Quantifying the Rate Copper Leaches From a Copper Drinking Vessel Into Simulated Beverages Under Conditions of Consumer Use

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Abstract **The Moscow Mule cocktail, which contains ginger beer, lime juice, and vodka, is commonly served in a copper mug. There has been increasing concern that copper can leach into the cocktail, given the acidic nature of the drink. Under the experimental conditions studied, copper does leach from the copper mug into the beverage. We observed copper leaching into the cocktail solution at a rate of 0.048 ± 7 x 10^{-4} ppm copper/min at room temperature. The leaching rate was found to be dependent on the acidity of the solution (increasing at lower pH) and molecular oxygen content. We quantified the copper concentration using inductively coupled plasma-atomic emission spectroscopy (ICP-AES). The rate of copper leaching into the Moscow Mule cocktail was found to be significant and accumulated copper concentration exceeds the U.S. Environmental Protection Agency standards for drinking water within 27 minutes (World Health Organization, 2004). Any risk posed by the accumulation of copper, however, can be mitigated by serving the Moscow Mule cocktail in a copper mug lined with stainless steel to avoid direct contact of the acidic liquid with the copper surface directly, as stipulated by the Food and Drug Administration model *Food Code*.**

Introduction

Identifying the conditions under which potentially hazardous chemical agents, such as metal ions, are released from surfaces in contact with foodstuffs and beverages is an important first step in assisting environmental health professionals as they promote consumer safety. Copper leaching from a food contact zone into foodstuffs remains an undercharacterized process despite the presence of copper and copper alloy surfaces in both a) industrial food and beverage production and b) municipal water supplies. There are several food products—notably cheese (Rodriguez et al., 2011), beer (Zufall & Tyrell, 2008), distilled spirits (Neves et al., 2007), and tea (Karak & Bhagat, 2010; Lv et al., 2013)—that are brought into contact with a copper surface during production. Copper leaching is especially problematic for foodstuffs with low pH. Ishiwata et al. (1986) found that after 24 hr at room temperature, a 4% acetic acid aqueous solution in a copper mug contained 103 ± 10 ppm copper compared with a pure water solution in a copper mug, which contained 1.7 ± 0.1 ppm copper. The rate and mechanism of the copper leaching, however, was not reported.

Copper leached into foodstuffs has various potential impacts on consumer health. The recommended dietary allowance of copper for adults is 900 µg/day (Institute of Medicine, 2001). Copper has known health benefits and is essential for the functioning of some enzymes (Festa & Thiele, 2011). Copper also has a low incidence of eliciting allergic reactions (Fage et al., 2014). Little is known, however, about the toxicity of extended copper intake, and more research is needed to determine if copper intake over a prolonged period of time poses a significant public health risk (Brewer, 2010; Patel & Aschner, 2021).

In this article we use a popular cocktail traditionally served in a copper vessel as a model system to study copper leaching under conditions of simulated consumer use. This cocktail, known as the Moscow Mule, contains vodka, lime juice, and ginger beer. Much lore surrounds the reason why the drink is served in a copper mug, but many argue that the taste is enhanced by the copper vessel. A study by Hong et al. (2009) indicates that interactions between copper and salivary proteins could play an important role in the perception of flavor. Despite the potential flavor enhancement, there has been increasing public health concern regarding the safety of using a copper mug for a beverage as acidic as the Moscow Mule cocktail (State of Iowa Alcoholic Beverages Division, 2017). To our knowledge, the amount of copper leaching into the Moscow Mule cocktail has never been quantified. In this article we report the rate, total amount, and mechanism of copper leaching from a copper mug into a Moscow Mule cocktail.

TABLE 1

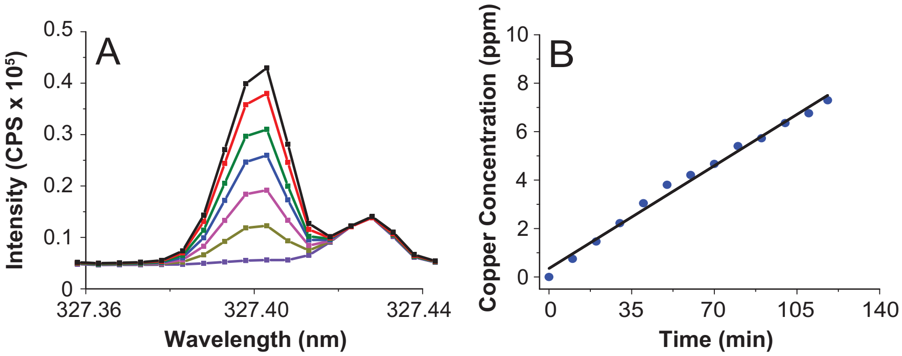
Ingredient Volumes and pH Values of the Moscow Mule Solution

Ingredient	Volume of Pure Ingredient Used (ml)[a]	pH of Pure Ingredient	pH of Ingredient After Dilution[b]
Lime juice	22	2.6	2.5
Ginger beer	133	3.0	3.2
200 proof ethanol	35.6	–	–
Deionized water	53.4	–	–

[a] The total volume of the Moscow Mule solution was 244 ml, which represents approximately one half of the volume of the copper mug.
[b] The pH of the ingredient after dilution to the final volume of 244 ml with deionized water; the pH of the Moscow Mule solution was 2.7.

FIGURE 1

Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AEC) and Copper Concentration From a Moscow Mule Solution Within a Copper Mug



Note. A) ICP-AES of copper from a Moscow Mule solution held within a copper mug at 0 min (purple), 20 min (dark yellow), 40 min (magenta), 60 min (blue), 80 min (green), 100 min (red), and 120 min (black). B) Copper concentration as a function of time for a Moscow Mule solution held within a copper mug. The copper concentration (blue circles) as a function of time was fit with a linear trendline (black). CPS = counts per second.

Methods

Moscow Mule Solution Preparation

All materials were used as received directly from the supplier. Cocktail ingredients were chosen to be representative of consumer use. Moscow Mule components included: lime juice, ginger beer, aqueous ethanol solution, and a 16-oz solid copper mug.

We prepared a Moscow Mule cocktail solution in a copper mug. Table 1 details the ingredients and their pH values. Ice was not used as an ingredient for any of the experiments conducted in this study. When analyzing the contribution each ingredient had on copper leaching, the individual ingredients were diluted with deionized (DI) water to the concentration typically found in a Moscow Mule cocktail. For the purposes of this study, vodka was replaced with 200 proof ethanol diluted to the appropriate concentration with DI water. For pH studies, an aqueous solution was brought to the desired pH using hydrochloric acid.

Measurements via Inductively Coupled Plasma-Atomic Emission Spectroscopy

All metal ion concentration measurements were performed using inductively coupled plasma-atomic emission spectroscopy (ICP-AES; PerkinElmer Instruments model Optima 2000 DV). Copper and gold ICP standards (GFS Chemicals, Inc.) were prepared in aqueous 1% nitric acid solution.

Internal Standards

We selected an internal standard of gold to quantify copper concentrations because the emission intensity is similar to copper, the emission maxima between copper and gold do not overlap, and any gold that might be present in the copper mug would not be expected to undergo a redox leaching process and contaminate the solution. For a given concentration, the copper emission at 327.393 nm was approximately 10 times more intense than the gold emission at 267.595 nm. We calculated the copper–gold response factor (*f*) for the ICP-AES instruments using the following equation:

$$\frac{\textit{Peak Area Copper}}{[\textit{Copper}]} = f\,\frac{\textit{Peak Area Gold}}{[\textit{Gold}]}$$

We calculated the copper–gold response factor over a range of concentrations to ensure minimal variance. The average response factor was 12.7 with a standard deviation of 0.1 over the concentration range investigated. Samples to be analyzed were taken from the copper mug at time intervals, transferred to volumetric flasks that had been cleaned with aqua regia (1:3 molar ratio of nitric and hydrochloric acid) to remove trace metals, spiked with 10 ppm gold, and then diluted to volume in preparation for ICP-AES analysis. The previous equation was used to calculate the copper concentration in the solution.

UV-Vis Measurements

All UV-Vis measurements were performed with an HP 8453 diode array UV-Visible spectrophotometer.

Scanning Electron Microscopy

All scanning electron microscopy (SEM) images were gathered using a Zeiss Supra 55VP field emission scanning electron microscope.

Results and Discussion

ICP-AES measurements demonstrated that copper does leach into a Moscow Mule solution in a copper mug. Figure 1A shows ICP-AES measurements of copper from a Moscow Mule solution in a copper mug at time intervals of: 0 min (purple), 20 min (dark yellow), 40 min (magenta), 60 min (blue), 80 min (green), 100 min (red), and 120 min (black). Figure 1B shows copper concentration as a function of time for a Moscow Mule solution in a copper mug; the copper concentration (blue circles) as a function of time was fit with a linear trend line (black).

We observed copper leaching into the solution at a rate of 0.048 ± 7 x 10^{-4} ppm copper/min at room temperature (Figure 1B). At this rate, the concentration of leached copper in a copper mug reaches 1.3 ppm in slightly over 27 min. The U.S. Environmental Protection Agency mandates that copper levels in drinking water that exceed 1.3 ppm must be reported (World Health Organization, 2004). The Food and Drug Administration (FDA) model *Food Code* prohibits foodstuffs with a pH < 6.0 from coming in contact with copper due to concerns of copper leaching (U.S. Department of Health and Human Services, 2017). The Moscow Mule solutions in our experiments had a measured pH of 2.7 and the pH did not change throughout the course of the experiment. Despite FDA regulations, Moscow Mule cocktails routinely are served in copper mugs in establishments all over the country.

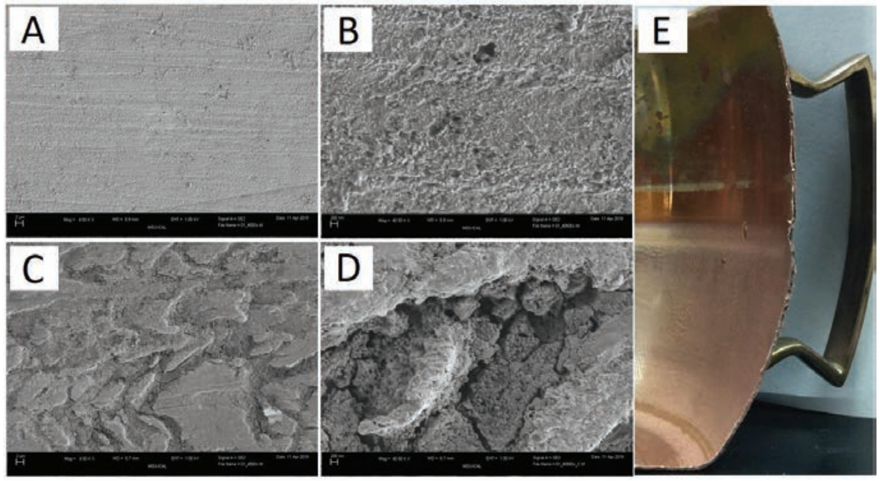
It is informative to consider the maximum daily allowance of copper that can safely be consumed. According to the World Health Organization (2004), a safe maximum consumption of copper is 10 mg/day. Thus, an individual would need to consume over 30 Moscow Mule cocktails (each containing 1.3 ppm of copper and a volume of 244 ml) to exceed the limit of 10 mg of copper per day. Given this information, acute copper toxicity from consumption of Moscow Mule cocktails in one sitting is unlikely. As mentioned previously, however, the long-term effects of elevated copper consumption are largely unknown (Brewer, 2010; Patel & Aschner, 2021).

We observed slight differences between the copper leaching rates for the mugs used in this study, but copper leaching was observed under all conditions studied. While it might not be possible to directly apply the specific leaching rate values presented here to a Moscow Mule cocktail prepared under other conditions, the overall trend of copper accumulation appears to hold true.

The difference in the copper leaching rate between the mugs did not appear to be correlated with any properties of the mug that could be assessed with the unaided eye. The geometric surface area and microscopic electrochemically active surface area could both be important factors that contribute to the difference in copper leaching rates among the mugs used in this study. The microscopic surface area of the mugs used in this study was characterized using SEM. Figure 2 shows SEM images of the copper mug surface with limited contact with the Moscow Mule cocktail at 2 **μm** (Figure 2A) and 200 nm (Figure 2B) scale. SEM images of the copper mug surface after exposure to

FIGURE **2**

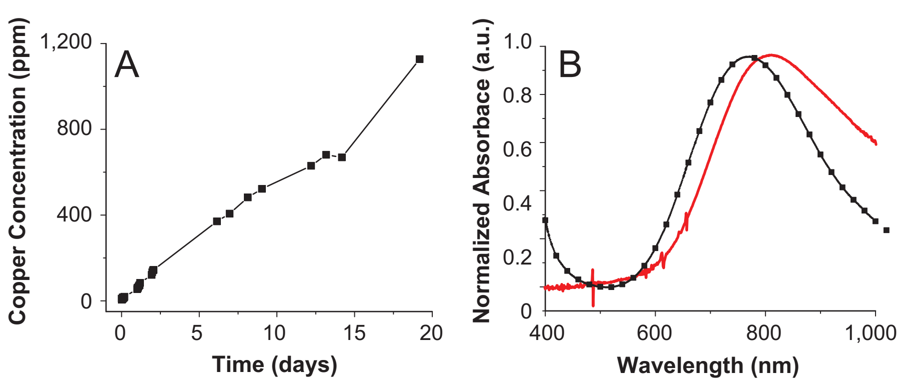
Scanning Electron Microscope (SEM) Images of Copper Mug Surface



Note. SEM images of copper mug surface with limited contact with the Moscow Mule cocktail at A) 2 μm scale and B) 200 nm scale. SEM images of copper mug surface after exposure to 26 Moscow Mule cocktails for a cumulative exposure time of 75 hr at C) 2 μm scale and D) 200 nm scale. E) Cross-sectional digital photograph of the copper mug; upper half of the mug had limited exposure and the lower half of the mug had exposure to 26 Moscow Mule cocktails for a cumulative exposure time of 75 hr.

FIGURE **3**

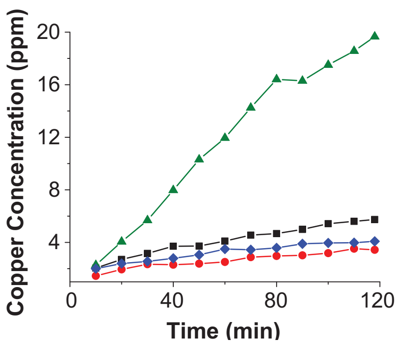
Copper Concentration as a Function of Time and Normalized UV-Vis Absorption Spectra



Note. A) Copper concentration as a function of time for a Moscow Mule solution held within a copper mug over the course of 20 days. B) Normalized UV-Vis absorption spectra of an aqueous copper(II) nitrate solution (red) and a Moscow Mule solution held within a copper mug for over 20 days (black squares).

FIGURE **4**

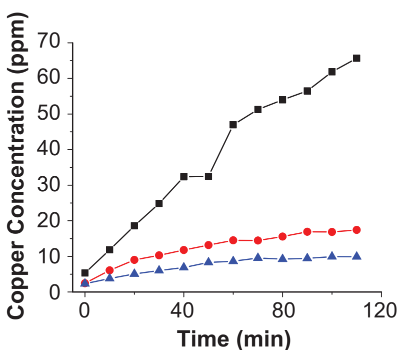
Copper Concentration as a Function of Time for Moscow Mule Ingredient Solutions Held Within a Copper Mug



Note. The Moscow Mule ingredient solutions include ginger beer (green triangles), lime juice (black squares), deionized water (blue diamonds), and ethanol (red circles).

FIGURE **5**

Copper Concentration as a Function of Time for Hydrochloric Acid Solutions of Varying pH

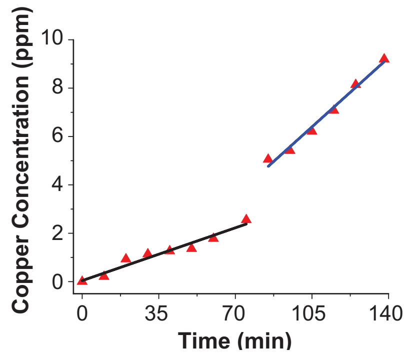


Note. The solutions had pH values of 1 (black squares), 2.9 (red circles), and 4.5 (blue triangles).

26 Moscow Mule cocktails for a cumulative exposure time of 75 hr are shown at 2 µm (Figure 2C) and 200 nm (Figure 2D) scale. Figure 2E shows a cross-sectional digital photograph of the copper mug. The upper half of the mug had limited exposure to the Moscow Mule solution and the lower half of the mug had a cumulative exposure time of 75 hr.

FIGURE **6**

Copper Concentration as a Function of Time for Moscow Mule Solutions Held Within a Copper Mug



Note. Each Moscow Mule solution initially was sparged with nitrogen gas to remove atmospheric oxygen and then placed in the copper mug under a nitrogen atmosphere. At 75 min, the solution was sparged with atmospheric gas for 15 min to replenish dissolved oxygen. Linear fits of the oxygen-free (black) and oxygen reintroduced (blue) regions are shown.

The oxidation of elemental copper to aqueous copper(II) ions results in both the leaching of copper(II) ions into the solution and the formation of microstructures as well as nanostructures on the copper surface. Outside of a controlled laboratory environment, the washing, polishing, and repeated use of copper surfaces likely has an effect on the electrochemically active microscopic surface area. Additionally, mechanical polishing is likely to obscure the visual evidence of the chemical etching, thus making it more difficult for consumers to realize that contaminates are being introduced into their beverage.

In our experiments, after being left undisturbed for several days in the mug, the cocktail solution turns a distinct turquoise color and we measured the copper concentration to be as high as 1,000 ppm. Figure 3A shows copper concentration as a function of time for a Moscow Mule solution in a copper mug over the course of 20 days. Normalized UV-Vis absorption spectra of an aqueous copper(II) nitrate solution (red) and a Moscow Mule solution in a copper mug for over 20 days (black squares) are shown in Figure 3B. The concentration of copper as a function of time is linear over the course of 20 days, consistent with a zero-order reaction mechanism (Figure 3A). Zero-order reactions have been encountered in other heterogeneous reactions where access to the surface limits the rate at which the reaction proceeds.

Due to the zero-order reaction kinetics, the copper is continuously accumulating in the Moscow Mule solution and does not equilibrate at a fixed value. Thus, while not directly applicable to the typical consumer experience with a Moscow Mule cocktail, the continuous leaching over the 20-day study highlights the importance of applying the FDA model *Food Code* prohibiting acidic foodstuffs coming in contact with any copper surface. More broadly, the accumulation of metal ions into acidic foodstuffs and drinking water with prolonged exposures to metal surfaces should not be overlooked by environmental health professionals. For example, water with a low pH in metal pipes was an important and preventable factor that contributed to the high levels of lead leached into the Flint, Michigan, water system in 2014 (Torrice, 2016).

The constant leaching rate of copper into the Moscow Mule solution contrasts with the time-dependent leaching rates observed from most other metal surfaces into foodstuffs or simulated foodstuffs. For the leaching of chromium, iron, and nickel from a stainless steel surface, the initial leaching rate was fastest and the leaching rate decreased with time (Herting et al., 2008; Kamerud et al., 2013). Additionally, the decrease in the leaching rate of tin from metal cans into foodstuffs was attributed to the eventual consumption of all the oxygen dissolved in the foodstuffs or trapped in the headspace (Parkar & Rakesh, 2014). Finally, chromium is unique among the other metals studied and was found to leach from a stainless steel surface at a constant rate on a 20-day time scale (Chiavari et al., 2014).

Solutions of the individual ingredients were diluted with DI water to the concentration found in a Moscow Mule cocktail (Table 1) to study the effect of each ingredient on the copper leaching rate. Figure 4 shows copper concentration as a function of time for solutions of ginger beer (green triangles), lime juice (black squares), DI water (blue diamonds), and 14% ethanol (red circles) in a copper mug. We observed copper leaching with all four ingredients investigated. The highest leaching rates were observed for ginger beer. Lime juice and ginger beer had the lowest pH values and fastest copper leaching rates.

We systematically investigated the effect of pH on the copper leaching rate by preparing hydrochloric acid of varying pH. Figure 5 shows copper concentration as a function of time for solutions of varying pH. The aqueous solutions had pH values of 1 (black squares), 2.9 (red circles), and 4.5 (blue triangles). As the pH of the aqueous hydrochloric acid solution decreased, the rate of copper leaching increased. The data in Figures 4 and 5 are consistent with pH being an important predictor of copper leaching rate, but it is not the sole contributor.

Interestingly, the lowest pH component (lime juice) of the cocktail solution does not result in the fastest leaching rate, suggesting that there are other species in solution that contribute to copper leaching. This result is consistent with studies (Agarwal et al., 1997) that showed that chromium and nickel leached from stainless steel vessels at a higher rate for foodstuffs than for pH-equivalent aqueous solutions of the predominate pure organic acids found in the foodstuffs. The ginger beer solution (133 ml diluted to 244 ml) is much more concentrated than the lime juice solution (22 ml diluted to 244 ml) once diluted to the total volume of the drink—thus any effect due to other species in solution could be more pronounced.

We investigated the mechanism by which metallic copper is transformed to copper(II) and found molecular oxygen to have a pronounced effect on the rate of copper leaching into the solution. Figure 6 shows copper concentration as a function of time for Moscow Mule solutions held within a copper mug (red triangles). The Moscow Mule solution initially was sparged with nitrogen gas for 15 min to remove atmospheric oxygen and then placed in the copper mug under a nitrogen atmosphere. At 75 min, the solution was sparged with atmospheric gas for 15 min to replenish dissolved oxygen. Linear fits of the oxygen-free (black) and oxygen-reintroduced (blue) regions are shown.

For the mug used in this experiment, copper leaches into the nitrogen-sparged Moscow Mule solution at a rate of 0.03 ± 0.003 ppm copper/min. Once oxygen was reintroduced, the copper leaches into the Moscow Mule solution at a rate of 0.08 ± 0.005 ppm copper/min. The 2.6-fold increase in the copper leaching rate is consistent with molecular oxygen acting as an oxidant in the copper leaching mechanism. Interestingly, the copper leaching rate is not zero under oxygen-free conditions, suggesting that the other ingredients in the Moscow Mule solution could contain compounds that act as oxidants under these conditions.

There are several important factors that must be taken into consideration before directly applying the findings here to a consumer setting. First, the copper leaching rate varied among different mugs. The electrochemically active surface area of a mug, and therefore the rate of copper leaching, is strongly dependent on the mechanical and chemical processes that mug has experienced. Second, the studies we conducted were at room temperature, whereas a Moscow Mule cocktail typically is served over ice. The slightly elevated temperature of the Moscow Mule solution in this study likely results in a lower dissolved gas concentration and a slower copper oxidation reaction rate constant.

Therefore, the rate of copper leaching in a Moscow Mule cocktail served to a consumer may be different than that reported here. Regardless, our results clearly demonstrate that copper leaching does occur at an appreciable rate under multiple solution conditions, and thus supports the discontinuance of serving an acidic cocktail such as the Moscow Mule in a copper mug.

Conclusion

In summary, under the conditions studied, copper leaches into the Moscow Mule solution at a constant rate. The zero-order copper leaching kinetics are consistent with a reaction mechanism that is rate limited by the microscopic surface area of the copper mug. We also found the leaching rate to be dependent on pH and dissolved oxygen concentration. Other ingredients in solution, however, might also act as oxidants or chelating ligands that could accelerate the copper leaching rate.

In this article, we provide an intriguing and relevant example to environmental health professionals and the public of a potentially hazardous substance that is common and at the same time extremely easy to avoid. Our study presents a clear alternative for environmental health professionals and the public, as fortunately copper mugs lined with stainless steel or other chemically inert materials are widely available for a similar cost. As such, the potential hazard posed by the direct contact between an acidic beverage—such as the Moscow Mule—and the copper surface could easily be mitigated.

While our study focused on one particular cocktail, the identified mechanism and rate of copper leaching can inform environmental health professionals of the "why" behind this regulation and enable them to effectively evaluate plan reviews and carry out inspections in related situations. In particular, review of large catered events where specialty drinks or other specialty foods might be served should prompt an environmental health professional to ask more questions and determine if vessels lined with stainless steel might be more appropriate. In addition to the acidity of the foodstuffs, the temperature and the amount of time the product is contained in the copper vessel could all impact safety implications. In agreement with the FDA model *Food Code*, individuals should avoid consuming foodstuffs with a pH lower than 6.0 that have come in contact with copper.

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