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Assessing Zinc Thresholds in Commonly Used Herbs in India and Associated Health Risks

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Abstract

Use of herbal medicines has greatly increased as complementary and alternative medicine for the ailment of many diseases. In the present study zinc content was determined in the commonly consumed raw herbs sold over the table in India (East, West, North, South Zones). Zinc is an important micronutrient and plays a vital role in regulation in antioxidant activity and acts as a catalyst. However, excessive absorption of zinc causes suppression of copper & iron absorption, causes GIT disorders and alterations in blood lipoprotein level. The aim of this study was to determine the Zinc concentration in the market samples of raw herbs by ICP-MS (Inductively Coupled Plasma-Mass Spectrometry) and to model the estimated daily intake (EDI).

Keywords: Medicinal Plants, Zinc, Estimated Daily Intake, ICP-MS

Introduction

The Indian form of alternative medicine originated about 2000 years ago which relies on medicinal plants and their formulations (1)(2). According to the World Health Organization (WHO), these plant-based medications are used as primary treatment by around 80% of the world's population (3). These alternative herbal medicines are prepared from medicinal plants and are generally considered safe for use (4). However, as the need for these therapies is increasing, the natural resources of these herbs are becoming depleted, and in order to fulfil the high demand, herbs are being grown on farms using standard agronomic approaches. Also, for getting high yield, use of chemical fertilizers and pesticides have been increased considerably. This in turn is affecting the overall nutrient status and enhancing the accumulation of some essential nutrients beyond the permissible limits. It is well documented that unethical farm practices and heavy load of chemicals results in the accumulation of toxic substances in the various parts of medicinal plants (5)(6). Apart from this, mining of ores and minerals, heavy industrial practices such as use of foundries, smelters, oil refineries, petrochemical plants, chemical industry, untreated sewage sludge and diffuse sources such as metal piping and combustion by-products from coal-burning power stations etc. are also the major contributors in increasing heavy metal contamination in soil and water (7)(8). This study discusses the zinc status in the commonly used herbs in India shortlisted API Volume I of AYUSH (9). Zinc is essential for overall homeostasis of human body and plays a crucial role in growth and development and acts a signalling factor and is one of the most common trace elements (10). It contributes to the regulation of chronic inflammatory status by

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decreasing inflammatory cytokines. Additionally, it is known to alleviate oxidative stress by taking part in the synthesis of antioxidant enzymes. It also acts as a catalyst for these enzymes, as well as is involved in lipid, protein, and carbohydrate metabolism (11)(12). Apart from acting as an essential micronutrient and supporting many metabolic processes, numerous studies on lipid and metabolic disorders are been reported with high zinc concentrations. Increased intake of zinc also impacts the immune system of body (13). Nausea, fever and weakness are also some associated side-effects with high zinc consumption (14).

Material and Methods

Study area

Medicinal plants samples in the raw form were procured from the local markets of East, West, North and South zones of India and the passport data for the same was generated. Plant identification and authentication was done from different authorised regional centres by expert botanists from "Regional Ayurveda Research Institute for Gastro-Intestinal Disorders (Guwahati), Regional Ayurveda Institute for Fundamental Research (Pune), Regional Ayurveda Research Institute (Jhansi) and Regional Ayurveda Research Institute for Metabolic Disorders (Bengaluru)"; for East, West, North and South respectively.

Sampling and metal analysis

Raw herbs sold over the table in the Indian market were chosen and collected from the local markets of each region for the analysis of zinc concentration. The names and part used of the respective herb is given in Table 1,2,3&4. The plants were tagged properly, stored in polyethylene bags, and brought to the laboratory.

Reagents

The reagents used were of Supra Grade. HPLC grade water (Rankem) was used. All the glassware soaked overnight in 1% HNO_3 solution and washed with deionized water were used after drying. Hydrogen peroxide (30%) & nitric acid (65%), Merck, Darmstadt, Germany was used.

Digestion of herb samples

The herb samples were grounded into powder and weighed (1g) in PTFE (Teflon) vessels. To this 8ml diacid mixture (2:5 v/v HNO_3 and H_2O_2) was added. The vessels were kept for few hours as such for cold digestion. Samples were digested in a microwave assisted digestion system; Anton Paar Multiwave 7000; with ramp time and hold time of (10, 5 and 5 minutes) for 3 cycles at 90^0C, 120^0C and 150^0C respectively. The digested material was filtered through 0.2 mm syringe filter and was made up to 50 ml in a volumetric flask. The digested samples were stored at 4°C for analysis.

Estimation of zinc concentration

Zinc content determination with the digested leaf samples was performed on a model ICP-MS (Inductively Coupled Plasma Mass Spectrometery) NexION 2000; Germany using Syngistix software; Inductively Coupled Mass Spectrophotometer under an optimized measurement condition with Argon and Helium gas flow. NIST (National Institute of Standards and Technology) certified 1000 ppm zinc standard was used to plot standard graph. The samples were analysed in triplicates and the average values of each sample was used as result.

Estimated daily Intake

Both the metal concentration in plants and the amount consumed of the respective plants influence the average estimated daily intake (EDI). The EDI of Zinc was calculated using Eq.1, recommended by the USEPA (15).

EDI = X (FIR/WAB) ………………………….(1)

where EDI is the estimated daily intake (mg/kg bw/day); X is the Zinc concentration

in the sample (mg kg^{-1}); FIR is daily

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consumption rate for adults. WAB is the average adult body weight (kg) which is considered to be 65 kg.

Results and Discussion

The data for zinc concentration is presented in Tables 1,2,3 & 4 and estimated daily intake in the respective herbs sampled is presented in Table 5. The concentration of zinc (Zn) in the analysed samples ranged between 5.657 and 88 mg kg^{-1} in East, 4.919 mg kg^{-1} and 78.089 mg kg^{-1} in West, 3.44 mg kg^{-1} and 95.962 mg kg^{-1} in North, 1.858 mg kg^{-1} and 78.336 mg kg^{-1} in South. The maximum concentrations of zinc in Yavani (*Trachyspermum ammi* Linn.); 58.475, 78.089, 95.962, 66.759 mg kg^{-1}, Svetajiraka (*Cuminum cyminum* Linn); 71.389, 60.226, 59.441, 76.145 mg kg^{-1}, Upakuncika (*Nigella sativa* Linn); 130.74, 64.759, 55.021, 57.703 mg kg^{-1} in all the four zones viz, East, West, North and South respectively and 7 samples in East, West & South Zones and 10 samples in North Zone were found be above permissible limit of 50 mg/kg (Figure 1-4). Overall, results revealed that of the samples analysed; 9.7% in East, 12.9% in West, 16.12% in North and 11.2% in South had concentrations higher than 50 mg kg^{-1} (which is the permissible limit (PL)) set for zinc in herbal medicines by FAO/WHO (16)(17). High concentration of Zinc (above 50 mg kg^{-1}) was observed in Mint, Basil, Parsley, Sage and Thyme (52.97, 50.10, 112.19, 58.78, 112.19 mg kg^{-1} respectively) sampled in UAE (18). Similar to present study, high concentration of zinc, 314.05 mg/kg was found in cumin sampled from Egyptian market (19). The samples of medicinal spices sampled from Polish market were reported with the zinc concentrations with the maximum permissible limits (20).

The EDI's of zinc is calculated on the basis of average concentration of zinc in the herbs and the consumption rate for adults (Table 5). According to the calculated data, the values of the daily intake of zinc were observed to be on the higher side for the medicinal herbs with high zinc concentrations in the respective zones; with the values ranging from 0.08- 3.29 mg/kg bw/day in east zone, 0.07-4.65 mg/kg bw/day in west zone, 0.05-4.22 mg/kg bw/day in north and 0.03-3.63 mg/kg bw/day in south zones across India. Zinc is a trace element that is essential for normal growth, blood clotting, thyroid function, and DNA & protein synthesis. Excessive zinc consumption has a negative impact on the immune system, blood lipoprotein levels, and copper level (18). Extreme intake of zinc also causes abdominal pain, diarrhoea, vomiting and nausea. High zinc uptake in medicinal plants is due to presence of heavy amount of zinc present in cultivation soil. Therefore, various biological, chemical, and physical strategies are employed to render soil free from zinc contamination. These involve use of hyperaccumulator plants such as *Arabidopsis halleri, Noccaea caerulescens* (21)*, Thlaspi ochroleucum* (22); apart from this *Dichapetalum subsp. Sumatranum and D. subsp. Pilosum* are strong zinc hyperaccumulators (23). Polyaspartate (PASP) synthesized from L-aspartic acid produced by modified thermal procedure has potential to chelate Zinc ions from contaminated soil (24).

Conclusion

The present research based on the zinc estimation in commonly used medicinal plants across four geographical locations across India, showed that the concentration of Zinc in some herbs were beyond the acceptable limits. The findings of the present research also highlight the significance of safety and hygiene practices from harvest to their reaching to consumer end. Therefore, it is utmost necessary to implement the regular monitoring of testing of the quality and mineral content in the raw herbs sold over the table in the Indian markets. It is also to be taken into consideration that the many of the herbs studied in the present research are incorporated in the daily meals. Future prospects regarding the presence of more toxic elements and their impact on overall human health and also on herbs must be studied. Also,

Table 1. Zinc Concentration in the Fruits and Flower Buds of the Herbs Sampled (ppm±SD)

Herb Name	Parst Used	East	West	North	South
Ajamoda (*Apium leptophyllum*	Fruit	**	**	43.323±1.1	78.336±0.9
Yavani(*Trachyspermum ammi*)	Fruit	58.475±1.12	78.089±1.1	91.35±1.2	66.759±0.9
Dhanyaka (*Coriandrum sativum)*	Fruit	40.49±0.9	60.49±1.12	40.876±0.9	45.809±0.9
Svetajiraka (*Cuminum cyminum*)	Fruit	71.389±1.23	60.226±1.54	59.441±1.32	76.145±0.9
Krsnajirak (*Carum carvi*)	Fruit	49.698±1	50.689±1.23	48.723±1.32	43.338±0.9
Amalaki (*Emblica officinalis*)	Dry Fruits	6.874±1.5	23.34±1.32	7.788±2.1	9.218±1.1
Kankola (*Piper chubeba*)	Fruit	32.25±0.9	17.13±1.1	65.09±0.9	31.12±0.8
Udumbara (*Ficus racemose*)	Fruit	8.38±1.12	26.5±1.1	13.23±1.21	13.42±1.1
Jatìphala (*Myristica fragrans*)	Fruit	20.85±1.56	9.701±1.2	14.491±1.1	9.16±1.1
Suksmaila (*Elettaria cardamomum*)	Fruit	38.502±1.32	12.984±0.9	51.008±1.2	47.66±1.1
Haritaki (*Terminalia chebula*)	Fruit	14.05±1.12	14.54±1.1	10.489±1.4	**
Misreya (*Foeniculum vulgare*)	Fruit	14.917±1.43	14.519±1.1	31.32±1.21	63.77±1.1
Bilva (*Aegle marmelos*)	Fruit pulp	**	4.919±0.9	13.34±0.9	17.65±0.8
Aragvadha (*Cassia fistula*)	Fruit pulp	**	7.137±1.32	17.1±0.9	25.85±0.9
Hingu (*Ferula foetida*)	Oleo-gum-resin	6.36±2.1	9.341±0.7	8.367±1.32	18.401±1.1
Lavanga (*Syzygium aromaticum*)	Flower bud	12.531±0.9	18.176±1.1	16.171±1.12	18.209±1.1

Table 2. Zinc Concentration in the Root and Rhizomes of the Herbs Sampled (ppm±SD)

Herb Name	Parst Used	East	West	North	South
Haridra (*Curcuma longa*)	Rhizome	14.248±1.1	11.637±0.9	**	**
Sunthi (Zingiber officinale)	Rhizomes	25.021±1.21	16.818±1.23	**	36.532±0.9
Citraka (*Plumbago zeylanica*)	Root	6.086±1.13	**	75.32±1.21	14.09±1.1
Aswagandha (*Witthania somnifera*)	Root	44.431±1.13	14.417±1.12	15.753±1.4	15.576±1
Arka (*Calotropis procera*)	Root	30.94±1.18	29.83±1.12	**	7.35±0.9
Eranda (*Ricinus communis*)	Root	28.48±1.15	19.85±1.5	14.51±1.3	6.389±0.9
Karavira (*Nerium indicum*)	Root	**	12.277±1.21	26.43±1.1	31.37±1.2
Nyagrodha (*Ficus benghalensis*)	Aerial Root	15.06±0.8	12.68±0.9	12.68±0.9	12.27±0.8
Pashanabheda (*Bergenia ciliate*)	Rhizome	39.96±1.1	15.07±2.1	46.06±0.9	35.77±1
Taamalaki (*Phyllanthus fraternus*)	Dried Root	**	47.03±0.9	39.41±0.9	**

**The herbs were not available in the respective markets

Table 3. Zinc Concentration in the Stem and Whole Plant of the Herbs Sampled (ppm±SD)

Herb Name	Parst Used	East	West	North	South
Kantakari (*Solanum surattense*)	Whole plant	**	**	**	29.374±0.8
Apaamaarga (*Achyranthes aspera*)	Whole Plant	17.67±0.9	23.6±1.45	30±0.9	20.81±0.9
Argavadha (*Cassia fistula*)	Stem Bark	**	12.1±0.9	5.294±1	14.26±1
Ashvattha (*Ficus religiosa*)	Bark	8.232±0.8	14.712±1.1	**	21.45±1.1
Bilva (*Aegle marmelos*)	Stem Bark	5.647±1.12	25.37±1.9	22.56±1.1	14.76±1.2
Guduci (*Tinospora cordifolia*)	Stem	14.39±1.32	52.65±1.12	13.031±1.1	31.09±1.21
Khadira (*Acacia catechu*)	Heart Wood	8.55±1.12	15.803±1.23	13.23±1.3	21.46±1.1
Kanchanara (*Bauhinia variegate*)	Stem Bark	16.49±1.15	6.585±1.45	8.353±1.1	14.16±1
Nyagrodha (*Ficus benghalensis*)	Stem Bark	13.86±0.6	12.7±1.65	12.246±0.9	19.33±1
Taamalaki (*Phyllanthus fraternus*)	Whole Plant	88.4±1.12	**	**	**
Taamalaki (*Phyllanthus fraternus*)	Stem	**	11.26±0.9	40.86±1.1	**
Udumbara (*Ficus racemose*)	Bark	37.89±1.11	14.64±1.2	12.29±1.11	21.14±0.9
Tvak (*Cinnamomum zeylanicum*)	Bark	14.27±0.9	7.326±1.32	18.07±1.1	8.905±1.2
Babbula (*Acacia nilotica*)	Stem bark	**	5.94±1.1	3.818±0.9	**
Lodhara (*Symplocos racemose*)	Stem Bark	**	**	3.44±1.1	1.858±1
Asoka (*Saraca asoka*)	Stem Bark	**	**	9.78±1.1	4.722±1.1

**The herbs were not available in the respective markets

Table 4. Zinc Concentration in the Seeds and Leaf Samples of the Herbs (ppm±SD)

Herb Name	Parst Used	East	West	North	South
Upakuncika (*Nigella sativa*)	Seed	130.74±0.9	64.759±1.12	55.021±1.1	57.703±0.9
Candrasura (*Lepidium sativum*)	Seed	**	49.305±1.21	**	58.696±0.9
Atasi (*Linum usitatissimum*)	Seed	27.14±1.15	53.53±1.11	72.12±0.9	43.72±1.3
Arkaparna (*Calotropis procera*)	Leaf	51.75±0.9	45.25±1.43	61.5±0.9	35.88±1.1
Eranda (*Ricinus communis*)	Leaf	23.97±1.11	**	**	14.49±1.1
Eranda (*Ricinus communis*)	Seed	43.69±1.19	63.76±1.3	73.48±1.1	63.75±0.9

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Gunja (*Abrus precatorius*)	Seed	20.22±2.1	29.87±1.21	37.43±1.2	22.03±0.9
Karavira (*Nerium indicum*)	Leaf	75.44±1.21	26.09±1.43	14.913±1.32	42.5±1.1
Tvakpatra (*Cinnamomum tamala*)	Leaf	39.125±0.8	32.132±1.32	32.224±1.21	28.857±1.2
Karanja (*Pongamia pinnata*)	Seed	**	23.95±1.32	**	36.35±0.8
Karpasa (*Gossypium herbaceum*)	Seed	**	11.95±1.21	34.46±1.1	27.82±0.9
Taamalaki (*Phyllanthus fraternus*)	Dried Leaf	**	45.62±0.8	12.514±1.1	34.51±1.2
Puuga (*Areca catechu*)	Seed	19.33±1.13	30.42±1.2	15.1±0.9	47.14±1.1
Kulattha (*Macrotyloma uniflorum*)	Seed	36.367±1.12	12.279±1.43	54.91±0.9	5.258±0.9
Vijaya (*Cannabis sativa*)	Leaf	54.79±1.13	**	**	**

**The herbs were not available in the respective market

Table 5. Calculated Daily Intake of Zinc in Respective Herbs

Herb Name	Part Used	Estimated Daily Intake (mg/day kg bw)			
		East	West	North	South
Ajamoda (*Apium leptophyllum*)	Fruit	**	**	1.33	2.41
Yavani (*Trachyspermum ammi*)	Fruit	0.90	1.20	1.48	1.03
Dhanyaka (*Coriandrum sativum*)	Fruit	3.11	4.65	3.14	3.52
Haridra (*Curcuma longa*)	Rhizome	1.10	0.90	**	**
Hingu (*Ferula foetida*)	Oleo-gum-resin	0.10	0.14	0.13	0.28
Lavanga (*Syzygium aromaticum*)	Flower bud	0.19	0.28	0.25	0.28
Tvakpatra (*Cinnamomum tamala*)	Leaf	0.60	0.49	0.50	0.44
Upakuncika (*Nigella sativa*)	Seed	2.01	1.00	0.85	0.89
Svetajiraka (*Cuminum cyminum*)	Fruit	3.29	2.78	2.74	3.51
Sunthi (*Zingiber officinale*)	Rhizome	0.77	0.52	**	1.12
Krsnajirak (*Carum carvi*)	Fruit	1.53	1.56	1.50	1.33
Kantakari (*Solanum surattense*)	Whole Plant	**	**	**	0.45
Candrasura (*Lepidium sativum*)	Seed	**	0.76	**	0.90
Aswagandha (*Witthania somnifera*)	Root	1.37	0.44	0.48	0.48
Amalaki (*Emblica officinalis*)	Dry Fruits	0.53	1.80	0.60	0.71
Atasi (*Linum usitatissimum*)	Seed	1.25	2.47	3.33	2.02
Arka (*Calotropis procera*)	Root	0.48	0.46	**	0.11
Arkaparna (*Calotropis procera*)	Leaf	0.80	0.70	0.95	0.55
Apaamaarga (*Achyranthes aspera*)	Whole Plant	0.27	0.36	0.46	0.32

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Argavadha (*Cassia fistula*)	Stem Bark	**	0.19	0.08	0.22
Ashvattha (*Ficus religiosa*)	Bark	0.13	0.23	**	0.33
Bilva (*Aegle marmelos*)	Stem Bark	0.09	0.39	0.35	0.23
Citraka (*Plumbago zeylanica*)	Root	0.09	**	1.16	0.22
Eranda (*Ricinus communis*)	Leaf	0.37	**	**	0.22
Eranda (*Ricinus communis*)	Root	0.44	0.31	0.22	0.10
Eranda (*Ricinus communis*)	Seed	0.67	0.98	1.13	0.98
Gunja (*Abrus precatorius*)	Seed	0.31	0.46	0.58	0.34
Guduci (*Tinospora cordifolia*)	Stem	0.66	2.43	0.60	1.43
Karavira (*Nerium indicum*)	Leaf	1.16	0.40	0.23	0.65
Karavira (*Nerium indicum*)	Root	**	0.19	0.41	0.48
Khadira (*Acacia catechu*)	Heart Wood	0.13	0.24	0.20	0.33
Kanchanara (*Bauhinia variegate*)	Stem Bark	0.25	0.10	0.13	0.22
Karanja (*Pongamia pinnata*)	Seed	**	1.11	**	1.68
Karpasa (*Gossypium herbaceum*)	Seed	**	0.18	0.53	0.43
Kankola (*Piper chubeba*)	Fruit	0.50	0.26	1.00	0.48
Nyagrodha (*Ficus benghalensis*)	Ariel Root	0.23	0.20	0.20	0.19
Nyagrodha (*Ficus benghalensis*)	Stem Bark	0.21	0.20	0.19	0.30
Pashanabheda (*Bergenia ciliate*)	Rhizome	0.61	0.23	0.71	0.55
Puuga (*Areca catechu*)	Seed	1.49	2.34	1.16	3.63
Taamalaki (*Phyllanthus fraternus*)	Whole Plant	1.36	**	**	**
Taamalaki (*Phyllanthus fraternus*)	Dried Root	**	0.72	0.61	**
Taamalaki (*Phyllanthus fraternus*)	Dried Leaf	**	0.70	0.19	0.53
Taamalaki (*Phyllanthus fraternus*)	Stem	**	0.17	0.63	**
Udumbara (*Ficus racemose*)	Fruit	0.90	2.85	1.42	1.45
Udumbara (*Ficus racemose*)	Bark	0.58	0.23	0.19	0.33
Vijaya (*Cannabis sativa*)	Leaf	0.84	**	**	**
Misreya (*Foeniculum vulgare*)	Fruit	0.46	0.45	0.96	1.96
Jatìphala (*Myristica fragrans*)	Fruit	0.16	0.07	0.11	0.07
Suksmaila (*Elettaria cardamomum*)	Fruit	1.18	0.40	1.57	1.47
Haritaki (*Terminalia chebula*)	Fruit	0.22	0.22	0.16	**
Tvak (*Cinnamomum zeylanicum*)	Bark	0.22	0.11	0.28	0.14
Kulattha (*Macrotyloma uniflorum*)	Seed	2.80	0.94	4.22	0.40
Babbula (*Acacia nilotica*)	Stem Bark	**	0.09	0.06	**
Bilva (*Aegle marmelos*)	Fruit Pulp	**	0.08	0.21	0.27
Aragvadha (*Cassia fistula*)	Fruit Pulp	**	0.11	0.26	0.40
Lodhara (*Symplocos racemose*)	Stem Bark	**	**	0.05	0.03
Asoka (*Saraca asoka*)	Stem Bark	**	**	0.15	0.07

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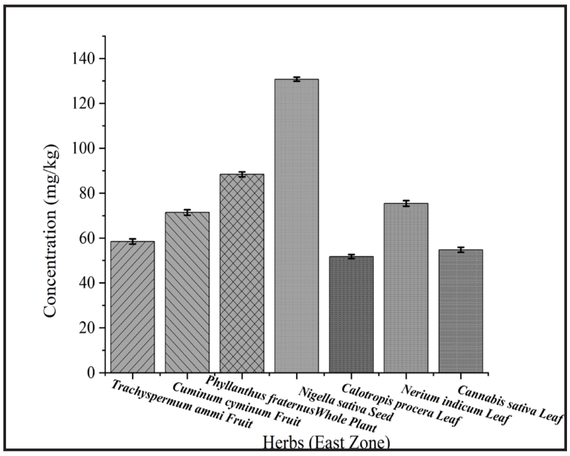

Figure 1. Zinc Concentration above PL in East Zone

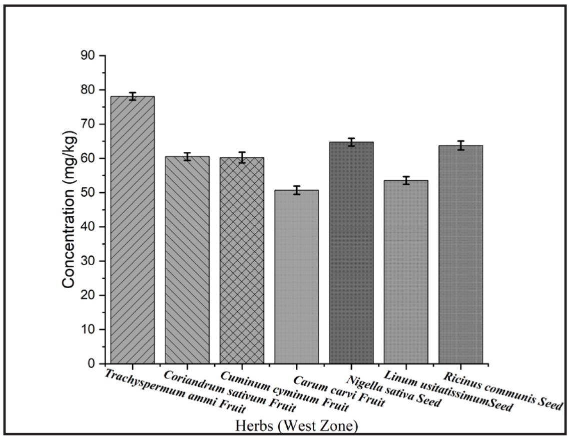

Figure 2. Zinc Concentration above PL in West Zone

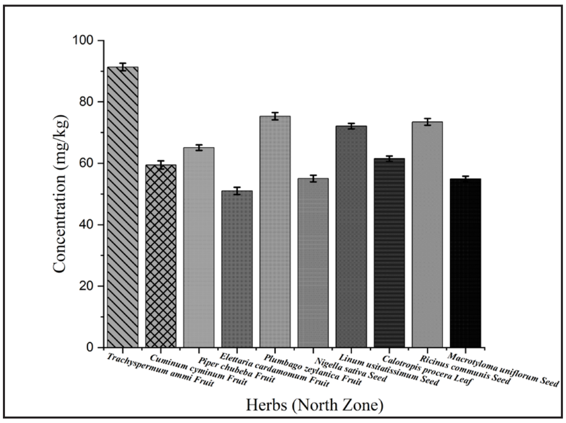

Figure 3. Zinc Concentration above PL in North Zone

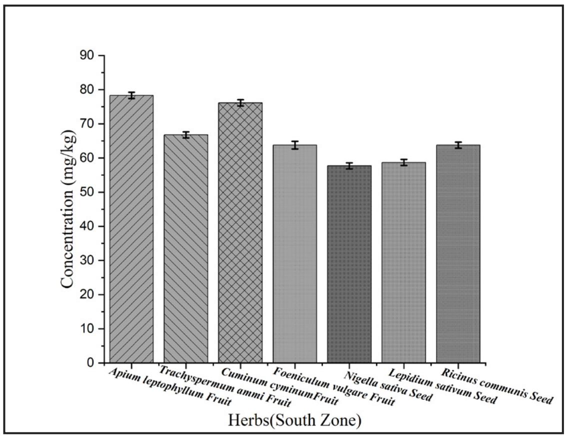

Figure 4. Zinc Concentration above PL in South Zone

remediation measures to reduce contamination of soil should be practiced.

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