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A Survey of Metals Found in Tattoo Inks

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Abstract

While the practice of tattooing has existed for thousands of years, it has recently begun growing in popularity in the US. With the increasing prevalence of tattoos, the methods and inks involved in the tattooing process have also developed. Tattoos now use many brightly colored inks, often made using metal-based pigments. There is concern that chemicals may be present in tattoo inks in concentrations that may lead to human health concerns either during application or removal of tattoos. Since exposure to metals has been linked to tremors, liver damage, memory loss, cognitive loss, and even death, there is concern about the prevalence of metals in tattoo inks in general. To this end, a survey of 226 commercial tattoo inks was performed and each ink was analyzed for the presence of heavy metals using two different x-ray methods: Particle Induced X-Ray Emission and Scanning Electron Microscopy/Energy Dispersive Spectroscopy. Fifteen metals were identified in various tattoo inks by these rapid x-ray methods, including chromium, manganese, nickel, copper, barium, and lead. Conclusions can be drawn about the prevalence of metals in some pigment colors and from some brands.

Keywords

Metals, Tattoo Inks, Tattoos, PIXE, SEM, X-Ray Analysis

1. Introduction

The percentage of Americans with at least one tattoo rose from 16% to 21% of the total population between 2003 and 2013 [1]. Since the 1970's, when tattoos began their rise in popularity [2], companies have been finding new formulas to create a wide array of colorful inks. The composition of many inks, however, may be a cause for concern, since studies have shown that metals and other compounds, both hazardous and unregulated, may be present in tattoo inks [3]

[4] [5] [6] [7].

Metals are often used to produce brightly colored pigments, many of which could be used in tattoo inks. In the process of tattooing, the needle is loaded with ink and injected into the skin, past the epidermis and into the dermis, where the ink is deposited. Some blood vessels are broken in this process, providing a pathway for the ink and any associated metals to enter directly into the bloodstream. Furthermore, the tattoo laser removal process (which is also increasing in popularity together with tattooing) degrades and injects most of the inks from the skin directly into the bloodstream allowing for even higher exposure to the metals contained in the inks during the removal process. These metals can accumulate in vital organs in the body, which may cause neurodegenerative diseases, disrupting important bodily functions, and even causing cancer [8]. For example, the presence of metals and other chemicals in some inks applied to tattoos in the lumbar region will often prevent the administration of an epidural anesthetic during childbirth [9] [10] [11]. While in the dermis, tattoo inks expose the body to small amounts of these metals for an extended period of time. Since many metals have well-known chronic and acute toxicity effects in humans, this particular form of consumer product may lead to metal poisoning of consumers in the absence of regulation in the US.

This study was designed to analyze the metal content of a large number of commercially available tattoo inks to assess the prevalence of various metals available to the US market. The methods used to examine these inks included two X-ray methods, Particle Induced X-Ray Emission (PIXE) and Scanning Electron Microscopy (SEM) paired with Energy Dispersive Spectroscopy (EDS) performed at Hope College.

2. Methods

From 2014 to 2016, 226 tattoo inks were purchased from 18 different manufacturing brands. Local tattoo artists were asked which brands of ink were the most popular in order to give a broad representation of inks most likely being used by tattoo artists and consumers in the US. The brands sampled in this survey include the following brands: Arcane, Colour King, Deep Colours, Dermaglo, Dynamic, Eternal, Fusion, Intenze, Kabuki, Kuro Sumi, Mom's, Prizm, Skin Candy, Stable Thin Line, Stable Traditional, Star Brite Colors, Waverly, and World Famous.

In preparation for x-ray analysis, each ink sample was thoroughly shaken, and one to three drops of the sample was placed onto a 17 mm × 20 mm rectangular substrate of standard copier paper (Spectrum brand/Georgia-Pacific Papers) and allowed to dry. This paper had previously been tested to be free of any heavy elements except calcium [12]. The average diameter for each ink aliquot was roughly 3.6 mm. Because thickness of the ink spots on the substrate varied due to the differing densities of the individual ink solutions, inks with lower densities were given multiple coats until sufficient thickness was achieved. This non-uniformity added a degree of quantitative uncertainty, but complete homogeneity in thickness and smoothness was unnecessary as a majority of the characteristic x-rays from the sample come from the outermost layer, approximately 100 - 200 μm into the sample for the most penetrating technique. Eight inks were typically loaded onto a single paper substrate and placed onto a solid aluminum target frame which held the paper in place with no other substrate in the path of the ion beam.

All of the samples were analyzed via Particle Induced X-Ray Emission spectroscopy using the Hope College Ion Beam Analysis Laboratory (5SDH Pelletron Accelerator, National Electrostatics Corp, Middleton, WI). Each sample was irradiated with approximately 1.5 - 2.0 nA of 3.4 MeV protons for 300 seconds in a high vacuum scattering chamber. The characteristic x-rays emitted from each sample were detected by a Si(Li) detector (Ortec, model SLP-10180-ST) located at 135˚ with respect to the beam axis. All samples were initially run with a 0.002” mylar filter placed between the target and X-ray detector to suppress back-scattered protons. This thin filter allowed for K_α and K_β x-rays to be recorded for elements heavier than sulfur. Approximately half of the samples were run again with a 0.015 aluminum filter replacement to suppress all x-rays from elements lighter than iron. This method was particularly useful when a large titanium peak was observed and the high x-ray production rate from that element masked other elements at lower concentrations. A lower current of 0.4 nA was used with the aluminum filter to reduce the spectral fit error. Elements with x-ray energies above 7.4 keV (>Ni) were primarily measured using the thicker aluminum filter, whereas those below 7.4 keV were primarily measured using the mylar filter. This allowed for greater accuracy of quantification in difference energy regions of the spectrum.

Samples were periodically tested against a blank copier paper substrate mounted in the target frame to ensure no background elements could be seen from the target ladder. Furthermore, to ensure no contamination was coming from the paper onto which the ink was painted, a blank piece of the standard copier paper was analyzed periodically and was found to be absent of all detectable heavy elements except calcium. Each sample was analyzed in relation to a NIST calibration standard (SRM 1412) with certified concentrations of various elements, which allowed for the absolute normalization of elemental concentrations measured in each sample. The beam current used to measure each sample was measured in a Faraday cup that was inserted before and after each run. At these beam energies and intensities, the PIXE analyses were entirely non-destructive for these samples. All elemental K_α x-rays were quantified for each sample using the thick-target option of the peak fitting program GUPIXWin [13] Using this software, the measured x-ray yields were converted into an effective concentration (in ppm) in the dried pigment in each sample.

To assess the analytical reproducibility of the method 10% of samples were replicated with PIXE two to three times. Two thirds of the samples were also analyzed with Scanning Electron Microscopy coupled with Electron Dispersal Spectroscopy. Similar to PIXE, SEM/EDS excites an element's electrons but instead uses a 15 keV electron beam. Because of a thinner filter SEM/EDS is more sensitive to lighter elements and PIXE, with its higher beam energy, is more sensitive to heavier elements. Even with these slight differences, the two methods produced very comparable results.

3. Results

Among the 226 inks analyzed, 15 metals and one halogen were present above the limit of quantification (LOQ). In order of prevalence, the elements found were titanium, iron, chromium, copper, zirconium, manganese, bromine, nickel, niobium, strontium, zinc, barium, molybdenum, lead, vanadium, and tungsten as shown in Figure 1.

Titanium was seen in high concentrations in almost 91% of the samples; titanium dioxide is a known brightener used in tattoo inks [14] and is not known to be toxic to humans even at high doses [15]. As titanium is mostly insoluble in water, the high presence of titanium dioxide nanoparticles renders inductively

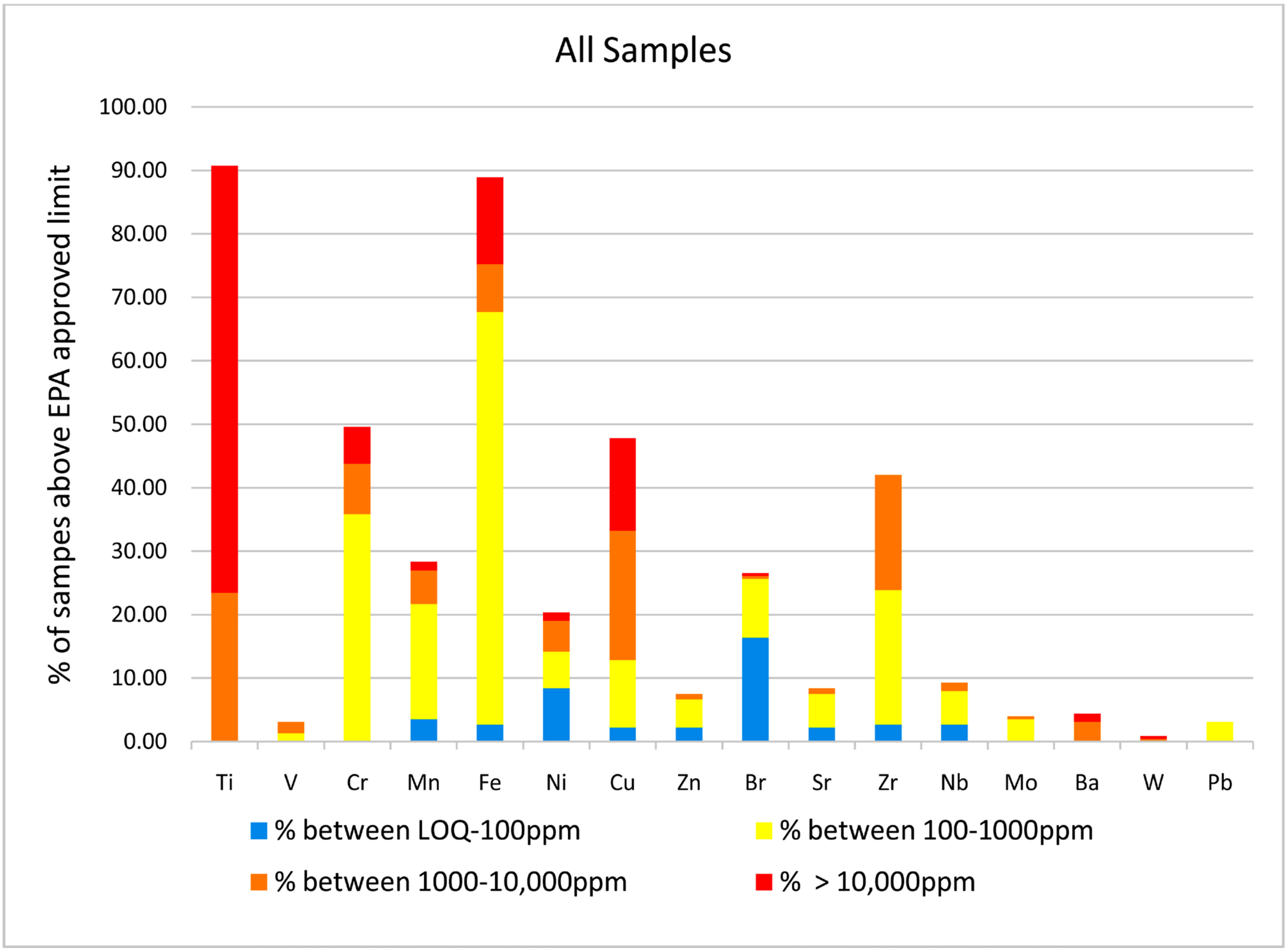


Figure 1. Percentage of inks containing elements above their LOQ with the percentage of inks in specific concentration ranges in order of atomic number.

coupled plasma mass spectroscopy (ICP/MS) analysis difficult without complete digestion, which is why two x-ray methods were chosen for analysis instead. Zirconium, presumably in the form of zirconium dioxide which is a pigment additive used to coat titanium dioxide particles [16], was found in over 40% of the inks, and is also considered relatively nontoxic. Iron was also observed in nearly 90% of the ink samples, and is suspected to be in the form of iron oxide which is a known darkener used in tattoo inks [14]. Iron is not usually considered as a toxin, however, some of the inks with exceptionally high iron concentrations may be a cause for concern as iron can be potentially harmful at very high concentrations and can cause problems to the cardiovascular and central nervous systems, kidney, liver and blood. [17]. The metals of highest concern, particularly chromium, nickel, copper, barium, and lead, can be seen below in **Figures 2-6**.

There were 18 different brands sampled in total. In **Figures 2-4**, only the data from the 10 brands with greater than 10 samples each was used because fewer samples did not seem like a sufficient subset of data per manufacturer. Most of the inks from the other 8 brands, however, fit the general trend. In **Figure 5** and **Figure 6**, the data encompassed all the samples from all 18 brands but there was neither barium or lead seen in any of the remaining 16 manufacturers' inks.

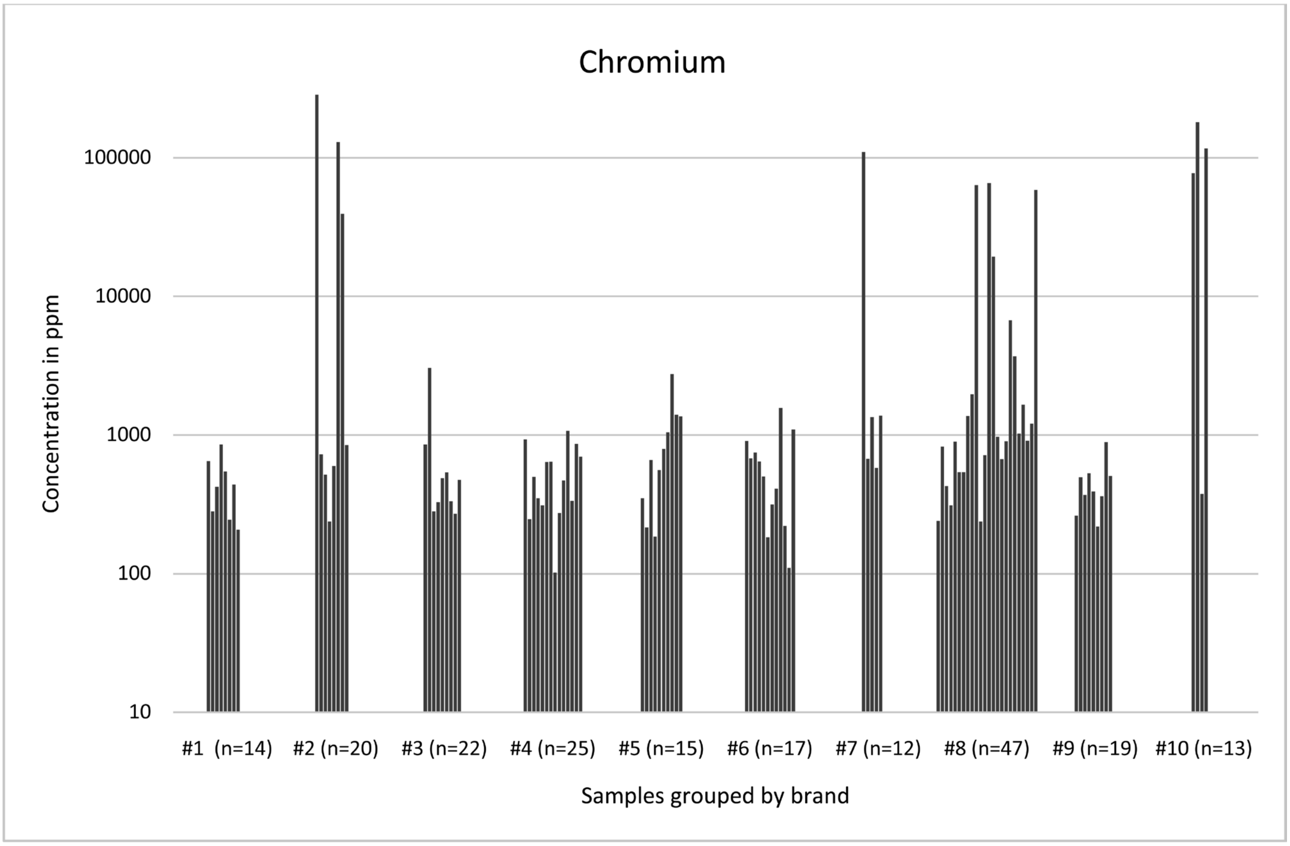


Figure 2. Ink samples containing chromium above the LOQ. Samples were grouped by brand and only brands with more than 10 ink samples were used, where n is the number of sampled inks by that manufacturer. Each bar represents an individual sample. Concentration is given on a logarithmic scale.

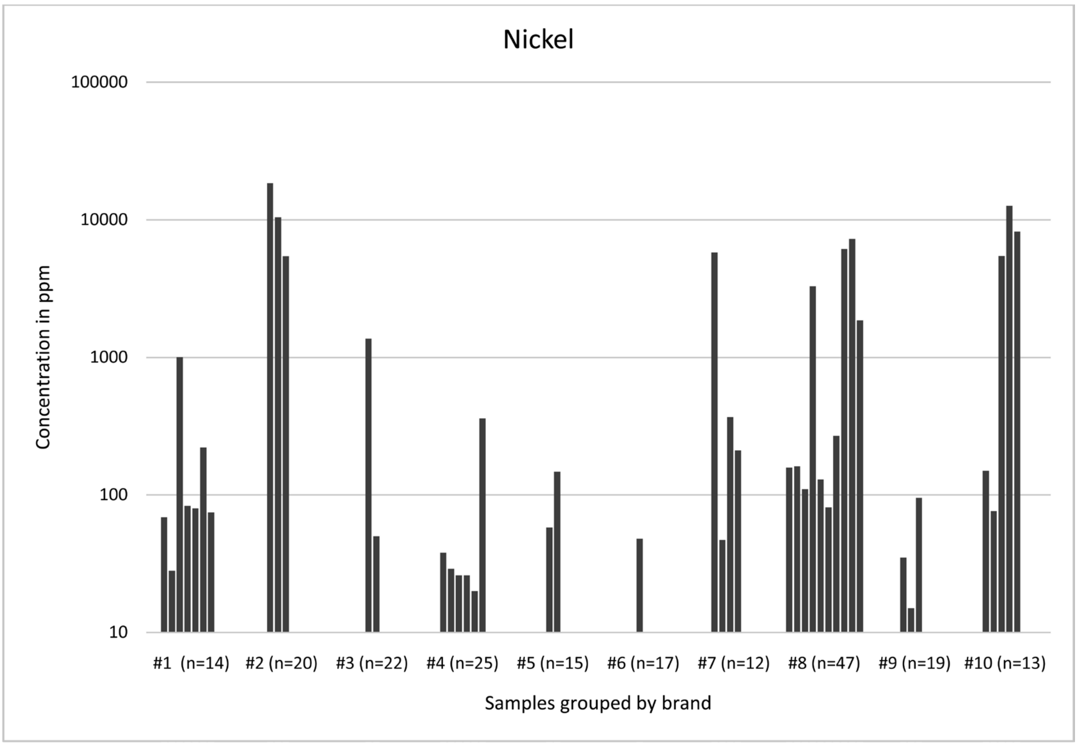


Figure 3. Ink samples containing nickel above the LOQ. Samples were grouped by brand and only brands with more than 10 ink samples were used, where n is the number of inks sampled by that manufacturer. Each bar represents an individual sample. Concentration is given on a logarithmic scale.

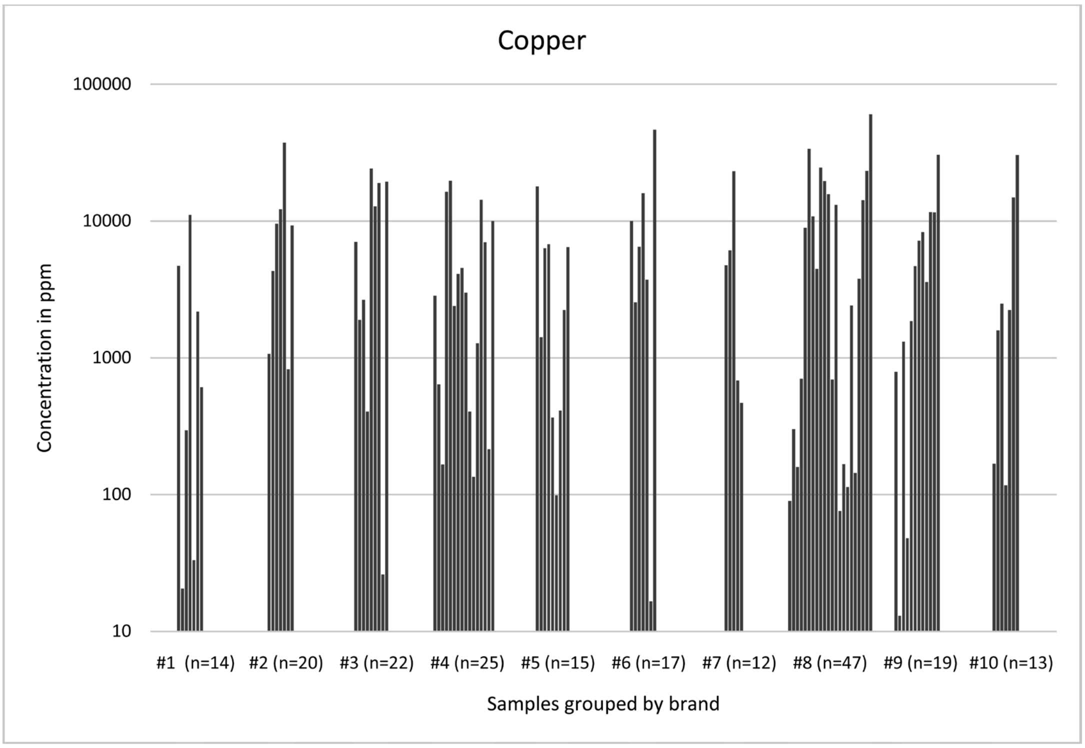


Figure 4. Ink samples containing copper above the LOQ. Samples were grouped by brand and only brands with more than 10 ink samples were used, where n is the number of inks sampled by that manufacturer. Each bar represents an individual sample. Concentration is given on a logarithmic scale.

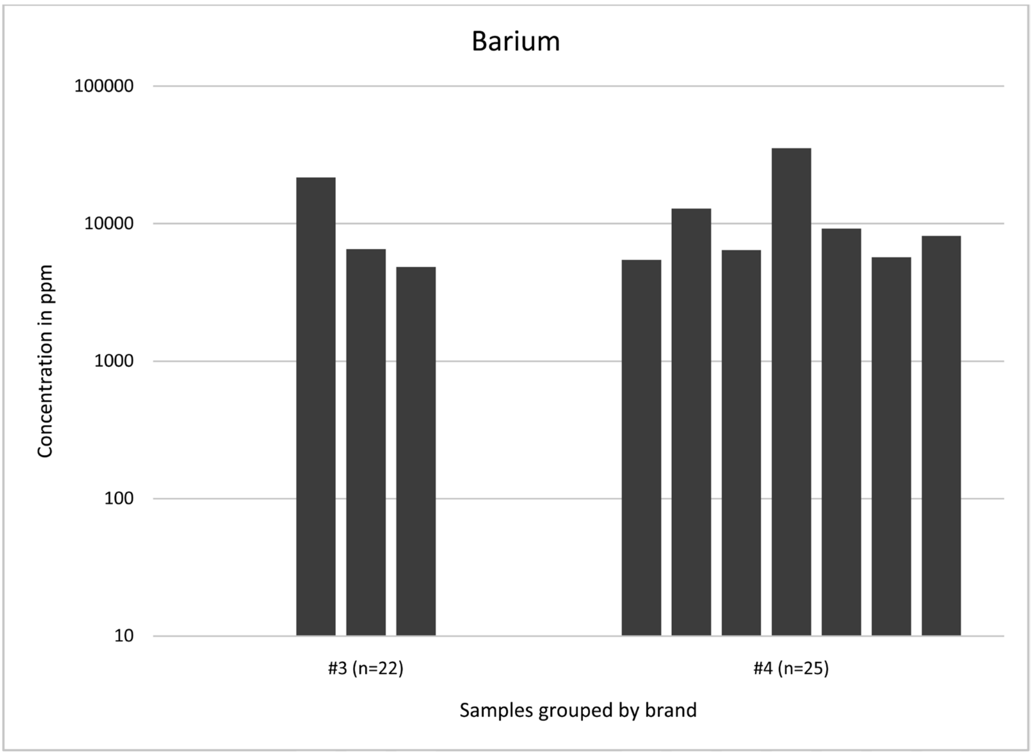


Figure 5. Ink samples containing barium above the LOQ. Samples were grouped by brand and n is the number of inks sampled by that brand. This includes all inks tested but only two brands contained any measurable barium. Each bar represents an individual sample. Concentration given on a logarithmic scale.

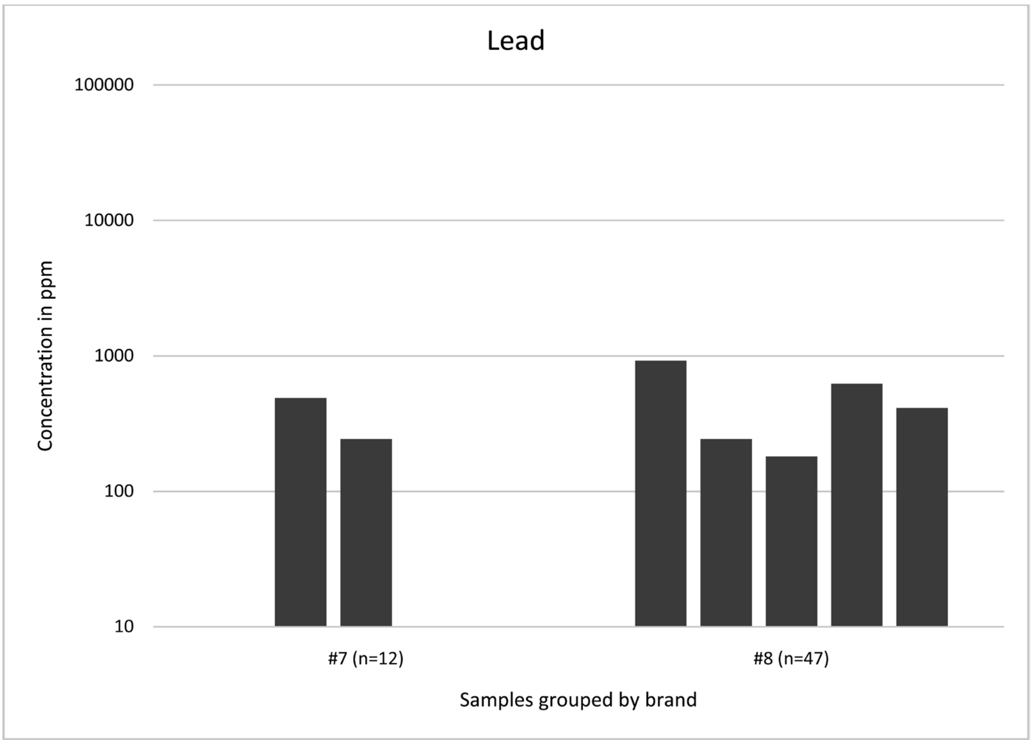


Figure 6. Ink samples containing lead above the LOQ. Samples were grouped by brand and n is the number of inks sampled by that brand. This includes all inks tested but only two brands contained any measurable lead. Each bar represents an individual sample. Concentration given on a logarithmic scale.

Figure 2 and **Figure 3** suggest that the quantities of chromium and nickel present in tattoo ink pigments vary significantly, as the inks from brands 2, 7, 8, and 10 contained concentrations of chromium and nickel that were 1 to 2 orders of magnitude higher than the other brands. Chromium is known to cause damage to the central nervous system [8] [18], and nickel is an allergen to approximately 20% of people and can cause skin sensitivity, rashes, and eczema [19].

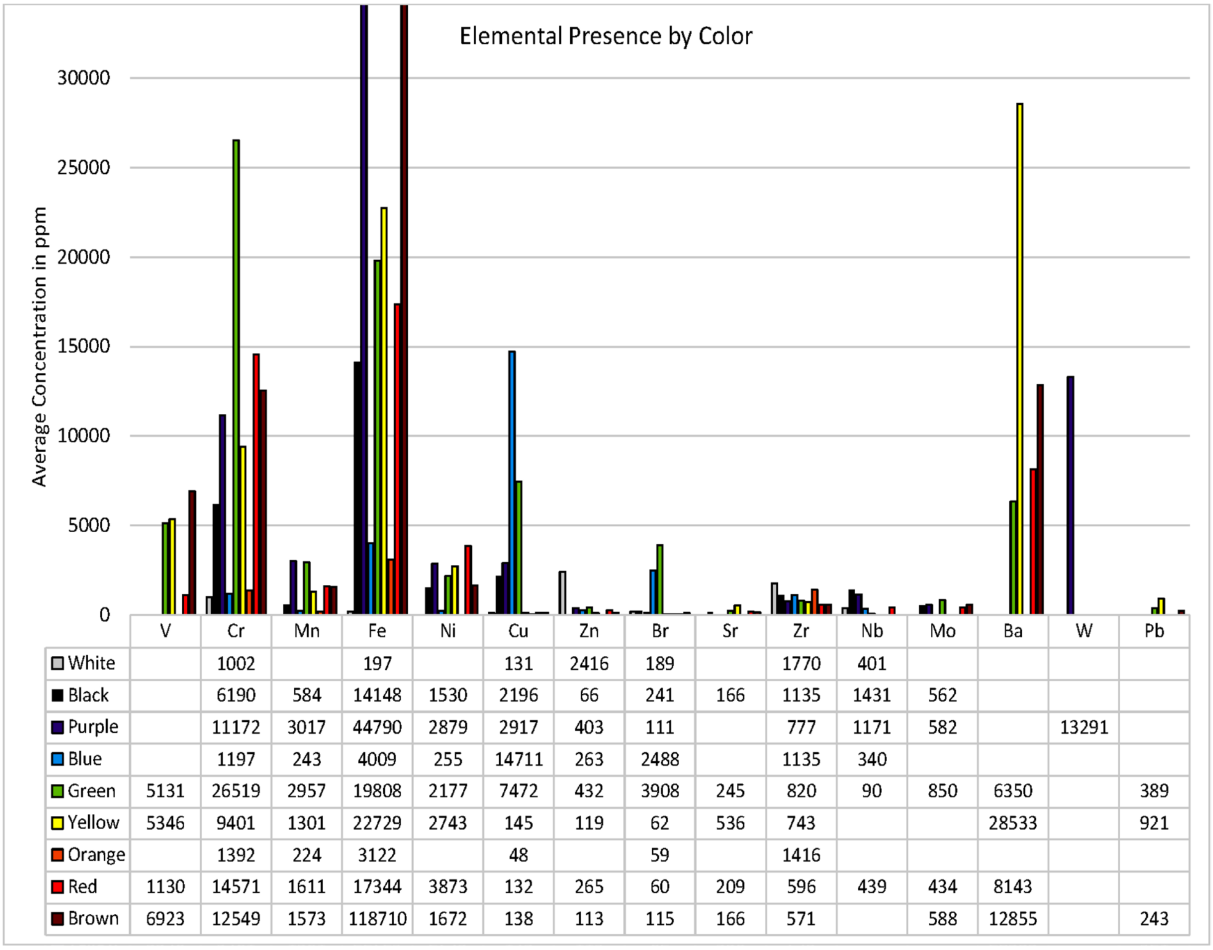
Figure 4 shows the copper distribution between brands, and shows more or less uniformity among manufacturers, compared to the previous figures. The widespread presence of copper in high concentrations in tattoo pigments is most likely associated with copper phthalocyanine blue BN, and phthalocyanine green—ink pigments that have been used in tattoos for almost 100 years. While copper can cause anemia, liver and kidney damage, as well as stomach and intestinal irritation [17] [18], it is thought to be relatively safe when it is bound to phthalocyanine. During tattoo removal procedures, however, this metal ion would be freed from its phthalocyanine "cage" and would dissolve into the blood stream.

Though very few of the inks contained barium, **Figure 5** is notable because of barium's toxicity and the high concentrations in the inks in which it was found. Barium can cause damage to the gastrointestinal tract and is a known skin sensitizer and can cause rashes and other skin problems with prolonged contact [8] [20]. It is expected that barium is present in the form of barium chromate, a bright yellow pigment, as each ink containing barium was found to be accompanied by chromium and was in its highest concentrations in yellow inks (**Figure 7**).

Figure 6 highlights the presence of lead found from two brands. Lead is a known toxin that can cause kidney failure, hypertension, neurological disorders, birth defects and liver damage [8]. Similar to barium, it is expected that the lead is present in the form of lead chromate, also a bright yellow compound, as each ink that contained lead also contained chromium and was observed exclusively in bright green and yellow inks (**Figure 7**).

There are other elements of concern that were observed frequently in the inks sampled. Among these are manganese and bromine. Manganese was seen in approximately 28% of the inks tested and is known to have adverse health effects. Overexposure to manganese in miners can cause a neurological disorder similar to Parkinson's disease, that causes tremors, difficulty walking, and muscles spasms [17] [21]. Bromine, which was found in over 26% of the samples, can cause severe skin damage such as blister formation, brownish discoloration, and ulcers, as well as neurological and kidney damage [22]. Bromine is likely to be part of organic pigments that were not the focus of this study.

The distributions of metals found in various tattoo inks sorted by color are shown in **Figure 7**. Bright green inks were found to contain high concentrations of chromium, manganese, iron, copper, bromine, and barium, as well as traces of lead. Yellow inks contain similarly substantial amounts of chromium, iron, nickel, and barium, and traces of lead. High concentrations of copper and bromine



	V	Cr	Mn	Fe	Ni	Cu	Zn	Br	Sr	Zr	Nb	Mo	Ba	W	Pb
⬜ White		1002		197		131	2416	189		1770	401				
⬛ Black		6190	584	14148	1530	2196	66	241	166	1135	1431	562			
🟪 Purple		11172	3017	44790	2879	2917	403	111		777	1171	582		13291	
🟦 Blue		1197	243	4009	255	14711	263	2488		1135	340				
🟩 Green	5131	26519	2957	19808	2177	7472	432	3908	245	820	90	850	6350		389
🟨 Yellow	5346	9401	1301	22729	2743	145	119	62	536	743			28533		921
🟧 Orange		1392	224	3122		48		59		1416					
🟥 Red	1130	14571	1611	17344	3873	132	265	60	209	596	439	434	8143		
🟫 Brown	6923	12549	1573	118710	1672	138	113	115	166	571		588	12855		243

Figure 7. Average concentration of each element (ppm) in ink samples by color. Titanium was excluded as it was found in very high concentrations for nearly every color.

were found present in blue inks. Red and brown inks both showed considerable amounts of chromium, iron, nickel and barium. Purple was the only color that showed high concentrations of tungsten, however, it only appeared in two inks which were both from brand 8. Purple also had extremely high concentrations of iron and substantial amounts of chromium. It was found that white and orange inks seemed to contain the fewest metals and these occurred at lower concentrations.

4. Discussion

This study showed that 16 elements, including 15 heavy metals, were present in tattoo inks in readily measurable amounts by rapid x-ray techniques. While a few of these elements are considered nontoxic, a majority of these findings are cause for concern because it is well established that chromium, manganese, nickel, copper, bromine, barium, and lead, all of which were commonly found in tattoo inks, have adverse health effects. Since nickel, barium, and bromine are known to cause skin damage such as rashes or eczema, it is suspected that these elements could contribute to infections and skin rashes that commonly occur around tattoos over time. More importantly, it appears that some manufacturers are able to provide tattoo inks that have much lower metal content than other manufacturers. These data suggest strongly that pigment design and use, as well as quality control still play an important role in the presence of metals in tattoo inks. In the absence of federal regulation of tattoo inks, we suggest that routine x-ray analysis of tattoo inks and tattoo ink pigments could help consumers avoid pigments with high metal concentrations. The purpose of this paper was not to identify specific brands that have fewer or more metals in their pigments; it was designed to bring attention to the fact that metals are being added to many tattoo inks and is a cause for concern. We do not wish to single out any manufacturer simply because our sampling was not comprehensive for any brands given the thousands of inks available commercially worldwide, and represents only a snapshot of some of the most popular brands in the US in 2014-2016. Details about specific inks or brands may be requested from the authors. Clearly further study is warranted on these inks and consumers that would like to avoid potential metal hazards in their inks should have their products tested regularly by any standard x-ray technique that measures metals. Similarly, the concentrations reported here are for dried pigment layers on paper, not actual inks applied to human skin, so the actual concentrations to which consumers are exposed remains unknown.

Another point of consideration is that the tattoo removal process is highly toxic to the body as the laser ablation sends all of the pigment directly into the bloodstream for the immune system to accommodate. Cases of blistering and rashes are common during tattoo removal, but recent cases have shown the deposition of carcinogenic compounds from the process as well [23]. Caution should be exercised with tattoo removal as heavy metal poisoning from the pigments is also plausible. The concentrations of metal ions in the bloodstream during tattoo removal is worthy of further study.

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