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Heavy metal contamination in South African medicinal plants: A cause for concern

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ABSTRACT

The quality and safety of herbal medicines is becoming a major concern worldwide particularly due to contamination by heavy metals. The present study quantified the levels of heavy metals in frequently used South African medicinal plants and determined the variations in certain biological activities and phytochemical compositions. Eleven plant species were obtained from both *muthi* shops (MS) (commercial outlets) and from open street markets (OSM) for comparison. Samples were dried, powdered and digested using microwave acid-assisted digestion. The digested solutions were analysed for heavy metals using Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES). Plants were classified based on their elemental composition using chemometric techniques. Powdered plant samples were extracted using 70% acetone and screened for antibacterial activity against *E. coli* and *S. aureus*. Phytochemical analyses were carried out to determine total phenolic and flavonoid content. Of the 22 samples analysed, *Bulbine natalensis* obtained from OSM and *Alepidea amatymbica* obtained from MS exhibited high levels of Al [5559 and 4392 mg/kg dry weight (DW)] and Fe (4164 and 4465 mg/kg DW) respectively. Levels of As and Hg were above the World Health Organization permissible limits in most of the samples analysed. Hierarchical cluster analysis classified the samples into four groups based on their metallic analyte concentrations. Group one having low metal content and group four having a high metal content. In general, plant samples with high levels of metals yielded greater antibacterial activity. However, antibacterial activity recorded in this study is not an indicator of high levels of heavy metal contamination as some samples despite the high levels of metal exhibited low antibacterial activity. The variations in the amounts of phenolics and flavonoids in the evaluated samples could have probably been that some of the plant samples may have been harvested from different localities or at different times of the year, perhaps plant age or degree of storage. The results highlighted the need for in-depth risk and quality assessments.

1. Introduction

The quality and safety of herbal medicines are major concerns worldwide due to increasing heavy metal contamination resulting from anthropogenic activity. Adverse effects of herbal medicines contaminated with heavy metals are well documented (Ernst, 2002; Steenkamp et al., 2006; Dargan et al., 2008). In South Africa, cases of metal poisoning associated with the use of traditional medicines are common, with arsenic (As), chromium (Cr) and magnesium (Mg), the most frequently implicated metals resulting in poisoning, morbidity and mortality (Steenkamp et al., 2002). Thus, the screening of traditional medicines for potentially harmful components has been recommended to protect consumers (Chang, 1995).

Some medicinal plants have the ability to accumulate heavy metals when grown in contaminated soils. The addition of heavy metals in herbal medicinal products through deceitful practices such as adulteration for alleged increase in therapeutic properties is also well documented (Ernst, 2002; Chan, 2003; Haider et al., 2004). Thus stringent legislation on the production and processing of herbal medicine as well as detailed information on herbal products is of paramount importance to protect consumers. One of the most common practices in South Africa is the collection of medicinal plants from wild populations (Zschocke et al., 2000). This not only threatens the ecological balance but also leads to safety concerns as a result of heavy metal contamination arising from industrial encroachment. In South Africa, medicinal plants are commonly sold at informal open street markets (OSM) or indoor *muthi* shops (MS). The OSM are usually positioned in the hub of the city centre to allow easy access to commuters. This exposes herbal material to various kinds of urban pollutions such as industrial and vehicular emissions. Numerous efforts have been made by the South African Medicines Regulatory Authority in terms of regularizing the practice of South African Traditional Medicine. However, issues regarding the procurement, processing and sale of herbal products are still major

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concerns. The source and quality of raw materials as described in good agricultural and collection practices (GACP) and good manufacturing processes (GMP) are essential steps for quality control of herbal medicine. These will play a pivotal role in guaranteeing the quality and stability of herbal products (WHO, 1998). According to WHO (2007), the concentration of trace elements must be controlled in medicinal plants in order to meet and improve quality assurance and safety.

Heavy metal contaminations in medicinal plants affect the phytochemical composition as well as the biological activities and thus influence the efficacy of medicinal plant products (Street, 2012). Zahid et al. (2011) reported that the presence of heavy metals in the medicinal plant *Marsilea minuta* reduced antimicrobial activity by two possible mechanisms either by changing the amounts of bioactive compounds in the plants or by deactivating the bioactive compounds through chelation of metal ions. There is insufficient information linking the quality, safety concerns and efficacy measures with regard to heavy metals in South African medicinal plants.

The aim of this study was to quantify heavy metals in some frequently used South African medicinal plants obtained from both herbal shops and outdoor markets and to classify the samples based on their elemental composition using chemometric techniques. Plant samples were also assessed for efficacy by screening the antimicrobial activity and quantifying phenolic and flavonoid contents.

2. Materials and methods

2.1. Sample collection and preparation

Eleven plant species were obtained from both MS and OSM in Pietermaritzburg, KwaZulu-Natal, South Africa. Voucher specimens were prepared and are deposited at the John Bews Herbarium, University of KwaZulu-Natal, Pietermaritzburg. Information with regards to the use of the selected medicinal plants is presented in Table 1. All plant materials were washed under running tap water, oven dried at 50 °C and ground into powders. The powdered samples were used for analysis.

2.2. Reagents and solutions

All reagents used were of analytical grade. Ultra-pure (UP) water was used for preparing the solutions and dilutions. Stock solutions of metals (1000 mg/L) were prepared from their nitrate salts.

2.3. Microwave acid-assisted digestion

A microwave acid-assisted system was used to digest plant material. Plant samples (0.5 g DW) were placed in Teflon vessels and 10 mL 55% HNO_3 added. The vessels were heated in a microwave system operating up to 1200 W. The temperature of the microwave system was programmed as follows: 1st stage, 2 min heating from ambient to 170 °C; 2nd stage, 3.30 min heating to 180 °C and 3rd stage, 9.30 min held at 180 °C. After complete digestion, the clear solutions were transferred to 50 mL volumetric flasks and made to volume with UP water. These samples were stored in high density polyethylene bottles until analysis. A blank containing 10 mL 55% HNO_3 was used.

2.4. Elemental analysis using ICP-OES

An Inductively Coupled Plasma-Optical Emission Spectroscopy (ICP-OES) (Varian 720-ES, Varian Inc, Palo Alto, CA, USA) instrument was used for elemental analysis. ICP-OES provides a multi-elemental analysis and supports a broad linear calibration range. ICP-OES is a powerful tool that is used in determining metal concentrations in a variety of different sample matrices (Hou and Jones, 2000). The operating conditions for the ICP-OES instrument were as follows: RF power 1.0 kW; viewing geometry axial; Argon gas used as plasma gas flow at the rate of 15.0 L min^{-1}; auxiliary gas flow rate 1.50 L min^{-1}; nebulizer gas flow rate 0.75 L min^{-1}; and replicate reading time 9.0 s. All analyses were performed in triplicate.

2.5. Sample preparation for antibacterial and phytochemical screening

The ground material (1 g DW) was extracted with 70% acetone in a sonication bath (Julabo GmbH sonicator) for 1 h. The plant extracts were then filtered using Whatman No. 1 filter paper. The filtrates were concentrated by a rotary evaporator and then air-dried under a stream of cold air. The dried extracts were kept in the dark at 10 °C until ready for use. The dried extracts were resuspended in 50% methanol and used for the phytochemical analysis.

2.6. Antibacterial microdilution assay

Antibacterial activity of the plant extracts was determined using the minimum inhibitory concentration (MIC) technique (Eloff, 1998; Ndhlala et al., 2009). Overnight cultures of a Gram-negative (*Escherichia coli* ATCC 11775) and a Gram-positive (*Staphylococcus aureus* ATCC 12600) bacterial strains were used. Neomycin was used as the positive control and 70% acetone was used as the negative control. The MIC was recorded as the concentration of the last well in which there was no bacterial growth. The assay was repeated twice in duplicate for each extract.

Table 1
Selected South African medicinal plants investigated in this study and their uses in treating various ailments.
Information presented in this table was sourced from Van Wyk et al. (2008) and Hutchings et al. (1996).

Plant family	Species	Voucher number	Plant part	Traditional uses
Celastraceae	*Cassine transvaalensis* (Burtt Davy)	A Okem 10 NU	Stem bark	Infusion is used for stomach pain and haemorrhoids and as enemas
Hypoxidaceae	*Hypoxis hemerocallidea* Fisch. & C.A. Mey.	A Okem 21 NU	Tuber	Immune booster of HIV patients, treating prostate hypertrophy and urinary tract infections, treating cancer and as emetics
Myrsinaceae	*Rapeana melanophleos* (L.) Mez	A Okem 14 NU	Stem bark	Decoction used as expectorants and emetics, stomach disorders, muscular pains and to strengthen the heart
Asphodelaceae	*Bulbine natalensis* A. Rich	A Okem 18 NU	Root	Back pain and kidney diseases, venereal diseases, diabetes, rheumatism and to purify blood
Passifloraceae	*Adenia gummifera* (Harv.) Harms	A Okem 20 NU	Stem	Infusion used as emetics, decoction for malaria
Apiaceae	*Alepidea amatymbica* Eckl.	A Okem 16 NU	Root	Decoction used for colds and chest complaints, as well as for asthma, influenza and abdominal cramps
Hyacinthaceae	*Drimia elata* Jacq. Ex Willd.	A Okem 19 NU	Bulb	Bulb scales are rubbed on chest for stabbing pain, infusion used as emetic, expectorants and diuretic
Lycopodiaceae	*Lycopodium clavatum* L.	A Okem 8 NU	Whole plant	Kidney stones, urinary tract infections, gastric inflammations, for lung and bronchial disorders and fevers
Lauraceae	*Ocotea bullata* (Burch.) E. Mey.	A Okem 9 NU	Stem bark	Bark infusion for headache, nervous disorders, stomach pain and urinary disorders
Hyacinthaceae	*Schizocarphus nervosus* (Burch.) Jessop	A Okem 15 NU	Bulb	Infusion for enema and rheumatic fever
Cucurbitaceae	*Momordica balsamina* Schumach.	A Okem 13 NU	Root	High blood pressure, diabetes, stomach disorders, malaria and hepatitis B