

Levels of Potentially Toxic Metals in Synthetic Industrial Essential Oils Utilized in Northern Nigeria.

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ABSTRACT

In view of the growing concern of the modes of application of synthetic industrial essential oils (SIEOs) in Northern Nigeria, twenty (20) typical samples were investigated for levels of potentially toxic metals (Al, Cd, Cr, Mn, Pb, and Zn). The standard calibration method of flame atomic absorption spectrophotometry (FAAS) was used to determine the metals. Ni ($1.93 \pm 0.62 \mu\text{g/g}$) showed the highest average concentration while Cr ($0.14 \pm 0.06 \mu\text{g/g}$) was the least. Inter-metal Pearson correlation coefficient (r) values between all seven metals determined revealed that the highest positive correlation ($r=0.73$) was recorded between Cd and Zn. Similarly, a high anti-correlation ($r = -0.63$) was observed between Zn and Al. All other metals were within the prescribed limits for cosmetic products except Ni, Pb, and Al. However concentration ranges are very likely to amplify when the entire range of products utilized are considered indicated higher health and environmental risk from the continuous direct application of this products in Northern Nigeria.

(Keywords: synthetic fragrance, direct application, toxic metals, human health, northern Nigeria)

INTRODUCTION

In view of the growing concern of the modes of application of synthetic industrial essential oils (SIEOs) in Northern Nigeria, there is the need also to analyze this product for levels of potentially toxic metals. Potential toxic metals are metals with harmful effects especially on living organism. Their harmful effects or toxicity usually arise from factors such as their presence in a system where they normally do not exist and unwanted or presence in larger concentrations than normal. Otherwise, these metals are natural

occurring, constituting more than half of the period table, besides which they are found in different human and environmental matrices to be useful. In other words, anthropogenic activities, in most part have led to their dislocations and accumulations to harmful levels. However, studies have revealed that while some of the metals are classified as essential trace metals (e.g. Cr, Fe, Co, Cu, Zn, Se, and Mo), some of them; Mn, Si, Ni, V, and Sn are probably essential trace metals; and further some of them As, Cd, Pb, Al, and Hg are considered potentially toxic, some possibly essential elements for animal and human life. In fact, all essential metals may also be toxic in animals and humans if ingested at sufficiently high levels and for a long enough period (Manahan, 2003; Myers, 2003; Aras and Ataman, 2006).

Due to their being natural components of the Earth's crust; they cannot be degraded or destroyed, therefore are dangerous because they tend to bioaccumulate, i.e. they accumulate in living things any time they are taken up and stored faster than they are broken down (metabolized) or excreted (Abdulla and Chmielnicka, 1990; Zedeck, 2004; Lenntech, 2006). The inorganic forms of most metals tend to be strongly bound by protein and other biologic tissue. Such binding increases bioaccumulation and inhibits excretion. There is a significant amount of tissue selectivity in the binding of metals which have also been reported to be responsible for carcinogenic, systemic, immunological, and developmental effects (Manahan, 2003; 2005). The toxicological profiles for each of the metals studied in this work have been well documented by the Agency for toxic substances and diseases registry (ATSDR). For Aluminum (ATSDR, 1999a), Cadmium (ATSDR, 1999b), Chromium (ATSDR, 2000a), Manganese (ATSDR, 2000b), Nickel (ATSDR, 2005a), Lead (ATSDR, 1999c) and Zinc (ATSDR, 2005b).

The sources of metals in essential oils may be from catalytic agents, complex organic compounds and as trace impurities from natural sources (Zheljzakov and Jekov, 1994). Traces of toxic metals have been found to be present as residues of catalytic industrial processes or as components of petroleum and petrochemical raw materials used in the production of synthetic materials (Al-Swaidan, 1993; Essoka et al., 2006). It was also suggested that where proper industrial recovery, removal or reduction procedures are not applied to such products, the likelihood of such heavy metals exceeding permissible limits may be pronounced.

The direct application of SIEOs in Northern Nigeria entails 'neat', straight; undiluted or unmodified use of the product in its full strength in personal and household care such as perfuming hair, body, clothes, bed, and floor and as room air freshener, bath and wood burning admixes. Through these various forms of application, toxicants can enter the human body via, especially inhalation and dermal absorption through the skin. On the other hand, volatilization, burning or combustion and domestic wastewater effluents, these toxicants can be released into the environment (Zedeck, 2004). Therefore this work intends to provide information of the presence and concentration levels of these toxic metals thereby elucidating the extent of risks associated with the frequent direct application of SIEOs in Northern Nigeria.

MATERIALS AND METHODS

Study Area, Samples and Sampling: The study area comprises the three major points of distributions of SIEO products within Northern Nigeria: Sokoto, Kano and Maiduguri. A total of 75 SIEO samples were collected for this study over a two year (2005 and 2006) period. This followed the detailed survey of the forms of applications of SIEOs in Northern Nigeria as described by Hati (2005a) and Hati *et al.* (2008). Three major categories of SIEOs applications were identified. These are socio-cultural, fashion and industrial formulations. Due to the wide range of these products, samples collected for this work was obtained from narrowed statistical data of volumes of annual output distribution of the products. This was obtained from manufacturers' distributors in the study area. Thus, only SIEO products ≥ 4500 tons/yr were collected for analysis. Twenty (20) typical SIEO products

qualified. For each of these, a composite (100ml) of five different batches of 20ml each was collected directly from product manufacturers distributors by decanting into well labeled sterile glass bottles.

Sample and Standard Preparations: The 20 SIEO samples were prepared according to the wet ashing procedures described by Vogel (2000) and SC (2000). To each sample, 5ml was placed in separate 100ml Kjeldahl flasks and heated at about 60°C for 1hr to reduce presence of volatile organic compounds. This was then treated with 6ml hot concentrated nitric acid (HNO₃, BDH, England) and later 4ml hot concentrated perchloric acid (HClO₄, BDH, England). After heating to a clear solution, the resulting content made up to the 100ml mark using distilled water. Standard calibration solutions of each metal analyte (Al, Cd, Cr, Mn, Ni, Pb, and Zn) were prepared according to SC (2000) from a stock concentration of 1000 μ g/g of each metal analyte to cover the optimum absorbance range for the standard calibration curve. Blank solutions were also prepared accordingly.

Flame Atomic Absorption Spectrophotometric Analysis (FAAS): The standard calibration method of FAAS was used to determine Al, Cd, Cr, Mn, Ni, Pb, and Zn in the 20 SIEO samples, using the Shimadzu AA-6800 equipped with ASC-6100 auto sampler and air-acetylene atomization gas mixture. Results obtained were statistically analyzed using coupled Microsoft Excel+Analyse-it® version 1.73 (2006). Variations were considered significant at $p < 0.05$.

RESULTS

The levels of the seven metals determined in the samples showed variant concentrations and in some cases were not detected. But over 20% of the samples showed significant ($p < 0.05$) variations in the metal contents. Table 1 shows the summary result of metal concentrations (Mean \pm SD) in the 20 typical SIEO samples analyzed. Ni (1.93 \pm 0.62 μ g/g) showed the highest average concentration while Cr (0.14 \pm 0.06 μ g/g) was the least. The Box-Whisker plot (Figure 1) shows graphically, the increasing order of concentration spread of each of the metals in the samples. It is evident from the plot also that Ni recorded the highest and least (4.847 \pm 1.002 and 0.002 \pm 0.000 μ g/g) concentrations and hence the widest in range of metal content. Table 2

describes the inter-metal Pearson correlation coefficient (r) values between all seven metals determined in the SIEO samples. This shows the extent of association of the metals in the samples. Thus the highest positive correlation ($r=0.73$) was recorded between Cd and Zn. Similarly, a high anti-correlation ($r = -0.63$) was observed between Zn and Al, while the least correlated, $r = 0.06$, was between Cr and Zn.

Table 1: Metal Concentration ($\mu\text{g/g}$) in SIEOs.

Metal	Mean	SD	%ND
Cr	0.14	0.06	11.11
Mn	0.39	0.07	0.00
Zn	0.52	0.08	11.11
Al	0.60	0.12	33.33
Cd	0.61	0.01	11.11
Pb	1.20	0.23	5.26
Ni	1.93	0.62	11.11

SD = Standard deviation
ND = Not detected

Table 2: Pearson Correlation Coefficient (r) Values Between Metals in SIEO Samples.

	Cr	Cd	Mn	Zn	Al	Pb	Ni
Cr							
Cd	-0.10						
Mn	0.47	0.20					
Zn	0.06	0.73	0.17				
Al	-0.36	-0.62	-0.41	-0.63			
Pb	0.12	-0.30	-0.14	-0.15	0.36		
Ni	-0.28	-0.43	-0.14	-0.26	0.24	0.31	

DISCUSSION

Generally, the results of metal contents in this study shows significant variations between the SIEO samples investigated.

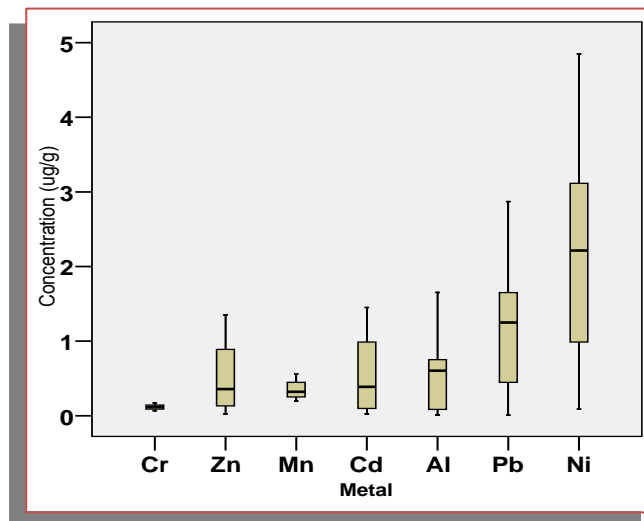


Figure 1: Box-Whisker plot of Metals Content in 20 Typical SIEO Samples.

These variations are very likely to amplify immensely from one sample to another when the large number of various products and versions used in Northern Nigeria are considered since only less than 50% of the total product range of SIEOs that can be found in Northern Nigeria market were investigated in this study. Significant factors such as differences in batch production (Byrne and O'Grady, 1990), essential oil formulations (RUK, 2004), which include the economic considerations in the proliferations of lower SIEO versions; physical and chemical changes through production, transportation, storage and exposure may be responsible for the observed variations.

As anticipated, the result of Ni and Pb showed the highest concentrations in the SIEO samples studied. The most probable reason for this is in their widespread applications in industrial catalytic processes, such as the hydrogenation of oils (Ni) and coloration of products (Ni and Pb) (Spectrum, 2006). However, the result of levels of Ni in this study show correspondence to that of Al-Swaidan (1993) but far less than that reported by Pera et al. (2003) and Zhejzakov and Jekov (1994) for heavy metals contents in natural essential oils.

However, with exception of Ni, Pb, and Al, all other heavy metals studied in this research were within the prescribed limits given for heavy metals in cosmetic products (NAFDAC, 2003).

Inter-metal correlation result showed high correlations between Cd and Zn, this perhaps represent commonalities in their concentration association, applications in the manufacturing or sources in the raw materials for SIEOs.

SIEOs have become highly acceptable due to the relatively low cost of production, continuously available alternative and the demand for large quantities of fragrant compounds for the growing industrial processes requiring the use of fragrant compounds. Both "nature identical" and newer fragrant compounds are being synthesized, rapidly reducing the dependence on natural sources in plants essential oils. Also, for carrier oils, petroleum products are now used as alternatives. Presently, more than half of the over four thousand raw materials available for fragrance production for industrial essential oils are now synthesized. They mostly serve as raw materials for the vast range of personal and household care products manufactured (Fortineau, 2004; Turin and Yoshii, 2003).

In Northern Nigeria These products are utilized in high volume as a source of fragrance materials. The socio-cultural forms of application, which include burning SIEOs in concoctions with other fragrant resins and wood and the prevalent practice of multi-product combinations commonly called Uhmra, which are usually allowed to stand for several days before direct application on the skin. However, through these various forms of application, toxicants can enter the human body via, especially inhalation and dermal absorption through the skin. On the other hand, volatilization, burning or combustion and domestic wastewater effluents, these toxicants can be released into the environment (Zedeck, 2004).

Thus brief notes are here given on the toxicological profiles of each of metals studied in this work as given by ATSDR.

Aluminum: Skin damage has been observed in female TF, Carworth mice, New Zealand rabbits, and Large White pigs following the application of 10% aluminum chloride (0.005-0.1 g Al) or aluminum nitrate (0.006-0.013 g Al) for 5 days; but not from aluminum sulfate, hydroxide, acetate, or chlorhydrate. The damage consisted

of hyperplasia, microabscess formation, dermal inflammatory cell infiltration, and occasional ulceration. These results suggest that the development of adverse dermal effects from exposure to aluminum depends upon its chemical form. Aluminum is not bioaccumulated to a significant extent. Notable exceptions include some herbs and the tea plant, which can accumulate aluminum to 3,000-4,000 ppm and to 10,000 ppm respectively. Aluminum does not appear to accumulate to any significant degree in cow's milk or beef tissue and is, therefore, not expected to undergo biomagnification in terrestrial food chains. Similarly, because of its toxicity to many aquatic organisms, including fish, aluminum does not bioconcentrate in aquatic organisms to any significant degree. In order to bioaccumulate in the food chain, a substance cannot be acutely toxic to links in the chain; otherwise, the bioaccumulation stops (ATSDR, 1999a).

Cadmium: According to ATSDR (1999b) Cadmium is widely distributed in the body, with the major portion of the body burden located in the liver and kidney. Animals and humans appear to have a similar pattern of distribution that is relatively independent of route of exposure, but somewhat dependent on duration of exposure. About 40-60% of the cadmium in inhaled smoke can pass through the lungs into the body. Higher amounts of cadmium can enter the body from cadmium in air or smoke (25 to 60% of the cadmium present) than from cadmium in foods (about 5-10% enters the body). The cadmium not taken into the body through the lungs is breathed out. The cadmium not taken into your body from food or water leaves your body in feces. The potential for cadmium to harm human health depends upon the form of cadmium present, the amount taken into the body, and whether the cadmium is eaten or breathed. Breathing air with very high levels of cadmium can severely damage the lungs and may cause death. Breathing air with lower levels of cadmium over long periods of time (for years) results in a build-up of cadmium in the kidney, and if sufficiently high, may result in kidney disease. Other effects that may occur after breathing cadmium for a long time are lung damage and fragile bones.

Chromium: Chromium (III) has been described as an essential nutrient that helps the body use sugar, protein, and fat (Hati et al., 2005b). However under certain environmental conditions (Awan et al., 2003) and certain metabolic transformations (ATSDR, 2000a), chromium (III)

may be readily oxidized to chromium (VI) compounds that are toxic to human health. In general, chromium (VI) is more toxic than chromium (III). Breathing in high levels (>2 µg/m³) chromium (VI), such as in a compound known as chromic acid or chromium (VI) trioxide, can cause irritation to the nose, such as runny nose, sneezing, itching, nosebleeds, ulcers, and holes in the nasal septum. Long-term exposure to chromium has been associated with lung cancer in workers exposed to levels in air that were 100 to 1,000 times higher than those found in the natural environment. Lung cancer may occur long after exposure to chromium has ended. Chromium (VI) is believed to be primarily responsible for the increased lung cancer rates observed in workers who were exposed to high levels of chromium in workroom air. Breathing in small amounts of chromium (VI) for short or long periods does not cause a problem in most people. However, high levels of chromium in the workplace have caused asthma attacks in people who are allergic to chromium (ATSDR, 2000a).

Manganese: According to ATSDR (2000b) Manganese and manganese compounds exist naturally in the environment as solids in the soil and as small particles in water. Manganese can be released into the air by industry and by the burning of fossil fuels and related substances such as SIEOs. Too much manganese may also cause serious illness. Most manganese compounds seem to cause the same effects, although it is unknown whether exposure to different manganese compounds results in slight differences in adverse effects. The symptom of excess manganese results in the disease called 'manganism.' Manganism occurs because too much manganese injures a part of the brain that helps control body movements. Some of the symptoms of manganism may improve upon certain medical treatments, but the improvements are usually temporary, and the brain injury is permanent.

Nickel: Detailed toxicological profile for nickel has been well documented by ATSDR (2005a). It is stated that the major sources of exposure to nickel are food, breathing air, drinking water, or smoking tobacco containing nickel. Skin contact with soil, bath or shower water, or metals containing nickel, as well as, metals plated with nickel can also result in exposure. The most common harmful health effect of nickel in humans is an allergic reaction. Approximately 10–20% of the population is sensitive to nickel. A person can

become sensitive to nickel when jewelry or other items containing nickel are in direct contact and prolonged contact with the skin. The most common reaction is a skin rash at the site of contact. In some sensitized people, dermatitis (a type of skin rash) may develop in an area of the skin that is away from the site of contact. Other serious harmful health effects from exposure to nickel, such as chronic bronchitis, reduced lung function, and cancer of the lung and nasal sinus, have occurred in people who have breathed dust containing certain nickel compounds while working in nickel refineries or nickel processing plants.

Lead: According to ATSDR (1999c), no proof that Pb causes cancer in humans but kidney tumors have developed in rats and mice given large doses of Pb. The animal studies have been criticized because of the very high doses used, among other things.

Zinc: Detailed toxicological profile for zinc is also well documented in ATSDR (2005b). Zinc occurs mostly as zinc sulfide ore and compounds are widely used in industry. Some of the uses of zinc compounds such as zinc acetate, zinc chloride, and zinc sulfate, are used in preserving wood and in manufacturing and dyeing fabrics. Others are used by the drug industry as ingredients in some common products, such as vitamin supplements, sun blocks, diaper rash ointments, deodorants, athlete's foot preparations, acne and poison ivy preparations, and antidandruff shampoos. Taking too much zinc into the body through food, water, or dietary supplements can also affect health. Skin irritation from exposure to these chemicals would probably occur in humans.

CONCLUSION

The levels of potentially toxic metals analyzed in SIEOs revealed varying concentration ranges. Ni, Al and Pb were above permissible limits. However concentration ranges are very likely to amplify when the entire range of products utilized are considered indicating higher health and environmental risk from the continuous direct application of this products in Northern Nigeria.

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