Using single particle ICP-MS for monitoring metal-containing particles in tap waters

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Water Impact statement

Engineered nanomaterials (ENMs) are used extensively in a variety of commercial products and applications. ENMs in tap water may originate from contaminated source waters or be generated in distribution systems (e.g., corrosion related), and hence it is essential to monitor the presence of metal-containing particles in drinking waters to understand their life cycle and associated human exposure risks.

Using single particle ICP-MS for monitoring metal-containing particles in tap waters

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Abstract

Engineered, natural or incidental colloidal sized materials in tap water may originate from source waters or be generated in distribution systems (e.g., corrosion related). An optimized single particle (sp)-ICP-MS technique was applied to tap waters (*n* = 50) collected from three buildings to analyze for Pb, Fe, Sn, Cu, Ag and Ti-containing particles. Pb, Sn and Fe-containing particles were detected at an average concentration (ng/L) of 1.2 (range: 0.06 – 4.8), 1.8 (range: 0.11 – 14), and 88 (range: 26 – 890), respectively, representing at a minimum 0.4 %, 10 %, and 15 % of the corresponding total dissolved metal concentrations. No particulate Ti and Ag was observed in samples. Pb concentrations in the first 125 mL fraction collected was on average three times higher than the subsequent samples. Detection of Cu particles required modification of the sample introduction system (direct self-aspiration into nebulizer) to reduce matrix interaction with auto-sampler tubing. Cu-particles were detected in 50 % of analyzed samples at an average concentration of 69 (range: 15 – 136) ng/L. While all metal concentrations were below health advisory levels, this study showcases the feasibility and first application of spICP-MS to monitor metal-containing particles in tap waters, and the results suggest that the particulate forms of the studied elements may represent a significant fraction of the bulk elemental concentration in tap waters.

Keywords: Tap water; Drinking water; Nanoparticles; Single particle ICP-MS; Lead.

Introduction

Engineered nanomaterials (ENMs) are reported in common consumer products such as cosmetics, toothpastes, and sunscreens [1] and they may pose unintended consequences upon release into the environment. As a consequence, there have been advances in analytical techniques to differentiate nano-sized particles from dissolved ions or micron to larger sized particles in water. Resulting from the focus on human exposure to ENMs, there is recognition that ENMs exist as a relatively small fraction of natural (and incidental) colloidal or nano-sized particles [2]. Nanoparticle concentrations in environmental waters are expected to be at the parts-per-trillion (ng/L) level [3]. It is challenging to detect such low concentrations of nanoparticles using analytical methods such as dynamic light scattering (DLS), differential centrifugal sedimentation (DCS), and nanoparticle tracking analysis (NTA), because the minimum concentration required to obtain a feasible particle size distribution is in the parts-per-billion (µg/L) range for these techniques [4]. Unlike DLS, DCS and NTA, single particle inductively-coupled plasma mass spectrometry (spICP-MS) method is capable of detecting and sizing nanoparticles at environmentally relevant concentrations (e.g., ng/L) in addition to providing chemical information [4-6]. Previous work has primarily focused on developing spICP-MS technique and comparing them to other methods capable of sizing and quantifying nanoparticles in environmentally relevant levels [4, 7-10].

Single particle ICP-MS is a time-resolved analysis that detects particles as pulses above the background elemental signal [11, 12]. Dissolved analytes are homogenously dispersed in a sample and determines the intensity of the background signal. Hence, the sensitivity and accuracy of spICP-MS to determine and size nanoparticles depends largely on the dissolved concentration since nanoparticle "pulses" are typically considered to be those with counts greater than three or four times the standard deviation (3σ or 4σ) of the background signal. For particles to be accurately sized, background dissolved concentrations would ideally be low so a clear distinction between background and small particle 'pulse' can be made. The size is determined from the intensity of each nanoparticle event and is binned in a particle size distribution (PSD) histogram to determine the average size of the particles and their polydispersity [4, 13]. Several studies have applied spICP-MS technique to study the fate of nanoparticles in fortified water samples [14, 15], with a few recent works featuring its application in monitoring non-fortified natural waters [6, 16]. Nano- and colloidal particles in tap waters, to the best of our knowledge, have not been monitored using spICP-MS and remain unregulated because of the lack of data confirming their presence and toxicity. In cities' distribution systems, metal-containing particles may occur from either the survival of particles through water treatment systems and/or particle generation from the treatment process. The distribution system may additionally contribute to the particles in tap water since piping is likely to corrode and/or solder could be freed by shear [17-19]. Pb, Fe and Sn particulates from distribution pipelines, solders, and fixtures have been reported to occur in tap and drinking waters [19-22]. Nanosized iron oxide particles released from the distribution system can further adsorb hazardous chemicals such as Pb and As, and hence the presence of such particles in distribution pipelines can become a health hazard [21, 22]. Prior research has used various analytical methods including filtration [23], magnetic measurements [22], and size-exclusion chromatography with ICP-MS [21] to detect and characterize such particles.

The aim of this study was to develop the capability to apply spICP-MS to tap water. Tap water samples from different buildings, collected over time, were analyzed using spICP-MS to detect and size Pb, Fe, Sn, Cu, Ag and Ti-containing particles. Ag and Ti containing particles were thought to be potentially related to ENMs that may have entered the distribution system after treatment [24]. Ti containing particles also occur naturally and may originate from the source water. The other metals were likely to be related to corrosion and scaling by-products in the distribution pipelines [18, 25]. It was necessary to implement careful equipment operating conditions to successfully avoid matrix interferences and apply spICP-MS for routine monitoring of metal-containing particles in tap waters.

Experimental

Reagents

The ionic elemental standards were purchased from VHG Laboratories (Manchester, UK). Trace metal grade nitric acid (70%) and hydrochloric acid (33–36%) were purchased from EMD Chemicals (OmniTrace, EMD Chemicals, Gibbstown, NJ, USA) and J.T. Baker (Ultrex II, JT Baker Inc., NJ, USA), respectively. Tannic acid stabilized Au reference nanomaterial (80 nm) was purchased from NanoComposix (CA, USA). All other chemicals were obtained from Sigma-Aldrich (MO, USA) unless specifically stated. Deionized water (18.3 MΩ cm, NANOpure Infinity, LA, USA) was used to dilute samples.

Sample Collection

Tap (drinking) water samples were obtained from sink faucets over the course of 4 days at 3 different sites in Phoenix, Arizona: (i) an apartment complex in Tempe, AZ (building 1) (ii) a new Arizona State University (ASU) building with a 2012 Leadership in Energy and Environmental Design (LEED) certification (referred to as building 2); and (iii) an older ASU classroom/office structure built in 1976 (building 3). All three locations have the same source water and are served by the same drinking water treatment plant. At each location and day, two sets of samples were collected: one during the morning (AM) and one during the afternoon (PM) to understand the impact on stagnant water overnight and during peak usage. For each sampling period an aliquot of 125 mL (referred to as F125) was obtained immediately when the faucet was turned on. This was done to investigate possible buildup of metal particles in the pipes over time. The faucet was then run for a full minute before obtaining a bulk sample of 500 mL (referred to as bulk). For Cu analysis, 14 additional tap water samples were collected from building 2 for analysis using a modified spICP-MS method (see results section). A total of 50 tap water samples were collected (36 + 14), stored in refrigerator and analyzed within seven days after collection.

spICP-MS Instrumentation and Data Analysis

Analysis by spICP-MS was performed using a Thermo Scientific (Waltham, MA) X-Series II ICP-MS in time-resolved analysis (TRA) mode [6, 13]. The TRA mode provides output signal as intensity (cps) versus time. An 80 nm Au reference nanoparticle standard at 200 ng/L was analyzed to determine transport efficiency using the particle mass method described elsewhere [13, 26]. Calibration standards were prepared in 2% HNO_3 for ^{208}Pb, ^{56}Fe, ^{118}Sn, ^{65}Cu, ^{49}Ti, ^{107}Ag, and 2% HCl for ^{197}Au. All elements, except ^{56}Fe, were measured in the normal mode using argon as the carrier gas. ^{56}Fe was measured in the collision cell technology (CCT) mode. Calibration standards were analyzed for 70 seconds and tap water samples were analyzed for 180 seconds. The instrument was tuned prior to each sample batch using a solution containing Li, In, Ce and U at a concentration of 10 μg/L and by monitoring for minimum oxide levels (CeO/Ce < 2%). A dwell time of 10 ms was used. Sample flow rate was measured by weighing the water mass pumped in one minute (mL/min) and was used for calculating the transport efficiency, i.e., the fraction of sample reaching the plasma and analyzed [26].

The most commonly used approach for sizing particles is to measure the average and standard deviation of the background signal through an iterative analysis, and considering anything above a chosen cut-off (mean + x * σ) as a particle detection event. Particulate pulses with instrument response greater than 4 standard deviations (4σ) from the background signal were counted as particle events in the present study. The pulse intensity above background was then converted to a mass via a dissolved calibration curve, and to diameter via particle density and spherical geometry. We assumed that all metal-containing particles are composed of single element and used the density of the corresponding element to obtain particle size distributions. To calculate particulate mass concentrations, the elemental mass corresponding to all pulse signals from a given run was divided by the sample volume analyzed, which was known based on the instrument flow rate and analysis time [4, 13]. The average background/baseline signal of the elements in spICP-MS for the samples (i.e., $<4\sigma$) was used to determine ionic elemental concentration and concentration of particles less than the size detection limit (SDL) of the instrument. In this paper, 'background' concentration is defined as the ionic elemental concentration plus metal-containing particles below SDL. The size detection limit for the particles will vary depending on both the instrument sensitivity and the standard deviation of the background signal (sample matrix). The instrument's minimum detectable size determined for Pb, Fe, Sn, Cu, Ti and Ag were 11.3, 55, 26, 40, 75 and 13 nm, respectively [13].

Transmission Electron Microscopy

About 50 mL tap water sample was sonicated for five minutes to suspend particles. A Ted Pella carbon type-B transmission electron microscopy (TEM) grid was placed at the bottom of the tap water sample in a centrifuge tube. The sample was centrifuged at 4,600 G for 4 hours to settle any metal-containing particles present on to the surface of the TEM grid. Microscopy was performed on a JEOL 2010F TEM (Peabody, MA, USA) with energy dispersive X-ray spectroscopy (EDS). EDS data is reported in a counts vs. energy (KeV) graph. Copper peaks are a result of the copper TEM grids used for analysis. Mean particle diameter was measured manually with ImageJ^{TM} software.

Results and discussion

Metal-containing particle detection and quantification in tap waters by spICP-MS.

For each sample, time-resolved spICP-MS data was obtained for Pb, Fe, Sn, Cu, Ti, and Ag. Pb, Fe, and Sn particles were detected in all samples with varying concentrations, with Fe being the most abundantly detected particles in tap water samples at an average concentration of 88 (range: 26 – 890) ng/L (Figure 1). Pb and Sn particles were detected at an average concentration of 1.2 ng/L (range: 0.06 – 4.8) and 1.8 ng/L (range: 0.11 – 14.1), respectively. Though particles (pulses) were detected in all samples, the particle concentrations for Pb (53 %) and Sn (44 %) were low (< 1 ng/L) in about half the samples analyzed. No particulate Ti and Ag was observed in any samples analyzed. Cu-containing particles were detected after method modification and the results are discussed in the section below.

Pb-containing particles. Concentration of particulate Pb was < 1 ppt in buildings 1 and 2, but higher in building 3 with an average concentration of 2.6 (range: 0.45 – 4.8) ng/L. The average background Pb-concentration (< 4σ) in buildings 1, 2, and 3 was 125 (range: 34 – 476), 441 (range: 181 – 690), and 876 (range: 177 – 2059) ng/L, respectively. To the best of our knowledge, there are no lead pipes in the buildings that we sampled; but older homes in the metro-Phoenix region might contain lead. Before enforcement of the Federal Lead Ban in 1988, solders were typically made of 40 to 50% lead [27]. Hence, the elevated Pb concentration in building-3 relative to other buildings could be attributed to the age of the structure (built in 1976), which could contain a piping system leaching Pb (Pb-based solder shearing off). The Environmental Protection Agency drinking water action level for lead is 15 µg/L [27], and all drinking water measurements in this study were below this level. Background and particulate-Pb concentrations were higher in the initial 125 mL samples compared to their secondary bulk samples; this observation was especially pronounced in building-3 for background Pb concentration (F125 = 1.53 ± 0.49 µg/L and bulk = 0.22 ± 0.04 µg/L) (Figure 1a and 1b). Mean background Pb concentrations in AM samples from the three buildings were generally higher than the PM averages (see supporting information (SI) Figure S1b); whereas no significant difference was observed for the particulate Pb form between AM and PM samples. These data are indicative of dissolved Pb and/or Pb particles smaller than the SDL building up in pipes close to the faucet over short time scales. Particulate Pb fraction detected in the present study was low at only 0.4 (range: 0.09 – 1.8) % of the total bulk Pb by mass (Figure 2). Prior research indicates that particulate Pb may enter the distribution lines from corrosion of lead bearing elements in the premise plumbing and/or via adsorption of dissolved Pb onto suspended solids [19, 20, 25].

Fe-containing particles. Buildings 1, 2 and 3 exhibited average Fe-containing particle mass concentration of 40.8 (range: 26 – 237), 65.1 (range: 51 – 890), and 63.0 (range: 34 – 213) ng/L as Fe, and corresponding background concentration of 615 (range: 595 – 1738), 703 (range: 336 – 1289), and 105 (range: 501 – 1088) ng/L as Fe, respectively. The average particulate fraction of Fe in the tap water samples was calculated to be 10 (range: 2 – 71) % (by mass) of the total bulk Fe, with one sample from building 2 featuring as high as 71% particulate-Fe in the initial 125 mL fraction (F125) collected during the morning of the first sampling day. Elevated particulate Fe content was detected in the F125 (Figure 1c) and most morning (AM) samples collected immediately after turning on the tap (SI Figure S1c). This can be likely attributed to stagnant water leaching Fe-particles from the distribution pipes [18]. The background Fe concentration, however, was mostly similar between the various sample fractions.

Sn-containing particles. Particulate Sn concentration was low and detected near or less than 1 ng/L concentration in buildings 1 and 3. Building 2 featured a relatively higher average concentration of 3.9 (range: 0.8 – 14) ng/L. The average background Sn concentration ($< 4\sigma$) in buildings 1, 2, and 3 was 5.2 (range: 2.7 – 7.9), 19.5 (range: 3.1 – 106), and 12.1 (range: 4.2 – 45) ng/L, respectively. No observable trends occurred in either dissolved or particulate Sn concentrations between F125 and bulk fractions. However, build-up of Sn particles overnight was noticed for building-2, but not for buildings 1 and 3 (SI Figure S1e). We expected similar results for Sn in building-3, since Pb levels were high and the presence of both Sn and Pb could then be associated with leaching from solder material in pipes. It is not clear what the sources are for these particles in tap waters and further research is needed. One study evaluated the presence of contaminants in scales collected from pipe specimens across the U.S. and categorized Sn as a 'moderate contaminant' based on the concentration detected in scale samples [25]. Sn was detected in 55% of the samples analyzed in that study at an average concentration of 1129 mg/kg. Hence, scaling in the pipelines in addition to leaching from solder material [19, 20], could likely be the source for the detected particles and is supported by the high particulate fraction observed for Sn in the present study (15 (range: 2.2 – 33) % of the total bulk Sn concentration; Figure 2).

Method modification for detection of Cu-containing particles

Detection of Cu particles was initially challenging due to fluctuation of baseline observed in all tap water samples (Figure 3a). We measured elevated Cu concentrations using conventional ICP-MS to be between 0.2 and ~1 mg/L in building-2, a Leadership in Energy and Environmental Design (LEED) certified building built in 2012. The building facility managers were able to reduce Cu levels in the building through daily flushing of the entire six-story building plumbing system. LEED certified buildings are designed to reduce water consumption (> 60%), but it appears building and plumbing codes do not require downsizing pipe diameters. As a result, long retention time in the pipes depletes chlorine residuals and influences copper corrosion [28].

To identify the cause for baseline interferences, we performed controlled experiments using CuO_2 as a model nanoparticle spiked into nanopure water. We were able to point out that the baseline interference was caused due to the interaction of background Cu (-complexes) with the sample introduction system (SIS) tubing of the ICP-MS instrument. However, we were not able to identify what the tap water constituent was that caused this interference. Multiple spICP-MS runs were performed using the model nanoparticle (~ppt range) in the presence of common tap water constituents (carbonates, bicarbonates, NOM etc.), but none of these contributed to the interference observed. In order to reduce the sample interaction with SIS tubing, we evaluated two modifications to the spICP-MS method: (i) addition of a surfactant/complexing agents to the sample; and (ii) reducing the length of sample tubing. For the first modification, we tested the addition of sodium dodecyl sulfate (SDS), EDTA and 18-crown-6 (1,4,7,10,13,16-hexaoxacyclooctadecane) to tap waters and nanopure water spiked with CuO_2 particles. The justification for using these compounds are as follows: EDTA and 18-crown-6 were used in order to complex with Cu to eliminate the background Cu interacting with tubing; whereas, SDS was used in order to keep the particles in suspension and thus reducing particle interaction with the tubing. EDTA, crown ethers, and SDS have been used for ICP-MS analysis in the past for preservation, speciation and separation purposes [29-31]. Complexation with EDTA and 18-crown-6 approach did not work, and the interference persisted in spICP-MS analysis (Figures 3b & 3c). The use of SDS at ~12 mM concentration in samples helped with stabilizing the baseline spectrum of Cu in tap water samples (Figure 3d). However, we observed an increase in the intensity of the baseline spectrum, thus significantly affecting the resolution and SDL of Cu-particles. In the second modification approach, to reduce the sample interaction with SIS tubing we bypassed the auto-sampler and introduced the sample directly into the ICP-MS nebulizer through a short tubing (SI Figure S2); i.e. the sample was allowed to ‘self-aspirate’ into the nebulizer. This modification stabilized the background signal for Cu and allowed us to detect and quantify Cu-containing particles in tap waters (Figure 3e). Additionally, this modification did not alter the background intensity of the spICP-MS spectrum and thus did not have any impact on the resolution of the method.

The self-aspiration technique was used to determine the Cu-containing particle concentration in samples. A new set of samples ($n = 14$) was collected from building 2 (after the pipelines were flushed and Cu concentrations were reduced) and analyzed using the modified method for Cu-particles in tap water (transport efficiency was calculated for self-aspiration using ionic calibration and reference Au standard). Cu-containing particles were detected in 50 % of the samples analyzed at an average concentration of 69 (range: 15 – 136) ng/L. We also noticed that there were no substantial differences between F125 and bulk, and between AM and PM samples for Cu concentrations. This could be as a result of the very high background Cu concentration (34 ± 28 μg/L) present in the water samples, that might have masked the instrument's ability to distinguish particulate events from dissolved Cu (see next section).

Sizing of metal-containing particles in tap waters and confirmation by TEM

In spICP-MS, detection of particles is primarily impacted by the intensity of the baseline signal caused by ionic/background fraction of the element analyzed. The background concentration, as determined by spICP-MS baseline spectrum (i.e. $< 4\sigma$), was in the ng/L range for Sn (11 ng/L; range: 2.7 – 106), Pb (421 ng/L; range: 34 – 2059), and Fe (836 ng/L; range: 336 – 1738). This enabled us to detect true nanoparticles (< 100 nm diameter) of these elements in the tap water samples as seen in the PSD plots (Figures 4a–c). The minimum particle size detected for Sn, Pb and Fe in the present study were roughly 45, 60, and 90 nm, respectively. The minimum size detected in the present study was higher than the minimum particle size (D_{min}) calculated previously for these elements using the same instrument at 26, 13, and 55 nm, respectively [13]. This was expected since the calculated D_{min} values in the latter study represented lower end sensitivity of the instrument under the best conditions, and the difference is very likely due to tap water matrix interferences. In case of Cu, the average background Cu concentration in the samples analyzed was 34 ± 28 μg/L and was almost two orders of magnitude higher than the other elements monitored. Hence, the minimum particle size detected in tap waters was ~140 nm, which is not nanoparticle by definition (Figure 4d). This was expected because with increasing ionic/background elemental concentration, the minimum particle size detected by the instrument increases linearly [6]. Hence it is very much possible that smaller Cu-particles may have been present in tap water samples which were binned along with ionic Cu as background concentration ($< 4\sigma$).

To confirm the presence of particles detected in the present study, select-samples were subjected to TEM and EDS. We used a Cu TEM grid and hence we were not able to identify Cu particles in samples. Sn and Pb particle concentrations were low in all samples as discussed above, and hence we couldn't identify those particles via TEM even after multiple attempts. However, Fe-containing particles were successfully detected and confirmed using TEM (Figure 5). Elemental analysis by EDS found Fe in tap water nanoparticles, along with C and O, Cu from the TEM grid, and trace amounts of other elements. The image revealed spherical particles of average diameter of 8 ± 2 nm, which was an order of magnitude lower than what was determined by spICP-MS sizing. This difference could be due to the fact that the spICP-MS was able to detect only particle aggregates and the size detection could have been impacted by the presence of high Fe background concentration (836 ng/L). Fe particle size determined by TEM in this study is comparable to previously reported particle size of corrosion-related Fe particles (3.2 – 5.6 nm) in tap water samples [22].

Study limitations and future work

Several studies list the criteria and requirements to obtain accurate size distributions of particles via spICP-MS [13, 32, 33]. The selection of instrument dwell time and a critical nanoparticle number concentration in samples are important considerations to control precision associated with the counting of particle events and in the generation of size distributions. In this study, we used a 10 ms dwell time and the number of particle events for Pb and Sn were low. By increasing the analysis time to > 3 minutes, we were able to capture sufficient particle events for Pb and Sn. The displayed PSD histograms (Figure 4) are from selected samples that featured the highest number of corresponding particles in samples analyzed. However, the background levels for Fe and Cu particles were relatively high and hence the 10 ms dwell time used in this study might have not been ideal for the detection of smaller particles. This effect can be observed in the PSD histograms, especially for Cu particles. Recent work has shown that extremely short dwell time (as low as 0.1 ms) can significantly improve instrument resolution enabling the detection of particles even at high background concentrations [33]. It was our goal to test the feasibility of using spICP-MS to detect common metal-containing particles in tap water samples and though we achieved successful detection, further modifications such as removal of interfering dissolved elemental forms (e.g. via ion-exchange [34]) may be needed for accurate sizing of nanoparticles in tap water. Additionally, spiking experiments (similar to CuO_2 used in this study) with other model nanoparticles will provide quantitative information about tap water matrix effects and nanoparticle recoveries for various elements.

Conclusions

We successfully demonstrated the feasibility of using spICP-MS for routine monitoring of common metal-containing particles in tap waters. Results show that the particulate form of Sn and Fe represent a significant fraction of the corresponding total bulk elemental mass (>10 %). Pb and Cu particles were also detected in tap water samples, likely due to leaching from pipelines. Method modification (self-aspiration) was necessary for successful detection of Cu-particles in tap water. To the best of our knowledge, this is the first application of the 'self-aspiration' technique for spICP-MS that may be extended to other challenging matrices showing similar interferences with the instrument SIS. Future improvements to further reduce nanoparticle size detection limits, potentially through shorter instrument dwell times and removal of interfering background ionic forms, will make it possible to resolve and monitor even smaller metal-containing particles in drinking waters. All metal concentrations detected were lower than the regulatory limits in drinking water, but the presence of particulate forms in tap waters will require additional research to evaluate possible human exposure risks. It is well known that toxic species such as Pb and As can adsorb on to Fe nanoparticles leading to elevated levels in water at the point of use [19, 22, 35]. Fe-containing particle concentration of up to 890 ng/L in tap water was detected in the present study. Hence, it is particularly important to evaluate the risks of such incidental nanoparticles (e.g. corrosion byproducts) released from distribution pipes, since the concentration of ENMs are typically low in tap waters [36].

Conflicts of interest

There are no conflicts to declare.

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Supporting information. Included in the SI as figures: (i) comparison of metal-containing particles between AM and PM samples; and (ii) instrumental set-up for spICP-MS of tap water.

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Figure Captions

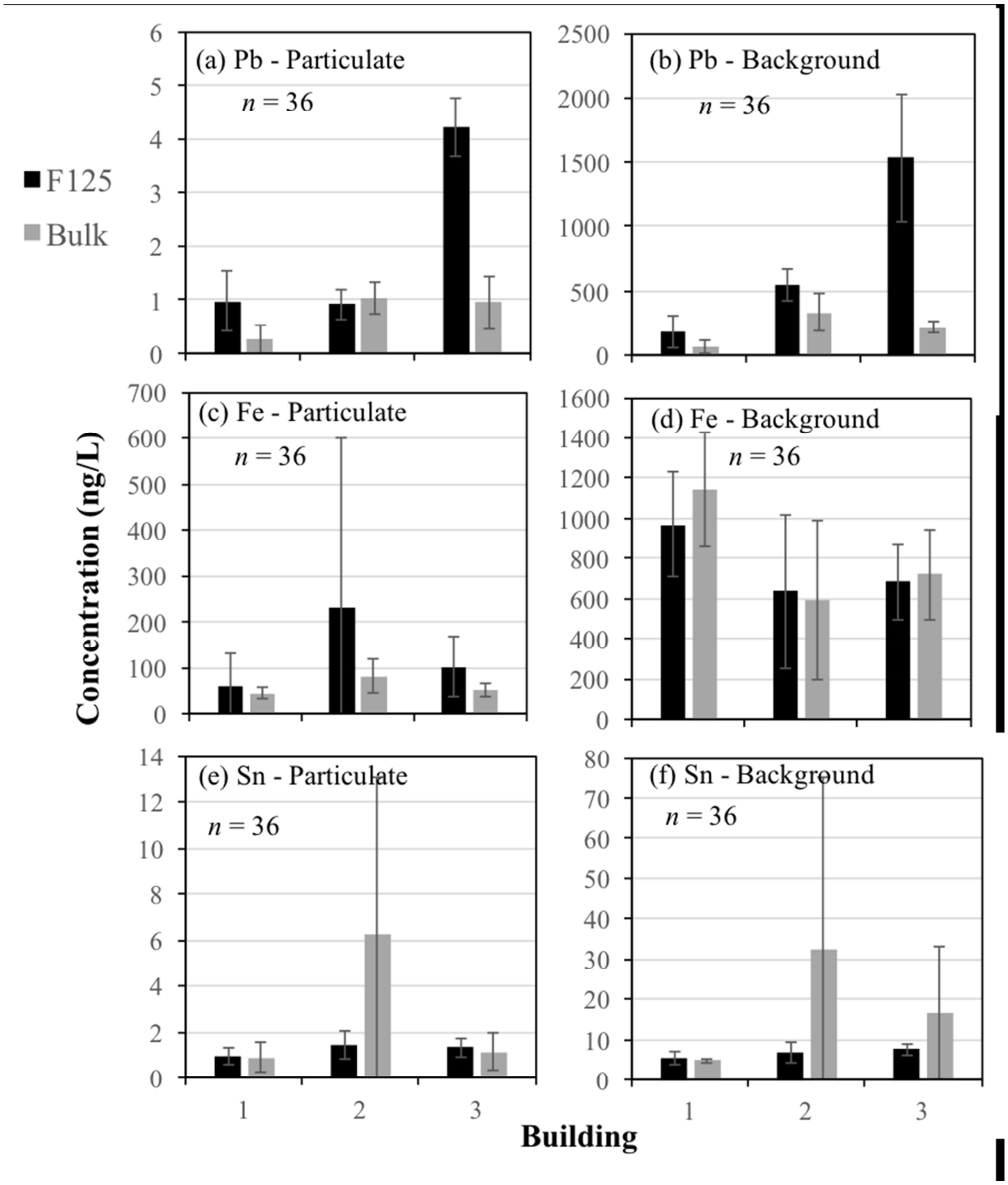
Figure 1. Average particulate (a – Pb; c – Fe; e – Sn) and background (b – Pb; d – Fe; f – Sn) elemental concentrations in the initial 125 mL fractions (black bar) and secondary 500 mL fractions (bulk; grey bar) of tap water collected from three buildings determined by spICP-MS. '*n*' represent the number of tap water samples analyzed. Error bars represent plus/minus one standard deviation.

Figure 2. Minimum fraction of metal-containing particles detected in tap waters. '*n*' represents the number of samples analyzed. Error bars represent plus/minus one standard deviation.

Figure 3. Raw signal spectra of Cu in tap water using (a) normal spICP-MS set-up; (b) normal set-up plus addition of 10 mM EDTA to sample; (c) normal set-up plus addition of 10 mM 18-crown-6 to sample; (d) normal set-up plus addition of 12 mM SDS to sample; and (e) self-aspiration technique.

Figure 4. Size distribution of metal-containing particles detected in tap waters collected from Phoenix, AZ. Shaded region represents particles sized at < 100 nm (i.e. nanoparticles by definition).

Figure 5. Identification by (a) TEM and (b) elemental analysis (EDX) of Fe-containing nanoparticles in tap water. Note: Cu detected is from TEM grid.



Average particulate (a – Pb; c – Fe; e – Sn) and background (b – Pb; d – Fe; f – Sn) elemental concentrations in the initial 125 mL fractions (black bar) and secondary 500 mL fractions (bulk; grey bar) of tap water collected from three buildings determined by spICP-MS. '*n*' represent the number of tap water samples analyzed. Error bars represent plus/minus one standard deviation.

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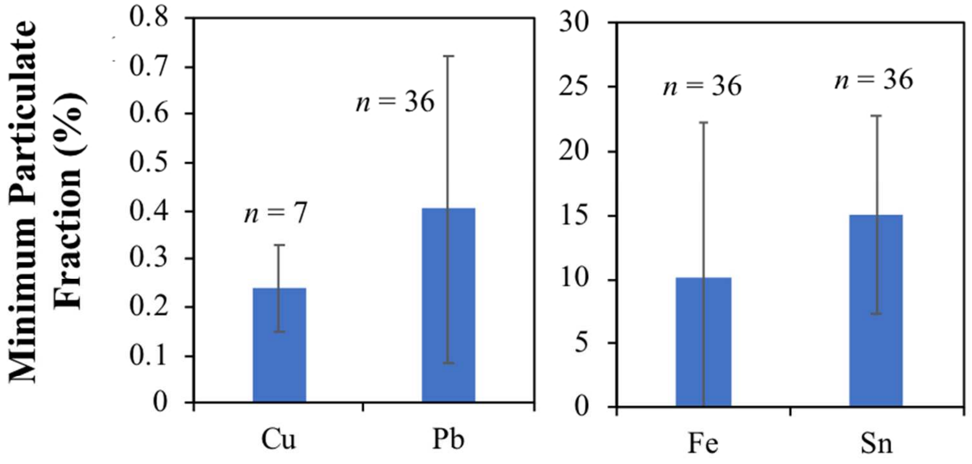

Minimum fraction of metal-containing particles detected in tap waters. '*n*' represents the number of samples analyzed. Error bars represent plus/minus one standard deviation.

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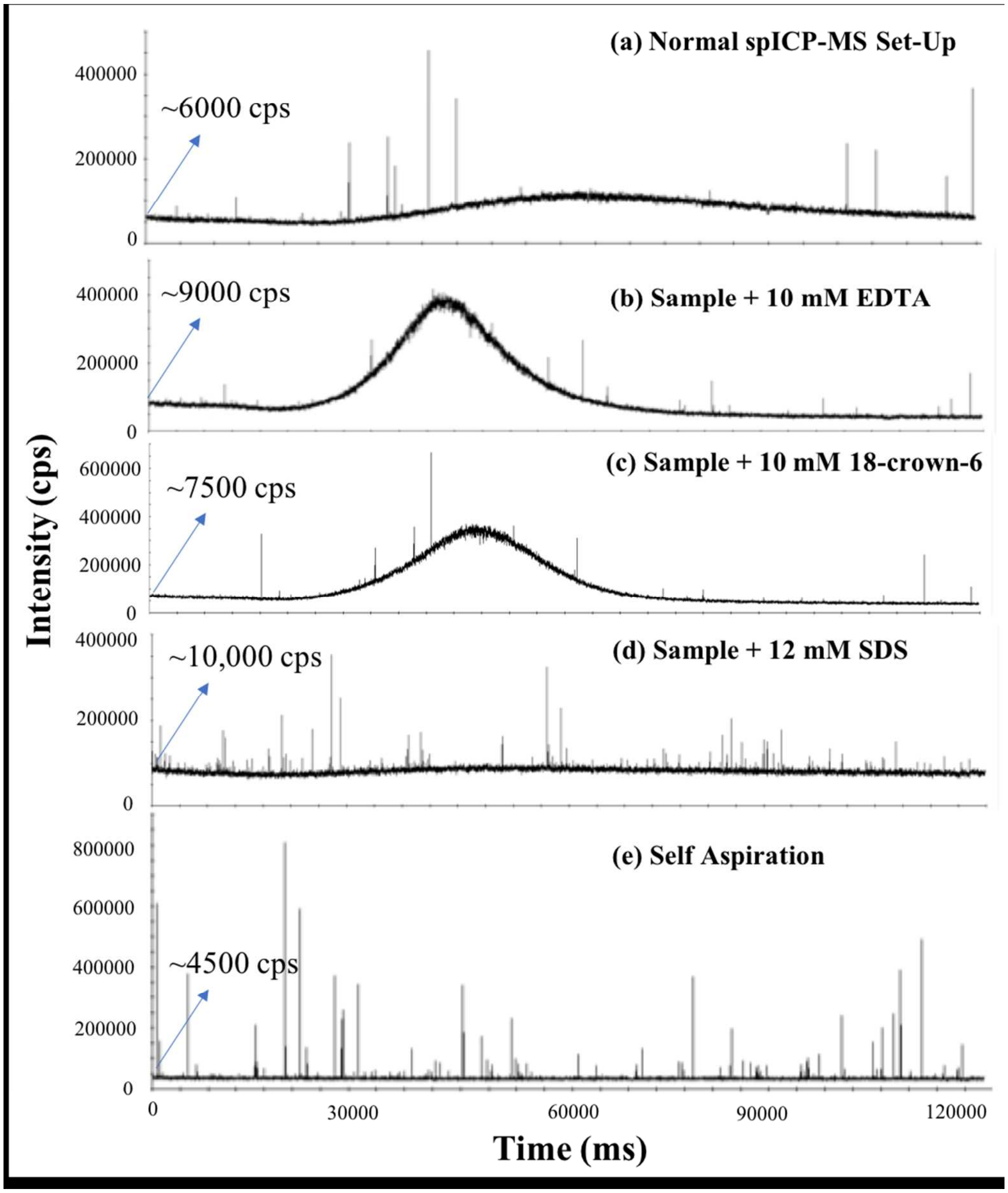

Raw signal spectra of Cu in tap water using (a) normal spICP-MS set-up; (b) normal set-up plus addition of 10 mM EDTA to sample; (c) normal set-up plus addition of 10 mM 18-crown-6 to sample; (d) normal set-up plus addition of 12 mM SDS to sample; and (e) self-aspiration technique.

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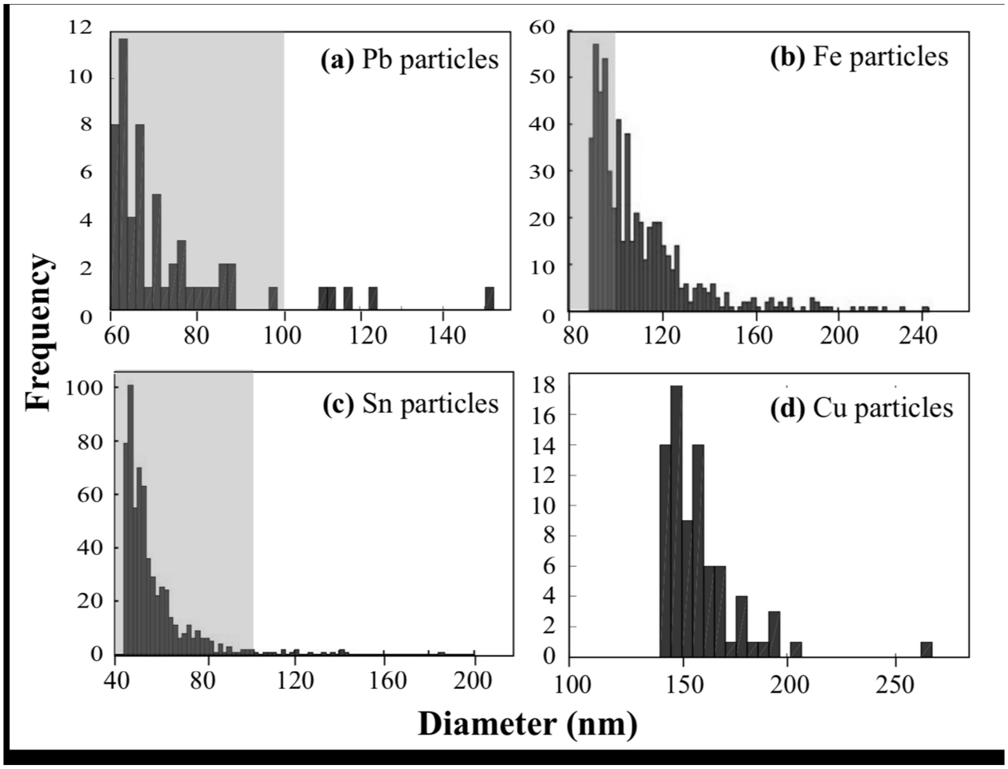
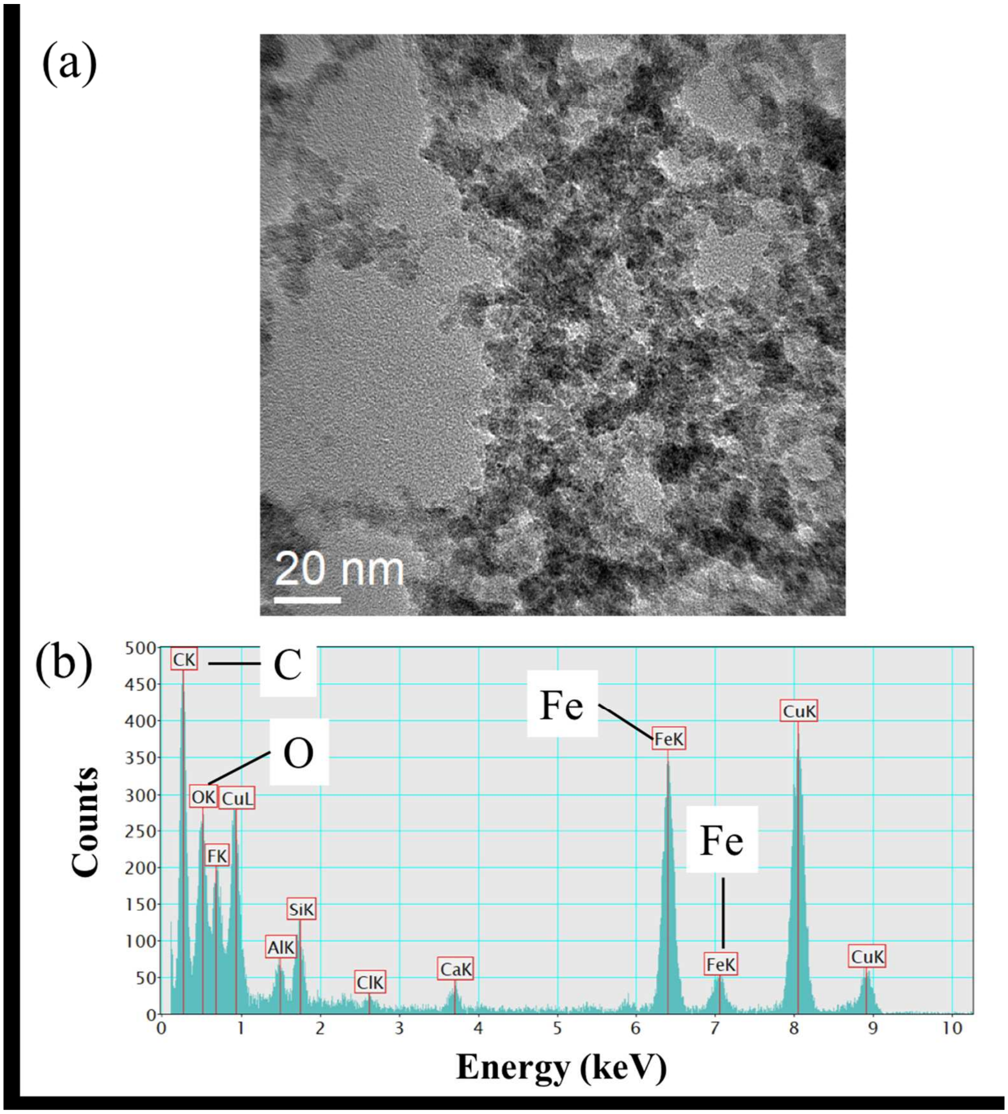


Size distribution of metal-containing particles detected in tap waters collected from Phoenix, AZ. Shaded region represents particles sized at < 100 nm (i.e. nanoparticles by definition).

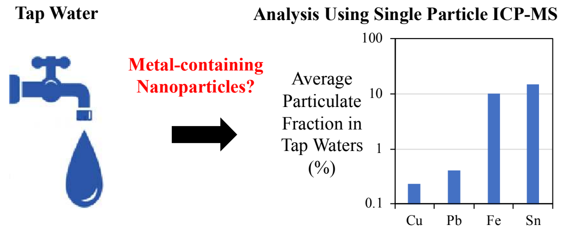
240x183mm (150 x 150 DPI)



Identification by (a) TEM and (b) elemental analysis (EDX) of Fe-containing nanoparticles in tap water. Note: Cu detected is from TEM grid.

191x211mm (150 x 150 DPI)

Table of Content Graphic



This study provides the feasibility of using single particle ICP-MS technique for convenient and routine monitoring of engineered nanomaterials in tap water.