for medicinal plants, must be followed to maintain heavy metal levels below the permissible limits in the finished herbal products. Further, during collection of herbal raw materials the protection of medicinal plants as the prime natural resources for their sustainable use should be the priority of all stake holders and end users.

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