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Human exposure to particles at the air-water interface: Influence of water quality on indoor air quality from use of ultrasonic humidifiers

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ABSTRACT

Ultrasonic humidifiers provide indoor relief to symptoms caused by dry air and produce aerosols containing both water and minerals that are present in the water that fills the humidifier. This study investigated the spatial distributions, concentrations, and metal and mineral composition of aerosols emitted when an ultrasonic humidifier was filled with deionized water (DI), low mineral tap water (LL), high total dissolved solids (TDS)/high hardness water (HH), and high TDS/low hardness water (HL). Aerosol/particle sizes and counts were obtained at six horizontal distances in both the plume and near floor for each water quality. Results are that water quality significantly affects particle size distributions which become uniform after 0.9 m from the humidifier outlet, and are independent of vertical distance from the humidifier. The mean count median diameters were 64 nm for DI, 129 nm for LL, 234 nm for HH, and 260 nm for HL; the particle counts and total mineral solids concentrations were 2,194 #/cm^3 (16 μg/m^3) for DI, 21,070 #/cm^3 (113 μg/m^3) for LL, 38,353 #/cm^3 (438 μg/m^3) for HH, and 43,880 #/cm^3 (521 μg/m^3) for HL. The μg/m^3 values for LL, HH, and HL exceeded $PM_{2.5}$ ambient air standards. Model predictions are that the deposition mass in the human respiratory system from inhaling particles emitted from HH and HL water exceed 135 μg for a 1 to 3-month old child and 600 μg for an adult over an 8-hr period. Mineral water quality significantly affects the distribution and concentration of emitted and inhaled indoor air particles. Consumers may unknowingly be degrading their indoor air quality when using tap water of acceptable drinking water quality as humidifier fill water.

1. Introduction

Human exposure to chemicals released at the air–water interface is an increasing global concern for human health as exemplified by the United States Environmental Protection Agency's conceptual model (USEPA, 2016) that considers inhalation of treated tap water as an important route of human exposure to contaminants. As tap water is mostly used by humans in indoor environments; e.g., cooking, showering, washing, humidification; and people spend about 90% of their time indoors (Amoatey et al., 2018), the indoor air-tap water interface is a potential source of contaminant exposure. Inhalation exposure to volatile organic compounds present in water and transmitted to air is well documented, especially for trihalomethanes (Lin and Hoang, 2000; Wang et al., 2007). An increasing exposure route is that of inhaling inorganic constituents in tap water emitted from portable ultrasonic humidifiers, which are an economic and convenient consumer product used globally by children and adults (Feng et al., 2018). Humidification alleviates low relative humidity levels that can cause different health problems, such as dry eyes, throat, and nose, as well as discomfort from lung injury or asthma. Doctors may recommend patients to use humidifiers if they have a cold or asthma and the relative humidity in their home is low (Mayo Clinic, 2019). Ultrasonic, or non-thermal, humidifiers are considered an essential household product for a new baby (Baby List, 2019). Approximately 17.2 million households in the USA use humidifiers (USEPA, 2012). Likewise, the demand and use of portable humidifiers is high and growing in China (Shen and Feng, 2019). While humidifiers can improve health, adverse health effects are also acknowledged. In Korea, approximately 8 million people were exposed through inhalation to disinfectants specifically marketed for humidifiers; the disinfectants caused lung-injuries and even death in children

Abbreviations: CMD, count median diameter; GSD, geometric standard deviation; MPPD, multiple-pathway particle dosimetry; ICP-MS, inductively coupled plasma with mass spectrometer; SMPS, scanning mobility particle sizer; DI, deionized water; LL, low TDS and low hardness water; HH, high TDS and high hardness water; HL, high TDS and low hardness water; TAS, total airborne solids

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and adults (Kim et al., 2016). A phenomenon known as "white dust" is associated with filling ultrasonic humidifiers with hard water; inhaling this white dust caused childhood lung injury (Daftary and Deterding, 2011). Ultrasonic humidifiers can create health risks due to inhalation of asbestiform-fibers when such fibers are present in tap water used as fill water (Roccaro and Vagliasindi, 2018). Humans can inhale the metals present in tap water, as approximately 85–90% of the metals and minerals present in tap water are emitted to indoor air by ultrasonic humidifiers (Sain and Dietrich, 2015; Sain et al., 2018), as well as 0.22–0.57 μm metal oxide particles (Yao et al., 2019).

Ultrasonic humidifiers use an ultrasonic transducer to create ultrafine water droplets, or aerosols. Thus, the aerosols and breathing air contain the chemicals present in the original water source, creating an intimate link between air quality and water quality. Millions of people around the world fill household humidifiers with water of varying quality. Many ultrasonic humidifier manufacturers recommend filling their 2.5–3 L units with tap water, while others make no recommendations, and a few recommend using distilled water (Table S1 in Supplemental Information). Water quality for both bottled (Marcussen et al., 2013) and tap water (Dietrich and Devesa, 2019) highly vary across the globe, with local geology being the most influential factor for chemical composition and treatment being the second. Conventional drinking water treatment little changes mineral constituents, thus a groundwater or surface water with initial high mineral content or hardness will be of the same water quality when delivered to the tap. While distillation and membrane processes such as reverse osmosis substantially remove minerals, the final tap water requires remineralization for pH and corrosion control (Dietrich and Devesa, 2019). Thus, exploring the relationship between mineral water quality and aerosols/particulates in indoor air is required to assess human exposure to humidified air.

Overall, mineral water quality is determined by the aggregate parameter of total dissolved solids (TDS). TDS is the sum of the mass per volume of all dissolved constituents in water and mostly comprised of cations and anions. TDS $<$ 1500 mg/L comprise freshwaters and the range can be wide, such as between 22 and 1589 mg/L TDS in the 100 largest USA cities (Van der Leeden et al., 1990). TDS in drinking water is not regulated for health reasons. For reasons of aesthetics and taste, the World Health Organization (WHO) and some governments recommend $<$ 1000 mg/L TDS while other global governments recommend TDS $<$ 500 mg/L (Dietrich and Devesa, 2019). Unfortunately, TDS in global freshwater is increasing due to anthropogenic causes and consumers may be unaware of these changes (Dietrich and Burlingame, 2015). Anthropogenic increases to TDS include hydraulic fracturing for gas recovery, urban runoff, chemicals in treatment processes, and piping in homes. Saltwater intrusion and mineral springs also contribute to water with higher TDS (WHO, 2011). In temperate zones, TDS increases in winter months when deicing road salts are used to remove ice and snow from roadways, presenting a correlation with winter time when increased humidifier usage occurs (Highsmith et al., 1988; Kaushal et al., 2005; Dietrich and Burlingame, 2015).

While TDS measures overall minerals, specific cations and anions vary across water sources with Ca^{2+}, Mg^{2+}, and Na^{+} being the most abundant metal cations in water and bicarbonate, sulfate, and chloride being the most abundant anions in water. Hardness measures aqueous divalent cations; calcium and magnesium are the two that occur in the highest concentrations in water and vary globally in association with limestone and dolomitic geologies (Dietrich and Burlingame, 2015; Dietrich and Devesa, 2019). Water source also influences the presence of calcium and magnesium with ground waters having more metals than surface waters. A North American study (Azoulay et al., 2001) reported median concentrations in different types of water: surface waters (e.g., lakes and rivers), 36 mg/L Ca^{2+} and 8 mg/L Mg^{2+}; ground waters, 48 mg/L Ca^{2+} and 12 mg/L Mg^{2+}; bottled waters, 6 mg/L Ca^{2+} and 3 mg/L Mg^{2+}. The ranges of these two cations were also quite large, with means and standard deviations of: surface waters (e.g. lakes and rivers), 34 $\pm$ 21 mg/L Ca^{2+} and 10 $\pm$ 8 mg/L Mg^{2+}; ground waters, 52 $\pm$ 24 mg/L Ca^{2+} and 20 $\pm$ 13 mg/L Mg^{2+}; bottled waters, 18 $\pm$ 22 mg/L Ca^{2+} and 8 $\pm$ 18 mg/L Mg^{2+}. Thus, like TDS, hardness can be highly variable in tap waters and this variability may contribute the phenomenon of "white dust".

Aerosols produced from ultrasonic humidifiers contain small particulate contaminants from the fill water, therefore aerosols can be used synonymously with particulate matter. The size of particulates is important to regulatory entities as size is the most relevant factor for inhalation health threats. Particulate size is the determining factor in the motion and deposition of aerosols, including in the human respiratory tract. For particles under 10 μm, inertial and gravitational forces contribute most to the deposition of particles (Dong, 2012). A previous study showed that resuspended particles from indoor air surfaces contributed to increased indoor air particle concentrations (Lai, 2002), and thus humans could inhale the resuspended particles produced from ultrasonic humidifiers.

This study focused on characterizing aerosols under 10 μm, specifically $<$ 0.25 to 2.5 μm as those are the particles that most penetrate into the gas-exchange portion of the lungs and can cause irritation and injury to the lungs depending on exposure and inhalation rates. As particle diameter decreases, surface area ratio increases and these particles are able to pass through the head and tracheobronchial region into the gas exchange (peripheral), or alveolar portion of the lungs. Inhalable coarse aerosol particles (between 2.5 and 10 μm), or aerosol particles with rapid velocities, are affected mainly by inertial forces and they impact airway walls as they are unable to follow in line with the airstream of inhalation. However, fine and ultrafine particles, aerodynamic diameter smaller than 2.5 μm and denoted as $PM_{2.5}$, especially at resting breathing rates, are able to travel with the airway and deposit or settle in the tracheobronchial portion of the lungs, and even smaller particles in the gas-exchange portion of the lungs (Newman, 1985).

In steady breathing, particles between 6 and 10 μm have a 52–60% chance of depositing in the oropharynx, a 21–28% chance of deposition in the tracheobronchial zone, and a 5–17% change of settling in the gas exchange region. Particles between 4 and 6 μm have 20–52%, 12–21%, and 17–42% chance of settling into the relative zones, and particles from 2 to 4 μm have a 0–20%, 2–12% and 40–50% chance of settling in each respective zone (Newman, 1985). Settling in the tracheobronchial zone or the alveoli of the gas exchange region can exacerbate and is linked to certain injuries such as asthma, respiratory inflammation, some cancers, and the jeopardization of lung function (Xing et al., 2016). Particulate inhalation more negatively affects the elderly, children, and those with respiratory injury. A child's respiratory health was shown to be compromised due to inhalation of the white dust produced by minerals present in an ultrasonic humidifier (Daftary and Deterding, 2011), and this "white dust" has been shown to expose lung tissues of mice to the contaminants of the water (Umezawa et al., 2013). Localized air quality is another important factor for exposure. Epidemiological evidence suggests that children living in a mining region have a higher relative risk of respiratory disease from exposure to metals in dust than children living nearby in non-impacted regions (Zychowski et al., 2019).

At present, there is great interest for assessing children's exposure to air pollution as they are a more vulnerable population (Mazaheri et al., 2016; Sloan et al. 2017). Both outdoor and indoor air pollution are a concern, and one study showed that even when outdoor $PM_{2.5}$ infiltration was lowered, indoor $PM_{2.5}$ remained high as indoor sources increased to maintain $PM_{2.5}$ exposure (Wichmann et al., 2010; Sloan et al., 2017). Residential indoor air quality is not currently regulated in many countries, however, there are regulatory limits for PM in workplace and ambient air. The USA National Ambient Air Quality Standard (NAAQS) for PM_{10} is 150 μg/m^3 averaged over 24-hr period for both the primary and secondary standard; the primary NAAQS for $PM_{2.5}$ is 35 μg/m^3 over a 24-hr period, and 12 μg/m^3 over 1-year period, while the secondary NAAQS for $PM_{2.5}$ is 35 μg/m^3 over 24-hr period, and 15

μg/m^3 over 1-year period (USEPA, 2015). The WHO (2006) has defined a guideline value for PM_{10} at 50 μg/m^3 averaged over 24-hr period, and 20 μg/m^3 averaged over 1-year period, and 25 and 10 μg/m^3 over a 24-hr and 1-year period for $PM_{2.5}$.

Humidifiers have been shown to release inhalable aerosols that carry contaminants from the original fill water placed in the humidifier (Highsmith et al., 1988; Sain and Dietrich, 2015; Sain et al., 2018; Yao et al., 2019). 85–90% of aerosols emitted from an ultrasonic humidifier are in the respirable range (Sain and Dietrich, 2015; Sain et al., 2018). Water quality has also been shown to affect particle size distribution as well as particle concentrations (Sain et al., 2018).

This project innovatively identifies and quantifies exposure to metals and TDS transferred from tap water to inhalable particles by ultrasonic humidifiers. Specific objectives are to evaluate: (1) how inorganic water quality, as measured by total dissolved solids and specific elemental content, affects the size and concentration distribution of the particles emitted to indoor air; (2) how the size and concentration of particles change over horizontal and vertical distances from the outlet of the humidifier; (3) inorganic chemical masses of particles emitted and influence of different TDS water qualities; (4) adult and child exposure from inhalation of indoor air particles emitted from ultrasonic humidifier.

2. Materials and methods

2.1. Ultrasonic humidifier and water quality

For this experiment we used a generic, popular, store brand ultrasonic humidifier. This humidifier was chosen as it most accurately mimics the type of humidifier that the average home would purchase and use. Four water matrices were used in this experiment. Nanopure™ water, which is membrane-filtered, deionized, and carbon filtered, was used as a control and the other three tap water qualities were (1) low TDS and low hardness (LL) water; (2) high TDS and high hardness (HH) water; and (3) high TDS and low hardness water (HL). The LL water was unfiltered tap water from Blacksburg, Virginia. HH and HL water were prepared by adding salts to the LL water. To 3 L of unfiltered Blacksburg tap water, 1.152 g of $NaHCO_3$, 0.616 g of $CaSO_4{\cdot}2H_2O$, 0.646 g of $MgSO_4{\cdot}7H_2O$, and 0.048 g of KCl were added to prepare the HH water, and 1.152 g of $NaHCO_3$, 0.9282 g of Na_2SO_4, 0.1266 g of K_2SO_4, and 0.048 g of KCl were added to prepare the HL water. Water samples were drawn or created on the morning of experimentation in order to prevent oxidation and precipitation. Alkalinity was tested for LL, HL and HH water using titration Standard Method 2320B (Water Environmental Federation and American Public Health Association, 2017). TDS of DI, LL, HH, and HL water was calculated from summing bicarbonate and elements measured by inductively coupled plasma-mass spectrometer. Table 1 shows water quality data of the LL, HH, and HL water.

2.2. Experimental methods

2.2.1. Instrumentation

Expelled aerosols were determined using two particle counters. An AeroTrak Handheld Particle Counter (AeroTrak 9306, TSI) was used for particles measuring 0.3–10 μm. The AeroTrak counts particles in the air according to 6 set size ranges, 0.3, 0.5, 1.0, 2.5, 5, and 10 μm; in our research it was used to classify particles between 1 and 10 μm. A scanning mobility particle sizer (SMPS 3936, TSI) measured particles 0.014–0.750 μm. Normalized concentrations (dN/dlogDp) were used in reporting and analyses as the instrument provides normalized concentration values that are independent of bin width, allowing values at the mode to be similar across instruments with different resolution (TSI Aerosol Statistics).

A cascade impactor classified particles present in a sample of air using 5 known size ranges: < 0.25 μm, 0.25–0.5 μm, 0.5–1 μm, 1–2.5 μm, and > 2.5 μm. As previously described, collected particles on filters in each size range were dissolved in dilute nitric acid and then quantified by Thermo Electron X Series inductively coupled plasma with mass spectrometer (ICP-MS) to metals and elements (Yao et al., 2019). Minimum reporting levels for constituents in ng/L are as follows: Na, Mg, P, Fe: 5.00; Ti, V, Cr, Mn, Co, Ni, As, Y, Ag, Cd, Sn: 0.10; Sc, Cu, Zn, Se: 1.00; Al, K, Br: 10, Si, Ca: 500; S: 5000, Cl: 1000. The term “total elements” is the sum of all individual metal and element mass concentrations from the ICP-MS measurements. As the ICP-MS cannot measure carbon, the bicarbonate in emitted particles was calculated from multiplying the ratio of sodium to bicarbonate in the fill water by sodium in emitted particles, for each water quality. The summation of bicarbonate and ICP-MS total elements in emitted particles is considered total airborne solids (TAS). TAS is analogous to TDS; since the aerosols lose water to become solid airborne particles, ‘dissolved’ does not apply. Indicator litmus paper (8.5 × 11 in. Goldenrod Paper SM-925) that would change color when in contact with water was used to qualitatively determine moisture deposition on the floor.

2.2.2. Test location

The room used for this experiment was a medium sized conference room of 5.6 × 4.3 × 3 m (L × W × H), and 72.2 m^3. The air exchange rate (AER) of the experimental room was 1.5 per hour when windows and door were closed. Before each experiment, the windows and door were kept open overnight, and the AER of 1.5 per hour ensures the

Table 1
Water quality of deionized water (DI), Low Hardness/TDS water (LL), High Hardness/High TDS water (HH), and Low Hardness/High TDS water (HL). Units in mg/L except for pH.

	Concentration, mean ± standard deviation, mg/L			
Parameter	DI	LL	HH	HL
Total Dissolved Solids	2.0 ± 0.67	95.8 ± 5.67	697 ± 34.2	772 ± 27.6
Alkalinity (mg/L as $CaCO_3$)	nd[a]	35 ± 2.03	256 ± 4.14	253 ± 4.01
Hardness (mg/L as $CaCO_3$)[b]	0.33 ± 0.18	44.2 ± 3.13	211 ± 5.07	38.3 ± 1.15
HCO_3^{-}[c]	nd[a]	36.5 ± 5.45	327 ± 15.85	330 ± 19.88
Na	0.073 ± 0.046	8.9 ± 0.28	108 ± 12	192 ± 6.0
Mg	0.016 ± 0.004	4.5 ± 0.35	21.7 ± 15.8	3.8 ± 0.11
K	0.090 ± 0.056	1.4 ± 0.06	8.9 ± 0.87	26.5 ± 1.66
Ca	0.105 ± 0.068	10.3 ± 0.68	48.9 ± 2.68	9.1 ± 0.28
SO_4^{2-}	1.2 ± 1.4	7.3 ± 0.64	153 ± 8.7	181 ± 1.78
Cl^-	nd[a]	13.5 ± 2.1	16.5 ± 1.35	16.7 ± 0.35
pH	7.5 ± 0.058	7.1 ± 0.15	7.7 ± 0.058	7.8 ± 0.058

[c]calculated from electoneutrality for LL tap water; for HH and HL, calculated from HCO_3^- added to LL water.
[a] nd: not detected
[b] calculated from calcium and magnesium concentrations

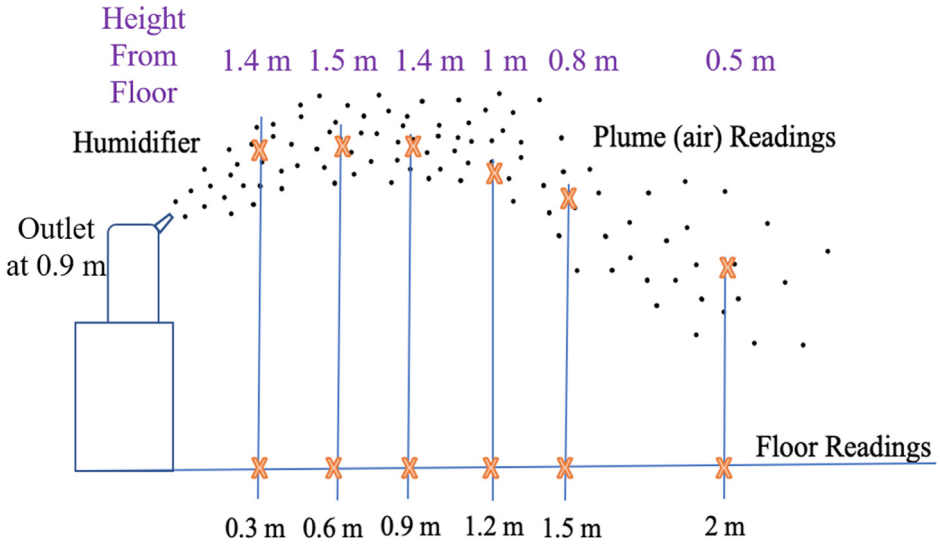

Fig. 1. Experimental room design. "X" represent the different sampling locations. 0.3 m, 0.6 m, 0.9 m, 1.2 m, 1.5 m, 2 m is distance away from the humidifier outlet. Figure is not to scale.

Table 2
Count median diameter, geometric standard deviation, and particle concentrations per water quality at 1.5 m in plume.

Water Quality	0.014–0.75 μm Particles, Scanning Mobility Particle Sizer (SMPS)		0.3–10 μm Particles, AeroTrak		Combined (SMPS + AeroTrak)	
	CMD (GSD) nm	Concentration #/cm^3	CMD (GSD) nm	Concentration #/cm^3	CMD (GSD) nm	Concentration #/cm^3
DI	64 (1.70)	2,153	902 (2.53)	41	64 (1.70)	2,194
LL	129 (1.74)	20,619	497 (1.95)	451	129 (1.75)	21,070
HH	175 (1.91)	38,110	460 (1.89)	243	234 (1.88)	38,353
HL	192 (1.91)	39,891	343 (1.69)	3,989	260 (1.91)	43,880

CMD: count median diameter; GSD: geometric standard deviation

indoor air and outdoor air were fully exchanged. Therefore, the indoor air is expected to be clean background before the beginning of each experiment. The windows facing outside had blinds drawn to block sunlight and assist in maintaining a constant temperature. The humidifier was positioned in the corner of the room facing to the center at 0.9 m in height (Fig. 1). A cascade impactor was positioned at the beginning of the experiment and set at 1.5 m away from the humidifier in the direct line of the plume. Goldenrod indicator litmus paper was placed on the floor, starting below humidifier, at the beginning of the experiment to track deposited wetness on the floor. The humidifier was operated full for two hours to establish steady-state conditions (Sain et al., 2018; Yao et al., 2019).

After operating the humidifier for two hours, sampling using SMPS and AeroTrak began. Samples were taken at twelve locations: six horizontal distances, 0.3 m, 0.6 m, 0.9 m, 1.2 m, 1.5 m, and 2 m from the diffuser outlet of the humidifier, and each distance had a location in the direct plume of the emissions as well as one meter below the plume, at ~ 2 cm above the floor. The SMPS scanned an air sample for 0.014–0.750 μm size distribution profile every 6 min, and the AeroTrak measured counts of particles 0.3–10 μm every 6 min. Upon finishing the 6-minute air sample measurement, the SMPS and AeroTrak were moved manually to the next sampling location. After sampling at all six horizontal locations in the plume and above the floor, the entire sampling process was repeated two additional times resulting in three replicate measurements at each horizontal and vertical location on each day. The experiment was repeated on three different days for each of four water qualities, resulting in nine replicates being taken at each horizontal and vertical location per water quality. The total SMPS and AeroTrak sampling time for each daily experiment was 3.6 h and the mean values of the particle concentrations at each sample location represent reported data.

2.3. MPPD model

Multiple-path particle dosimetry (MPPD) model (v3.04, ARA) was used to calculate particle deposition within the human respiratory tract. The MPPD model comprises eight airway morphologies for humans, and two of them include exposure analysis for age-specific groups. Yeh/Schum 5-lobe (Yeh and Schum, 1980) and age-specific 5-lobe (Phalen et al., 1985; Mortensen, 1988) airway morphology models were selected in this study for adult exposure and 1 to 3-month child exposure, respectively. Default settings for the airway models were used. A standard "adult" weighs 70 kg and breathes 20 m^3/day (U.S. Environmental Protection Agency, 2011), and a 1 to 3-month "child" weighs 6 kg and breathes 3.5 m^3/day (USEPA, 2008). Aerosols inhaled were assumed spherical and of 1.7 g/cm^3 density, which is representative of aerosolized inorganic mineral salts (Sarangi et al., 2016). Other measured properties (i.e. aerosol median diameter, standard deviation of aerosol size distribution, and mass concentration) were manually entered for the four water qualities. Deposited mass was generated assuming 8-hr exposure period.

2.4. Statistical analyses

Count median diameter (CMD) and geometric standard deviation (GSD), used to describe the central tendency and spread of a particle size distribution, were calculated using equations in Supporting Information. SMPS and AeroTrak data were combined and the CMD and GSD were calculated for each triplicate experiment at each location. 3-way ANOVA, with variables of water quality, horizontal distance, and vertical distance, followed by Tukey's honest significance difference (HSD) test, were used to determine statistically similar distributions among all sampling locations. Linear regression of mineral content in

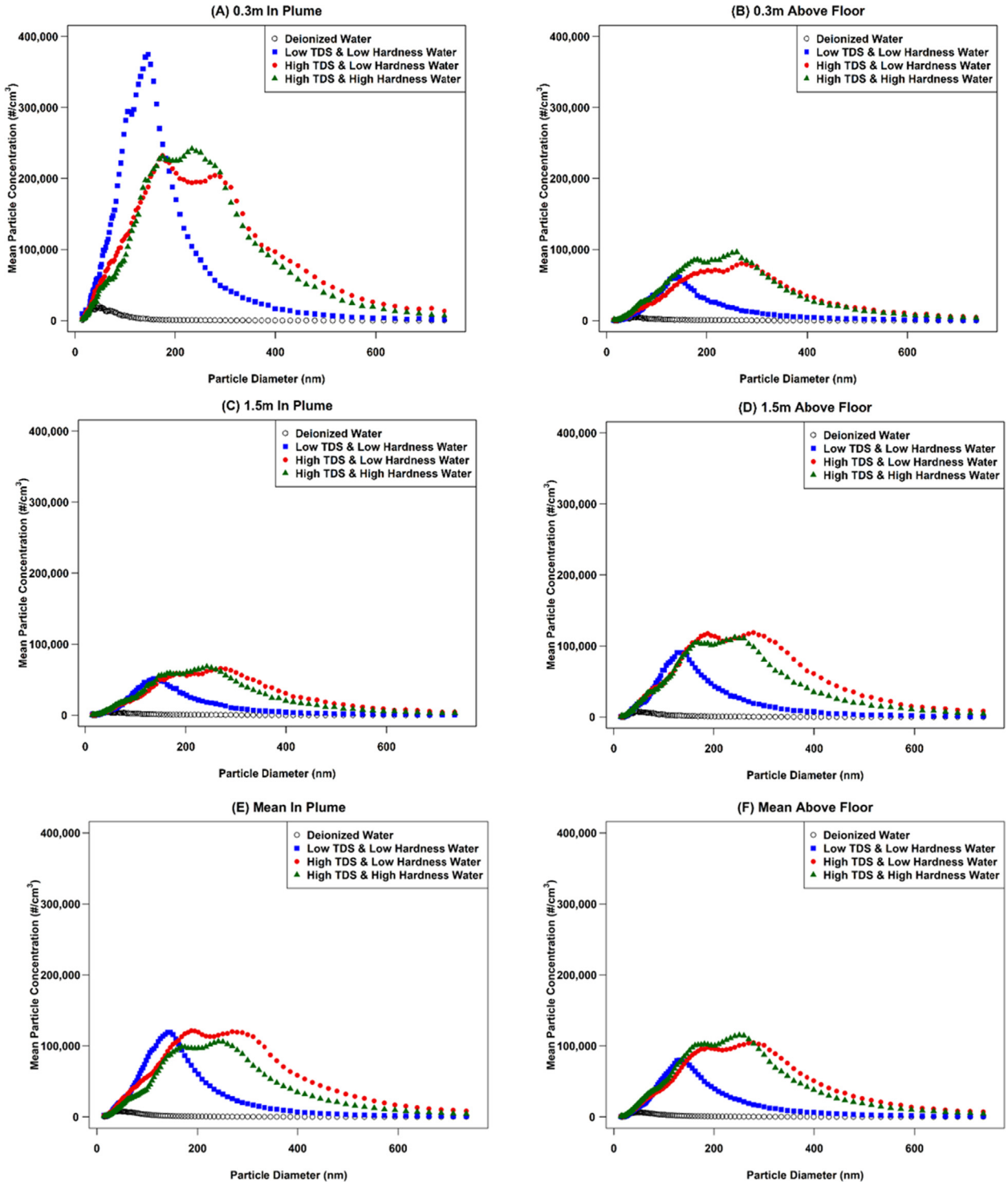

Fig. 2. Particle size distribution for each water quality at 0.3 m in plume and above floor, 1.5 m in plume and above floor, and mean values across all sampling locations in plume and above floor.

fill water and mineral content in emitted particles was performed. Alpha value for all statistical analysis was 0.05 and all analysis was performed in R version 3.6.2 (R R Studio, 2019).

3. Results

3.1. Inorganic water quality affects the size and concentration distribution of particles emitted to indoor air

Ultrasonic humidifier consumption rates for water were similar despite changes in water quality. Approximately 300 mL of water was consumed by the humidifier per hour, and the average water consumption for the 8-hour experiment was 2.4 L. Table 2 and Fig. 2 show

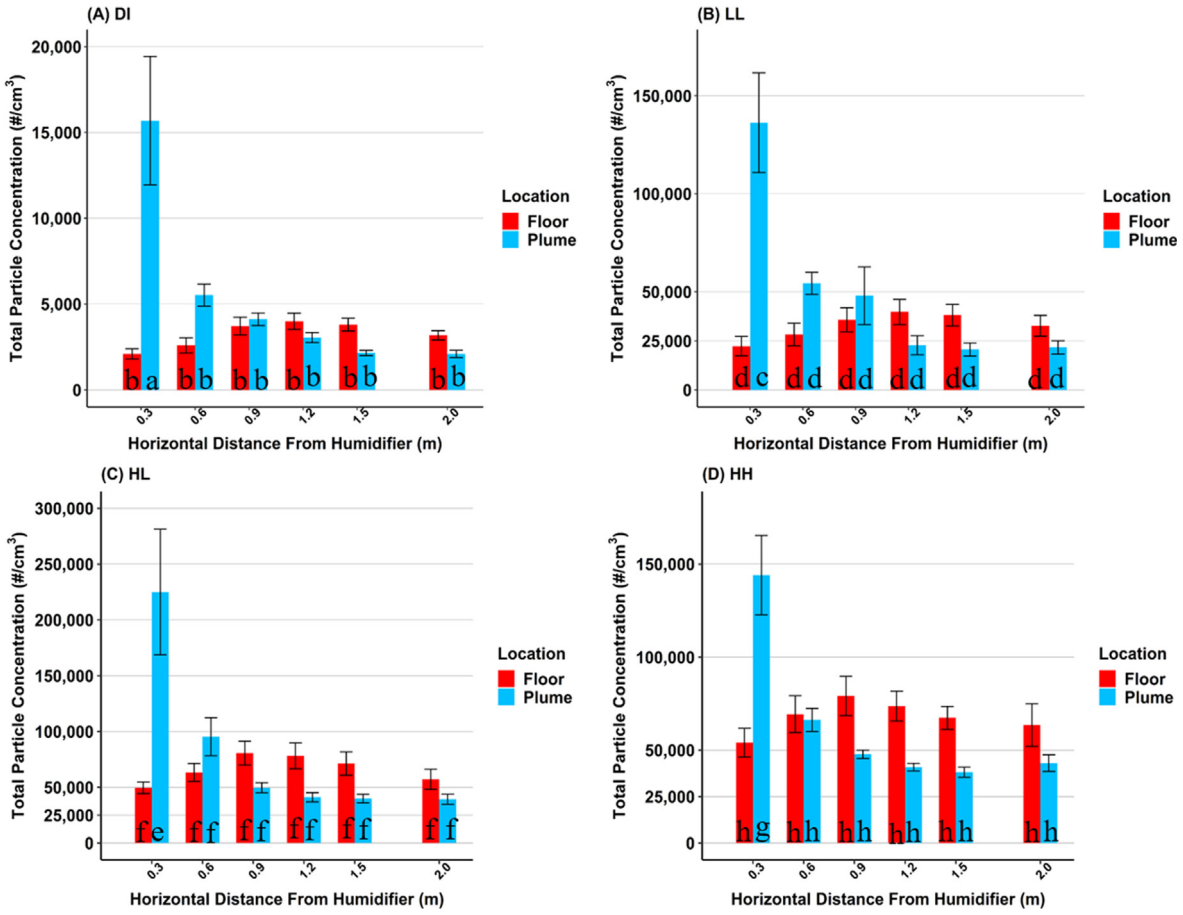

Fig. 3. Total particle concentrations per water quality and sampling locations. A: deionized water (DI). B: low hardness and low TDS water (LL). C: low hardness and high TDS water (HL). D: high hardness and high TDS water (HH). Different letters (a-h) on the bars indicate statistically different groups (alpha = 0.05). Error bars are standard errors. Note: y-axes are different scales for each water quality.

that water quality had significant effects on particle concentration and size distribution, with deionized water having the smallest concentrations and diameters. LL tap water had a slightly higher median diameter, while HH water and HL water had distributions with a wider spread and a higher count median diameter. Concentration readings at 0.3 m in the plume were significantly higher for LL and HH water. Figures S1-S4 in Supporting Information show the concentration to diameter distribution curves for each water quality on the same scale to visually describe effects of water quality and horizontal and vertical distances.

The larger diameter particles measured by the AeroTrak were at much lower concentration than the small diameter particles measured by SMPS. Therefore, similar count median diameters were calculated from SMPS and SMPS + AeroTrak combined, as seen in Table 2. The mean count median diameters were 64 nm for DI water, 129 nm for LL water, 234 nm for HH water, and 260 nm for HL water, which indicates that particle size increases with increased TDS. Distribution shapes varied, though all distributions were polydisperse with GSD > 1.25.

As TDS concentration increased, so did particle size and number concentration, as shown in Fig. 2. HH and HL waters had the highest concentrations and median diameters. Water qualities with higher TDS produced curves that had higher concentrations, were shifted right to wider diameters, and were more dispersed and bimodal. DI and LL waters displayed shorter, narrower curves with one predominant mode as shown in Fig. 2. HH and HL water displayed taller, wider curves with visible bimodality and the distributions were slightly skewed. Similar particle diameters and distributions were observed in the plume and above the floor.

3.2. Size and concentration of particles with respect to horizontal and vertical distances from humidifier outlet

Fig. 3 includes the total particle concentrations for all sampling locations within each water type. The particle concentrations for the four water qualities were statistically different from each other. Water with higher TDS emitted particles at higher concentrations than waters with lower TDS. In addition, in-plume samples were similar to above-floor samples beyond 0.9 m away from humidifier. Particle concentrations were highest at 0.3 m in the plume for DI, LL, and HH water, and highest at 0.6 m in the plume for HL water. Aside from 0.3 m and 0.6 m in plume, the rest of the distances and plume/above-floor locations followed a similar pattern despite water quality, with corresponding above floor readings below the plume being slightly higher than the readings in the plume, as shown in Fig. 3. While the data in Fig. 3 appear to suggest that the particle counts are higher above the floor than in the plume after 0.9 m, based on the results of 2-way ANOVA and Tukey's HSD test, total particle counts were highest at 0.3 m in plume and statistically the same in the other horizontal and vertical locations for the four water qualities. Fig. 2 and Fig. 3 reveal that emitted particles were higher in quantities and larger in size when closer to the humidifier outlet.

Fig. 4 shows the count median diameters at each location per water quality. Water with higher TDS produced larger particles, however, horizontal location did not have a significant effect on particle diameter. For low-mineral water (i.e., DI & LL), in-plume and above-floor samples had similar CMD, while the CMDs were statistically much higher in-plume than above floor for high-mineral water (i.e., HL & HH). A possible explanation is that the wetness in LL-water produced

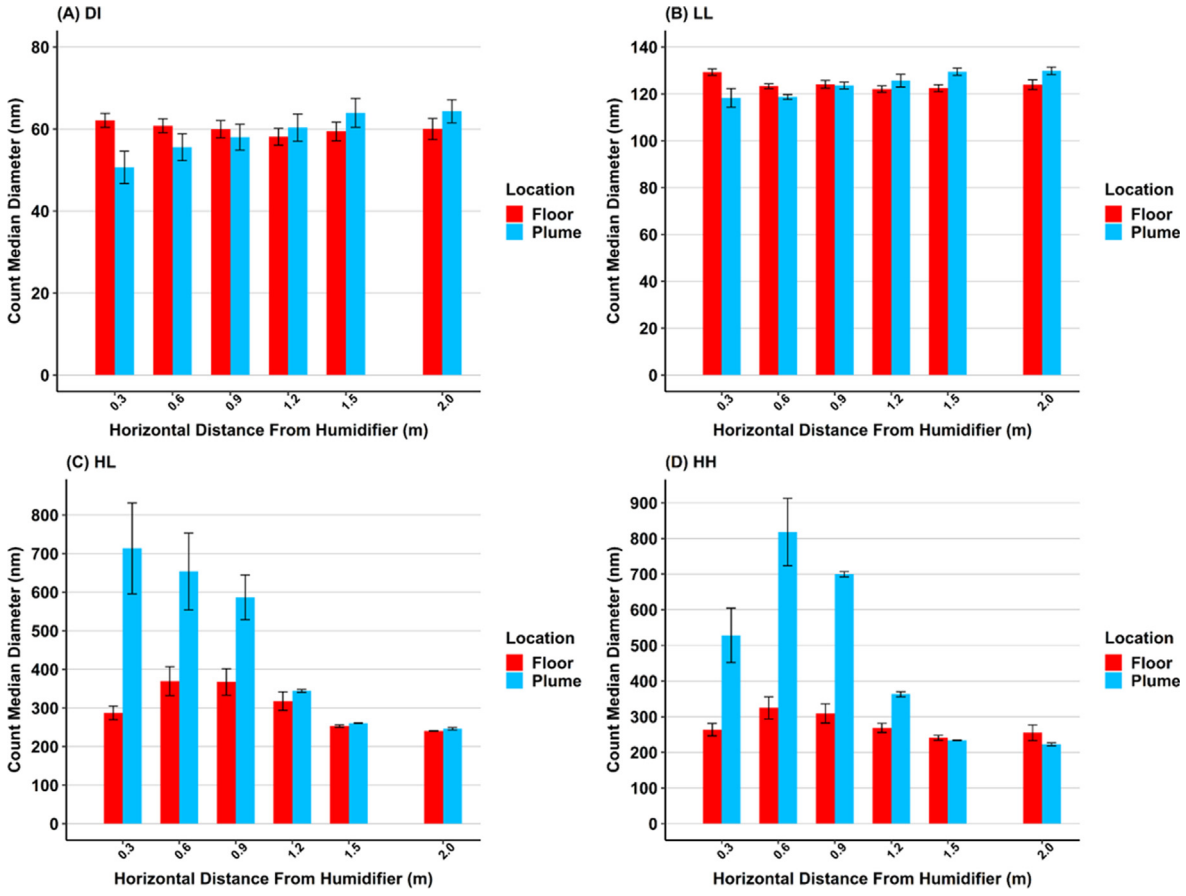

Fig. 4. Count median diameters per water quality and sampling locations. A: deionized water (DI). B: low hardness and low TDS water (LL). C: low hardness and high TDS water (HL). D: high hardness and high TDS water (HH). No statistical difference in CMDs in plume and above floor for DI or LL water (alpha = 0.05). Error bars are standard errors. Note: y-axes are different scales for each water quality.

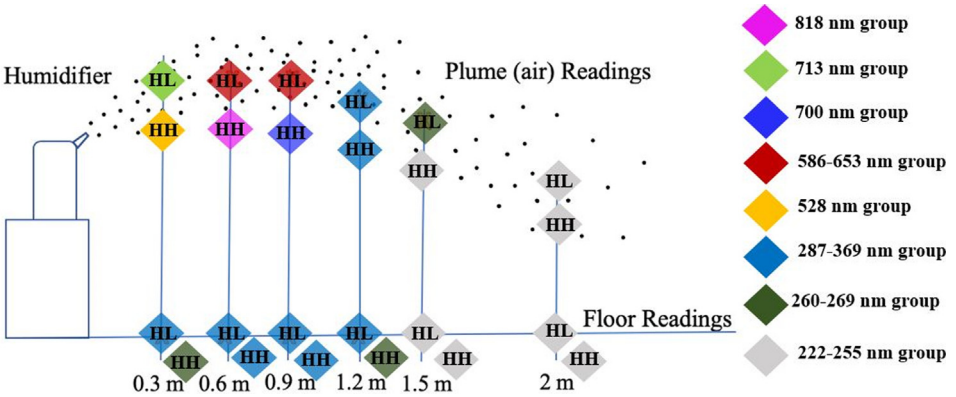

Fig. 5. Count median diameters (CMD) differentiated in significant groups for HL (high TDS, low hardness) and HH (high TDS, high hardness) water. Data generated from scanning mobility particles sizer and AeroTrak were combined. CMDs of LL (low TDS, low hardness) water is statistically identical across all locations with range of 118–130 nm. CMDs of DI water is statistically identical across all locations with range of 50–64 nm. No statistical difference in CMDs in plume and above floor for DI or LL water (alpha = 0.05).

particles evaporated quickly due to larger surface-to-volume ratio. The larger particles with smaller surface-to-volume ratio produced from HL and HH waters evaporate relatively slower, possibly also due to the anions and cations retaining waters of hydration. Thus, larger particles contain more moisture in the plume while smaller evaporated particles are present above floor. In Figs. 3 and 4, higher variability, indicated by larger standard errors, exists at the locations closer to humidifier outlet (0.3 m and 0.6 m) because the total particle concentrations and the CMDs are larger in the two locations. At locations closer to humidifier outlet, evaporation may cause the existence of aerosolized particles in a wider size range; larger particles are "wet", and smaller particles are relatively dry.

Due to multiple relationships being examined across many variables, 3-way ANOVAs followed by Tukey's HSD test were used to assess the relationships between water quality, in-plume or above-floor location, and horizontal distance from the humidifier (see Table S2 in Supplemental Information for grouping of all water qualities). Many interactions of water type, location, and distance from humidifier affect the CMDs comparisons. Water quality is always a significant factor influencing the HSD comparison results. Groups with different colors are significantly different from one another, as shown in Fig. 5. Fig. 5 shows that water quality has significant effects on emitted particles' CMD, and particles were "well-mixed" beyond 0.9 m away from humidifier. Each water quality produced size distributions statistically different from one another when comparing water qualities and distances.

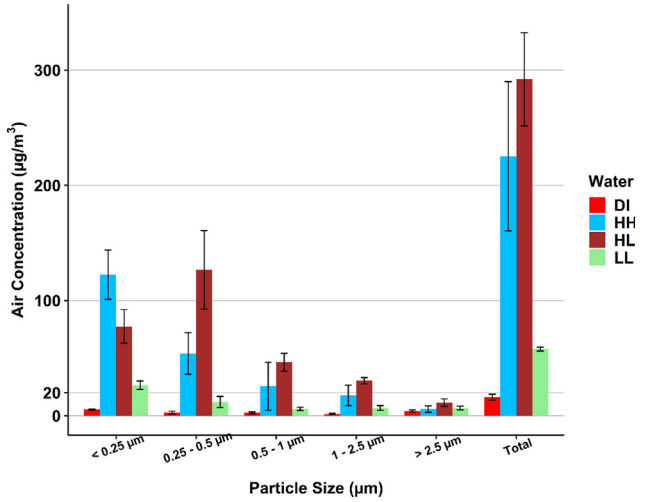

Fig. 6. Particle mass concentration in air (based on total airborne solids) as a function of particle diameter and water quality. DI- deionized water; LL- low TDS and low hardness water; HH– high TDS and high hardness water; HL- high TDS and low hardness and water. Error bars indicate standard errors.

Wetness was not observed on the indicator paper located on the floor. Stoke's Law was utilized to determine settling probability on the floor (all CMDs calculated and presented in Table S2 in Supporting Information) under the conditions of this experiment with a 5.6 × 4.3 × 3 m (L × W × H; volume = 72.2 m^3) room and air exchange rate of 1.5 hr^{-1}. Settling times for smaller particles characterized by the SMPS were > 290 h for all locations and water qualities and many were > 1100 h (see Table S3 in Supplemental Information). Settling times were shorter for the larger particles measured by the Aerotrak, ranging from 0.7 to 42 h, with the shortest time exhibited by DI water. Thus, most particles would remain suspended in the air and few settle on the floor under the conditions of our experiments. The air vent is positioned at the ceiling of the experimental room, which may allow floor deposition from its occasional, automatically on and off ventilation. In addition, larger particles closer to the humidifier have shorter settling times, which may contribute to floor deposition. The results indicated that most emitted particles from ultrasonic humidifiers produced by low-TDS and high-TDS waters were cleared by air exchange before they gravity settled on the floor. The above floor samples have similar size distribution and total particles as in plume samples (Figs. 2 & 3), indicating that emitted particles are present at ~ 2 cm above floor.

3.3. Inorganic chemical mass of emitted particles and influence of different TDS water qualities

As expected, the greatest concentration of metals and minerals on cascade impactor filters were for water qualities with higher TDS, with HL water showing the largest concentration of particles sized between 0.25 and 2.5 μm. Fig. 6 reveals the individual concentrations on each filter size bin per water quality. Particle mass concentrations, based on total airborne solids (TAS) that combine total ICP-MS elements and bicarbonate, are: 16 μg/m^3 for DI, 113 μg/m^3 for LL, 438 μg/m^3 for HH, and 521 μg/m^3 for HL water. Concentration in the air increased as concentration in the water increased. Approximately 90% of the emitted particles were in the inhalable 0.25–0.5 μm and < 0.25 μm size bins.

Fig. 7 reveals the linear regression models of metal/element content in fill water and in emitted particles for the four water qualities. Consistently, across all water qualities, air concentrations of individual metals and elements were proportional to their corresponding water concentrations. The R^2 of total ICP-MS elements for LL water, HL water, and HH water 0.977, 0.999, and 0.999, respectively, indicating that more metals/elements in the water to fill ultrasonic humidifier, more metals/elements are emitted in the air. The slopes for total ICP-MS elements of HL and HH waters are similar, approximately 0.6–0.7 μg/m^3 increment in TAS level per 1 mg/LTDS increase in fill water. Fig. 7 corroborates Fig. 6, and particles < 0.5 μm have larger slopes (purple and yellow lines in Fig. 7), suggesting most emitted particles were in the inhalable range of < 0.5 μm.

3.4. Adult and child exposure from inhalation of indoor air particles emitted from ultrasonic humidifier

A uniqueness of our research is comprehensively characterizing properties of emitted particles from humidifiers to calculate deposited mass of particles within the human respiratory tract. The deposited mass was generated from the properties of emitted particles, i.e., CMD and GSD for each water quality in the plume at 1.5 m, TAS mass concentrations of airborne particles on impactor filters, and an 8-h exposure time representing sleeping overnight in a humidified room when the indoor environment was at steady-state. Fig. 8 shows the deposited mass in each respiratory tract region relative to each water quality for an adult (70 kg, 20 m^3/d inhalation) and 1 to 3-month child (6 kg, 3.5 m^3/d inhalation). Larger particles emitted from ultrasonic humidifiers filled with high TDS waters (i.e., HL & HH) had higher mass deposition in the head region due to greater inertia. DI water was predicted to have a slightly higher deposition fraction in the pulmonary region than head and tracheobronchial regions based on particle size, but it represented lower inhalation exposure risk because almost no metals or elements were detected in the DI water and the particle counts were very low. Higher mass concentration produced by high TDS water resulted in higher mass deposited in the deeper lung, and thus posing higher inhalation exposure risk to humans.

Adults inhale more emitted particles and more particles deposit in the deeper lung, while children inhale less mass. Because children have lower body mass, they are exposed at a higher daily per body weight (BW) dose than adults even though there is less mass deposited in their respiratory tract. From the model results for 8 h exposure, children are exposed to 23.1 μg-total particles/kg-BW via inhaling the particles produced from ultrasonic humidifier filled with HH water, and adults are exposed to 8.7 μg-total particles/kg-BW. Under the scenario when using HL water to fill the humidifier and operate for 8 h, children are exposed to 28.8 μg-total particles/kg-BW, and adults are exposed to 11.2 μg-total particles/kg-BW. Children experience approximately 2.5 times higher exposure doses than adults.

4. Discussion

This innovative study is the first to calculate deposited mass of particles within the human respiratory tract when children and adults are exposed to particles from an ultrasonic humidifier using various water qualities. The comprehensive particle data for four water qualities used to fill the humidifier were used to model inhalation for children and adults. A general rule from the results is that as the TDS increases in fill water, proportionally more metals/elements are emitted as inhalable particles, greatly increasing particulate matter levels within indoor environments. High TDS water disproportionately increases < 0.25 μm particles compared to low TDS water. The increased PM levels lead to more particles deposited in human respiratory system, especially for head and pulmonary regions (Fig. 8). Taking low hardness and low TDS water as baseline, the higher TDS in HL and HH waters produce more fine particles from an ultrasonic humidifier, especially particles smaller than 0.25 μm (Fig. 6), which is consistent with previous studies (Highsmith et al., 1988; Sain et al., 2018; Yao et al., 2019).

The indoor air quality particulate values for LL tap water of 21,070 #/cm^3 and 113 μg/m^3, are consistent with high $PM_{2.5}$ mass

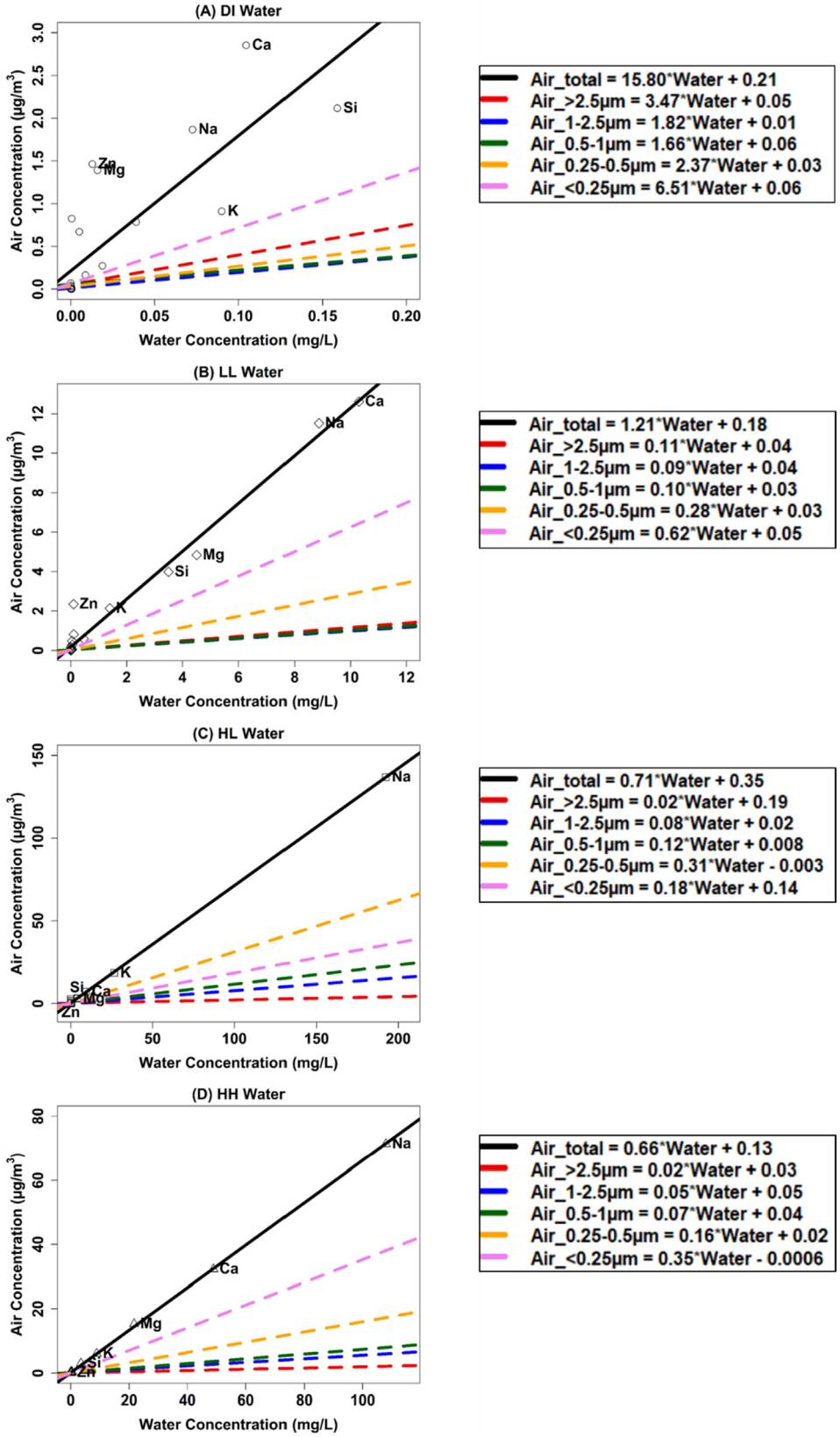

Fig. 7. Linear regression models of metals/elements in fill water and in air. A: deionized water (DI). B: low hardness and low TDS water (LL). C: low hardness and high TDS water (HL). D: high hardness and high TDS water (HH). Note: y-axes are different scales for each water quality. Sodium, magnesium, potassium, calcium, silicate, and zinc are labeled.

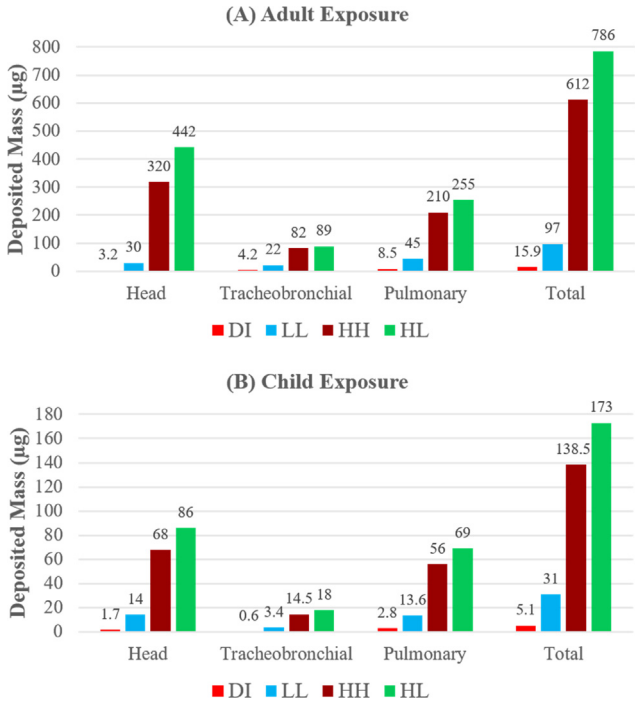

Fig. 8. MPPD model predicted deposited mass (based on total airborne solids) in human respiratory system of 1to 3-month old child and an adult from inhaling particles emitted by ultrasonic humidifier filled with different water qualities and 8-h exposure time. DI- deionized water; LL- low TDS and low hardness water; HH– high TDS and high hardness water; HL- high TDS and low hardness and water.

concentrations that infants were exposed to in their homes. A study of 0–1 year old infants investigated outdoor and indoor air exposure to ICP-MS elements in $PM_{2.5}$ particles to find that the indoor locations had higher $PM_{2.5}$ counts and mass concentration (Sloan et al., 2017). For three of the infants, their night time exposure concentrations reached 99, 140, or 176 μg/m^3 in their individual bedrooms. The authors attributed these high bedroom concentrations to the use of humidifiers filled with local tap water.

Prior research found 0.34 μm as count median diameter from measuring 0.2–5 μm particles emitted from ultrasonic humidifier filled with 303 mg/L TDS and 145 mg/L hardness water in a room of 25 m^3 with 1.16 per hour AER (Highsmith et al., 1988) Our results showed consistency in that the count median diameters of emitted particles from humidifiers filled with 697 mg/L TDS and 211 mg/L hardness water (HH), and 772 mg/L TDS and 38 mg/L hardness (HL) water were 0.35 μm and 0.38 μm at 1.2 m in plume, respectively (Fig. 4). In addition, a ratio of $PM_{2.5\text{-}10}/PM_{2.5}$ of 0.11 was previously reported under the conditions described above (Highsmithet al. 1988), and our study has the ratio at ~ 0.125 for HH water and ~ 0.06 for HL water, which confirms that emitted particles were dominantly < 2.5 μm particulates.

Highsmith et al. (1988) performed linear regression for mineral concentrations in fill water and in air, and the slopes were 1.92 for $PM_{2.5}$ and 0.17 for $PM_{2.5\text{-}10}$, which means per 1 mg/L TDS increase in fill water, the total increase of PMs in air was 2.09 μg/m^3. A later Highsmith study (1992) reported mass concentration of < 2.5 μm particles at 6307 μg/m^3 and 2.5–10 μm particles at 771 μg/m^3 (total at 7078 μg/m^3) from ultrasonic humidifier filled with 303 mg/L TDS water in a room of 25 m^3 with 1.16 per hour AER. Another study had an increment of 2.8 μg/m^3 per 1 mg/L increase in mineral concentration in fill water within a smaller room of 33.5 m^3 volume with AER of 0.87 per hour (Sain et al., 2018). Our study showed total increase in PMs in air is 1.21 μg/m^3 for LL water, 0.71 μg/m^3 for HL water, and 0.66 μg/m^3 for HH water, per 1 mg/L increase in TDS of fill water (Fig. 7) in a 72.2 m^3 room (AER = 1.5 h^{-1}). Smaller increment per 1 mg/L increase

Table 3
Comparison of steady state concentrations and number of emitted particles emitted from ultrasonic humidifier and current regulatory limits.

Water Quality	Exceeding Standard or Guidance			
	USEPA[a] (2015) (for ambient air)	WHO[b] (2006) (for ambient air)	US OSHA PEL[c] (2019) (for indoor, workplace air)	
	$PM_{2.5}$ 35 μg/m^3 PM_{10} 150 μg/m^3	$PM_{2.5}$ 25 μg/m^3 PM_{10} 50 μg/m^3	Respiratory fraction[d] 5 mg/m^3 or 530 #/cm^3	Total dust 15 mg/m^3 or 1,765 #/cm^3
This study[e]				
DI:2,194 #/cm^3	No $PM_{2.5}$	No $PM_{2.5}$		No particle mass
16 μg/m^3	No PM_{10}	No PM_{10}		Yes particle count
LL:21,070 #/cm^3	Yes $PM_{2.5}$	Yes $PM_{2.5}$		No particle mass
113 μg/m^3	No PM_{10}	Yes PM_{10}		Yes particle count
HH:38,353 #/cm^3	Yes $PM_{2.5}$	Yes $PM_{2.5}$		No particle mass
438 μg/m^3	Yes PM_{10}	Yes PM_{10}		Yes particle count
HL:43,880 #/cm^3	Yes $PM_{2.5}$	Yes $PM_{2.5}$		No particle mass
521 μg/m^3	Yes PM_{10}Yes PM_{10}	Yes PM_{10}		Yes particle count
Sain et al. (2018)[e]				
DI:12,100 #/cm^3	na[f] $PM_{2.5}$	na $PM_{2.5}$		No particle mass
	na PM_{10}	na PM_{10}		Yes particle count
LL:30,700 #/cm^3	Yes $PM_{2.5}$	Yes $PM_{2.5}$		No particle mass
118 μg/m^3	No PM_{10}	Yes PM_{10}		Yes particle count
HH:46,500 #/cm^3	Yes $PM_{2.5}$	Yes $PM_{2.5}$		No particle mass
495 μg/m^3	Yes PM_{10}	Yes PM_{10}		Yes particle count
HL:59,100 #/cm^3	Yes $PM_{2.5}$	Yes $PM_{2.5}$		No particle mass
966 μg/m^3	Yes PM_{10}	Yes PM_{10}		Yes particle count

a. standards are for ambient air and not indoor air, value is 24-hr average
b. guidelines for 24-hr average and stricter than regulatory limits of European countries
c. PEL = permissible exposure limits, value is 8-hr time weighted average for inert and nuisance dust or particulates not otherwise regulated
d. respiratory fraction determined by cyclone allowing passing for PM_{10}
e. DI- deionized water; LL- low TDS and low hardness water; HH– high TDS and high hardness water; HL- high TDS and low hardness and water.
f. na: not applicable

in aqueous mineral concentration in fill water is due to different experimental conditions, such as humidifier model, water consumption rate, room size, and air exchange rate (e.g., room size and AER drive concentrations of indoor air particles).

Previous research reported number and mass concentrations of emitted particles for four water qualities similar to this study, but in a smaller room (volume = 33.5 m^3; AER = 0.87 h^{-1}) and the values were comparable (Sain et al., 2018), as shown in Table 3. Through comparing regulatory limits and guidelines of air quality and concentrations of emitted particles in Table 3, it is apparent that the levels of residential indoor airborne particles produced by ultrasonic humidifier pose adverse health effects to humans, especially when using high-mineral content to fill the humidifier. Moreover, $PM_{2.5}$ and PM_{10} regulatory limits are based on health risk evaluation for ambient air and not for indoor air, which should be taken into account in future legislation.

Implications of this study are that humidification of indoor environments simultaneously results in emission of inhalable mineral particles when ultrasonic humidifiers are used. The increased particulate matter in indoor air when using tap water with minerals can exceed ambient air $PM_{2.5}$ and PM_{10} standards and will be most influenced by water quality, which globally and regionally varies greatly in TDS and hardness content. TDS and hardness for tap water do not have WHO or government health-based regulatory guidance or standards because they are unlikely to have adverse health effects when consumed by ingestion. In fact, calcium and magnesium hardness are beneficial for cardiovascular health and, therefore, are desirable in drinking water (Cotruvo and Bartram, 2009). The challenge to global consumers and societies is that typical regulations/guidance for drinking water are based on ingestion, the regulations/guidance for air quality are based on inhalation, and there is no integrated regulations/guidance for inhaling of water constituents during routine consumer use, such as using humidifiers or showering. Consumers may unknowingly be degrading their indoor air quality when using tap water of acceptable drinking water quality. For humidifiers, degradation of air quality by water quality can be avoided by using distilled water. A much higher particle concentration exists at 0.3 m in-plume location, which indicates the inhalation exposure may be much higher than the deposited mass reported in this study. Further research on the inhalation exposure when ultrasonic humidifier is placed very close to human infants and adults is required. This study expands the knowledge scope of inhalation exposure to aerosolized minerals from ultrasonic humidifiers, and links exposure doses with water quality and resulting particle mass concentration in air.

5. Conclusions

This research demonstrates that indoor air μg/m^3 concentrations for particles emitted from ultrasonic humidifiers filled with tap water containing minerals exceed ambient air concentrations for $PM_{2.5}$ and/or PM_{10}. When inhaled during an 8-hr exposure time, and depending on mineral water quality, humidifier aerosols can deposit up to 100 s of μg minerals in the human child respiratory tract and 3–4.5 times more μg of minerals in human adult respiratory tract.

Water quality has the greatest impact on the size and concentration distribution of emitted particulates from an ultrasonic humidifier. Water with higher total dissolved solids produced more and larger particles from ultrasonic humidifiers than waters with lower TDS. The HL water produced had more TDS than the HH water, hence why it produced larger and more particles. Distance in the plume from point of emission has a minor effect and only results in a significant difference in particle concentration distributions closer to the humidifier outlet in the plume, while particle distributions in the plume and about a meter below the plume were the same. Higher TDS results in greater lung dose of particles, especially for children. Distilled water should be used whenever possible to prevent respiratory irritation (USEPA, 1991), although there is little indication that consumers are aware of or follow this consideration.

CRediT authorship contribution statement

Wenchuo Yao: Conceptualization, Methodology, Writing - original draft, Writing - review & editing. **Rachael Dal Porto:** Investigation, Methodology, Writing - original draft. **Daniel L. Gallagher:** Formal analysis, Writing - review & editing. **Andrea M. Dietrich:** Conceptualization, Writing - review & editing, Supervision.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2020.105902.

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