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Phytoaccumulation of heavy metals and pesticide residues in the leaves of twelve selected malaueg medicinal plants

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Abstract

The study aimed to determine the phytoaccumulation of heavy metals and pesticide residues in the leaves of twelve selected medicinal plants commonly used by the Malaueg community of Rizal, Cagayan, namely: *Tabernaemontana pandacaqui* Lam. (bicarud), *Alocasia* sp (biga), *Imperata cylindrica* (L.) Raeusch (gabut), *Curcuma* sp (kunig), *Ficus septica* Burm. f. (liwliw), *Cymbopogon citrattus* (DC.) Staph (lubigan), *Hyptis brevipes* Poit. (matalagdaw), *Chromolaena odorata* (L.) R.M.King & H.Rob. (NPA), *Peperomia pellicida* (L.) Kunth. (pansit – pansitan), *Mollugo* sp (papait), *Eleusine indica* (L.) Gaertn. (tarattut), and *Ehretia microphylla* Lam (wild tea). Specifically, it aimed to (a) determine the heavy metal content of the soil and the leaves of the twelve selected Malaueg medicinal plants, (b) determine the pesticide residue content of the soil and the leaves of the twelve selected Malaueg medicinal plants and (c) determine the phytoaccumulation potential of the twelve selected Malaueg medicinal plants. The non – accumulation of heavy metals and pesticide residues by the plants points out to the safety of the leaves of the plants for use in preparation of products like personal and home care products, herbal ointment, cream, & lotion, functional food, etc. It is recommended that a test for the presence of other pesticide residues like organophosphates, carbamates, and the like be also conducted.

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Introduction

Soil (one of the principal substrata of life) is the biologically active, porous medium that has developed in the uppermost layer of Earth's crust, serving as a reservoir of water and nutrients, as a medium for the filtration and breakdown of injurious wastes, and as a participant in the cycling of carbon and other elements through the global ecosystem and it has evolved through weathering processes driven by biological, climatic, geologic, and topographic influences (Sposito, 2018). A soil is simply a porous medium consisting of minerals (the largest component, 45-49%), water (the second basic component, 2-50%), organic matter (the third basic component, 1-5%), gases (the fourth basic component, 2-50%), and microorganisms (final basic soil component, in very high numbers but less than 1%) (DeGomez & Kleinman, 2015).

Soil contamination refers to the destruction of land that could be used constructively by human activities, either directly or indirectly and the developing countries are also steadily but surely moving toward this direction which would lead to the devastating effects soil pollution on the environment (Bethany, 2017). Bethany also stated that soil pollution is caused by mindless human activity such as: industrial waste; deforestation; excessive use of fertilizers and pesticides & garbage pollution which brings about climate change, loss of soil fertility, and impact on human health.

Heavy metals constitute an ill-defined group of inorganic chemical hazards, and those most commonly found at contaminated sites are lead (Pb), chromium (Cr), arsenic (As), zinc (Zn), cadmium (Cd), copper (Cu), mercury (Hg), and nickel (Ni) (Wuana & Okieimen, 2011). Growth reduction as a result of changes in physiological and biochemical processes in plants growing on heavy metal polluted soils has been recorded and bioremediation is encouraged because it is an environmentally friendly approach achieved via natural processes aside from its being economical (Chibuike & Obiora, 2014). The Stockholm Convention on Persistent Organic Pollutants specifically banned the use of or unintentional release of "the dirty dozen", including nine chlorinated pesticides, viz Dichlorodiphenyl-1, 1, 1-trichloroethane (DDT), lindane (γ-hexachlorocycohexan [γ-HCH]), aldrin, endrin, dieldrin, chlordane, heptachlor, toxaphane, mirex and hexachlorobenzen (HCB), one chemical for industrial use viz polychlorinated biphenyls (PCB) and polychlorinated dibenzo -pdioxins and polychlorinated dibenzofurans produced as by-products during incineration. The SC has a mechanism for banning additional chemicals and has recently included, e.g., hexachlorocyclohexanes (HCHs) and nine other (POPs). The SC became international law on 17 May 2004 (http://chem.pops.int/default.aspx) and has focused the attention of governments on the need to formulate practices on the elimination, restriction in use and management of POPs in the environment. (Abongo *et al.,* 2015) Despite the official ban or restriction on these pesticides, they are still being detected in the environment.

Currently the use of some of the organochlorine compounds is allowed under the convention, for health purposes. For example, lindane is a traditional component of lotion, creams and shampoos for the control of lice and mites in humans and in veterinarian products against ectoparasites [3]. DDT is used in the public health sector for malaria vector control. Environmental fate of these pesticides has become an issue many developing and developed countries. (Abongo *et al.,* 2015).

Phytoremediation basically refers to the use of plants and associated soil microbes to reduce the concentrations or toxic effects of contaminants in the environment and it is widely accepted as a cost-effective environmental restoration technology (Greipsson, 2011). It is a form of bioremediation and applies to all chemical or physical processes that involve plants for degrading or immobilizing contaminants in soil and groundwater (Phillips, 2018). According to Phillips, there are six types phytoremediation: phytosequestration or phytostabilization, rhizodegradation, phytohydraulics, phytoextraction or phytoaccumulation, phytovolatilization and phytodegradation.

Phillips also stated phytoextraction or phytoaccumulation is the process by which plants take up-or hyper-accumulate - contaminants through their roots and store them in the tissues of the stem or leaves and the contaminants are not necessarily degraded but are removed from the environment when the plants are harvested. This is particularly useful for removing metals from soil. The researchers aimed to look into the phytoaccumulation of soil contaminants like heavy metals and pesticide residues in twelve selected medicinal plants commonly used by the indigenous people of Rizal, Cagayan North, Northern Philippines in order to assess the safety of the use of the medicinal plants.

Material and methods

Collection of Samples

Collection of Soil. Composite samples from 10-15 cores were collected from the sampling areas. The mixed samples were transferred to separate containers and were sent to the laboratory. Each sample was labelled using waterproof marking pen and the following data were recorded: sample identification code; date samples and time sampled. Undried soil samples were sent in plastic bags to prevent drying soil and they were stored in a refrigerator at 4°C until the analyses are completed.

Mature leaves were collected for extraction purposes. In the collection area, the leaves were wiped clean and stored in vacuum sealed bags for transport to the laboratory. Collection was done in previously geotagged areas where the plants that were authenticated were collected.

Preparation of Samples

The soil samples were oven dried at 105°C- 110°C to constant weight (24 hours). After drying, the soil were crushed and pulverized and ground using a heavy duty grinder, PX – MFC 90 D by Polymix, to obtain representative sample. For long term storage samples were thoroughly dried, sealed and placed under refrigerated conditions (4°C) until required analysis was completed. Plant leaves were brushed briskly to remove visible soil and dust particles by deionized water as quick as possible. Leaves were dried and water was blotted of with filter paper, weighed and place it in an oven at 40°C for 12 to 72h. Dried samples are grinded finely in a heavy duty grinder PX - MFC 90 D by Polymix. Samples are kept in vacuum sealed bags, labeled properly and stored prior to analysis. Containers were kept in a cool dry place. For long term storage samples were thoroughly dried, sealed and placed under refrigerated conditions (4°C) until required analysis was completed.

Heavy Metal Analysis

Both soil and plant material was subjected to HNO_3 digestion using the Multiwave GO Microwave Digestion System. The presence of cadmium, chromium, & lead was tested using the Flame Atomic Absorption Spectrometry equipment by Perkin Elmer. The detection of arsenic was sourced out to the CRL Environmental Corporation.

Pesticide Residue Analysis

The test for the presence of organochlorate pesticide residue was sourced out to the CRL Environmental Corporation.

Result and discussion

Arsenic was detected above the Method Detection Limits (MDL) in the soil of the collection areas of all twelve selected Malaueg medicinal plants except in the soil of the collection area of lubigan. There was no detection of cadmium in eight out of twelve plants but the detection level of cadmium in the four plants, namely, kunig, lubigan, NPA & pansit – pansitan was below the MDL. The collection areas of all plants contained chromium above the MDL. Lead was detected above reporting levels (RL) in the collection areas of all plants except biga, liwliw, and tarattut. From the data it can be gleaned that the soil in the collection areas are contaminated with heavy metals.

No heavy metals were detected in the leaves of the plants despite the presence of these heavy metals in the soil of the collection area. All plants have the ability to absorb heavy metals and some may absorb through any part of the plants (Sabine & Griswold, 2009).

Arsenic is generally not absorbed or limited to the roots since the leaves of the plants were tested then no bioaccumulation of arsenic was observed. Uptake of cadmium is possible in plants, however, not one of the four plants with low levels of chromium absorbed it. Chromium absorption is possible in plants and all twelve medicinal plants did not absorb chromium that was above detection limits in the soil where they were collected. Above reporting limits of lead in the collection area of nine of the twelve medicinal plants showed no uptake by the plants.

It can be inferred that the plants do not have phytoaccumulation or phytoextraction abilities. Although these metals have crucial biological functions in plants and animals, sometimes their chemical coordination and oxidation-reduction properties have given them an additional benefit so that they can escape control mechanisms such as homeostasis, transport, compartmentalization and binding to required cell constituents and these metals bind with protein sites which are not made for them by displacing original metals from their natural binding sites causing malfunctioning of cells and ultimately toxicity (Jaishankar et. al., 2014). It can be deduced that the leaves of the plants may be used for medicinal purposes because no heavy metals were detected.

Table 1. The Arsenic, Cadmium, Chromium, and Lead Composition of the Soil in the Collection Area of the 12 Selected Malaueg Medicinal Plants.

Plant	Arsenic: MDL = 0.1	Cadmium: MDL = 0.4	Chromium: MDL = 1.0	Lead: RL = 1.0
Bicarud	3.6	ND	26	3.7
Biga	2.5	ND	15	ND
Gabut	2.3	ND	26	3.0
Kunig	3.2	0.2	33	12
Liwliw	2.8	ND	22	ND
Lubigan	ND	0.2	11	1.9
Matalagdaw	4.2	ND	16	12
NPA	2.4	0.2	27	18
Pansit pansitan	1.7	0.2	21	7.7
Papait	1.9	ND	36	6.9
Tarattut	1.5	ND	18	ND
Wild Tea	1.4	ND	12	1.8

Legend: MDL = Method Detection Limit/s RL = Reporting Limit/s ND = No Detection.

Table 2. The Arsenic, Cadmium, Chromium, and Lead Composition of the Leaves of the 12 Selected Malaueg Medicinal Plants.

Plant	Arsenic: MDL = 0.1	Cadmium: MDL = 0.4	Chromium: MDL = 1.0	Lead: RL = 1.0
Bicarud	ND	ND	ND	ND
Biga	ND	ND	ND	ND
Gabut	ND	ND	ND	ND
Kunig	ND	ND	ND	ND
Liwliw	ND	ND	ND	ND
Lubigan	ND	ND	ND	ND
Matalagdaw	ND	ND	ND	ND
NPA	ND	ND	ND	ND
Pansit pansitan	ND	ND	ND	ND
Papait	ND	ND	ND	ND
Tarattut	ND	ND	ND	ND
Wild Tea	ND	ND	ND	ND

Legend: MDL = Method Detection Limit/s RL = Reporting Limit/s.

Only three organochlorine pesticide residues were detected in the soil of the collection areas of the 12 selected Malaueg medicinal plants, namely: 4,4' - DDD, 4,4' – DDE, & 4,4' - DDT. The organochlorine pesticide residues absent in the soil of the collection areas are the following: Aldrin, alpha – BHC, alpha - Chlordane, beta - BHC, Chlordane (tech), delta - BHC, Dieldrin, Endosulfan I, Endosulfan II, Endosulfan sulfate, Endrin, Endrin aldehyde, Endrin ketone, gamma - BHC (Lindane), gamma - Chlordane, Heptachlor, Heptachlor epoxide, Methoxychlor & Toxaphene. 4,4' - DDD was detected above reporting limits (RL) in the collection areas of 6 out of 12 plants, 4,4' - DDE was detected above RL in the collection areas of 8 out of 12 plants, and 4,4' - DDT was detected in the collection areas of 6 out of 12 plants. No pesticide residues were detected in the soil of gabut and lubigan. 4,4' - DDD, 4,4' - DDE & 4,4' - DDT were detected in kunig, pansit pansitan, and tarattut soils. 4,4' - DDD & 4,4' - DDE were detected in bicarud, matalagdaw, and papait soils. 4,4' - DDE & 4,4' - DDT were detected in wild tea soil. Liwliw soil only had 4,4' - DDE while biga and NPA soils had only 4,4' - DDT. According to Aktar, Sengupta & Chowdhury in 2009 stated that organochlorine (OC) insecticides, used successfully in controlling a number of diseases, such as malaria and typhus, were banned or restricted after the 1960s in most of the technologically advanced countries. The soil data shows that organochlorines, particularly; DDD, DDE, & DDT, had been used in the past in the collection area. According to Agbeve, Osei – Fosu, & Carboo in 2014, OCPs have long residual action and persist in the environment. The researchers cannot make a conclusion that it is still being used today after being banned or restricted.

Table 3. The DDD, DDE, & DDT Composition of the Soil in the Collection Area of the Twelve Selected Malaueg Medicinal Plants.

Organo chlorate	Bd	Ba	G	K	Li	Lu	M	N	PP	P	T	WT
4,4'-DDD: RL=0.83	1.2	ND	ND	1.4	ND	ND	1.9	ND	1.6	1.2	4.0	ND
4,4'-DDE: RL=0.83	1.3	ND	ND	1.4	1.6	ND	2.5	ND	5.5	1.0	19	1.6
4,4'-DDT: RL=0.83	ND	1.3	ND	0.97	ND	ND	ND	0.85	3.1	ND	14	5.4

Legend: Bd = Bicarud K = Kunig M = Matalagdaw P = Papait Ba = Biga Li = Liwliw N = NPA T = Tarattut G = Gabut Lu = Lubigan PP = Pansit Pansitan WT = Wild Tea RL = Reporting Limit Unit = µg/kg.

Table 4. The DDD, DDE, & DDT Composition of the Leaves of the Twelve Selected Malaueg Medicinal Plants.

Organo chlorate	Bd	Ba	G	K	Li	Lu	M	N	PP	P	T	WT
4,4'-DDD: RL=0.83	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
4,4'-DDE: RL=0.83	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
4,4'-DDT: RL=0.83	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

Legend: Bd = Bicarud K = Kunig M = Matalagdaw P = Papait Ba = Biga Li = Liwliw N = NPA T = Tarattut G = Gabut Lu = Lubigan PP = Pansit Pansitan WT = Wild Tea RL = Reporting Limit Unit = µg/kg.

The results show that there were no organochlorine pesticide residues detected in the leaves of the plants. This indicates that the plants do not bioaccumulate the detected pesticide residues in the soil despite their long residual action and persistence in the environment. This shows that even if the pesticides persists in the environment, the twelve Malaueg medicinal plants. Its non – accumulation also indicates that the plants identified earlier and their extracts & products are safe for human consumption.

Pesticide residue determination in medicinal plant parts used in traditional medicine ensures or guarantees the quality and safety of herbal medicines in terms of its pesticide residue contamination. OCPs are characterized by high persistence, diffusion in the environment, low polarity, low aqueous solubility, high lipid solubility (lipophilicity), bioaccumulation and biomagnification in the food chain. The lipophilic nature of OCPs coupled with its low reactivity with respect to light and chemicals as well as its low biological degradation rates have led to OCPs accumulation in biological tissues and subsequent biomagnification in organisms progressing up the food chain (Agbeve *et al.*).

Conclusion

The non – accumulation of heavy metals and pesticide residues by the plants points out to the safety of the leaves of the plants for use in preparation of products like personal and home care products, herbal ointment, cream & lotion, functional food, etc.

Recommendation

It is recommended that a test for the presence of other pesticide residues like organophosphates, carbamates, and the like be also conducted. Furthermore, a test for the presence of heavy metals and pesticide residues be also conducted on the roots and the stems of the plants.

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