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Systematic Investigation of Heavy Metal Assays in Indian Fashion Jewellery

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Abstract:
The assessment of heavy metals viz., aluminium, cadmium, chromium, cobalt copper, iron, lead, mercury, molybdenum, nickel, tungsten and zinc, present in fashion jewellery widely used in SAARC Countries were analyzed by Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) method. The observed ICP-OES results indicate that most of the fashion jewellery items contain significant levels of highly toxic heavy metals, which are in excess of WHO, USEPA and CPCB standards. This can cause adverse toxicological impact on human health upon prolonged exposure/ contact.

Keywords: *Heavy metals, fashion and fancy jewellery, health hazards, ICP –OES*

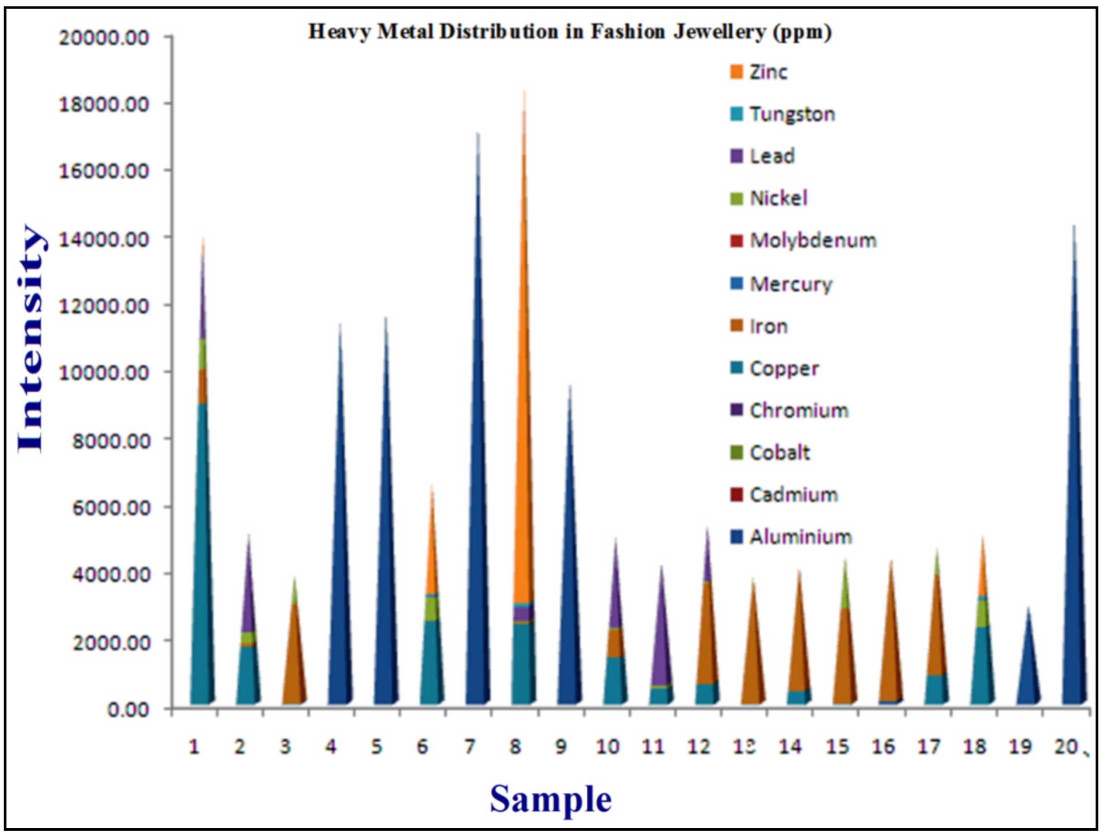

Figure 1

1. Introduction

Ever since human civilization began, jewellery has been one of the fascinating aspects of Indian women. Indian women are much fond of wearing chains, studs, hairpins, rings, bangles, etc. Jewellery made with noble metals such as gold, silver, platinum and diamonds have become very popular due to their luster and rust free nature their can be worked into ornaments of almost any shape. In modern days, people have inclined towards the fashion jewellery manufactured using various heavy metals. These metals are cheaper and also they are alternative to noble metals. Indian women desire for fashion jewellery is unmatched although they are unaware of the ill effects caused on human health. In the recent past, fashion jewellery has moved into the limelight of environmental pollution and occupational health hazards, which greatly focuses on the allergic effect of the heavy metals.

The studies on heavy metals in food, cosmetics, fashion jewelleries, etc., are of interest due to their inherent toxic nature. Heavy metals like cadmium, lead, nickel, chromium, mercury, etc., are cumulative poisons and are exceptionally toxic at certain levels. (Turkdogan et al. 2003; Kawata et al. 2007; Liu et al. 2009). Even at lower concentration, some of these elements cause damage to internal organs of the living species. Various forms of mammalian cancers, respiratory diseases, organ failures and retardation of the intellect had been edified as due to metal poisoning (Hall 2002; Davydova S.L 1999; Brams et al.1989). For instance, an increased incidence of certain kinds of cancer would have perhaps occurred as a result of direct inhibition of DNA which mismatches remediation by cadmium (Mc Murray et al. 2003; El-Safty et al.2007). The toxicity of lead at high levels of exposure is well known, but a major concern as of today is the possibility that continual exposure to relatively low levels of lead may entail adverse health effects like damage of nervous system (Bergback et al. 1992; Koller et al. 2004). Lead and cadmium are two potentially harmful metals that have aroused considerable concern. In fact, lead has been described as the most severe environmental contaminant to arise in human civilization (Smith et al 1995). Zinc has been reported to cause the same signs of illness. Although individual metals exhibit specific signs of their toxicity, the following have been reported as general signs associated with cadmium, copper and zinc poisoning, namely gastrointestinal disorders, diarrhea, stomatitis, tremor, ataxia, paralysis, vomiting and convulsion, depression, and pneumonia when vapors and fumes are inhaled (Duruibe, et al., 2007). Human exposures to these xenobiotics from diverse sources have been widely investigated. These usually involve industrial and medical wastes (Dorigo et al 2004) pesticide, petroleum by-products (Mowat et al. 2001) beverages (Maduabuchi et al.2008), snacks and confectioneries (Narin et al. 2005; Gopalani et al. 2007) foods (Mahaffey et al. 1975) and cosmetics (Ayenimo et al. 2010)

These reports indeed discussed the potential health implications of heavy metals through ingestion and inhalation of contaminated air, foods or drinks. Information on the exposure to metal toxins through dermal contact is very scanty, and few or no data exists on the personal care products concentration of cadmium, chromium, copper and zinc in the world. These metals are cumulative poisons and exceptionally toxic (Ellen et al. 1990). However, a few investigations have been carried out on the domestic and household sources of metal exposure in the developed countries (Friberg et al.1986; Alloway 1995; Narin et al. 2005; Fiala Hr. 2006; Gopalani et al. 2007; Lin et al. 2009). The findings were based on corrosion of metal plumbing fittings, galvanized roofs, health care products, saucepans, utensils, recycled papers, children ski helmets and housing paint as significant sources of metal exposure. No attempts have been made to investigate fashion jewellery items, which affects directly on human body. The belief is that human exposure to heavy metals is usually from contaminated air, soil, water or food. The presence of trace organic compounds such as polychlorinated biphenyls (Storr – Hansen et al. 1988) and organosilicone compounds (Horii and Kannan 2008) in personal care and house hold products is some indirect evidence of possible heavy metals additives in these products. Heavy metals and trace of organic compounds have been identified as common pollutants in diverse environmental matrices (Malawska.and Wiłkomirski 2001). Hence, we herein report the heavy metal (aluminium, cadmium, chromium, cobalt copper, iron, lead, mercury, molybdenum, nickel, tungsten and zinc) analysis of fashion jewellery products like studs, hairpins, chains, rings and bangles by ICP-OES method.

2. Materials and Methods

The commonly used fashion jewellery samples were purchased from some domestic market in India. The samples include hair pins, earrings, rings, bangles, chains. Twenty different fashion jewellery samples acquired were subjected for the analysis. These items were taken to the laboratory for chemical analysis. All the reagents used were of analytical grade (BDH, India) chemical.

The samples were made into small pieces and exactly 2g of each sample are digested three times in 50 ml of nitric acid. The residue obtained due to the digestion was dissolved in 6% HNO_3 and made up to 100 ml. The solution contains 20000 ppm of elements, since 2g of acid digested sample is made up in 100ml. The sample solution were analyzed for heavy metals such as aluminium, cadmium, cobalt, chromium, copper, iron, mercury, molybdenum, nickel, lead, tungsten and zinc using ICP OES available at the SAIF IITM, Chennai.

3. Results and Discussion

Heavy metal exposures from fashion jewellery have largely been overlooked. This study is to evaluate metallic concentration in fashion jewellery, which has direct contact with human body. From the ICP-OES studies it is reported that the samples contain considerable levels of metals like aluminium, cadmium, cobalt, chromium, copper, iron, mercury, molybdenum, nickel, lead, tungsten and zinc. Results indicate wide range of concentration for these elements within each class placed in Table 1.

From the ICP-OES data some of the samples contain non hazardous additives which escaped during the digestion process

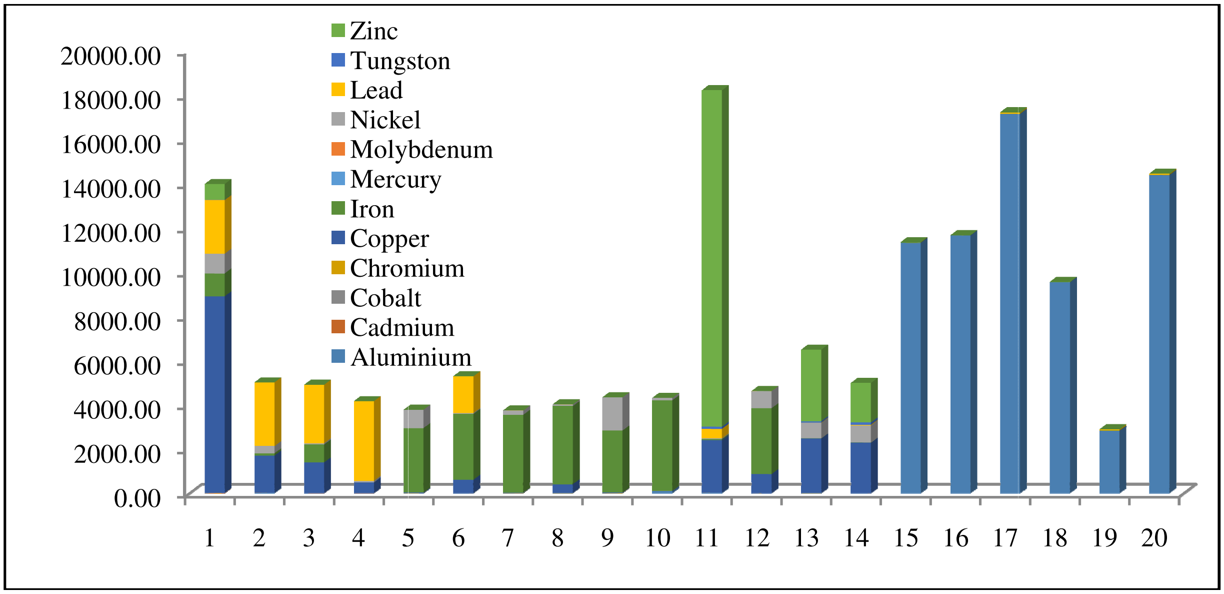

Figure 2: Indicative figures of Heavy Metal Distribution in Fashion Jewellery in ppm.

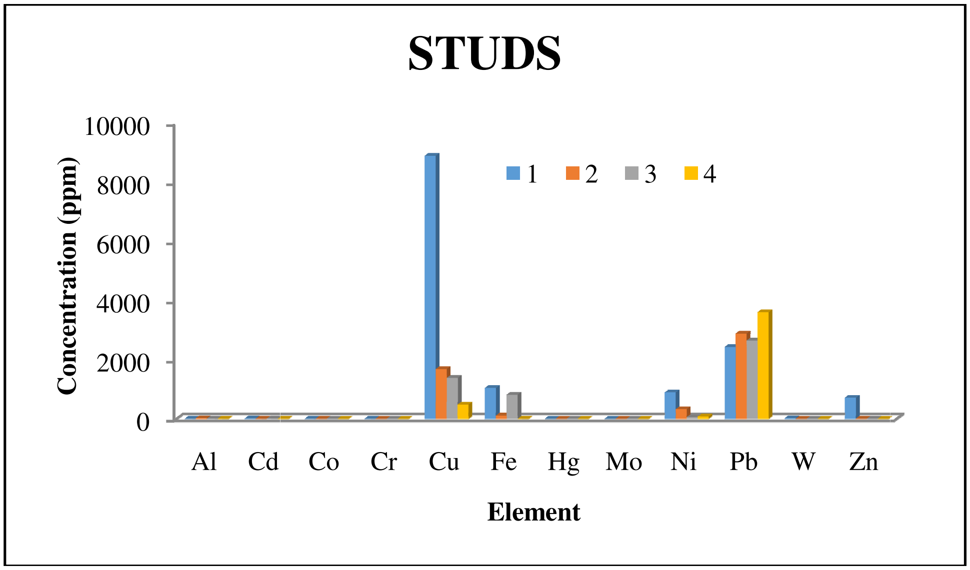

Figure 3: Heavy metal contents in Stud Samples (1 – 4)

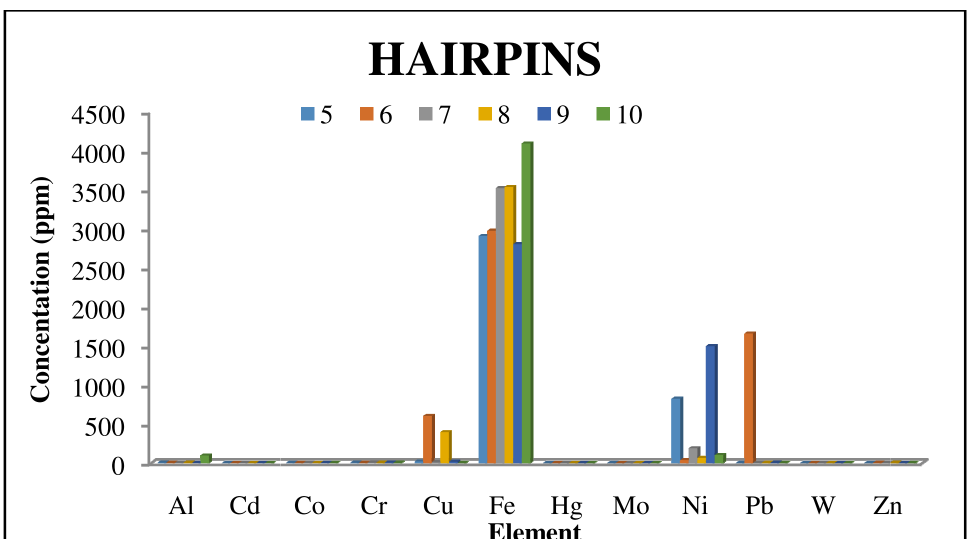

Figure 4: Constituent of major Elements in Hairpin Samples (5 – 10)

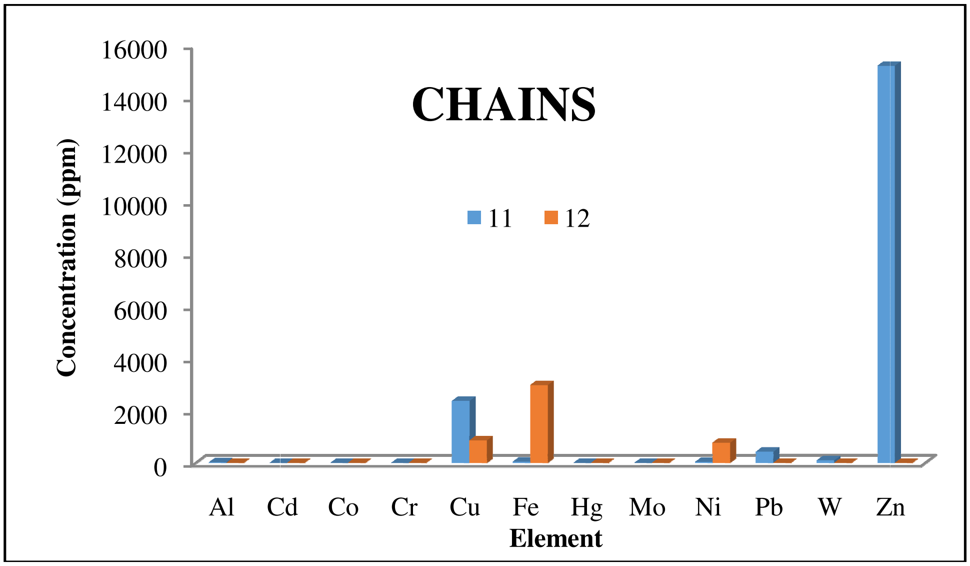

Figure 5: Zinc Dispensation in Chains (11 – 12).

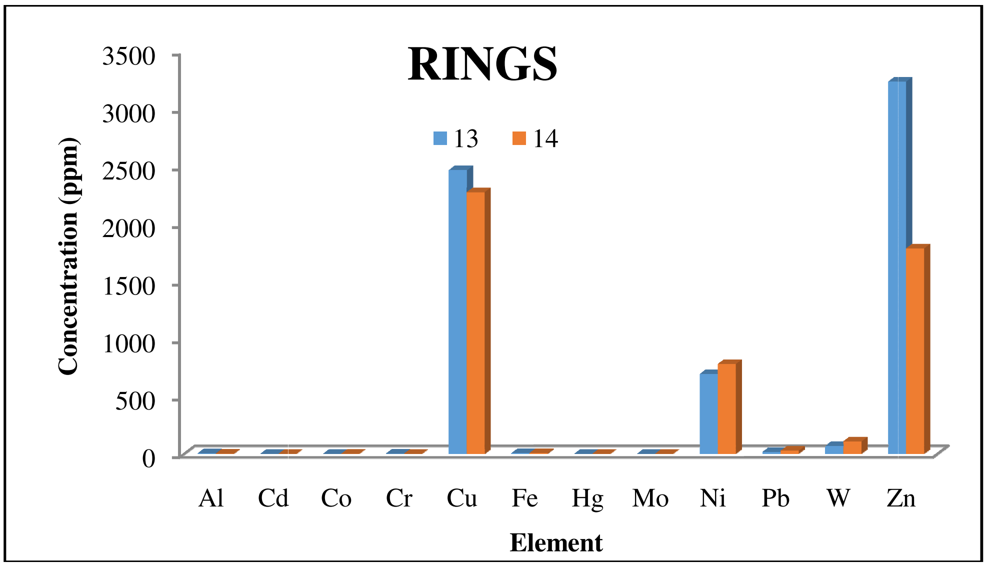

Figure 6: Schematic view of Copper, Nickel and Zinc Metals Present in Rings (13 – 14).

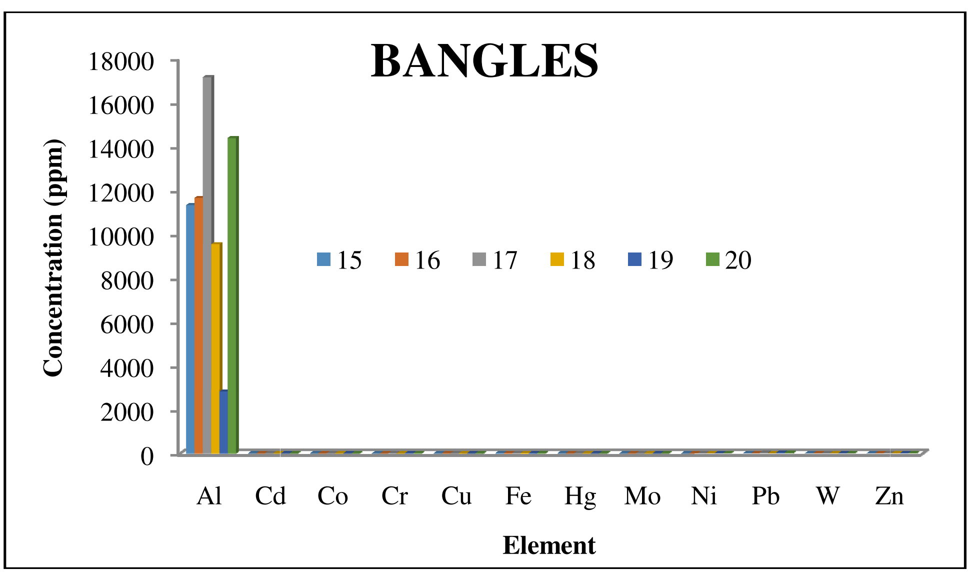

Figure 7: Indication of Aluminium Content in Bangles (15 – 20).

The fashion jewellery has been characterized as a) Aluminium based, b) steel based, c) brass/ bronze based items. The Aluminium based fashion jewellery items found to be free from mercury and the other hazardous heavy metals. the steel based items contains nickel, lead, copper metals to enhance the color and appearance. The brass based fashion jewellery contains lead as one of the major constituent. All the categories contain lead invariably to increases the color, boost the glittering appearance and reduce the production cost.

3.1. Aluminium

It is an inexpensive, lightweight and very malleable metal that is silver-white in color used extensively to create jewellry. Aluminum jewellry is durable and exhibits aesthetic appearance. It looks great with both casual and dressy attire. But it can accumulate in the muscle, skin etc. Depending on the source of exposure, aluminum cases hazardous effect to the human beings. Evidence suggests that inhaled aluminum may contribute to the development of pulmonary fibrosis and, to a lesser degree, pulmonary granulomatosis and neurodegenerative diseases (Turkdogan et al. 2003).

Aluminium present in the samples range from 0.41 ppm to 17145ppm out of 20000 ppm. The average amount of aluminium in twenty samples is 3348.97 ppm. The highest amount is found in bangles and varied from 2816 ppm to 17145.0 ppm. Average value of aluminum composition in the bangle is 11139 ppm. Its composition varies from 0.41 to 14.9 ppm and average amount of aluminium 4.16 ppm in stud samples. Its concentration lies between 0.72 ppm and 96.38 ppm in hairpin samples. Minimum quantity of aluminium is found in chain and ring samples

3.2. Cadmium

Cadmium is used in various applications such as manufacture of solder, electrical supplies, batteries, in the production of televisions, etc. Moreover, sewage sludge which contains Cadmium and other heavy metals is often applied to agricultural land as a fertilizer material (David et al 2008). Cadmium is present, typically in trace amounts, along with the metal components, such as zinc or tin, used to make fashion jewelleries. It has been a component in solders used to join jewellery parts. But it is one of the most toxic metals, children's jewelry which exceeds 0.0075 percent cadmium would be illegal based on laws. Once it is absorbed to the human body it is efficiently retained throughout life. Lower levels of exposure are mainly of concern with respect to toxicity to the kidney and metabolic variations with pathologic consequences (Benoff et al. 2000; Ye et al. 2000; Suwazono et al. 2000; Jarup et al 2000).

Cadmium ranges from 0.004 ppm to 6.86 ppm. More amount of cadmium found in studs than other samples whose concentration lies between 0.56 ppm and 6.86 ppm in studs. Most of the hairpins samples don't have cadmium and / or very less quantity of cadmium observed in bangle, chain and ring samples.

3.3. Cobalt

Cobalt is a hard, lustrous, grey metal. The cobalt- based colors and pigments have been used since ancient times for making jewellry. Cobalt is not only hypoallergenic; it is bio-compatible to the body. Cobalt is a basic element that exists naturally.

The concentration of cobalt varies from 0.009 ppm to 1.56 ppm. High amount of cobalt present in Hair pin samples than the other samples. Its concentration varied from 0.05 to 0.82 ppm in studs and 0.30 ppm 1.58 ppm in hairpins. The concentrations of cobalt in bangles were very less. Other samples such as rings and chains contain less than 1 ppm.

3.4. Chromium

Chromium is a metal that occurs naturally in the environment in the form of metal-ores combination with other elements as chromium salts, some of which are soluble in water. Being a metal, chromium does not degrade nor can it be destroyed. Named for its colored compounds, chromium has also been used to make dyes and pigments for paints, and to make bricks for furnaces, tan leather, and preserve wood Chromium allergy can either be caused due to the chrome covering metals or from the other sources of chrome. The EPA has developed toxicity values to estimate the risk of developing cancer or experiencing other adverse health effects as a result of inhaling or ingesting chromium. The toxicity value for estimating the risk of acquiring cancer following inhalation exposure is called an inhalation unit risk (UR), which is an estimate of the chance that a person will be prove to cancer from continuous exposure to a chemical in air at a unit concentration of 1 mg/m^3 .

The amount of chromium varies from 0.02 ppm to 11.28 ppm. The average concentration of chromium in the twenty samples is 2.73 ppm. Considerable quantity of chromium is found in bangles which varies from 0.12 ppm to 11.28 ppm. 0.02 to 0.58 ppm of chromium is found in ear ring sample and 1.6 ppm to 5.94 ppm in hairpin samples. A lesser amount of chromium, that is below 2 ppm observed in chain and ring samples

3.5. Copper

Mass-produced fashion jewellery often contains copper or copper alloys and it can be fatal if ingested and cause dermatitis if it has regular skin contact. Copper fumes can cause metal fume fever. The minimum concentration of copper found in bangle is 0.49 ppm and the maximum is 8890 ppm in stud. Considerable amount of copper present in studs and quantifiable amount of copper also have been found in ring and chain samples. 0.87 ppm to 603 ppm of copper is estimated in hairpins. Comparatively very less quantity of copper is observed in bangles.

3.6. Iron

It is an excellent and versatile material for construction of strong, tough, easily formed and worked jewellery, and also cheap compared to other alternatives. The amount of iron varies from 0.43 ppm to 4096 ppm. and the average composition is 1246 ppm. The highest quantity was found in hairpin and minimum was found in ear rings. Comparatively less amount of iron present in ring samples.

3.7. Lead

Lead is a soft, heavy metal, which is malleable and inexpensive. It is for these reasons that lead is used in consumer products in particular children's jewellery. Jewellery marketed specifically for children has been found to contain unsafe levels of lead because of the tendency of children to place objects in their mouths. A study conducted in the US found that, out of a total 139 jewellery items, "almost half (42.6%) of the items assayed were heavily leaded, exceeding 80% lead by weight. Average lead content for all items tested was 44.0%, and one or more heavily leaded items were found in samples from each retail store and each geographic location (Dymphna Povey 2010: Bergback et al. 1992; Koller et al 2004). When absorbed into the body, it results in damage to the cognitive development of children and damage to the nervous system. Concentration of lead is varied from 0.14 ppm to 3601 ppm. The average lead in the samples is found to be 690 ppm. Stud samples have considerable amount of lead. Except one hairpin sample others have less quantity. Chain and bangle samples also have lead content to a lesser extent. National legislation and regulation concerning lead and its hazardous properties in a variety of consumer products is described for India need to be actively implemented, and policy shall be framed for the welfare of consumers since nervous damages due to lead are known.

3.8. Mercury and Molybdenum

The amount of mercury in jewellery items can vaporize and reach levels that may be very harmful to health. Exposure may lead to behavioral changes like irritability, nervousness, tremors, impaired vision or hearing, memory problems. High exposures can result in permanent brain or kidney damage. The severity of harm depends on the level of mercury vapors, the duration of exposure, and individual sensitivity. Mercury can also be absorbed by skin contact. The Molybdenum metal is silvery white, very hard transition metal and a valuable alloying agent. It improves the strength of steel at high temperatures but it exposure can cause joint pains in the knees, hands, feet, articular deformities, erythema, and edema of the joint areas.

Mercury and molybdenum are present in comparatively less quantity. These vary from 0.086 ppm to 3.995 ppm (Hg) and 0.02 ppm to 0.606 ppm (Mo) respectively. Mercury present in the ring samples is less than 0.2 ppm. In bangle it is found to be 3.995 ppm. Molybdenum is not found in ear rings and chains. In bangles few samples only contain very less extent of molybdenum.

3.9. Nickel

Allergy to nickel is a fact which has received great attention in recent years, mainly due to the introduction of cheap fashion jewellery in which the underlying metal layer mainly comprises of nickel. Women are more susceptible to nickel allergy. It is observed that, 10 to 12% of the female population and 6% of the male population are found to be allergic to nickel. Inexpensive jewellery (below 14 Karat) is the main reason for allergy since the cheaper base metals often have high nickel content. The range of Nickel is from 0.7 ppm to1497 ppm. In bangles nickel content is less than other fashion jewellery sample. Four stud sample and one hairpin sample has much higher nickel content. (approximately 323 to 1497 ppm)

3.10. Tungsten

Tungsten is extremely hard and dense. Combined with carbon and other elements, it becomes tungsten carbide. Tungsten carbide jewellry allows for maximum hardness and rigidity without sacrificing tensile strength. Tungsten jewelleries were roughly 10 times harder than 18 karat gold and four times harder than titanium. In addition to tungsten carbide jewellry's design and high polish, its attraction to consumers is its assuring long lasting beauty. For people with sensitive skin tungsten carbide is the perfect jewellry metal. Tungsten carbide jewelry comprising tungsten carbide rings, tungsten rings etc can cause hypoallergenic. However, tungsten rings containing Cobalt will develop a negative reaction to the skin. After a short time period the ring will develop oxidation spots that cannot be removed or polished out. Concentration of Tungsten found between 0.05ppm and 108.08 ppm. Chain and ring sample contain more amount of tungsten than other samples. 0.05ppm to 0.14 ppm of tungsten is observed in hairpin samples. A lesser quantity is observed in bangles.

3.11. Zinc

It is widely used in jewellery making because of its unique properties. Zinc improves the melting and casting behavior of jewellery alloys due to its property of high vapor pressure and affinity for oxygen. Zinc is also employed as a whitener in white gold alloys. Gold alloys that comprise zinc, are yellow and white gold. In the work place environment, zinc contamination can lead to a flu-like condition known as metal fever. This condition will pass after two days and is caused by over sensitivity. Zinc can be a danger to unborn and newborn children. New born Children may be exposed to it through blood or milk of their mothers. Variation of zinc metal is from 0.11 ppm to 15215 ppm, the maximum is found in chain sample and one stud sample. Ring sample also have much amount of zinc. It lies between 0.25 ppm and 2.62 ppm for hairpins.

4. Conclusions

The observed ICP – OES results reveal that the widely available fashion jewellery items in SAARC countries contain hazardous heavy metals in alarming level. Though the selected stud items are of copper based alloys and are free from aluminium, cadmium, cobalt, mercury, chromium, molybdenum, tungsten, etc., they do contain lead and nickel moieties invariably in addition to iron and zinc metals. The hair-pin items are of black / grey metal based and do contain high level of the above. The selected chain items appeared lustrous white, made of copper – zinc alloy, which contain trace quantities of lead and nickel. However, the rings and bangles are of free from such elements.

Hence the utility and the composition of heavy metal in fashion jewellery is the crux of this paper. The determination of heavy metals in the fashion jewellery product has become an active area of research in the field of chemical and manufacturing industries. This research is important for the evaluation and characterization of sources of human and environmental exposure. This study determined the levels of Pb, Cd, Cr, Cu and Zn in some frequently used fashion jewellery in India. The valuable data generated will create awareness about the risks associated with indiscriminate use of fashion jewellery items and also provide baseline information for further research on epidemiological studies. This paper also focuses on the standard composition of jewellery alloys and their adverse effects of the alloy metals such as copper, zinc, cadmium, nickel and lead. From the discussion, it is observed that, both nickel and lead present in the samples can pose threat to humans on prolonged skin contact. This research might open up the need for standardization of acceptable levels of elemental constituents in fashion jewellery produce.

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	Studs				**Hairpins**					
Element/ Sample	1	2	3	4	5	6	7	8	9	10
Aluminium	0.69	14.85	0.70	0.41	4.19	1.63	1.28	2.92	0.72	96.40
Cadmium	6.86	0.56	3.16	1.67	0.00	0.01	0.00	0.00	0.00	0.00
Cobalt	0.82	0.32	0.05	0.05	1.58	0.61	0.73	0.30	0.90	0.41
Chromium	0.58	0.13	0.07	0.02	3.09	1.60	3.67	2.46	5.94	2.89
Copper	8890.00	1681.00	1382.00	478.00	19.59	602.60	11.10	395.20	16.20	0.87
Iron	1037.00	102.90	810.00	0.43	2911.00	2978.00	3524.00	3538.00	2806.00	4096.00
Mercury	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Molybdenum	0.00	0.00	0.00	0.00	0.61	0.00	0.02	0.00	0.23	0.00
Nickel	888.40	323.80	56.20	73.30	824.80	38.78	189.00	66.29	1497.00	101.00
Lead	2429.00	2879.00	2644.00	3601.00	1.55	1659.00	1.53	2.29	2.52	0.93
Tungsten	13.22	0.00	0.00	0.00	0.00	0.13	0.05	0.14	0.00	0.11
Zinc	707.80	1.41	0.21	0.11	1.25	2.41	1.18	2.62	0.25	0.45
Total in ppm	13974.37	5003.97	4896.39	4154.99	3767.65	5284.78	3732.56	4010.23	4329.76	4299.06
Total quantity of Heavy Metals in grams	1.40	0.50	0.49	0.42	0.38	0.53	0.37	0.40	0.43	0.43
% of Heavy Metal Constituent	70	25	24.5	21	19	26.5	18.5	20	21.5	21.5
Loss of materials on digestion* in ppm	6025.63	14996.03	15103.61	15845.01	16232.35	14715.22	16267.44	15989.78	15670.24	15700.94
Loss of materials on digestion * in g	0.60	1.50	1.51	1.58	1.62	1.47	1.63	1.60	1.57	1.57

Table 1: ICP – OES Measurement Values of fashion jewellery for heavy metal Assays in ppm.

	Chains		**Rings**		**Bangles**					
Element/Sample	11	12	13	14	15	16	17	18	19	20
Aluminium	18.55	0.47	3.88	1.73	11315.00	11645.00	17145.00	9535.00	2816.00	14375.00
Cadmium	0.24	0.00	0.00	0.04	0.00	0.01	0.01	0.01	0.00	0.00
Cobalt	0.18	0.50	0.17	0.71	0.00	0.00	0.01	0.00	0.01	0.01
Chromium	0.15	0.22	1.49	0.38	1.44	0.12	8.19	0.19	11.28	10.64
Copper	2374.00	858.60	2464.00	2273.00	1.57	1.67	0.49	1.06	0.91	1.50
Iron	46.30	2978.00	5.48	6.37	16.06	9.17	15.52	9.39	8.65	19.21
Mercury	0.00	0.00	0.17	0.09	4.00	0.00	0.00	0.00	0.00	0.00
Molybdenum	0.00	0.00	0.02	0.03	0.00	0.00	0.03	0.00	0.06	0.05
Nickel	37.40	769.40	693.20	779.60	0.26	0.16	0.23	0.07	0.22	0.31
Lead	428.00	2.56	16.42	27.09	5.36	0.00	33.05	0.14	40.30	29.06
Tungsten	94.50	0.00	69.20	108.08	0.00	0.00	0.25	0.00	0.14	0.13
Zinc	15215.00	0.57	3234.00	1785.00	3.97	2.50	13.05	3.52	6.34	9.17
Total in ppm	18214.32	4610.32	6488.03	4982.13	11347.67	11658.62	17215.83	9549.38	2883.91	14445.08
Total quantity of Heavy Metals in grams	1.82	0.46	0.65	0.50	1.13	1.17	1.72	0.95	0.29	1.44
% of Heavy Metal Constituent	91	23	32.5	25	56.5	58.5	86	47.5	14.5	72
Loss of materials on digestion* in ppm	1785.68	15389.68	13511.97	15017.87	8652.34	8341.38	2784.17	10450.62	17116.09	5554.92
Loss of materials on digestion * in g	0.18	1.54	1.35	1.50	0.87	0.83	0.28	1.05	1.71	0.56

Table 1: ICP – OES Measurement Values of fashion jewellery for heavy metal Assays in ppm.
**water soluble, volatile and other metal / nonmetal components*