



Profiling of heavy metal and pesticide residues in medicinal plants

Narendra Kumar¹ · Mahiya Kulsoom¹ · Vertika Shukla¹ · Dhananjay Kumar¹ · Priyanka¹ · Sanjeev Kumar² · Jaya Tiwari³ · Neetu Dwivedi⁴

Received: 12 January 2018 / Accepted: 16 August 2018 / Published online: 22 August 2018
© Springer-Verlag GmbH Germany, part of Springer Nature 2018

Abstract

Application of medicinal plant to cure ailments has been practiced by several civilizations. Nowadays, contamination of heavy metals and pesticide residues in medicinal plant is a serious concern, due to toxic effects on human health. The present study was designed with an aim to quantify the heavy metals and pesticide residues in the 20 medicinal herbs, frequently sold in the local market as raw material without any quality assurance. The concentrations of the elements are as follows: copper (2.42–19.14 $\mu\text{g g}^{-1}$), cadmium (0.01–2.10 $\mu\text{g g}^{-1}$), chromium (17.63–58.63 $\mu\text{g g}^{-1}$), iron (7.61–322.6 $\mu\text{g g}^{-1}$), and lead (13.00–54.47 $\mu\text{g g}^{-1}$), whereas total metal concentration ranged between 44.73 and 385.15 $\mu\text{g g}^{-1}$. Among the organic pesticides, HCH (1.63–6.44 $\mu\text{g g}^{-1}$) and DDT (0.63–7.14 $\mu\text{g g}^{-1}$) isomers were found to be present in medicinal plant material. Result showed that lead and chromium concentrations in the herbs were above the permissible limits set by WHO. These herbs should be regularly checked for quality assurance before using raw or as a herbal formulation to avoid chronic exposure of metal and pesticides to human being.

Keywords Bioaccumulation · DDT · Metal · Medicinal plants · Pesticide residues

Introduction

Use of herbal medicine is a traditional way to cure ailments and has been applied for more than five millennia in several civilizations (Petrovska 2012). Even today, plant material continues to play a major role in primary health care as therapeutic agents in many developing countries. For example, *Ocimum sanctum* and *Azadirachta indica* are known since ages to cure diseases (Dimitrova 1999; Wiart 2006; Petrovska 2012). Besides using herbal medicines directly to treat many diseases, natural products have contributed enormously to the development of important therapeutic drugs used currently

in modern medicine (Govindaraghvan and Nikolaus 2015). It is estimated that about 25% of all modern medicines are directly or indirectly derived from higher plants (Bodeker et al. 2005; Ziarati 2012). Rapid industrialization and unorganized urbanization may cause accumulation of toxic substances such as metals and pesticides in soil, water, and air (Kishan et al. 2014; Kumar et al. 2015; Rodriguesa et al. 2017). Heavy metals can enter the body either through food, air, or water and bio-accumulate over a period of time on chronic exposure (Srogi et al. 2002; Duruibe et al. 2007; Neha et al. 2017; Kumar et al. 2018). The relative toxicity of heavy metals to living beings follows the following pattern: $\text{Hg} > \text{Cu} > \text{Zn} > \text{Ni} > \text{Pb} > \text{Cd} > \text{Cr} > \text{Sn} > \text{Fe} > \text{Mn} > \text{Al}$ (Anna et al. 2015). Plants are sessile and rely on their environmental conditions for growth and development. They accumulate heavy metals in their harvestable parts via root uptake, foliar adsorption and decomposition of specific elements (Kishan et al. 2014). Contamination and accumulation of toxic elements in herbal medicines depends on factors like species, harvesting time, level and duration of contaminant exposure, cultivation, processing, topography, geographical origin, and storage of the plant material (Arpadjan et al. 2008). Heavy metals enter into food chain through their accumulation in plants where they find place through uptake and transport via transporters of essential elements according to their structural analogy (Chauhan et al. 2017). The uptake of contaminants in plants

Responsible editor: Philippe Garrigues

✉ Narendra Kumar
narendrakumar_lko@yahoo.co.in

- ¹ Department of Environmental Science, Babasaheb Bhimrao Ambedkar University, Lucknow 226025, India
- ² Center for Environmental Sciences, School of Earth Sciences, Central University of Jharkhand, Brambe, Ranchi, Jharkhand 8352 05, India
- ³ Department of Environmental Science, Asian Institute of Public Health, Bhubaneswar, Odisha, India
- ⁴ Department of Environmental Science, Sri Aurobindo College, University of Delhi, New Delhi 110017, India

occurs through the root system, which provides a wide surface area for the absorption (Ouyang 2002). The transfer of bound ions from the extracellular space to the hydrophobic environment of the membrane into the cell is facilitated by the transmembrane structures. Translocated metals become complexed and sequestered in cellular structures of shoots (Lasat et al. 1998). Translocation of metal-containing sap occurs from the root to the shoot in a process that is mainly controlled by root pressure and leaf transpiration (Khatoon et al. 2017). However, pathways involved in the transfer of POPs from air to plants involve gaseous exchange, particle-bound deposition, and wet deposition (McLachlan and Horstmann 1998). Conditions such as hydrophobicity, water solubility, and vapor pressure govern penetration of POPs into the vegetation. In addition, environmental characteristics, such as temperature, organic content of the soil and plant species, also influence the pollutant uptake mechanism (Khatoon et al. 2017).

The possibility of transmission of toxic heavy metals to humans and animals through the application of herbs grown in polluted area is a major concern. To get desirable therapeutic benefits, quality of the raw herbs should be ensured in terms of metal contamination. WHO (2007) advocates that herbs and herbal products should not be used without qualitative and quantitative analysis of their heavy metal contamination (Kishan et al. 2014). Physicochemical properties, safety, and efficiency of the herbal commercial products can be augmented by following good agriculture and collection practice (GACP), good manufacturing practice (GMP) before and during the manufacturing processes, and good laboratory practice (GLP) (Govindaraghavan and Nikolaus 2015).

Apart from metals, herbal drugs may also contain pesticide remainders, through faulty agricultural practices, such as spraying, handling of soils during farming, and administering fumigants during storage (Kunle et al. 2012; Mishra et al. 2007). Pesticides are used in agriculture to protect plants from insects, pests, and pathogens and to improve the quality and amount of harvest, but the accumulation of pesticide residues in edible parts of plants may cause toxic and allergenic effects on human health (Srivastava et al. 2006a, b). The prime adverse effects associated with the chronic exposure to organic pollutants are nervous disorders, including headache, dizziness, tremor, discoordination, and/or convulsions (Shaban et al. 2016). The maximum residue limits (MRLs) for pesticides have been set in the Commission Regulation and the Pesticide database of the European Union (Reinholds et al. 2017). There are quite meager evidence available on the quality and safety of medicinal herbs and their products sold in the local market. This study aims to determine the level of heavy metal and pesticides contamination in some frequently used herbs.

Material and methods

Sample collection Twenty raw medicinal plant materials (Table 1) were collected from local market of Lucknow, U.P., India. Plant materials were washed with tap water followed by distilled water and after drying ground manually and subjected to analysis.

Metal analysis The samples were digested in solution of nitric acid and perchloric acid mixture (1:3; v/v). One gram accurately weighed ground sample and 20 ml of aquaregia solution was placed into a 50-ml conical flask. The samples were heated on a hot plate at 70–80 °C until a clear solution was obtained. 0.1 N nitric acid was prepared for volume make-up of the digested sample. The metals content were analyzed on an atomic absorption spectrophotometer (AA 240 FS, Varian).

Pesticides estimation Ten grams of the sample was suspended in 50 ml of petroleum ether-acetone mixture (1/1 v/v) and shaken on an Excella E24 incubator shaker series for 3 h. The extract was filtered and concentrated to exactly 1 ml by using rotatory evaporator IKA RV 10 and nitrogen stream respectively. The samples were dissolved in a mixture of acetone and *n*-hexane (1:1; v/v). Column cleanup was done with anhydrous sodium sulfate and Florisil (activated magnesium silicate). 50 ml of eluting solvent (*n*-hexane: ethyl acetate: dichloromethane in the ratio of 70:15:15) was used for each sample. The samples were run on GC (Agilent 7890 A) equipped with ECD detector.

To control analytical quality, reagent blanks and sample replicates were used during the analysis to assess contamination and accuracy. Stock standards were used from Merck, India, traceable to the National Institute of Standards Technology (NIST) to establish calibration curves. Recovery studies of metal determination were performed using CRMs to display the efficiency of the methods used. The recovery rates were ranged between 86 and 103%.

Result and discussion

Range and mean concentration of the five metals viz. cadmium, chromium, copper, iron, and lead analyzed in 20 medicinal plants has been summarized in Table 2. The total metal concentration in selected plant material ranged between 44.73 and 385.15 $\mu\text{g g}^{-1}$. In most of the samples, the metal concentration was found to be beyond the WHO (2007) standards. Results revealed the expected influence of ambient environmental conditions (soil and air) on the total metal profile as the lower level of concentration of investigated metals was found in herbs growing in relatively clean regions than in those growing in polluted areas. The concentration of Pb and Cr in various medicinal herbs ranged between 13.26–54.47 and 17.63–58.63 $\mu\text{g g}^{-1}$ which was far above the standards prescribed

Table 1 Medicinal plants investigated for metal and pesticides contamination and their medicinal uses (Rastogi and Mehotra 1991, 1993)

S. No.	Botanical name	Medicinal uses
1	<i>Allium sativum</i> L.	Antibacterial, anti-viral, and anti-oxidant lowering blood pressure
2	<i>Aloe vera</i> L.	Skin treatment, diabetes, osteoarthritis, burns, stomach ulcers, diabetes
3	<i>Asparagus racemosus</i> L.	Anti-ulcerogenic, bronchitis, tuberculosis, diabetes, indigestion, antiseptic,
4	<i>Azadirachta indica</i> L.	Antiseptic, antipyretic, antidiabetic, antibacterial, detoxify the blood, and balance blood sugar levels, dental diseases, skin diseases, fevers, cough etc.
5	<i>Bacopa monnieri</i> L.	Improve memory capacity, intellectual activity and enhance the immune function by increasing immunoglobulin production etc.
6	<i>Calotropis gigantean</i> L.	Antivenom, rheumatic and stomach disorders, pain killer etc.
7	<i>Cannabis sativa</i> L.	Asthma, cystitis, AIDS, Dysentery, gonorrhea, gout, epilepsy, malaria, fevers, anti-inflammatory etc.
8	<i>Catharanthus roseus</i> L.	Leukemia, muscle pain, depression of the central nervous system
9	<i>Curcuma longa</i> L.	Cancer, Alzheimer's disease, diabetes, allergies. Boost immunity, anti-inflammatory and anti-oxidant, fever, anemia, skin, hypertension
10	<i>Datura stramonium</i> L.	Asthma, analgesic during surgery or bone setting baldness, malaria.
11	<i>Eclipta prostrate</i> L.	Hepatitis, anti-inflammatory, eye infections, cirrhosis, stress, tension
12	<i>Hibiscus sinensis</i> L.	Lowers hypertension, analgesic, nausea, ulcers and bladder infection
13	<i>Lantana camara</i> L.	Antiseptic, cancers, chicken pox, measles, asthma, ulcers, swellings
14	<i>Ocimum tenuiflorum</i> L.	Healing, fever and common cold, coughs, sore throat, respiratory disorder, kidney stone, stress, headaches, etc.
15	<i>Rauwolfia serpentine</i> L.	Hypertension, nervous system, treating cancer, anti-tumor
16	<i>Rosa rubiginosa</i> L.	Dysentery, diarrhea and gastro enteritis, constipation and urinary problem
17	<i>Solanum nigrum</i> L.	Liver diseases, diuretic, antispasmodics, vasodilator, antipyretic, etc.
18	<i>Tagetes patula</i> L.	Leukemia and melanoma cancer cells, menstrual problems, eye infections, inflammations, and for wound healing, etc.
19	<i>Tinospora cordifolia</i> L.	Increase platelet count, used in chronic fever, gout, vomiting, immunity disorders, stomach ulcer, liver protection
20	<i>Withania somnifera</i> L.	Diuretic, sedative, astringent, arthritis, memory, stabilizes blood sugar, stress reduction, anxiety treatment, depression and mood, relaxation.

by WHO, i.e., 10 and 2 µg g⁻¹ respectively. The concentration of Pb and Cr found in medicinal plants warns about the risk associated with the ingestion of contaminated herbal medicines (Haider et al. 2004). Correlation coefficient of lead with

chromium (R^2 0.7611) indicates the common source of origin of these metals. The presence of lead in plant material indicates the probable influence of vehicular activity on metal concentration in ambient environment (Khan et al. 2008).

Table 2 Heavy metal concentration (µg g⁻¹) in different medicinal plants (mean ± SD)

S. No.	Plants	Fe	Cu	Cd	Pb	Cr
	WHO standards	20	150	0.3	10	2
1	<i>Allium sativum</i>	226.22 ± 1.19	3.28 ± 0.05	0.62 ± 0.06	16.77 ± 0.60	17.63 ± 2.07
2	<i>Aloe vera</i>	18.780 ± 0.27	2.42 ± 0.05	1.16 ± 0.08	19.07 ± 0.62	21.61 ± 3.10
3	<i>A. racemosus</i>	322.620 ± 0.14	19.14 ± 0.21	0.68 ± 0.07	20.3 ± 0.50	26.72 ± 3.14
4	<i>Azadirachta indica</i>	252.11 ± 2.02	1.88 ± 0.10	0.38 ± 0.03	35.74 ± 0.10	27.14 ± 3.41
5	<i>Bacopa monnieri</i>	7.610 ± 0.01	4.95 ± 0.06	0.037 ± 0.00	13.47 ± 0.30	18.66 ± 2.36
6	<i>Calotropis gigantean</i>	271.87 ± 0.16	6.02 ± 0.06	0.07 ± 0.05	24.57 ± 0.60	31.08 ± 4.11
7	<i>Cannabis sativa</i>	51.36 ± 0.25	5.28 ± 0.06	0.23 ± 0.08	31.17 ± 0.30	48.41 ± 6.31
8	<i>Catharanthu sroseus</i>	57.41 ± 0.32	3.81 ± 0.11	0.69 ± 0.09	54.47 ± 0.62	47.36 ± 5.63
9	<i>Curcuma longa</i>	308.25 ± 0.14	6.17 ± 0.15	0.46 ± 0.01	30.07 ± 1.25	38.16 ± 5.44
10	<i>Datura stramonium</i>	62.41 ± 0.10	3.41 ± 0.08	0.52 ± 0.14	25.57 ± 1.21	34.49 ± 4.18
11	<i>Eclipta prostrate</i>	279.57 ± 0.60	3.93 ± 0.06	0.010 ± 0.00	33.37 ± 0.46	47.67 ± 5.67
12	<i>H. rosa-sinensis</i>	114.27 ± 0.22	5.6 ± 0.08	0.001 ± 0.00	40.77 ± 0.80	48.63 ± 5.77
13	<i>Lantana camara</i>	255.170 ± 0.76	4.95 ± 0.12	0.41 ± 0.11	34.37 ± 0.46	44.19 ± 5.54
14	<i>Ocimum tenuiflorum</i>	138.09 ± 1.14	14.58 ± 0.06	0.64 ± 0.05	13 ± 0.40	18.63 ± 2.04
15	<i>Rauwolfia serpentine</i>	267.340 ± 0.38	4.120 ± 0.06	0.79 ± 0.08	54.27 ± 0.30	58.63 ± 6.74
16	<i>Rosa rubiginosa</i>	224.11 ± 0.75	8.08 ± 0.05	0.74 ± 0.11	51.77 ± 1.05	47.18 ± 5.66
17	<i>Solanum nigrum</i>	99.27 ± 0.40	3.52 ± 0.06	0.65 ± 0.06	36.47 ± 0.06	45.91 ± 5.16
18	<i>Tagetes platula</i> L.	44.650 ± 0.24	7.41 ± 0.06	2.1 ± 0.08	52.87 ± 0.17	48.84 ± 6.04
19	<i>Tinospora cordifolia</i>	292,121 ± 0.64	5.81 ± 0.08	0.61 ± 0.08	25.67 ± 0.46	28.81 ± 4.04
20	<i>Withania somnifera</i>	144.520 ± 0.23	6.36 ± 0.11	0.97 ± 0.08	19.87 ± 0.62	24.77 ± 3.01

Results are expressed as mean of five replicates ± standard deviation (i.e., $n = 5 \pm SD$)

Table 3 Concentration and range of DDT and its metabolites ($\mu\text{g g}^{-1}$) detected in medicinal plant materials

S. No	Plants	ppDDE	opDDT	ppDDD	ppDDT	Total DDT
	WHO standard					0.1
1	<i>Allium sativum</i>	0.08 ± 0.007	6.44 ± 0.42	1.46 ± 0.204	0.78 ± 0.048	5.43 ± 0.56
2	<i>Aloe vera</i>	BDL	2.67 ± 0.14	BDL	3.14 ± 0.29	3.78 ± 0.56
3	<i>Asparagus racemosus</i>	0.18 ± 0.03	0.48 ± 0.06	BDL	0.45 ± 0.01	0.63 ± 0.16
4	<i>Azadirachta indica</i>	0.13 ± 0.05	8.23 ± 0.46	2.07 ± 0.304	0.61 ± 0.03	4.01 ± 0.44
5	<i>Bacopa monnieri</i>	0.1 ± 0.06	0.97 ± 0.15	BDL	0.45 ± 0.06	1.87 ± 0.17
6	<i>Calotropis gigantean</i>	BDL	2.44 ± 0.17	1.68 ± 0.20	4.36 ± 0.54	5.17 ± 0.67
7	<i>Cannabis sativa</i>	0.34 ± 0.06	5.12 ± 0.36	BDL	1.47 ± 0.76	5.78 ± 0.49
8	<i>Catharanthus roseus</i>	0.21 ± 0.04	4.73 ± 0.26	BDL	0.71 ± 0.11	4.44 ± 0.57
9	<i>Curcuma longa</i>	0.06 ± 0.01	4.04 ± 0.52	0.97 ± 0.186	0.58 ± 0.14	3.78 ± 0.47
10	<i>Datura stramonium</i>	0.08 ± 0.02	4.86 ± 0.35	BDL	1.87 ± 0.27	6.13 ± 0.57
11	<i>Eclipta prostrata</i>	0.37 ± 0.05	2.76 ± 0.38	2.64 ± 0.302	0.86 ± 0.17	3.63 ± 0.40
12	<i>Hibiscus rosa-sinensis</i>	0.74 ± 0.09	3.34 ± 1.42	BDL	0.98 ± 0.14	4.01 ± 0.56
13	<i>Lantana camara</i>	0.66 ± 0.18	6.42 ± 0.89	BDL	4.68 ± 0.57	7.14 ± 0.54
14	<i>Ocimum tenuiflorum</i>	0.46 ± 0.06	10.76 ± 1.22	3.68 ± 0.49	0.94 ± 0.18	5.76 ± 0.43
15	<i>Rauvolfia serpentina</i>	0.18 ± 0.03	4.48 ± 0.56	1.91 ± 0.26	5.16 ± 0.39	5.78 ± 0.31
16	<i>Rosa rubiginosa</i>	0.28 ± 0.08	1.83 ± 0.89	BDL	3.68 ± 0.19	5.02 ± 0.45
17	<i>Solanum nigrum</i>	1.02 ± 0.06	1.21 ± 0.06	0.12 ± 0.014	0.63 ± 0.03	2.71 ± 0.68
18	<i>Tagetes platula L.</i>	0.47 ± 0.09	5.77 ± 0.38	BDL	6.86 ± 0.45	6.98 ± 0.89
19	<i>Tinospora cordifolia</i>	BDL	1.77 ± 0.34	2.14 ± 0.14	1.04 ± 0.09	3.14 ± 0.35
20	<i>Withania somnifera</i>	0.08 ± 0.05	3.13 ± 0.89	BDL	1.14 ± 0.79	4.21 ± 0.59

Results are expressed as mean of five replicates ± standard deviation (i.e., $n = 5 \pm \text{SD}$)

Among the pesticides analyzed, the total DDT found in medical plants ranged between 0.63 and 7.14 $\mu\text{g g}^{-1}$ (Table 3). The maximum concentration of total DDT was found in *Lantana camara* (7.14 $\mu\text{g g}^{-1}$), the minimum in *Asparagus racemosus* (0.63 $\mu\text{g g}^{-1}$). Industrial effluents and waste material from pesticides factories release DDT to terrestrial and aquatic environments. Usually, DDT evaporates from soil and surface water into air, while some is broken down by photodegradation or by the microorganism in soil or surface

water. When DDT is broken into soil, it usually forms DDE or DDD (Hellawell 1988). Thus, the presence of DDT or its metabolites shows that the sites where these herbs were grown were heavily contaminated with DDT, whereas the availability of DDE and DDD indicates soil contamination due to spraying of pesticides for pest control which subsequently underwent photodegradation.

Among the different HCH isomers characterized, it has been observed that α -HCH predominates the total HCHs

Table 4 Concentration and range of HCH isomers ($\mu\text{g g}^{-1}$) detected in different medicinal plant materials

S. No.	Plants	α -HCH	β -HCH	γ -HCH	δ -HCH	Total HCH
	WHO standard					0.3
1	<i>Allium sativum</i>	4.46 ± 0.50	0.27 ± 0.04	1.42 ± 0.21	0.27 ± 0.04	4.63 ± 0.53
2	<i>Aloe vera</i>	1.64 ± 0.18	0.14 ± 0.06	1.64 ± 0.21	BDL	2.08 ± 0.22
3	<i>Asparagus racemosus</i>	0.31 ± 0.06	0.16 ± 0.02	0.56 ± 0.12	1.26 ± 0.18	2.02 ± 0.34
4	<i>Azadirachta indica</i>	3.74 ± 0.42	0.24 ± 0.06	2.64 ± 0.30	BDL	6.02 ± 0.77
5	<i>Bacopa monnieri</i>	0.71 ± 0.07	0.74 ± 0.09	2.89 ± 0.31	BDL	3.86 ± 0.42
6	<i>Calotropis gigantean</i>	1.67 ± 0.26	0.21 ± 0.06	2.77 ± 0.37	0.21 ± 0.21	4.16 ± 0.53
7	<i>Cannabis sativa</i>	2.76 ± 0.33	0.28 ± 0.05	1.87 ± 0.25	BDL	3.63 ± 0.40
8	<i>Catharanthus roseus</i>	1.86 ± 0.26	0.16 ± 0.06	2.42 ± 0.32	0.63 ± 0.42	3.84 ± 0.42
9	<i>Curcuma longa</i>	2.86 ± 0.35	0.18 ± 0.08	1.64 ± 0.21	BDL	3.63 ± 0.40
10	<i>Datura stramonium</i>	3.63 ± 0.41	0.27 ± 0.06	1.63 ± 0.21	BDL	3.66 ± 0.41
11	<i>Eclipta prostrata</i>	0.98 ± 0.14	0.32 ± 0.05	0.64 ± 0.12	BDL	1.66 ± 0.23
12	<i>Hibiscus rosa-sinensis</i>	0.72 ± 0.12	0.68 ± 0.11	1.08 ± 0.21	1.14 ± 0.18	2.16 ± 0.32
13	<i>Lantana camara</i>	2.63 ± 0.31	0.37 ± 0.21	2.08 ± 0.28	0.18 ± 0.11	2.96 ± 0.33
14	<i>Ocimum tenuiflorum</i>	4.01 ± 0.52	0.46 ± 0.32	1.94 ± 0.22	0.57 ± 0.08	5.42 ± 0.62
15	<i>Rauvolfia serpentina</i>	3.14 ± 0.37	0.42 ± 0.13	3.06 ± 0.43	0.48 ± 0.12	3.91 ± 0.44
16	<i>Rosa rubiginosa</i>	1.92 ± 0.21	0.14 ± 0.04	3.62 ± 0.45	1.27 ± 0.21	6.44 ± 0.76
17	<i>Solanum nigrum</i>	0.57 ± 0.08	0.88 ± 0.10	1.72 ± 0.19	0.28 ± 0.17	3.04 ± 0.42
18	<i>Tagetes patula L.</i>	1.78 ± 0.22	0.47 ± 0.07	1.86 ± 0.25	0.42 ± 0.12	3.41 ± 0.38
19	<i>Tinospora cordifolia</i>	1.08 ± 0.22	0.18 ± 0.15	0.86 ± 0.13	0.98 ± 0.23	1.63 ± 0.25
20	<i>Withania somnifera</i>	2.98 ± 0.37	0.29 ± 0.07	2.14 ± 0.35	0.67 ± 0.14	4.14 ± 0.57

Results are expressed as mean of five replicates ± standard deviation (i.e., $n = 5 \pm \text{SD}$)

profile (Table 4). Lindane and other HCH isomers are highly resistant to microbial and chemical degradation and thus persist in the environment for prolonged duration. The range of total HCH found in medicinal plants was ranged between 1.63 and 6.44 $\mu\text{g g}^{-1}$. The maximum (6.44) and minimum (1.63 $\mu\text{g g}^{-1}$) concentrations of total HCH were found in *Rosa rubiginosa* and *Tinospora cordifolia* respectively.

For the concentration of heavy metals, DDT and HCH detected in the samples were well above the permissible limits prescribed by WHO (2007). Findings warrant urgent attention of the quality assurance agencies to detect the level of toxic content in the herbal plant material before processing it for herbal formulation.

Overall, the results of heavy metal and pesticide analysis showed that the contaminants are present in varied concentrations in the 20 medicinal herbs commonly sold in the local market of Lucknow. Variation in the metal and pesticide accumulation may be assigned to the different anatomical and chemical characteristic of particular plant species including stage of growth, soil type, and the type of metals absorbed (Verma et al. 2007; Olowoyo et al. 2012; Orisakwe et al. 2012). Furthermore, contamination could occur during storage and/or at the point of sale. Therefore, it is not only the growth conditions but harvesting and processing also adds to the metal and pesticides contamination.

Results of the study revealed that the content of heavy metals and pesticides detected in the medicinal plant samples were above the permissible limits. Sources of heavy metal and pesticide contamination in herbs could be linked to water used in irrigation, polluted soils, fertilizers, pesticides, industrial emissions, transportation, harvesting, and storage processes. It is evident that there is an urgent need to implement a regular monitoring and testing program on the quality of the local and imported herbs sold in the market. Awareness among the suppliers and consumers should be disseminated to prevent collection of medicinal herbs growing near contaminated sites to prevent health risk associated with their consumption.

Acknowledgements The authors would like to thank The Head, Department of Environmental Science, BBAU, Lucknow, India, for providing infrastructural support.

References

- Anna F, Marzanna K, Edward S (2015) Determination of toxic metals by ICP-MS in Asiatic and European medicinal plants and dietary supplements. *J Trace Elem Med Biol* 30:54–58
- Arpadjan S, Celik G, Taskesen S, Gucen S (2008) Arsenic, cadmium and lead in medicinal herbs and their fractionation. *Food Chem Toxicol* 46:2871–2875
- Bodeker C, Bodeker G, Ong CK, Grundy CK, Burford G, Shein K (2005) WHO global atlas of traditional, complementary and alternative medicine. World Health Organization, Geneva
- Chauhan R, Awasthi S, Singh AP, Srivastava S, Pande V, Tripathi RD, Kumar A (2017) Heavy metal tolerance in crop plants: physiological and biochemical aspect. In: Shukla V et al (eds) *Plant adaptation strategies in changing environment*. Springer Nature, Singapore, pp 253–268. https://doi.org/10.1007/978-981-10-6744-0_11
- Dimitrova Z (1999) The history of pharmacy. *St Clement of Ohrid, Sofija*, pp 13–26
- Duruibe JO, Ogwuegbu MOC, Egwurugwu JN (2007) Heavy metal pollution and human biotoxic effects. *Int J Physical Sci* 2(5):112–118
- Govindaraghavan S, Nikolaus JS (2015) Quality assessment of medicinal herbs and their extracts: Criteria and prerequisites for consistent safety and efficacy of herbal medicines. *Epilepsy Behav* 52:363–371
- Haider S, Naithani V, Barthwal J, Kakkar P (2004) Heavy metal content in some therapeutically important medicinal plants. *Bull Environ Contam Toxicol* 72:119–127
- Hellawell JM (1988) Toxic substances in river and streams. *Environ Pollut* 50:61–85
- Khan CS, Cao Q, Zheng YM, Huang YZ, Zhu YG (2008) Health risks of heavy metals in contaminated soils and food crops irrigated with wastewater in Beijing, China. *Environ Pollut* 152(3):686–692
- Khatoun H, Pant A, Rai JPN (2017) Plant adaptation to recalcitrant chemicals. In: Shukla V et al (eds) *Plant adaptation strategies in changing environment*. Springer Nature, Singapore, pp 269–290. https://doi.org/10.1007/978-981-10-6744-0_11
- Kishan PS, Bhattacharya S, Sharma P (2014) American-Eurasian assessment of heavy metal contents of some Indian medicinal plants. *J Agric Environ Sci* 14(10):1125–1129
- Kumar N, Kumar S, Baudhh K, Dwivedi N, Shukla P, Singh DP, Barman SC (2015) Toxicity assessment and accumulation of metals in radish irrigated with battery manufacturing industry effluent. *Int J Veg Sci* 00:1–13
- Kumar D, Bharti SK, Anand S, Kumar N (2018) Bioaccumulation and biochemical responses of *Vetiveria zizanioides* grown under cadmium and copper stresses. *Environ Sustain* 1:133–139. <https://doi.org/10.1007/s42398-018-0009-z>
- Kunle OF, Egharevba HO, Ahmadu PO (2012) Standardization of herbal medicines: a review. *Int J Biodivers Conserv* 4(3):101–112
- Lasat MM, Fuhrman M, Ebbs SD, Cornish JE, Kochian LV (1998) Phytoremediation of radiocesium-contaminated soil: evaluation of cesium-137 bioaccumulation in the shoots of three plant species. *J Environ Qual* 27(1):165–169
- McLachlan MS, Horstmann M (1998) Forests as filters of airborne organic pollutants. *Environ Sci Technol* 32:413–420
- Mishra C, Sharma S, Kakkar P (2007) A study to evaluate heavy metals and organochlorine pesticide residue in *Zingiber officinalerosc.* collected from different ecological zones of India. *Bull Environ Contam Toxicol* 79:95–98
- Neha KD, ShuklaP KS, Baudhh K, Tiwari J, Dwivedi N, Barman SC, Singh DP, Kumar N (2017) Metal distribution in the sediments, water and naturally occurring macrophytes in the river Gomti, Lucknow, Uttar Pradesh, India. *Curr Sci* 113(8):1578–1585
- Olowoyo JO, Okedeyi OO, Mkolo NM, Lion GN, Mdakane STR (2012) Uptake and translocation of heavy metals by medicinal plants growing around a waste dump site in Pretoria, South Africa. *South Afr J Bot* 78:116–121
- Orisakwe OE, Nduka JK, Amadi CN, Dike D, Obialor OO (2012) Evaluation of potential dietary toxicity of heavy metals of vegetables. *J Environ Anal Toxicol* 2(3):136–139
- Ouyang Y (2002) Phytoremediation of modeling plant uptake and contaminant transport in the soil plant atmosphere continuum. *J Hydrol* 266(1–2):66–82

- Petrovska BB (2012) Historical review of medicinal plants' usage. *Pharmacogn Rev* 6(11):1–5
- Reinholds I, Pugajeva I, Bavrins K, Kuckovska G, Bartkevics V (2017) Mycotoxins, pesticides and toxic metals in commercial spices and herbs. *Food Addit Contam Part B* 10(1):5–14
- Rodrigues AAZ, De Queiroz MELR, De Oliveira AF, Heleno AAFF, Zambolim L, Freitas JF, Morais EHC (2017) Pesticide residue removal in classic domestic processing of tomato and its effects on product quality. *J Environ Sci Health Part B* :1–8
- Shaban NS, Abdou KA, Hassan NEY (2016) Impact of toxic heavy metals and pesticides residues in herbal products. *Beni-Suef Uni J Basic Appl Sci* 5:102–106
- Srivastava SK, Rai V, Srivastava M, Rawat AKS, Mehrotra S (2006a) Stimulation of heavy metals in different *Berberis* species and its market samples. *Environ Monit Assess* 16(1–3):315–320
- Srivastava LP, Kumar N, Gupta KP, Raizada RB (2006b) Status of HCH residues in Indian medicinal plant materials. *Bull Environ Contam Toxicol* 76:782–790
- Srogi K, Wlochowicz A, Szczepanik K (2002) Determination of heavy metal contents in samples of medicinal herbs. *Pol J Environ Stud* 11(5):467–471
- Verma P, George KV, Singh HV, Singh RN (2007) Modeling cadmium accumulation in radish, carrot, spinach and cabbage. *Appl Math Model* 31(8):1652–1661
- Wiart C (2006) *Ethnopharmacology of medicinal plants*. Humana Press, New Jersey, pp 1–50
- World Health Organization (2007) WHO guidelines for assessing quality of herbal medicines quality of herbal medicines with reference to contaminants and residues. Geneva
- Ziarati P (2012) Determination of some heavy metals in popular medicinal plants of Tehran's market. *J Pharma Health Sci* 1(3):31–36