## Article



# The evaluation of total mercury and arsenic in skin bleaching creams commonly used in Trinidad and Tobago and their potential risk to the people of the Caribbean

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#### Significance for public health

The lightening of one's skin to achieve a more attractive and successful appearance has become part of the culture of the Caribbean, and the use of low priced skin lightening creams is widespread. The dangers posed by these products is unknown to many of its users and for those better informed, the use of so called Mercury free creams are preferred. This study revealed that none of the products investigated were Mercury free and introduces the lesser known danger of Arsenic exposure from these creams. This study therefore serves to increase awareness of the dangers of these products to public health, and provides valuable information to the medical practitioner that may allow a more timely and accurate diagnosis of potential Arsenic and Mercury poisoning.

# Abstract

*Background.* Skin lightening is very popular among women and some men of the Caribbean, and its popularity appears to be growing. The lightening of skin colour is done to produce a lighter complexion which is believed to increase attractiveness, social standing and improves one's potential of being successful.

Design and Methods. Fifteen (15) common skin lightening creams found in pharmacies and cosmetic retailers throughout Trinidad and Tobago were evaluated for Mercury by Cold Vapor Atomic Absorption Spectrophotometry (CVAAS) and Arsenic by Hydride Generation Atomic Absorption Spectrophotometry (HGAAS). The results obtained were compared to global standards and previous research.

*Results*. Fourteen (14) of the fifteen samples analysed contained Mercury in the range of 0.473  $\mu$ g/g to 0.766  $\mu$ g/g. One sample had a Mercury content of 14,507.74±490.75  $\mu$ g/g which was over 14,000 times higher than the USFDA limit for mercury in cosmetics of 1  $\mu$ g/g. All samples contained Arsenic in the range 1.016  $\mu$ g/g to 6.612  $\mu$ g/g, which exceeds the EU limit for cosmetics of 0  $\mu$ g/g.

*Conclusions.* All the samples analysed contained significant amounts of Mercury and Arsenic and none of them can be considered *safe* for prolonged human use. The samples that contained Mercury levels which were lower than the USFDA limit contained Arsenic levels which exceeded the EU standard of 0  $\mu$ g/g in cosmetics. The popularity of these skin lightening creams in the Caribbean region places the population at elevated risk of chronic Mercury and Arsenic poisoning and possibly acute Mercury Poisoning.

# Introduction

## Skin bleaching to enhance beauty and social standing

Skin bleaching is the process of depigmentation of the skin via artificial means by the use of creams, soaps, lotions, injections and home-made products. Depigmentation is a procedure by which the melanin produced in the skin is reduced. Melanin is produced by special cells called melanocytes which are found mainly in the skin, eyes and hair bulbs. The enzyme responsible for melanin production is tyrosinase. Bleaching agents typically inhibits tyrosinase production by actively competing with copper in a key process leading to the production of melanin. The end product is the lightening of the skin.<sup>1</sup>

The islands of Trinidad and Tobago are the southernmost of the Caribbean Islands. The population is 1.3 million persons, of which 37.6% are of East Indian Descent, 36.3% are of African Descent, 24.2% are of mixed African/Indian Descent, 0.3% are of Chinese Descent, and 0.15% are of Caucasian Descent.<sup>2</sup> From this data, the population of Trinidad and Tobago is 98.1% African and East Indian Descendants.

Skin lightening practices such as the use of skin lightening or bleaching creams have been commonly used by individuals of darker skin tones from the African and East Indian populations.<sup>3</sup> In these populations, and among many others, a lighter complexion is deemed more beautiful, signifies a higher social status and enhanced economic mobility.<sup>4</sup>

A study by Drescher *et al.*<sup>5</sup> conducted in Barbados in 2010 revealed that it was skin bleaching and not fish consumption that was responsible for elevated mercury levels in middle aged women working in the fishing industry with the accompanying symptoms of chronic mercury poisoning being observed.

Studies show that skin bleaching is on the rise and is more prevalent among females.<sup>6</sup> This can be explained by the perception that women appear more sophisticated or more beautiful, gain more attention and are perceived to be of a higher social economic standing.<sup>4</sup>

Research have also found that skin bleaching is more prevalent among women with higher academic achievement,<sup>7</sup> who are more prone to seek professional employment. This is further supported by studies in hiring practices where lightly coloured individuals have a statistically higher chance of being hired than darker coloured persons.<sup>8</sup>

The use of skin bleaching creams in the Caribbean is not restricted to women only, but also to men who are seeking a lighter skin complexion. Recently, a famous regional singer launched his own line of skin whitening products<sup>9</sup> to add to the already popular men's line of skin whitening creams. Lighter skin





not only enhances the perception of greater attraction but also enhances the appearances of tattoos on skin as the darker coloured tattoos contrasts better with light skin than dark skin. In the case of men of East Indian decent, a recent study found that women of East Indian descent prefer men of a lighter complexion.<sup>10</sup>

### Mercury and arsenic as skin bleaching agents

Mercury used in cosmetics commonly exists in two forms; organic, which is used as a cosmetic preservative, and inorganic, which is commonly used as a skin lightening agent. These inorganic compounds are typically a combination of Mercury with elements such as Chlorine, Sulphur and Oxygen, and the compounds of Mercury commonly used in skin creams are typically Mercury (II) Chloride and Ammoniated Mercury.<sup>11</sup> Recent publications have reported that mercury is still being used in many skin lightening products found in Mexico, 12 Africa, 13 The United States 14 and Asia<sup>15</sup> Many of these studies have revealed levels of mercury in skin bleach exceeding the limits set by WHO.<sup>16</sup> Mercury based skin lightening creams have been found to elevate the body's Mercury levels in non-users of the product who share the same household with users of Mercury based skin lightening products.<sup>17</sup> Research by the CDC in 2010 revealed that Mercury vapours emanating from Mercury based skin lightening products was responsible for elevated Mercury levels in resident non-users of skin lightening products.17

Inorganic Mercury applied as creams on the skin can enter the body readily via absorption through the skin, inhalation or orally,<sup>18</sup> and long-term exposure due to repeated applications can result in a number of damaging effects which include kidney damage, skin discoloration, rashes, peripheral neuropathy, and psychosocial issues.<sup>11</sup>

Unlike Mercury, research on Arsenic and its use in skin bleaching creams is sparse despite arsenic's well-known toxicity. Arsenic is a naturally occurring element that is found in conjunction with inorganic or organic substances to create various compounds. The element has been linked to the cosmetic world as far back as the 19<sup>th</sup> Century, where Syrian and Austrian peasants ate arsenic to clean and soften their complexion.<sup>19</sup> Currently, Arsenic is found in pigments used in cosmetics and is regulated by the USFDA.<sup>20</sup>

Arsenic is present in a range of cosmetic products such as eye shadows, lotions and lipsticks, and only the European Union is known to have strict regulations against the metal, banning it from use in cosmetics.<sup>21</sup> It is reported that the presence of Arsenic in cosmetic products, may be due to the existence of the substances in the raw material used in the manufacturing of the products or the poor use of the raw materials. In addition, as contamination can be as a result of unintentional migration from the metal devices used in the manufacturing process.<sup>22</sup>

Exposure to Arsenic, in the inorganic form, has various chronic side effects such as vomiting, diarrhoea and nausea, as well as skin disorders and cancers.<sup>23</sup>

Despite the well-known toxicity of Mercury and Arsenic, creams containing Mercury can be easily found in pharmacies, beauty stores and beauty salons throughout the Caribbean. These products can also be available through the internet and ethnic markets<sup>.24</sup> The use of skin lightening products is particularly popular among the East Indian and African Populations where there are perceived socio-economic benefits of having a lighter skin complexion.<sup>11,25</sup> Even in Caribbean diaspora in countries such as The United States, the use of mercury based skin lightening creams is popular.<sup>14</sup>

Cosmetics are not required by local laws to list all their ingredients so many preparations do not list their content of mercury or arsenic in the ingredient listing or package label, and researchers have found that many skin whitening products do contain some level of mercury although none were declared in the ingredient list or labelling.<sup>26</sup>

## Research objective and outcome

This paper seeks to establish the Total Mercury and Total Arsenic contents of skin bleaching creams used commonly in the Caribbean region and is in keeping with international regulations and guidelines for cosmetics. No differentiation between organic and inorganic metals are addressed in this paper.

The information presented in this paper is intended to raise awareness of the hazards of continued unregulated usage of skin bleaching agents and to contribute to the body of knowledge that will be used to guide the implementation of regulatory standards for cosmetic products in the region.

# **Materials and Methods**

#### Sample collection

Fifteen commonly used skin lightening creams were purchased from various pharmacies and cosmetic outlets throughout the island of Trinidad in October 2016 and analyses in November and December 2016. It was observed that the diversity of skin bleaching products was limited mostly to products manufactured in the EU, India and Jamaica, and were manufactured between June 2014 to August 2016. Sample selection was done to ensure the widest diversity of samples and to capture the most popularly purchased samples.

#### Apparatus and equipment

All solutions were prepared in Millipore-purified water (conductivity 18M $\Omega$ -cm). All chemicals used were of ACS reagent grade or better. Nitric Acid (65%), Hydrochloric Acid (37%) and Perchloric Acid (70%) were all ACS grade, Tin (II) Chloride and Sodium Hydroxide were reagent grade (>98%) and Sodium Tetraborate was reagent plus grade (>99.5%), all obtained from Sigma-Aldrich. Analytical Mercury and Arsenic standards (1000 ug/mL) were certified NIST traceable obtained from Inorganic Ventures of Christiansburg VA.

All glassware used in this study were grade A quality. They were washed using laboratory detergent followed by rinsing with tap water then by distilled water. Glassware were then soaked in a 10% (v/v) Nitric Acid solution overnight and rinsed multiple times with ultrapure water before being allowed to air dry

Digestion was performed using a VWR Dry Block Heater capable of maintaining 150°C with digital control. Analysis was performed using the Varian AA 800 Atomic Absorption Spectrophotometer equipped with a VGA 77 Vapor Generation Accessory and Ultra AA lamps.

#### Digestion of samples for mercury and arsenic analysis

Fifteen skin bleaching creams were purchased from various pharmacies and cosmetic outlets throughout the country. The samples were digested for analysis of mercury and arsenic using a modified method of Hepp *et al.*<sup>20</sup> Approximately 0.5 g of a sample was weighed in triplicate into cleaned, dried 50 mL thick walled boiling tubes. 10 mL of a 1:1 mixture of Nitric and Perchloric acids were added to each sample and digested on a heating block at 130°C for 8 hours until a clear solution was obtained. This was allowed to cool to room temperature and further inspected to ensure clarity of solu-

tion. The clear digests were filtered through hardened ash less filter paper (Whatman No. 542) into grade A acid washed 50 mL volumetric flasks. 5 mL of concentrated Hydrochloric Acid was added to stabilize the metals,<sup>27</sup> and the solution made up to the volume with ultra-pure water. All fifteen (15) samples as well as blanks and spike samples were digested in this manner. This solution was utilized for the analysis of total Mercury by Cold Vapor Atomic Absorption Spectrophotometry (CVAAS).

For Arsenic, analysis by Hydride Generation Atomic Absorption Spectrophotometry (HGAAS) required that all Arsenic were converted to the  $As^{3+}$  species. This is achieved by reduction with 1%KI.<sup>27</sup> A 5.0 mL aliquot of the digested sample was pipetted from the 50mL volumetric flask into a grade A 25 mL volumetric flask. To this flask, 5 mL of a 5% KI solution was added along with 5 mL of a 1M HCl solution, and the volume made up with ultrapure water. The solutions were left at room temperature overnight to allow for the complete conversion of  $As^{5+}$  to  $As^{3+}$ . This process was repeated for all samples, blanks and spike samples. These solutions were analysed by HGAAS for total Arsenic.

#### Mercury and arsenic determination

Mercury was analysed using a Varian SpectrAA300 Atomic Absorption Spectrophotometer equipped with a VGA77 Hydride Generator Accessory, Mercury flow through quartz sample cell and Ultra AA lamps. The reductant solution used was SnCl<sub>2</sub> (25%w/v) in HCl (20%v/v)(27), and the acid solution used was Ultra-pure water. Mercury was measured in the cold vapor mode at 253.7 nm with deuterium background correction and a delay time of 70 seconds to compensate for desorption of mercury from the flow tubes. Calibration was performed using 1 µg/L, 10 µg/L, 20 µg/L, 30 µg/L and 50 µg/L standard. Detection limit was determined using the method described by Miller and Miller.<sup>28</sup>

Arsenic was analysed on the identical instrument as for Mercury except that the instrument was used in the Hydride Generation mode using an Air/Acetylene flame and Quartz flow cells. The lamp used was an Ultra AA lamp and measurement was done at 189.0nm with Deuterium background correction and a delay time of 45 seconds. Calibration was performed using 5  $\mu$ g/L, 10  $\mu$ g/L, 20  $\mu$ g/L, 30  $\mu$ g/L and 50  $\mu$ g/L standard. The reductant solution used in the generation of the stable hydride was NaBH<sub>4</sub> (0.6%w/v) made up in NaOH (0.5%w/v), and the acid used was 5M HCl.

#### Method validation

Method validation and quality control was ensured using Method Blanks, Spiked Recoveries and Reference Materials (DORM-4). The DORM-4 Certified Reference Material is a fish protein reference certified by the National Research Council of Canada. Spiked recoveries were determined using Papaya White samples spiked with 50 µg/kg and 100 µg/kg Mercury and Arsenic respectively. The recoveries are given in Table 1.

The validity of the results obtained is supported by the recoveries obtained from the analysis of DORM-4 CRM and spiked samples. The certified concentrations of Mercury and Arsenic were 412.0 $\pm$ 36.0 $\mu$ g/kg and 6870 $\pm$ 440  $\mu$ g/kg respectively. Triplicate analysis of DORM-4 gave Mercury and Arsenic levels of 403.8 $\pm$ 39.7  $\mu$ g/kg and 6941.8 $\pm$ 486.1  $\mu$ g/kg respectively. These results agreed with the certified values by 98% and 101% for Mercury and Arsenic respectively and with standard deviations of less than 10%, the precision of the method is also acceptable. The spiked Papaya White samples also supported the method validation. Papaya White was found to contain an average of 575.34  $\mu$ g/kg of Mercury and Arsenic working standard were pre-



pared by adding an appropriate amount of standard to the sample to yield an increase of 50  $\mu$ g/kg and 100  $\mu$ g/kg of both Mercury and Arsenic. The spiked samples were homogenized and analysed in triplicate. In all cases, recoveries ranged from a low of 97.2% to a high of 103.8% with standard deviations of <5%. Detection limits were determined to be 0.17  $\mu$ g/kg for Mercury and 0.21  $\mu$ g/kg for Arsenic. The analytical method used in this study is both accurate and precise and is fit for purpose.

# **Results and Discussion**

Fifteen skin-bleaching samples were obtained from pharmacies and cosmetic outlets throughout the island. Nine of the samples collected were manufactured in the EU, India manufactured four of the samples, the UK one and Jamaica one. These samples were analysed as per the validated method and the results of those analyses are summarized in Table 2.

## Mercury

All fifteen samples contained Mercury, with Deluxe Silken having the highest concentration of 14,507.741±490.75 µg/g. Under the USFDA guidelines,<sup>18</sup> the acceptable level of mercury in cosmetics other than products intended for use in the region of the eye is <1 µg/g. The sample of Deluxe Silken manufactured in Jamaica greatly exceeded the  $<1 \mu g/g$  value. This is due to the fact that mercury is the active ingredient in the formulation. This product has on its label a content of 3% Ammoniated Mercury [Hg(NH2)Cl], which contains 24,000 µg/g of elemental Mercury. Our analysis determined the concentration of mercury in Deluxe Silken to be 14,507.741 µg/g or 1.8% Ammoniated Mercury. This value correlates well with the findings of Hamann et al.24 who found 13,546 µg/g in Deluxe Nadinola Bleaching Cream from Jamaica, the predecessor product of Deluxe Silken. Deluxe Silken is approximately 14,000 times the recommended limit for Mercury and use of products of this kind can lead to symptoms of Mercury poisoning including Nephrotic Syndrome as have been documented in the US,14,29 Barbados<sup>5</sup> and Indonesia.<sup>30</sup> In addition, the Mercury concentration found in this particular product also poses a threat to fellow householders<sup>17</sup> co-workers and persons in close physical proximity to the users and who are exposed to Mercury vapours emanating from the product container and by extension the skin of the user.

Of the fifteen samples analysed, fourteen contained Mercury levels less than 1  $\mu$ g/g, with levels ranging from a low of 0.473  $\mu$ g/g to a high of 0.766  $\mu$ g/g. These values were below the USFDA recommended limit of <1.0  $\mu$ g/g, but exceeds the EU limit of 0  $\mu$ g/g. Mercury levels as low as 0.319  $\mu$ g/g in skin lightening

Table 1. Total mercury	and total	arsenic	recoveries	for	CRM and
spiked samples.					

Sample	Expected value µg/kg	Actual value µg/kg	% Recovery
Mercury DORM-4 Spike-50 Spike-100		$403.8\pm 39.7$ $607.7\pm 30.9$ $658.2\pm 26.6$	98.0 97.2 97.5
Arsenic DORM-4 Spike-50 Spike-100		6941.8±486.1 1099.1±51.0 1167.0±58.6	101.0 102.4 103.8



creams have been found to cause histopathological changes to brain, liver and kidney tissue in mice.<sup>31</sup> Low levels of Mercury based skin lightening creams used during pregnancy and lactation have been found to impair infant development.<sup>32</sup> Therefore, none of the samples analysed can be considered safe for use.

The Caribbean population was previously believed to be under threat of mercury poisoning due to its higher than average fish consumption, however recent research including work by Drescher *et* al,<sup>5</sup> Mckelvey *et al*.<sup>14</sup> and this current study indicates that the threat of Mercury poisoning is indeed real in the Caribbean, but may be more due to its cultural use of skin lightening creams than fish consumption.

#### Arsenic

Limited research exists on the Arsenic content of cosmetics, particularly skin bleaching creams. Adepoju-Bello *et al.* in 2012<sup>33</sup> found very low levels of Arsenic in cosmetics including skin bleaching creams in the range of 0.006  $\mu$ g/g to 0.013  $\mu$ g/g. Hepp *et al.* in 2014<sup>20</sup> found traces of Arsenic in body lotions ranging from trace levels to 0.2 ug/g. The levels of arsenic found in this current study exceeds those previously reported by those researchers by up to 30 times the amount, with the lowest value being 1.016  $\mu$ g/g (Fair & White) and the highest being 6.612  $\mu$ g/g (Symba Crème Skin Lite N Smooth). The results obtained however compared well with previous research by Alquadami *et al.* in 2013,<sup>34</sup> who found Arsenic levels in thirty-four (34) skin whitening creams ranging from a low of 0.34  $\mu$ g/g to a high of 14.76  $\mu$ g/g.

The results obtained from this study showed that all the samples analysed contained Arsenic. The results obtained also showed that the levels of Arsenic found in the skin lightening creams were higher than the levels of Mercury in all samples except Deluxe Silken where the active ingredient listed was Ammoniated Mercury. The Arsenic concentrations were as much as 10 times higher than the corresponding Mercury concentrations in some of the creams analysed. There was no clear pattern that could be observed that distinguishes the Arsenic levels in the creams by place of manufacture. Neither were any association between Arsenic and Mercury content observed. It should be noted that arsenic was not listed as a component of any of these products on the labels.

The presence of Arsenic in these skin lightening lotions, may

be due to the existence of the substances in the raw material used in the manufacturing of the products as well the use of lower quality materials in its manufacture particularly fillers and pigments.<sup>23</sup> Arsenic contamination may also be as a result of unintentional migration of the metal from wear and corrosion of metallic devices and machinery used in the manufacturing process.<sup>22</sup>

Although past studies have shown that Arsenic is present in a range of cosmetic products such as eye shadows, lotions and lipsticks, only the European Union is known to have strict regulations against the metal, banning it from use in cosmetics.<sup>35</sup> Neither the USFDA nor the WHO have defined acceptable levels for Arsenic in cosmetics.

The levels of Arsenic observed can enter the body via inhalation, ingestion and via the skin.<sup>36</sup> When in the body Arsenic can accumulate in hair, skin and nails and result in chronic health effects. Long term chronic exposure to arsenic can lead to multisystem failure as a result of various cancers.<sup>36</sup> Lifelong exposure to arsenic levels as low as 10 µg/L in drinking have been shown to elevate an individual's risk of cancer.<sup>37</sup> Other symptoms may include hyperpigmentation, palmar and solar keratosis, diarrhoea and vomiting, skin changes and neuropathy,<sup>36</sup> ischemic heart disease,<sup>38</sup> confusion and memory loss,<sup>23</sup> respiratory disease and a higher risk of diabetes.<sup>39,40</sup>

## Conclusions

This study revealed that all samples analysed did contain significant level of Mercury and Arsenic. One sample contained over 14,000 times the regulatory limit for mercury of 1 ppm maximum while the other 14 samples contained levels below the 1 ppm level, but were still very significant. In addition, this study revealed significant amounts of Arsenic in all the samples. In fourteen of the fifteen samples, the levels of Arsenic exceeded those of Mercury. Only the EU has an outright ban on Arsenic and its compounds in cosmetics, and it was interesting to note that ten of the fifteen samples originated from the EU.

The act of skin bleaching is very prominent in the Caribbean region and the diaspora of the Caribbean regions, as it is engrained in the culture. The popularity of skin lightening has always been

Name of product	Origin	Hg ± S.D µg/g	As ± S.D µg/g
TopiClear: Lemon Cream	EU	$0.627 {\pm} 0.055$	$2.024 \pm 0.289$
Crusader	EU	$0.475 \pm 0.045$	$4.044 \pm 0.214$
Caro Light	EU	$0.550 \pm 0.040$	$4.582 \pm 0.198$
Precious Beauty	EU	$0.516 \pm 0.061$	$4.763 \pm 0.359$
African Formula	EU	$0.519 \pm 0.040$	$5.219 \pm 0.125$
Metasol Medicated Skin-Lightening Cream	EU	$0.653 \pm 0.016$	$5.260 \pm 0.443$
Idole Crème Medical	EU	$0.740 \pm 0.035$	$6.163 \pm 0.072$
Xtreme Brite	EU	$0.584 \pm 0.012$	$6.471 \pm 0.148$
Fair & White	EU	$0.766 \pm 0.010$	$1.016 \pm 0.094$
Symba Crème Skin Lite N Smooth	UK	$0.508 \pm 0.043$	$6.612 \pm 0.072$
Papaya White	India	$0.575 \pm 0.055$	$1.023 \pm 0.143$
Fair & Radiant	India	$0.657 \pm 0.014$	$2.824 \pm 0.151$
Fair & Handsome	India	$0.473 \pm 0.045$	$5.291 \pm 0.095$
Naturally Fair Pearls	India	$0.565 \pm 0.044$	$5.721 \pm 0.604$
Deluxe Silken	Jamaica	$14,507.741 \pm 490.75$	$5.154 \pm 0.030$



high, and appears to be increasing with both men and women. Based on this study, the risks associated with the practice is very high, not just for both male and female users, but also members of their associated households, co-workers, and persons in close physical proximity. Chronic usage could lead to mercury and arsenic poisoning as well as physical, emotional and social complications resulting from such.

Cosmetics are not regulated in the region and manufacturers are not required by law to display the components of their products on the labels. Such lack of regulation could lead to severe health defects within the population and present burdens on both the health system and social support services of the region.

## Recommendation

Mercury and Arsenic levels present in skin lightening products do pose a risk to public health mostly due to its prolific use throughout the Caribbean region as well as the levels of the metals found in the more commonly used skin lightening creams. It is necessary that these products are regulated at the national level. Many of the countries of the Caribbean region are either signatories to the Minamata Convention or, are in the process of signing. The provisions of the convention require that states regulate mercury containing products including cosmetics. This research therefore will serve to guide public health policy and contribute towards conformance to the Minamata convention.

Further research is required to assess the levels of heavy metals in other cosmetic products including indigenous products to evaluate their risk to public health, and use this information to guide public health policies.

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