Received: 14 June 2022 | Revised: 5 October 2022 | Accepted: 17 October 2022
DOI: 10.1111/ina.13157

ORIGINAL ARTICLE

WILEY

Resuspension of particles deposited by nano-enabled consumer sprays: The role of product type, flooring material, and resuspension force

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Funding information
U.S. Consumer Product Safety Commission, Grant/Award Number: CPSC-S-16-0057; USDA-NIFA through NJAES at Rutgers, The State University of New Jersey, Grant/Award Number: USDA-NIFA-1008687.NJAES-07215

Abstract

With the development of nanotechnology, an increasing number of nano-enabled consumer products are introduced into the market. The release, deposition, and resuspension of particles from such products could be an important contributor to indoor air pollution and adverse health effects. Our study tested the spray and resuspension of seven nano-enabled consumer products and investigated how flooring material and resuspension force affected the resuspension of particles from these products. Results show that resuspension rates can range from 10^{-4} to 5×10^{-1} h^{-1}, depending on the product, flooring material (e.g., carpet and vinyl), and resuspension force (e.g., a walking adult and a moving child; the latter was simulated by a robotic sampler). The resuspension rate from the carpet was statistically significantly higher than that from vinyl flooring, while the resuspension rate by the adult was statiscally significantly higher than that by the robot. In addition, the interaction of investigated factors also played a role in particle resuspension rate. For a subgroup of products based on copper (Cu), silver (Ag), and zinc (Zn) nanomaterials, the resuspension rate reached as high 5×10^{-1} h^{-1}, rates higher than those reported in existing studies with house dust or Arizona Road Dust.

KEYWORDS
consumer spray, flooring material, human walking, indoor air quality, particle resuspension, robotic sampler

1 | INTRODUCTION

Particulate matter (PM) is a known indoor air pollutant, and it can cause adverse effects, including decreased lung function, increased respiratory symptoms such as bronchitis and asthma, and increased morbidity and mortality due to cardiovascular diseases.[1–4] The presence of airborne particles indoors is caused by outdoor PM and indoor activities such as cooking, cleaning, use of sprays, as well as candles and incense.[5–8] In addition, a large fraction of indoor particles eventually settles on surfaces which then can then be resuspended back into the air by human activities.[9–13]

The resuspension of particles is an important indoor particle source. Resuspension of different types of particles (e.g., silica particles,[14] potassium chloride [KCl] salt particles,[15] alumina powder,[16] cigarette smoke particles,[17] Arizona Test Dust,[18,19] biological particles,[10,20] and dust from real homes[5,21]) has been investigated.

Particles released from the ever-increasing nano-enabled consumer products are becoming an additional important indoor pollution source. More than 5000 nano-enabled consumer products from 50 countries have been commercialized,[22] and the number is growing.[23] Studies have shown that consumers could be exposed to airborne nanoparticles and their agglomerates released from such products during various product lifecycle stages.[24,25] Nanosized particles are also released from consumer products not labeled as nanotechnology-based.[25–27] Particles in the nanoscale size range (1–100 nm) have the potential to be more injurious compared with $PM_{2.5}$ and PM_{10},[28–30] because of their ability to enter the body's cells and damage proteins, cell membranes, and DNA, induce acute phase response, oxidative stress, and inflammation.[28,31,32] Exposure to nanoparticles is associated with vascular dysfunction, adverse acute respiratory, and cardiovascular effects, and histopathological (microscopic tissue) changes in the liver and kidney.[29,33,34] At the same time, concerns about the toxic potential of silver (Ag), zinc oxide (ZnO), and titanium dioxide (TiO_2) and other nanosized components commonly used in consumer products make it more important to understand the behavior of particles released from nano-enabled consumer products.[35–39] When such particles are released indoors, a certain fraction of them will settle on surfaces and could be resuspended, thus resulting in occupant exposures. However, there is currently a lack of data on the resuspension of particles from nanotechnology-based consumer products.

According to existing research, the resuspension of deposited particles depends on particle properties, flooring materials, human activity, and environmental conditions. Particle properties including size, shape, morphology, surface roughness, ionic strength, and chemistry can affect the particle–fluid, particle–particle, and particle–surface interactions, thus impacting particle detachment from the surface.[13,40] Henry and Minier[40] indicated that resuspension mechanisms were different for different particle sizes; so, nanosized particles may have different resuspension mechanisms and show different resuspension behaviors compared with larger particles.

One of the main modes of particle resuspension from the floor is walking, where air jets are created between shoes and the floor,[12] thus lifting the deposited particles. The characteristics of flooring material affect particle-surface interactions, thus affecting particle resuspension.[13,18,41] Previous studies have tested particle resuspension from multiple materials. For example, Gomes et al.[10] observed that resuspension rates for a linoleum flooring reservoir were greater than for artificial grass carpet for both quartz and roach dust, but no significant differences were observed for dust mites resuspended from low pile carpet and linoleum flooring. Qian and Ferro[18] found lower particle resuspension rates from a hard floor compared to new carpet when Arizona Road Dust was resuspended by a walking adult. Mukai et al.[15] found that more particles were resuspended from carpet than linoleum flooring and galvanized sheet metal when KCl salt particles were resuspended by turbulent flows. Tian et al.[42] showed that carpets provided significantly higher resuspension fractions (e.g., the fraction of surface dust resuspended per step) than hardwood and vinyl flooring for particles between 3.0 and 10.0 μm. Also, Benabed et al.[16] indicated that hardwood flooring provided higher resuspension of alumina powder than linoleum flooring for all investigated size ranges.

Human activity type is also a factor affecting particle resuspension. Studies demonstrated that particle resuspension varied by walking styles and speed as well as shoe types.[18,42–46] Besides the complex effects of different walking styles on particle resuspension, the differences in resuspension caused by an adult and a child should also be considered. While the personal exposure of an adult could be monitored by personal sampling devices, such devices are most often not feasible for use with young children. Thus, robotic samplers, such as the Pretoddler Inhalable Particulate Environmental Robotic (PIPER), could be used to simulate child movements and estimate their exposure.[47–50] In another study, Hyytiäinen et al.[51] used a custom-built 4-kg mechanical crawling unit to mimic the belly crawl of an infant and measured the resuspended microbiota in the infant's breathing zone.

Environmental conditions also greatly affect the resuspension of particles.[13] Increases in relative humidity (RH) can suppress particle resuspension.[52,53] An increase in RH decreases the resuspension rates of hydrophilic particles, while hydrophobic particles are less sensitive to the change of RH.[54] For example, high RH decreased particle resuspension for an old carpet, but enhanced resuspension for a new carpet.[19] Furthermore, particle loading, the contact time between a particle and its substrate, and the electrostatic interactions also affect particle resuspension.[13]

We hypothesize that the above factors will also affect the resuspension of particles released from nanotechnology-based consumer products. Thus, this study investigated the resuspension of particles released from 7 different consumer nanotechnology-based sprays, including silver (Ag)-based, zinc (Zn)-based, and copper (Cu)-based spray products. Overall, the objectives of this study were as follows: (1) to characterize the resuspension of particles that were deposited due to the use of nano-enabled consumer sprays; (2) to determine the differences in particle resuspension rates among different products; (3) to examine the effect of flooring materials on particle resuspension; and (4) to determine the particle resuspension rate caused by a walking adult and a moving child, with the latter simulated by a robot.

2 | MATERIALS AND METHODS

2.1 | Consumer sprays

In the initial phase of this project, an Ag-based shoe deodorizer (code S10), an Ag-based surface cleaner (code S13), and two Zn-based skin protectants (code Z2 and Z4) were purchased and investigated. These products were selected based on our earlier study investigating the release of nanoparticles from consumer sprays,[26] and the laboratory codes for these products are the same as in our previous work. Both S10 and S13 are described as nanotechnology-based;

both Z2 and Z4 were not labeled as nanosprays,[26] but they produced nanosized particles once sprayed.[26,27] The micrographs of nanoparticles in liquid-borne and airborne states of those products are provided in Calderón et al.[26] A complete suite of investigations (e.g., particle resuspension was investigated as a function of spray type, flooring material, and resuspension force) was performed with these four products. In the later stages of the project, three more products that had not been investigated in our earlier work were acquired: a Cu-based skin toner (code Cu1), an Ag-based immune support hydrosol (code S14), and a Zn-based immune system defense booster (code Z7). These three products were labeled by their manufacturers as nanotechnology based. Their laboratory codes were assigned sequentially following our earlier work and the numbering of products that had already been investigated. For these three products, we investigated only the particle resuspension from the carpet by an adult, as this combination resulted in the highest resuspended particle mass concentration according to our initial measurements. The description and advertised contents of all seven investigated products are presented in Table 1.

2.2 | Flooring materials

A carpet and vinyl surface were used. The carpet was an ordinary household level-loop carpet (Texture Carpet Model EF286-311-1200, the Home Depot). The vinyl flooring (Residential Vinyl Sheet Model C1100405K509G14) was acquired from the same source. A new carpet and a new vinyl sheet were used for each product. Each flooring material, with dimensions 2.8 m × 1.5 m = 4.2 m^2, was laid in the middle of the experiment chamber described below.

2.3 | Resuspension force

Particle resuspension was examined as a function of resuspension force (i.e., the weight of the experimenter): a walking adult (70 kg) and a moving ReCon Programmable Rover (3.7 kg with instruments, version 6.0, SmartLab Toys), which was used to simulate a child moving on the floor. This particular robot was successfully used in our earlier study investigating the resuspension of pesticides indoors.[55] The robot moved autonomously inside the chamber during the experiments based on its programming.

The same adult performed all resuspension experiments since the walking behavior, shoe type, and the experimenter's weight can affect particle resuspension.[18,44] In addition, the shoes were covered by disposable shoe covers.

2.4 | Experiment chamber

To investigate the deposition and resuspension of particles emitted from the sprays, a completely new chamber was created by partitioning an existing room, as shown in Figure 1. The chamber simulated real household conditions and was similar in size to a small room, with dimensions of 2.8 m length × 1.6 m width × 2.4 m height = 10.75 m^3. The chamber accommodated all measurement equipment, provided enough space for spray application, and was conducive for changing flooring type between carpet and vinyl. The chamber was constructed using plastic strip curtains, and the inner surfaces of the chamber were covered with aluminum foil to reduce static effects. In order to accommodate the existing room layout, the chamber was built between two air supply diffusers. The room entrance door and diffusers were sealed during experiments to avoid active ventilation and minimize particle penetration. Thus, the measured particles were only due to spraying and resuspension. After each day of experiments, the diffusers were opened to control the temperature and humidity inside the room and the chamber. In addition, between experiments, an air purifier (HA202 series, Honeywell Inc.) was operated in the room to minimize background particle concentration. The temperature and humidity inside the chamber were measured and recorded for each experiment.

The size distribution and concentration of the sprayed and resuspended particles were measured at 1.1 m above the floor for each experiment. The layout of the instruments is shown in Figure 1. The EPA recommended 1.1 m height for conducting indoor air investigations[56] was used as has been with earlier indoor air quality measurements.[57,58]

TABLE 1 Investigated spray products.

Product code	Advertised contents
Initially acquired products	
S10	Colloidal Silver
S13	1.21 µg/ml silver and silver nanoparticles[26]
Z2	Zinc Pyrithione
Z4	Zinc Pyrithione
Additional products	
Cu1	Colloidal copper
S14	Bio-active silver ions and silver nanoclusters
Z7	Liquid nano-suspension zinc gluconate. Particle size less than 200 nanometers

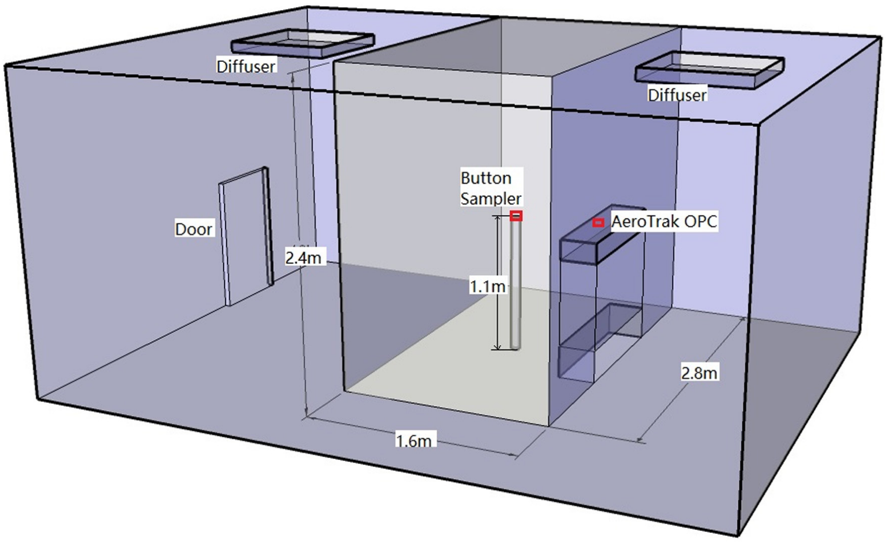

FIGURE 1 Design of the test chamber (the structure in the middle of a room with dimensions of 2.8 m length × 1.6 m width × 2.4 m height) and placement of equipment used in resuspension experiments.

2.5 | Equipment

2.5.1 | Measurement of particle mass concentrations

In order to calculate particle resuspension rates (described in section 2.7), floor mass loading and airborne particle mass concentrations were calculated. When using nanotechnology-enabled consumer sprays, the vast majority of the released particle mass is represented by larger particles (i.e. > 3 μm).[26] Therefore, we used filter-based particle mass concentrations to represent the overall mass concentrations.

Twelve 37 mm PTFE filters (SKC Inc.) were evenly distributed across the floor before each spraying experiment and were collected after spraying to determine the mass of deposited particles. The floor mass loading in g/m^2 was calculated based on the average deposited particle mass on those filters.

A Button Aerosol Sampler (SKC Inc.) with a 25 mm PTFE filter (SKC Inc.) and an AirChek XR5000 Air Sample Pump (SKC Inc.) that provided a 4 L/min flow rate were placed in the middle of the chamber at 1.1 m height (Figure 1) to capture resuspended particles and determine their mass concentration.

In order to determine the mass of particles captured on PTFE filters (SKC Inc.) (both for floor loading and airborne concentration), the filters were kept at a constant temperature (20–23°C) and relative humidity (30%–40%) for at least 3 days before weighing pre- and post-sampling. The filters were weighed using an MT-5 Microbalance (Mettler-Toledo). Quality control filters in the weighing room and on-site control filters (e.g., blanks) were used for each experiment and weighing session.

2.5.2 | Measurement of airborne particle number and mass concentrations

An Aerotrak optical particle counter (OPC; Model 8220, TSI Inc.) was used to monitor the number concentration of released and resuspended particles from 0.3 μm to greater than 10 μm. The device was placed at the edge of the flooring material at the height of 1.1 m (Figure 1). To estimate airborne particle mass concentrations for background measurements, we used the average value of the OPC measurement channel for particle size and assumed that particles were spheres with a density of 1 g/cm^3.

2.5.3 | Particle composition analysis

A 25 mm nuclepore membranes with a pore size of 1 μm (Whatman, Marlborough) were used to collect sprayed and settled particles for their composition analysis. One filter was left at the center of the chamber while spraying each product. The collected samples were coated with a 10 nm gold layer and then examined with Zeiss Sigma Field Emission Scanning Electron Microscope (FESEM) with an Oxford INCA PentaFETx3 Energy Dispersive X-ray Spectroscopy (EDS) system (Model 8100, ZEISS Microscopy) to determine the morphology and chemical composition of the particles. As a quality control measure, we examined a blank filter using the same techniques, and the blank filter's image coated with gold and a spectrum of its composition are presented in Figure S1. The presence of carbon, oxygen, and gold was observed.

2.5.4 | Personal protective equipment (PPE)

Protective clothing (Coverall with Hood, DuPont™ Tyvek®), respirator (Model 62023HA1-C, 3M Company), and disposable foot covers were worn during experiments to reduce the contribution of particles due to shedding from skin and clothing.

2.6 | Experimental process

The experimental process timeline is shown in Figure 2 and described below.

2.6.1 | Particle background control

Before each measurement, the air purifier was operated overnight to ensure low airborne particle background in the chamber. Immediately before spraying, the carpet or vinyl flooring was thoroughly vacuumed (Dyson ball compact allergy+, Dyson Ltd.). Following vacuuming, an adult wearing personal protective equipment walked in the chamber for 5 min to determine if any residual particles could be resuspended, based on aerosol mass measurements by the AeroTrak OPC. Once the airborne particle mass concentration was ~1.5 μg/m^3 or less, the flooring was considered "clean." If the concentration was higher, the surface was vacuumed and walked on again until the resulting airborne background concentration was ~1.5 μg/m^3. An example of background mass concentration is shown in Figure S2, and in this particular case, the total mass concentration was 1.3 μg/m^3. During this period, any measured airborne particle concentration was removed from the experiment-related particle concentration during the data analysis step. After the final vacuuming, any remaining particles were allowed to settle for an hour before spraying. For resuspension experiments, there was no vacuuming, but the background concentration was measured before each experiment.

2.6.2 | Procedure to measure sprayed and settled particles

Before spraying, the experimenter wearing PPE entered the chamber and evenly distributed twelve 37-mm PTFE filters on the floor for deposited particle collection. The AeroTrak OPC and Button Sampler remained stationary, as described in section 2.5. At the beginning of the experiment, the experimenter started the AeroTrak and the Button Sampler pump and then used a spray product (e.g., sprayed the product) parallel to the floor at the height of about 1.1 m while walking across the chamber at a regular pace for 6 min. The spray was activated manually, about once per second. After the spraying, the Button sampler's pump was stopped immediately, and the experimenter left the room to allow the particles to settle. The AeroTrak was left running for about 2 h until the remaining mass concentration was close to the background level. The 37-mm filters were then collected from the floor. One hour later, when the vast majority of particles had settled, the air purifier was turned on until the next day's resuspension experiment.

2.6.3 | Measurement of particle resuspension

The deposited particles were resuspended either by an adult (70 kg weight) walking on the surface or by the movement of a Recon Programmable Rover (3.7 kg, including the carried devices).

In the initial phases of the project, the resuspension experiments were repeated at 24, 48, and 72 h post-spraying to determine the time between the spraying and resuspension that would result in the highest resuspended particle concentration. These preliminary results are shown in Table 2. The time to reach the highest resuspended particle concentration for vinyl flooring was 24 h for all products, while the time varied among products to reach the maximum resuspension from carpet flooring. For each product, the time between spraying and resuspension yielding the highest resuspension was used in the subsequent experiments.

During the resuspension experiments, the experimenter wearing PPE walked in the chamber for 11 min (Figure 2). The airborne particles were sampled by a Button Sampler at 1.1 m, as shown in Figure 1. The Button sampler was stopped immediately after the 11 min resuspension period was finished, and the experimenter left the room allowing the particles to settle without further perturbation. The AeroTrak continued measuring until the airborne number concentration decreased to close to the background level, approximately 2 h.

Since 3–6 days were needed to perform each resuspension experiment, the S10, S13, Z2, and Z4 samples were resuspended twice by the adult and the robot from both carpet and vinyl floorings. The resuspension experiments with products Cu1, S14, and Z7 were performed twice and only by an adult walking on the carpet.

2.7 | Determination of product particle resuspension rate

Once particles resulting from spraying had settled and then stayed deposited for 2 h, the volatile components from each product evaporated, and the mass of solid material remaining on filters was

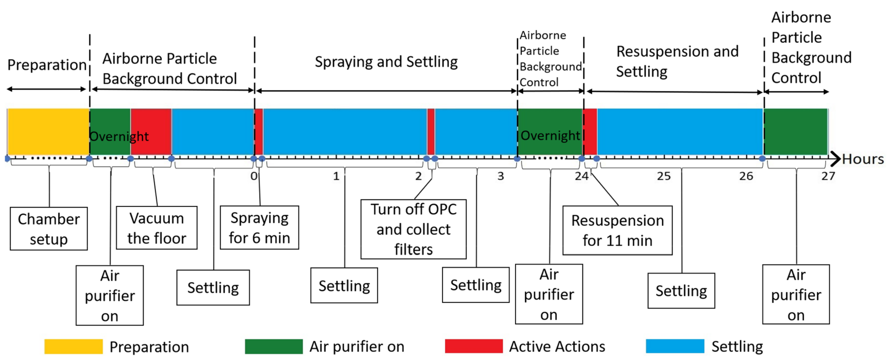

FIGURE 2 Timeline of each experiment.

TABLE 2 Number of 24-h intervals (e.g., days) post-spraying that resulted in maximum resuspended particle mass concentration.

Product code	Resuspension from carpet by an adult	Resuspension from vinyl by an adult	Resuspension from carpet by a robot	Resuspension from vinyl by a robot
Initially acquired products				
S10	1	1	1	1
S13	3	1	3	1
Z2	3	1	3	1
Z4	3	1	3	1
Additional products				
Cu1	2			
S14	2			
Z7	2			

determined by weighing. The measured floor loading by the deposited material and the mass concentration of resuspended particles captured on filters at 1.1 m height were used to calculate the resuspension rate for each product as a function of resuspension variables, which are listed in Table S1. Assuming a well-mixed air volume and evenly distributed particle loading on the floor, the resuspension rate for particles, *r*, can be estimated by Equation (1):[18]

$$r = \frac{V}{AL}\left(\frac{dC}{dt} + (a+k)C\right) \quad (1)$$

By integrating both sides of Equation (1) from time *0* to a certain time *t*, *r(t)* can be estimated by equation (2).[18] The volume where particles are resuspended is given in Equation (3):

$$r(t) = \frac{V}{AL(t)}\left(\frac{C(t)-C(0)}{t} + (a+k)C(t)\right) \quad (2)$$

$$V = AH \quad (3)$$

where: r, Resuspension rate (h^{-1}); V, Air volume where particles are resuspended (m^3); A, Resuspension surface area, for example, carpet or vinyl surface area (m^2); L, Floor loading (particle/m^2); C, Concentration inside the chamber (particle/m^3); a, Air exchange rate (h^{-1}); k, Deposition rate (h^{-1}); H, Chamber height, for example, the height where filter samples were collected (m).

In our study, the calculation of the resuspension rate is based on particle mass deposited and captured on filters: thus, the floor loading (L (t)) was measured as g/m^2, and the resuspended particle mass concentration (C) inside the chamber was measured as g/m^3. The concentration inside the chamber at t = 0 (C [0]) was measured before experiments and constituted the background level. Since there was no ventilation in the chamber, air exchange *a* is zero. Also, particle deposition and the resulting deposition rate (k) could be ignored since our resuspension experiments are short in duration. Combining Equations (2) and (3) and replacing number concentration with the mass concentration, the resuspension rate can then be derived from the simplified Equation (2) as:

$$r(t) = \frac{H}{L(t)}\left(\frac{C(t)-C(0)}{t}\right) \quad (4)$$

In our experiments, the chamber height H was 2.4 m, and the resuspended particle mass concentration C (t) was measured at 1.1 m height. The floor loading L was calculated based on the average mass loading (g/m^2) of twelve 37 mm filters evenly positioned on the floor during spraying. Figure S3 shows the average floor loading for each experiment and its standard deviation based on those twelve 37 mm filters. The relative standard deviation across experiments ranged between 21% and 79%, and the average relative standard deviation was 43%.

2.8 | Statistical analysis

A one-way ANOVA was used to examine the difference in sprayed mass concentrations among the products; a three-way ANOVA test examined whether the resuspension rate was statistically significantly affected by product, flooring material, and resuspension force. A post hoc Tukey's HSD (honestly significant difference) test was used to analyze the difference between each pair of variables. All statistical analyses were run by SPSS software (IBM).

3 | RESULTS

3.1 | Deposited particles

Representative images of particles from each product that settled on the floor after spraying are shown in Figure 3, and the composition spectra of these particles are shown in Figure S4 as a weight percent (Wt%) of total components. The composition spectra for a blank filter were also analyzed, and C, O, and Au were observed (Figure S1). The C and O are shown in all samples as they were detected in both the filter material and the deposited particles. Au was used for coating and not present in the consumer sprays, and thus, it was removed from the composition chart (weight percent) in Figure S4.

Particles from S10 (Figure 3A) ranged from about 30 nm to 300 nm in size, and both single particles and agglomerates particles can be seen. Also, it seems that the filter is covered by an emulsion that forms a solid layer on top of the filter surface. The remnant is likely part of the product matrix minus the evaporated volatile components. The examined particles (Spectrum 1) showed 1.7% silver (Figure S4A).

Figure 3B shows individual particles of S13 samples. S13 had an even higher emulsion amount than S10; once the volatile components evaporated, the remaining layer even covered some filter pores (see image on the left). While single particles from this product were hard to find, those that were observed were smaller than 100 nm (Figure 3B) and contained 0.1% silver (Figure S4B), which is one order of magnitude lower than the amount of silver in S10. When S10 and S13 were examined in bulk using ICP-MS,[26] their silver concentrations were 3.9 and 1.2 µg/ml, respectively.

Particles from Z2 (Figure 3C) differ entirely from silver-based S10 and S13 products. Z2 samples contained single and agglomerated particles, with single particles ranging from about 150 nm to larger than 5 µm in diameter. The composition of the examined Z2 particle (Spectrum 1 in Figure 3C) showed 7.5% of zinc by weight.

Particles from Z4 samples (Figure 3D) looked similar to those from Z2, but they appeared to be compacted into layers. The shapes of individual particles appear to be dominated by particles of platelet shape, for example, particles where two dimensions dominate, and the particles of Z2 were more three-dimensional. Individual particles from Z4 ranged from about 100 nm to larger than the 10 µm in diameter, and the largest agglomerated particles were visible to the naked eye. The Z4 particles (Spectrum 1 in Figure 3D) contained 9.9% zinc by weight (Figure S4C). Both Z2 and Z4 samples contained nitrogen, zinc, and sulfur (Figure S4C,D).

Cu1 samples (Figure 3E) contained both single and agglomerated particles. However, the shape of the particles is completely different from what we saw with other products. Typical Cu1 particles ranged from 100 nm to 300 nm in length and from 50 to 100 nm in width. The agglomerates were approximately 0.5 µm in size, with individual particles within the agglomerates easy to discern. The examined Cu1 particles (Spectrum 2 in Figure 3E) showed 1.9% of copper by weight (Figure S4E).

Images of Ag-based S14 samples (Figure 3F) also show both single and agglomerated particles, with individual particles ranging in size from 20 nm to 500 nm. Individual S14 particles resemble those of the S10 product—they seem to have a shape similar to a cube. The concentration of Ag in this agglomerate was 12.7% (Figure S4F).

Particles from Z7 are shown at large and small scales (Figure 3G). It is apparent that the particles from this Zn-based product look very different from particles from Z2 and Z4. The filter seems to be covered by emulsion drops and thin plate-type particles visible to the naked eye. Some of the larger particles resemble a flower or an ornament. Both single and agglomerated particles were found in the samples, and the individual particles were as small as 700 nm, while the agglomerated particles were as large as 80 µm. As shown in Figure S4G, the examined particles had 36.2% of zinc, a much larger Zn fraction than what was measured in Z2 and Z4 particles.

The single particles and their agglomerates observed in Figure 3 confirm findings in previous studies[25–27] that the use of nano-enabled consumer products can release airborne particles, these particles can settle on the floor, and the size of deposited particles can be as small as ~20 nm, even when the consumer products are not labeled as nanosprays by their manufacturers (e.g., Z2 and Z4).

3.2 | Particle size distribution during spraying

The size distributions of particles produced by spraying the seven products are shown in Figure 4. It is apparent that all products produced more particles of smaller sizes than larger sizes. All products show number concentrations higher than $10^7/m^3$ at the particle size of 0.4 µm, but the concentrations decrease as particle size increases. The number concentrations of S13, Z2, and Z4 are relatively steady as a function of diameter; they decrease only approximately one order of magnitude as the particle size increases from 0.4 to 15 µm. For comparison, the number concentrations of other products decrease by two to four orders of magnitude over the same size range.

Although the size distributions of nanosized particles were not measured due to the instrument's detection limit, the concentration of the released particles trended higher with decreasing particle diameter. Moreover, the particle number concentrations of different products became similar as the particle size decreased and ranged from 10^7 to 10^8 particles/m^3 in the 0.3–0.5 µm range. Similar results were observed by Nazarenko et al.[27] that during manual spraying of nanosprays, the particle number concentrations in the 14–500 nm range were similar for different nanosprays and ranged from 10^8 to 10^9 particles/m^3. The difference in the range of observed particle number concentration between studies is likely due to different examined particle sizes, spraying frequency, and different testing environments (e.g., ours was a walk-in chamber while Nazarenko et al.[27] used a biosafety cabinet).

3.3 | Particle mass concentration produced by spraying

The filter-based mass concentrations of particles released during spraying are presented in Figure 5. Z7 produced the highest mass concentration of 1.39×10^4 µg/m^3, while S14 produced the lowest mass concentration of 2.19×10^2 µg/m^3.

ANOVA test indicated a statistically significant difference among products (p-value < 0.05), and the post hoc Tukey's HSD test results are shown in Table 3. Z7 was statistically significantly different from all the other products, likely due to the high amount of emulsion released and deposited by Z7, as shown in Figure 3G. No statistically significant difference was observed between Cu1 and the other products, except Z7. In addition, no statistically significant

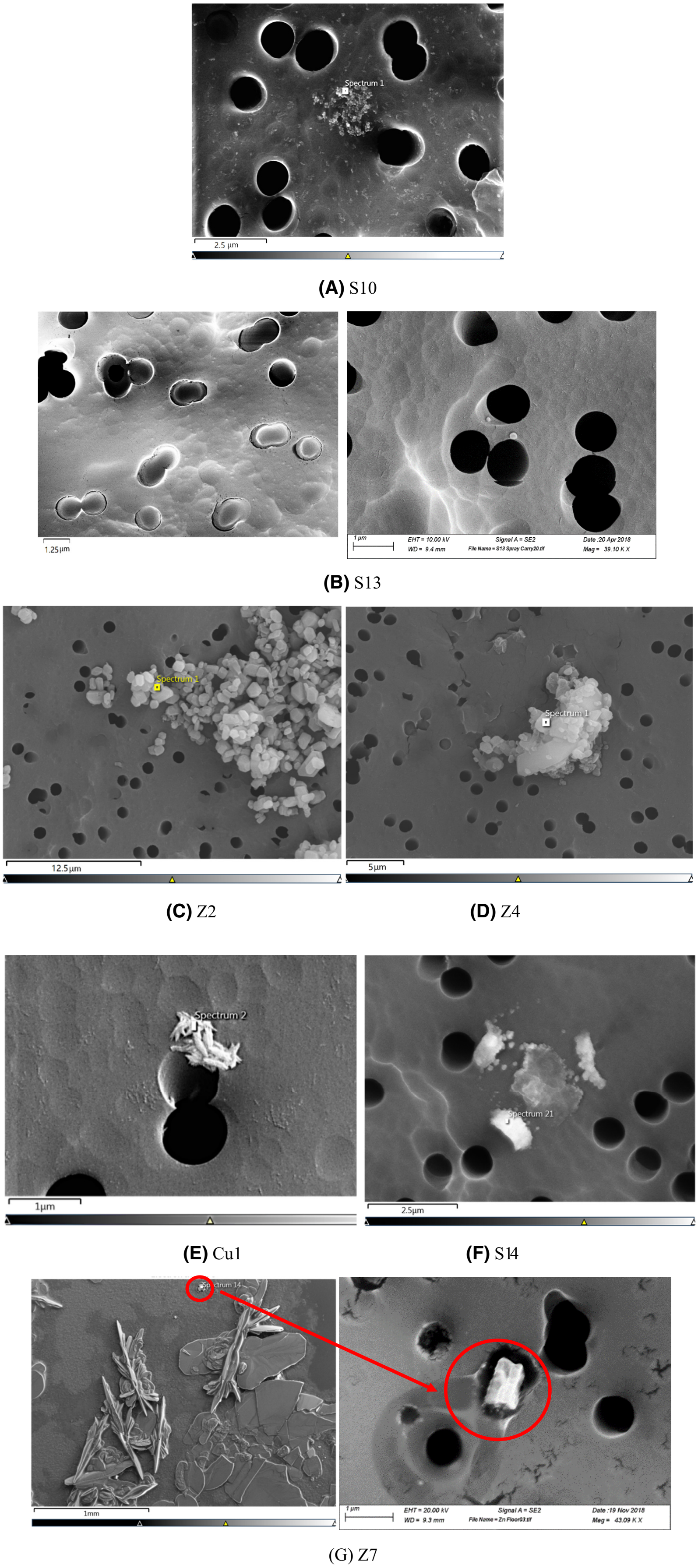

FIGURE 3 FESEM pictures of particles that were released during spraying and then deposited on filters placed on the floor. (A) S10, (B) S13, (C) Z2, (D) Z4, (E) Cu1, (F) S14, (G) Z7.

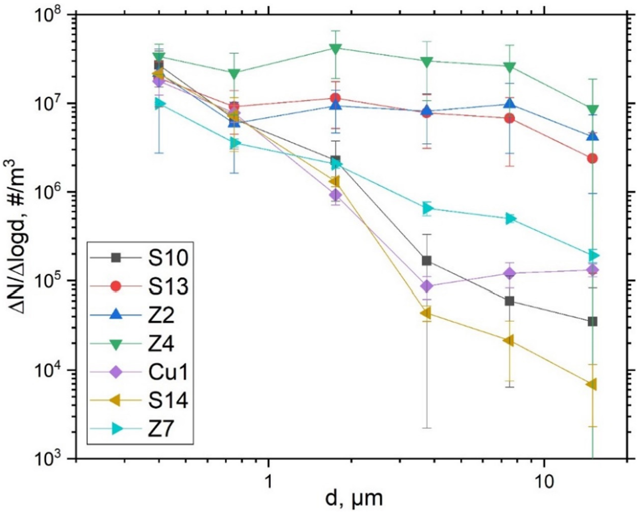

FIGURE 4 Size distributions of particles released during spraying and measured at 1.1 m height using Aerotrak OPC. The S10, S13, Z2, and Z4 data are averages of 8 repeats, and the Cu1, S14, and Z7 data are average of 2 repeats. The error bars represent standard deviations. *x*-axis and *y*-axis are in log scale.

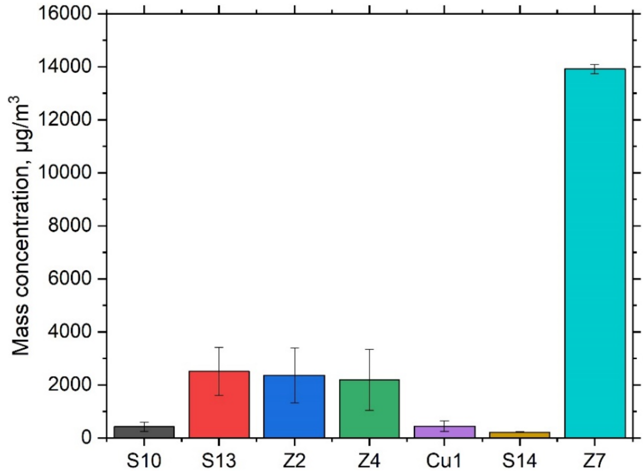

FIGURE 5 Mass concentrations of particles released during spraying and measured at 1.1 m height using a Button sampler (SKC Inc) with 25 mm PTFE filters. The S10, S13, Z2, and Z4 data are averages of 8 repeats, and the Cu1, S14, and Z7 data are average of 2 repeats. The error bars represent standard deviations.

differences were observed among S10, Cu1, and S14 and among S13, Z2, Z4, and Cu1.

3.4 | Resuspended particle size distribution

3.4.1 | The effect of different flooring materials and experimenters

The particle size distributions of S10, S13, Z2, and Z4 products resuspended from the two flooring types and by two different experimenters are shown in Figure 6. The number concentration of particles smaller than 0.75 μm in diameter is similar for the same product regardless of the flooring type (carpet vs. vinyl) or experimenter (adult vs. robot). When particles larger than 0.75 μm are considered, the highest number concentrations are resuspended from a carpet by an adult. When the flooring type is the same, higher particle number concentrations were produced due to resuspension by an adult compared to the robot in most size ranges, regardless of the product. When the adult resuspended the particles, higher particle number concentrations were typically resuspended from the carpet than vinyl flooring, especially larger particle sizes. When products are resuspended by the robot, products S13 and Z2 show higher resuspended number concentrations from carpet than vinyl in most size ranges, while the other products show similar results for both flooring types. The difference between the carpet and the vinyl flooring is not as pronounced due to the lower mass of the robot compared to the adult. However, it is important to stress that particles are still resuspended from both surfaces by the robot weighing only 3.7 kg. Higher particle concentrations are likely to be resuspended by a child playing on the floor, resulting in a child's exposure to those particles, especially due to the proximity of their breathing zone to the floor.

It is known that the deposited particles can be resuspended by human activities such as walking.[59–61] Meanwhile, the resuspended particle number concentrations tend to be higher for smaller particles than larger particles.[18,62] While the AeroTrak OPC could not measure particles smaller than 0.3 μm, the particle number concentrations seem to increase with decreasing particle diameter, especially when particles smaller than 0.75 μm in diameter are considered. We can speculate that if this trend continues, there is a high likelihood that nanosized particles were also resuspended during all experiments. Although the number concentration of resuspended particles increases with decreasing particle size, the resuspension rate for those smaller particles is likely to decrease due to increased particle adhesion forces, which tend to keep particles attached to the floor surface.[16,63–65] This aspect of particle resuspension will be examined in a separate paper. The tested sprays are relatively new and studying the resuspension of the particles from these products could help understand the behavior of these particles and provide the information needed to control exposure to such particles.

3.4.2 | The resuspension of particles from all products from the carpet

In addition to S10, S13, Z2, and Z4, three other products (e.g., Cu1, S14, and Z7) were also resuspended from the carpet, and the resulting size distributions for all seven products are shown in Figure 7. The number concentrations at 0.35 μm ranged from 10^6 to 10^7 particles/m³, and then the concentrations decreased by approximately two orders of magnitude as particle size increased from 0.35 to 15 μm. Z7 produced the lowest resuspended particle

TABLE 3 Tukey's HSD test *p*-values for the sprayed mass concentrations.

	S10	S13	Z2	Z4	Cu1	S14	Z7
S10	–						
S13	0.001*	–					
Z2	0.001*	1.000	–				
Z4	0.004*	0.988	1.000	–			
Cu1	1.000	0.063	0.103	0.169	–		
S14	1.000	0.029*	0.049*	0.086	1.000	–	
Z7	0.000*	0.000*	0.000*	0.000*	0.000*	0.000*	–

*Statistically significant at 0.05 level.

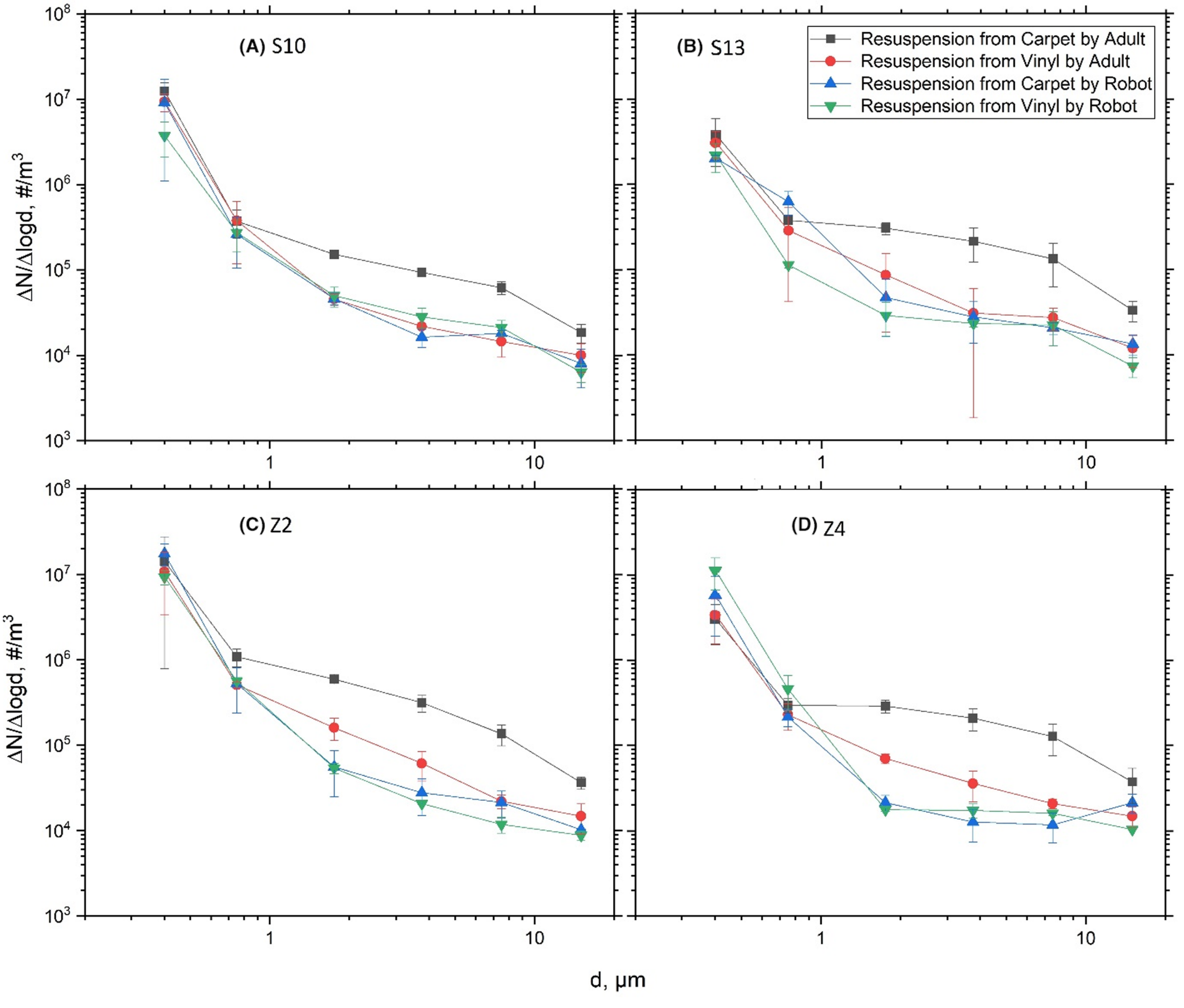

FIGURE 6 Particle size distributions during resuspension as measured by Aerotrak OPC at 1.1 m height. The data are averages of 2 repeats, and error bars represent standard deviations. (A) S10, (B) S13, (C) Z2, (D) Z4. *x*-axis and *y*-axis are in log scale.

number concentrations for all size ranges, and Z2 produced the highest resuspended particles for most size ranges. The size distributions produced by Cu1 and S14 were similar to those of S10, S13, and Z4.

3.5 | Resuspended particle mass concentrations

The airborne mass concentrations of resuspended particles from all products are shown in Figure 8. In general, the highest mass concentrations were resuspended from the carpet by the adult, and the lowest mass concentrations were resuspended from vinyl flooring by the robot. Additionally, adult resuspended higher particle mass concentrations than the robot for the same flooring type. Here, the mass concentrations were measured by a Button sampler with the 25 mm PTFE filter at 1.1 m height.

The resuspended mass concentrations for products S10, S13, Z2, and Z4 were analyzed using three-way ANOVA with the product, flooring material, and resuspension force as independent variables. The *p*-values of the variables and their interactions are shown in Table 4, while the *p*-values for each pair-wise comparison are shown in Table S2.

Overall, statistically significant differences could be found for each independent variable (p-value < 0.05). For example, the resuspended mass concentration from the carpet was statistically significantly higher than from vinyl flooring, and the mass concentration resuspended by an adult was statistically significantly higher than that resuspended by the robot; and Z2 and Z4 showed statistically significantly higher resuspended mass concentration than S10 and S13 (Table S2A). In addition to the 4 products mentioned above, Cu1, S14, and Z7 were also resuspended from the carpet by the adult. According to a one-way ANOVA, there was a statistically significant difference among these three products (p-value < 0.05; Table 4).

Additionally, the interaction of two variables and all three variables (i.e., flooring material, resuspension force, product) showed statistically significant effects on the resuspended mass concentrations. For example, a statistically significant difference was found in the interaction of flooring material and resuspension force (Table 4). However, as shown in Table S2B, this difference did not appear for all groups; the particle mass concentration resuspended from the carpet by the adult was statistically significantly higher than those resuspended from vinyl flooring by the adult, from the carpet by the robot, and from vinyl flooring by the robot. In contrast, no statistically significant difference was found among the latter three groups. Similar results were found with the interaction of flooring material and product, where a statistically significant difference was observed. For example, S13, resuspended from vinyl flooring, showed a statistically significantly lower mass concentration than Z2 and Z4, resuspended from carpet (Table S2C). The interaction of resuspension force and product also showed statistically significant effects on resuspended mass concentration, and more details are shown in Table S2D.

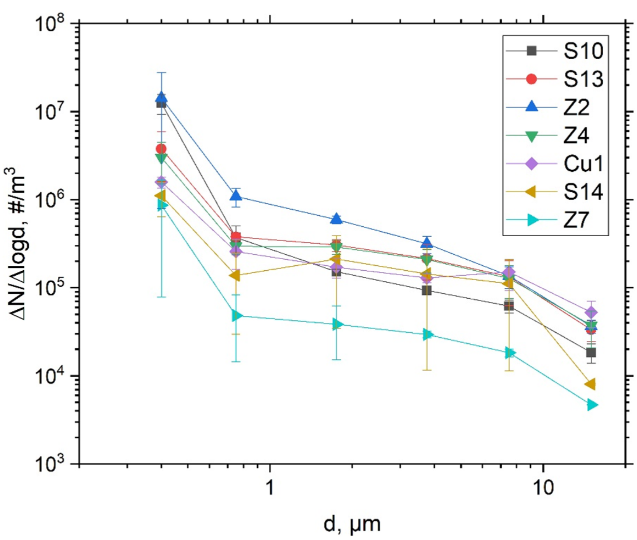

FIGURE 7 Size distributions for particles resuspended from carpet by an adult as measured by Aerotrak OPC at 1.1 m height. The data are averages of 2 repeats, and error bars represent standard deviations. *x*-axis and *y*-axis are in log scale.

3.6 | Particle resuspension rates

3.6.1 | Effect of product types on resuspension rates

We calculated resuspension rates of products for different resuspension scenarios using Equations (1–4) and the results are presented in Figure 9. The resuspension rates of S13, Z2, Z4, and Z7 were similar and ranged from 10^{-4} to 10^{-3} h^{-1}, while the resuspension rates of S10, Cu1, and S14 were higher and ranged from 10^{-1} to 1 h^{-1}. In fact, the resuspension rates for S10, Cu1, and S14 were 1–3 orders of magnitude higher than those for other products. This fact requires further investigation.

The effect of product type is easily observed when comparing the resuspension rates for the same flooring material and resuspension force. For example, when an adult resuspended particles from the carpet, Z7 showed the lowest resuspension rate (e.g., 1.19×10^{-4} h^{-1}) in our experiments. The low resuspension rate is consistent with FESEM images of the filter covered by emulsion from Z7 (Figure 8): the emulsion strongly adheres to the filter surface, and it could have covered individual particles, thus making it hard to resuspend the particles.

When the resuspension rate data of the products S10, S13, Z2, and Z4 were analyzed statistically by the ANOVA test (Table 5), statistically significant differences were observed among the products. *p*-values for the post hoc Tukey's HSD test are shown in Table S3; here, S10 produced a statistically significantly higher resuspension rate than S13, Z2, and Z4, while the latter three showed no statistically significant difference in resuspension rate (Table S3A).

In addition to the 4 products mentioned above, Cu1, S14 and Z7 were also resuspended from the carpet by a walking adult; thus, a one-way ANOVA test was used to compare the resuspension rate of all 7 products for this resuspension scenario, and a statistically significant difference was found among products ($p < 0.05$; details in Table 5).

The observed difference in resuspension rates among products could be explained by different properties of particles in the products[13] and differences in product matrix properties. For example, high particle roughness results in higher resuspended particle concentrations.[66] For different particle and surface types, the thermodynamic work of adhesion is different, thus, different

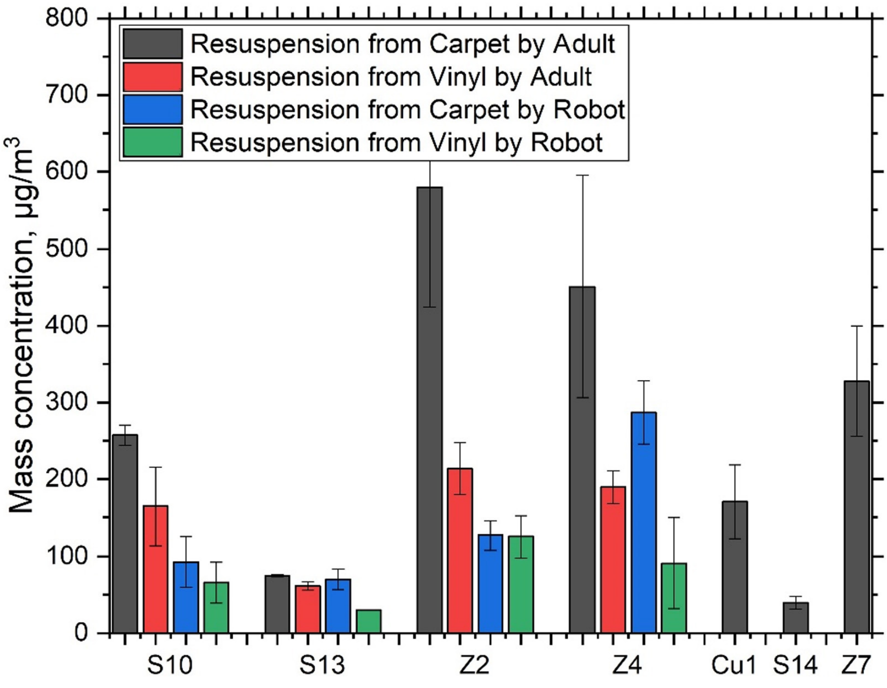

FIGURE 8 Mass concentrations from S10, S13, Z2, and Z4 during resuspension and measured at 1.1 m height by Button samplers (SKC, Inc) with 25 mm PTFE filters. The data are averages of 2 repeats, and error bars represent standard deviations.

TABLE 4 *p*-values from the ANOVA test for the resuspended mass concentrations

Variables	Three-way ANOVA *p*-value for S10, S13, Z2, and Z4	One-way ANOVA *p*-value for the resuspension from carpet by adult
Flooring material	0.000*	
Resuspension force	0.000*	
Product	0.000*	0.004*
Flooring material × Resuspension force	0.017*	
Flooring material × Product	0.013*	
Resuspension force × Product	0.008*	
Flooring material × Resuspension force × Product	0.032*	

Note: Three-way ANOVA test was applied to S10, S13, Z2, and Z4, and one-way ANOVA test was applied to particle resuspension from carpet by an adult for S10, S13, Z2, Z4, Cu1, S14 and Z7.

*Statistically significant at 0.05 level.

particles need to be provided a different amount of energy to be resuspended from the floor surfaces.[63] Also, as was illustrated in micrographs, the product matrix plays an important role. Therefore, depending on a product, it might be the matrix (e.g., additives) and not the particle type (e.g., metal) that governs the resuspension.

3.6.2 | Effect of flooring material on resuspension rates

According to the ANOVA test (Table 5), statistically significant differences were observed for resuspension from different flooring materials. Here, the resuspension rate from the carpet was statistically significantly higher than the resuspension rate from vinyl flooring.

Previous studies have shown that the flooring material affects the resuspension of particles,[10,13,15,18,41] including our earlier studies examining the resuspension of dust particles in participants' homes.[48,67] Our study examined particles from nano-enabled consumer sprays and came to the same conclusion: particle resuspension rates were statistically significantly higher for carpet than vinyl flooring. Since the particles were sprayed and then resuspended after 24 h, without any other activities on the flooring materials in between, the deposited particles should be located closer to the surface of the carpet fibers instead of being embedded deep into the carpet fibers. In addition, human walking can create air flow and vibration,[15] and particles on the carpet surface fibers could be subjected to a higher velocity than particles on smooth materials such as vinyl flooring, leading to a higher resuspension rate from carpet.[68] Moreover, the carpet piles act like springs, and once the experimenter's foot is lifted from the carpet, their potential energy is converted into kinetic energy propelling the deposited particles upward, thus promoting the resuspension from the carpet.[42,68] The difference between carpet and vinyl flooring may also be due to their differences

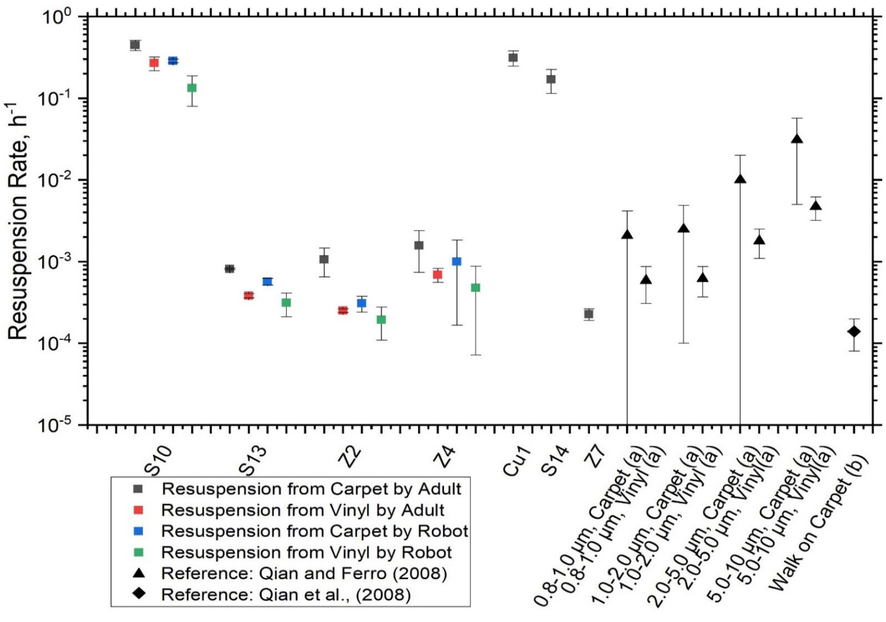

FIGURE 9 Resuspension rates of all seven products investigated in this study. The resuspension rates are the averages of 2 repeats. Error bars represent standard deviations. Y-axis is in log scale. Data from references (a) Qian and Ferro,[18] (b) Qian et al.[12]

TABLE 5 *p*-values from ANOVA test for particle resuspension rates.

Variables	Three-way ANOVA *p*-value for S10, S13, Z2, and Z4	One-way ANOVA *p*-value for the resuspension from carpet by adult
Flooring material	0.005*	
Resuspension force	0.010*	
Product	0.000*	0.000*
Flooring material * Resuspension force	0.784	
Flooring material * Product	0.000*	
Resuspension force * Product	0.001*	
Flooring material * Resuspension force * Product	0.976	

Note: Three-way ANOVA test was applied to S10, S13, Z2, and Z4, and one-way ANOVA test was applied to particle resuspension from carpet by an adult for S10, S13, Z2, Z4, Cu1, S14 and Z7.

*Statistically significant at 0.05 level.

in composition, which affects their static charge resulting in resuspension rates.[42]

Since we used only one type of carpet and vinyl flooring, further studies are needed to examine how different types of the same flooring material may affect the resuspension of deposited particles from nano-enabled sprays.

3.6.3 | Effect of resuspension forces on resuspension rates

As shown in Table 5, statistically significant differences were observed for different resuspension forces ($p < 0.05$), for example, adult vs. robot. Our study found that a walking adult caused statistically significantly higher resuspension rates than a robotic sampler that simulated a child. This is due to different moving strategies and the higher body weight of the adult experimenter compared with the robot, which resulted in higher resuspension force. In a typical walk by an adult, a tip vortex is developed at the edge of the shoe, and the flow from under the shoe is squeezed and ejected as a wall jet in front of and from both sides of the shoe.[63] Two counterrotating vortices are formed as the shoe touches the floor; they translate horizontally on the ground and slowly decay.[63] When the shoe moves upward, a strong suction flow is generated between the sole and flooring, leading to the transport of generated vortices.[62] These vortices are considered to be the main mechanism to resuspend particles into the environment. The resuspension process is affected by airflow characteristics around the moving person, especially by the wakes created by the person's movement.[13] However, when it comes to the robotic sampler's movement (e.g., rolling), it cannot provide as much flow and vibration as the adult, thus leading to lower resuspension rates.

Also, a child weighing more than 3.7 kg of the robot's weight is likely to cause a greater particle resuspension rate than the robot. In addition, a child's breathing zone is closer to the floor than the used sampling height of 1.1 m, and higher resuspended particle concentrations might be observed in the child's breathing zone than we measured in this study. For example, studies with robotic (PIPER) samplers examining exposures of children between the ages of 6 months to 3 years investigated breathing zones between 0.2 and 1.0 m.[47–50] Personal exposures to resuspended particles, including young children's exposures, will be examined in a separate paper.

3.6.4 | Interaction of different variables affected resuspension rates

For each product, the resuspension from the carpet by an adult showed the highest resuspension rate, while the resuspension from vinyl flooring by a robot showed the lowest resuspension rate. Meanwhile, the interaction of different variables played a role in the resuspension rate (Table 5); the post hoc Tukey's HSD test *p*-values of all variable pairs are shown in Table S3. Here, the interaction of flooring material and product and the interaction of resuspension force and the product showed a statistically significant effect on the resuspension rate. On the contrary, neither the interaction of flooring material and resuspension force nor the interaction of all three variables showed a statistically significant effect on resuspension rate.

When considering the interaction of different variables, it is important to mention that the statistically significant difference was only observed among some pairs. For example, the interaction of flooring material and product showed that the resuspension rate of S10 resuspended from the carpet was statistically significantly higher than from vinyl flooring, and they were both statistically significantly higher than other groups. In contrast, the other groups showed similar results (Table S3B). A similar result was observed for the interaction of resuspension force and product, where the resuspension rate of S10 resuspended by the adult was statistically significantly higher than by the robot, and they were both statistically significantly higher than other groups (Table S3C).

3.6.5 | The comparison with resuspension rates observed in other studies

Although nanosized particles were observed in our study based on image analysis, they likely did not dominate the mass concentration. Thus, the resuspension rates calculated based on particle mass concentration could be compared with other studies that did not consider nanoparticles. In Figure 9, along with our study's resuspension rates, we included particle resuspension rates reported in the literature.[12,18] The resuspension rates of Z2, Z4, and S13 were lower than the resuspension rates of 2.0–5.0 and 5.0–10.0 μm particles reported by Qian and Ferro[18] and closer to the resuspension rates of 0.8–1.0 and 1.0–2.0 μm particles reported by the same study. Since we sampled all particles without differentiating them by size, the calculated resuspension rate likely reflects the presence and contribution of smaller particles in our samples. On the contrary, the resuspension rate of Z2, Z4, and S13 products measured due to an adult walking on the carpet was higher than the resuspension due to walking on the carpet, as reported by Qian et al.[12]

The differences in particle properties and product matrix (e.g., solvent) could partly explain the difference between the resuspension rate in our study and other studies shown in Figure 9. We tested particles from nano-enabled consumer products while Qian and Ferro[18] used Arizona Road Dust, and Qian et al.[12] tested residential dust. Additionally, the difference in resuspension rate could also be explained by the difference in ventilation schemes, resuspension durations, human activities, and sampling height.

Our experiments were performed without active ventilation. However, passive ventilation was likely present in our chamber, as in any building. This is typical in residential homes during shoulder seasons when central or window HVAC is not used, but ventilation is achieved through passive means. If active ventilation had been used, it would have diluted the resuspended particles, thus decreasing the resuspension rate. On the contrary, an active airflow would have decreased the airborne particle concentration during spraying, thus decreasing the floor loading. Furthermore, if the incoming airflow is not sufficiently filtered, any particles delivered to the chamber by the active flow would have to be considered when calculating the resuspension rate.

Two studies referenced in our paper were performed under different ventilation strategies (e.g., sidewall supply ventilation and window ventilation). As Qian and Ferro[18] mentioned, the resuspension rates for the sidewall supply system are likely underestimated, and results could be different across ventilation schemes; however, the extent of the effect of different ventilation schemes was not clear. Human activity is another factor that affects the resuspension of particles. In our study, particles were resuspended by the walking adult or moving robot, while in the other two studies, particles were resuspended by multiple experimenters and activities (e.g., walking and sitting). Differences in walking style, the weight of the experimenters, and their shoe size and type can lead to a difference in resuspension results. In addition, the duration of the resuspension experiment may also lead to differences in resuspension rates. Our study resuspended particles for 11 min, while the other two studies resuspended them for 30 min. Moreover, the sampling height can also affect the resuspension rate. Our samples were collected at 1.1 m, the EPA recommended height for indoor air investigations,[56] while the other two studies chose 1.5 m to represent the entire room. The effect of sampling height on the resuspension rate will be addressed in a separate manuscript.

3.7 | Study limitations

3.7.1 | Uncertainties due to the experimental setup

We recognize that our study also has some limitations. First, the determination of particle background level was a challenge. Particles present inside the chamber before experiments and particles penetrating from outdoors during experiments could affect the results.

We did our best to seal the chamber, remove particles from the flooring material before experiments, and reduce the background particle concentration using an air purifier. Although some particles were left in the chamber and some particles penetrating from outdoors, leading to fluctuations in particle background level during the spray and resuspension experiments, the mass concentration of such particles was low and did not have a material effect on the resuspension rate. Plus, the background concentration was removed from our calculations.

Second, there was likely some loss of particles to the chamber walls due to electrostatic effects during spraying and resuspension, but the extent of the loss is difficult to quantify. To minimize the static effect and potential particle loss, the inner walls of the chamber were covered with aluminum foil. In addition, the experimenter wore low-lint protective clothing to minimize particle shedding. However, some resuspended particles could have attached to the clothing due to diffusion or electrostatic effects. The loss of resuspended particles would have lowered the resuspended particle concentration and the resulting resuspension rate.[42]

Third, the wall-boundary flow is known to affect particle resuspension. In wall-bounded turbulent flows, the characteristic timescale of turbulent structures decreases progressively as the flow structures lie closer to the wall.[69] Lighter particles experience wall turbulence dynamics more than heavier ones. They tend to have greater particle concentration than heavier particles in the near-wall region. In contrast, heavier particles have a larger time scale and filter out the effects of the smaller fluid scales.[70] Particles that have acquired enough momentum may coast through the accumulation region and deposit by impaction directly at the wall; otherwise, particles can deposit under the action of turbulent fluctuations.[71,72] Such particles are bound to remain in the viscous wall layer and accumulate in near-wall regions.[69] However, since the resuspension rates in our experiment were measured immediately after 11 min of walking in the center of the chamber, any particles accumulated near walls should not have had a substantial effect on the resuspension rate.

3.7.2 | Measurement uncertainties

The calculation of resuspension rates is based on weighing the material captured on filters; here, the low solid fractions of S10, S14, and Cu1 resulted in low filter weight, which increased measurement uncertainty. However, we used several different internal weight controls (e.g., standard weights and blank filters in the weighing room) to ensure accurate weighing results.

Due to the time constraints and limited availability of resources, only two repeats of each experiment were performed. However, even with this limited number of repeats, the variability among the repeats is not excessive and similar to or lower than in similar studies, as shown in Figure 8. Because of multiple investigated variables, a total of 38 spraying/resuspension experiments were performed. The data demonstrate the statistically significant effect of product, surface, and experimenter on the particle resuspension rate.

3.7.3 | Uncertainties due to the fluctuation in relative humidity

Variability in relative humidity (RH) could have affected particle resuspension. The RH in our study was between 35% and 55%, a common range for other resuspension studies.[18,45] However, You and Wan[68] suggested that a 20% change in RH could have a strong effect on the resuspension rate. They found that the resuspension rates of Arizona Road Dust at 41% RH were three times higher than that at 63% RH for both carpet and vinyl flooring. On the contrary, Rosati et al.[19] showed that the resuspension of particles was not significantly affected by RH for a new carpet when they changed the RH from 40% to 80%. Qian et al.[13] hypothesized that when the RH increases, the capillary force increases, but the electrostatic force is reduced for the resuspension from the new carpet, thus resulting in little change in the total adhesion force. Since we did not examine the effect of RH in our study and the observed effect of RH on particle resuspension is not entirely clear from other studies, we can only assume that the RH range between 35% and 55% should not have substantially affected particle resuspension.

4 | CONCLUSION

This project focused on the floor deposition of particles released from nano-enabled consumer sprays, followed by the examination of the resuspension of these particles as a function of the product, flooring material (e.g., carpet and vinyl), and resuspension force (e.g., adult walking and robot moving). All these variables showed statistically significant effects on the resuspension rate. The use of carpet resulted in statistically significantly higher resuspension rates than vinyl flooring, and an adult walking on surfaces showed statistically higher resuspension rates than a robot. The resuspension rates depended on the product. Meanwhile, the interactions of flooring material and product and resuspension force and product also affected the particle resuspension rates.

Our study helps fill knowledge gaps regarding the resuspension of particles from nano-enabled consumer sprays. This information will help understand and minimize exposures to particles from consumer sprays. The next step in our investigation is to focus on the size distribution of nanosized particles from those sprays and identify the resuspension behavior of the nanoparticles.

AUTHOR CONTRIBUTIONS

Ruikang He contributed to conceptualization, data curation, formal analysis, investigation, methodology, original draft, review, and editing. Jie Zhang contributed to the methodology and experiments. Gediminas Mainelis contributed to conceptualization, formal analysis, funding acquisition, methodology, project administration, resources, supervision, review, and editing. All authors have approved the manuscript.

ACKNOWLEDGMENT

This study was funded by Consumer Product Safety Commission CPSC-S-16-0057 and USDA-NIFA Hatch Multistate project 1008687 through NJAES project NJ07215 at Rutgers, the State University of New Jersey. The opinions expressed in this paper are those of the authors and do not necessarily represent the views of the Commission, USDA, or Rutgers University.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

How to cite this article: He R, Zhang J, Mainelis G. Resuspension of particles deposited by nano-enabled consumer sprays: The role of product type, flooring material, and resuspension force. *Indoor Air*. 2022;32:e13157. doi: 10.1111/ina.13157