A mini-Review on Transportation and Fate of Silver Nanoparticles Released from Consumer Products: Ecological Risk Assessments

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A mini-Review on Transportation and Fate of Silver Nanoparticles Released from Consumer Products: Ecological Risk Assessments

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Abstract: *Over the last few decades, engineering nanomaterials (ENMs) have been incorporated into a variety of commercially-available consumer products (CPs). Silver Nanoparticles (AgNP) are the most used ENMs in CPs, mainly due to their antibacterial properties. To better understand the fate, transport, and environmental impacts of AgNPs, many studies have been conducted. However, this research has been focused mainly on pristine AgNPs either lab-synthesized or purchased commercially. While these studies have contributed greatly to a better understanding of AgNP toxicology, they have not addressed the environmental concerns associated with AgNP release from CPs. This review assesses previously published work on routes of environmental exposure, characterization techniques, potential ecological risks of AgNPs, and the identification of available toxicity assays for AgNPs in CPs. A major objective of this review is to summarize and identify key issues, knowledge gaps, and potential directions for future research related to AgNPs released from CPs.*

Keywords: Environmental fate of nanomaterials; Aggregation; Dissolution; Silver nanoparticles toxicology

1. INTRODUCTION

Silver is well-known for its antimicrobial activity [1]. Silver Nanoparticles (AgNPs) are fine particles of metallic silver with a size less than 100 nm in at least one dimension. AgNPs may exhibit increased antimicrobial activity compared to bulk-Ag due to their large surface area [2]. Therefore, the applications and exploitations of AgNPs have witnessed a dramatic increase over the past decades. According to the nanotechnology consumer product inventory (CPI), AgNPs are currently the most widely used nanoparticle [3]. These products include textiles, personal care products, medical equipment, food packages, cook wares, antimicrobial agents, toys, baby products, paints, inks, air and water filters, vacuum cleaners, humidifiers, washing machines, electronics, refrigerators, dietary supplements and medicines [3-5]. AgNP-containing consumer products (CPs) have received considerable attention, but their potential health and ecological risks have yet to be fully understood. To date, extensive research has been conducted on the synthesis and characterization of AgNPs [6-8], their physicochemical behaviors under different conditions [9-13] and their toxicity to specific microorganisms [14-17]. The wide variety of applications has led to a wide variety of usage scenarios that also impact the characteristics of AgNPs. For instance, AgNPs used in textiles may be transformed into different Ag-species due to the interactions with the laundry detergents [18, 19]. These observations highlight the importance of investigating the use of AgNP-containing CPs in environmental exposure and health risk assessments. Only a few of the commercially-available AgNP-containing CPs have been evaluated. Therefore, the main objectives of this review are to summarize the major findings of past research related to AgNPs released from CPs, to identify key issues and knowledge gaps and to provide potential directions for future research.

2. CHARACTERIZATION OF AgNPs IN CPs

2.1 Characterization Techniques and Sample Collection

2.1.1 Solid Materials

There are two types of solids containing AgNP: Solid AgNP-containing CPs, and sediments, soils, biosolids in which AgNPs released from CPs may have accumulated. Physical characterization of AgNPs in solid CPs is considerably easier than that of environmental samples, which may require sample separation techniques. Samples assayed for Ag content are usually digested prior to analysis with inductively coupled plasma mass spectrometry (ICP-MS). The most common acid digestion procedures use HNO_3 [20], a HNO_3-HCl mixture or a HNO_3-H_2O_2 mixture [21-23]. An X-ray Fluorescence (XRF) analysis can be used to determine the Ag content when acid digestion does not result in complete digestion. However, sample pre-treatment is required for this analysis as well. As reported by Lorenz et al., textile samples were incinerated at 600°C in a muffle furnace prior to creating the pellets for XRF analysis [22]. Electron Microscopy techniques such as Transmission Electron Microscopy (TEM) and Scanning Electron Microscopy (SEM) are commonly used to characterize AgNPs. In these methods, a representative portion of the sample is paced on a TEM/SEM grid for imaging. This technique is effective for observing the AgNPs on the surface of the material but is not applicable for identification and characterization of AgNPs embedded in CPs. For particles embedded in solid matrices, thermal ashing has been used to break down the solid matrix, allowing more precise identification of AgNPs [24-26]. Although morphological and physicochemical changes may occur during the ashing procedure, subsequent measurements of size, shape, and aggregations may not be representative of the AgNPs present in the original CPs [26]. In comparison to other speciation analysis techniques such as X-ray Photoelectron Spectroscopy (XPS) and X-ray Diffraction (XRD), X-ray Absorption Spectroscopy (XAS) is the most sensitive and sophisticated method for the identification of AgNPs speciation in CPs or environmental relevant matrices [27]. The use of XAS for AgNP characterization in solid matrices has been reported in several studies. However, some difficulties have been reported in differentiation of the Ag species of interest due to similarities in X-ray near edge spectroscopy (XANES) [18, 27, 28].

2.1.2 Aqueous Materials

In aqueous phase materials, AgNPs are separated and characterized using a variety of instruments and techniques. These techniques have been used to characterize AgNPs in CPs such as disinfectant sprays, antimicrobial solutions, cosmetic products (shampoo, deodorants, and antiperspirants), inks, and paints. Dynamic light scattering (DLS) is commonly used to measure particle size distributions (PSD) of AgNPs in liquid matrices [29-31]. Various fractionation strategies are commonly used to separate AgNPs from liquid matrices based on their size, including field flow fractionation (FFF), sedimentation flow fractionation (SdFFF), and centrifugal-FFF. In addition to separation techniques, other analytical tools could be used to characterize AgNPs (e. g., ICP-MS, UV-visible light spectrometers (UV-vis), TEM, SEM, and atomic force microscopy (AFM)). Cascio et al., characterized AgNPs in commercial liquids by coupling FFF and UV-vis analyses (FFF-UV-vis) [29, 32]. They noted that sample heterogeneity in terms of size and shape may alter the surface plasmon resonance (SPR) band position resulting in inaccurate results. It is only reliable for identifying AgNPs in liquids with known background components since the UV-vis spectra are affected by the presence of various constituents in the AgNP suspension. AgNPs have been separated from environmental water samples using basic filtering techniques [20, 23, 24, 34]. Since most of these techniques use forced separation methods to separate AgNPs, further characterization of the separated AgNPs may not provide a true representation of the original suspension. Cloud point extraction (CPE) using Triton X-114 (TX-114) is another method used to separate AgNPs from antimicrobial solutions [34]. Since AgNPs are concentrated in the Triton X-114 phase, this method separates AgNPs from the aqueous matrix without altering their size or shape. As soon as the AgNPs in the Triton phase are digested with microwaves, they are characterized with ICP-MS, TEM/energy-dispersive X-ray spectroscopy (EDX) or UV absorption. Aqueous solutions contain all forms of Ag^+, including free dissolved Ag^+, Ag^+ adsorbed on AgNPs, and Ag^+ associated with matrix. An analysis of photon correlation spectroscopy can be used to measure the hydrodynamic particle size distribution of any nanoparticle suspended in a liquid medium with a different refractive index. Measurements have been made of the hydrodynamic particle size distribution of AgNPs in commercial spray solutions by combining a zetaPLS 90 with an induced particle sizing software [35, 36]. Stuart et al., were able to detect AgNPs in colloidal silver sprays by using an electrochemical technique known as particle-impact voltammetry. This technique measures and applies the oxidation potential of AgNPs for direct detection and size measurements [37].

2.1.3 Aerosols

The detection and characterization of AgNPs in aerosols require special techniques and instruments. The most practical approach for studying AgNPs in aerosols is by conducting experiments inside of a glove box. The primary method of collecting aerosol samples for characterization is through single-stage impactor filters. Particles with a size up to 30 nm can be fractionated using an electrical low-pressure impactor (ELPI). Particles can also be precipitated electrostatically based on their electrical mobility. It may be more appropriate, however, to use an online technique like SMPS (scanning mobility particle sizer) to obtain sufficient samples for subsequent analyses. This is especially true for short-term applications such as spraying mechanisms. A differential mobility analyzer (DMA) followed by a condensation particle counter (CPC) has been used to investigate the PSDs and number counts of released AgNPs from aerosols. In this method, particles are separated based on their electrical mobility, followed by a particle count of each fraction associated with SMPS [22, 31, 37, 38, 39]. To perform multiple analyses simultaneously, several instruments were connected to a glove box. In the first step, the aerosol was dried with a thermo-desorber before being analyzed with SMPS and electrostatic samplers simultaneously. To assess AgNP morphologically using TEM/SEM and to analyze their composition using EDX, TEM grids were placed on electrostatic samplers. Different types of AgNP-containing sprays were used to characterize an aerosol containing AgNPs (anti-odor spray for hunters, disinfectant spray, and throat spray) [33]. In this experiment, SMPS and CPC were used in addition to optical particle counters (OPCs) and diffusion chargers (DCs) for measuring AgNP concentrations and surface areas. Losert et al., have considered this issue and suggested a SMPS-ICPMS coupling technique to analyze both PSD and physicochemical characteristics of the released NPs simultaneously. As a result of this technique, AgNPs released from commercial spray solutions have been successfully analyzed [31, 40].

3. PAST RESEARCH ON AgNP-CPs

3.1 Solid Phase

Many solid-phase CPs contain AgNPs, however, many of the past studies have focused on textiles such as antibacterial socks, T-shirts, trousers, bed sheets, medical clothes, medical masks, and wound dressings. Previous studies have used textiles of different brands, fiber compositions (100% cotton, 100% polypropylene, and combinations of polyamide, elastane, and wool), origins (Germany, Japan, Switzerland, USA), Ag species (Ag^+, Ag(0), AgCl), concentrations (1.5 mg/kg - 2925 mg/kg), and usage scenarios [20-28]. Textiles and products used for medical purposes typically contained more Ag than ordinary products [41, 42]. Washing machines and humidifiers are the only items of household equipment that contain AgNP (humidifiers, vacuum cleaners, hair dryers, washers, water and air filters, and refrigerators). By releasing AgNPs into the washing solution, these washing machines provide antibacterial capabilities. Farkas et al., reported the presence of AgNPs in washing machine effluent. Additionally, the effluent showed strong bactericidal effects on the natural bacterial community [43]. The use of AgNP-emitting humidifiers for air sanitation is like that of washing machines. The disposal of humidifiers in landfills, however, may result in both aerosol and liquid contamination [20, 41]. Further, food storage containers with embedded AgNPs are widely available and are of high quality. Due to their direct exposure to human food, they are concerned. A number of studies have been conducted on commercially-available AgNPs containing food storage containers. In these studies, AgNP release and incorporation into food were influenced by temperature, storage time, and food chemistry. Other studies assessing the fate of AgNPs released from toothpaste and shampoo have been reported [20]. The toothpaste and the shampoo used in that study were claimed to contain colloidal Ag as well or a mixture of colloidal and Ag^+, respectively. The major objective of the study was to characterize the Ag particles in the product before and after being released to the environment through washing with tap water. However, this experiment does not simulate the real usage of these products. For instance, the influences of water quality and interactions with saliva (in the case of toothpaste) on the AgNPs were not considered.

3.2 Liquid/Aerosol phase

The most broadly studied and reported AgNP-containing CPs are antibacterial solutions. These antibacterial solutions are primarily applied by spraying, producing AgNP aerosols. It is challenging and therefore limited to conduct experiments on AgNPs released in aerosols due to difficulties in collecting samples. Even so, several studies have characterized AgNP aerosols released by antimicrobial sprays [22, 31-33, 36, 38, 39]. Two studies confirmed that commercial sprays released both individual and aggregated AgNPs [36, 39]. Various factors have been found to affect the characteristics of the Ag-NPs released from the bottle, including both the characteristics of the solution and the spraying mechanism [33, 36]. In implementing rules and regulations for AgNP-containing CPs in the aerosol phase, dispersion composition, storage mechanisms, dispensing mechanisms, and nozzle specifications must be considered. Research conducted by Benn et al., used two humidifiers made of plastic containing AgNPs to evaluate the fate of released AgNPs [20]. Although the product claimed to contain AgNPs, SEM/EDX analysis could not confirm the presence of AgNPs in the humidifier due to the instability of the plastic sample. Despite finding AgNPs in the water vapors released from humidifiers, their concentrations were too low (1.1 ± 0.4 µg Ag L^{-1} at 100 mL/hr flow rate and 0.19 µg Ag L^{-1} at 420 mL/hr flow rate) to allow adequate characterization. Quadros et al., simulated the use of a humidifier and a disinfectant spray in a furnished room with carpeted floors, painted walls, a door, and a window [41]. As a result of their study, the condensed vapor released from humidifiers contained an Ag concentration of (2.3 ± 0.7 ppb). Most of the previous research reported on AgNP-CPs are summarized in Table 1.

4. POTENTIAL ECOLOGICAL OF AgNPs

4.1 AgNPs Transformations in Aquatic Environments

All bodies of surface water may potentially be contaminated by AgNPs released from CPs. For instance, textiles contain high concentrations of AgNPs which have demonstrated to be released into water during washing. Humidifiers, detergents, paints, inks, washing machines and water filters can directly release AgNPs into water sources. Furthermore, the quality of water plays an important role in the potential transformations (size, shape, and speciation) of AgNPs; a number of studies have reported such transformations. According to research conducted on environmental exposure of AgNPs from CPs, the following possible scenarios have been investigated:

4.1.1. Usage and Disposal Scenarios

4.1.1.1. Release into water from washing of textiles

Textiles incorporated with AgNPs can release them into the water during washing [21, 22, 24, 27, 34]. Simulation of washing conditions was used to assess the impact of water quality (tap water versus ultrapure water) on AgNP release from socks. According to the source material, Ag releases contained both Ag^+ and AgNPs [24]. Compared to tap water, ultrapure water releases more Ag due to its corrosive nature. A similar study on socks containing AgNP found that AgCl was formed when hypochlorite (bleach)/detergent solutions were applied [18]. Since the detergent used in this study contained high hypochlorite concentrations, it was not representative of typical laundry detergents. Although the results demonstrate that oxidizing and chlorinating the fabric will convert a significant fraction of the AgNPs to AgCl. Geranio et al., conducted a detailed washing experiment using different types of AgNP-containing textiles in 2009 [21]. During that experiment, pH, surfactants, and oxidizing agents were tested on AgNPs; Ag^+ release was higher at pH 7 than pH 10. The presence of bleach accelerated the release of Ag^+ following the chemical reaction given.

$$Ag\ (s) + H_2O_2 \longrightarrow Ag^+\ (aq) + OH^{\cdot} + OH^-\ (aq) \quad (1)$$

The produced hydroxyl radicals promote further oxidation of metallic Ag and Ag^+ release.

$$Ag\ (s) + OH^{\cdot} \longrightarrow Ag^+\ (aq) + OH^-\ (aq) \quad (2)$$

Mechanical abrasion also affected Ag^+ release during washing in this experiment. After washing with a washing machine, most textiles released coarse AgNPs (>450 nm). Under wastewater treatment conditions, AgNP aggregates, precipitates, and textile fibers embedded with AgNP may exhibit significantly different behaviors. According to Lorenz et al., textiles with different compositions and types of AgNPs released different forms of AgNPs (Ag, AgCl, Ag nanowires) [22]. Besides aggregated AgNPs, nano-AgCl and nano Ag_2S may also be released from textiles under wastewater treatment conditions, which further emphasizes the importance of studying the fate of different forms of AgNPs. In addition to releasing AgNPs from textiles, washing procedures also altered the Ag speciation remaining on the textiles. It is important to take this phenomenon into consideration, as AgNPs may completely change after prolonged use in a CPs. As a result of these changes, the antibacterial properties of the products may be affected, resulting in unknown environmental effects. Using XAS analysis, Lombi et al., studied the transformation of Ag species in textiles before and after washing [27]. This experiment investigated textiles products containing different species of Ag and AgNPs (metallic AgNPs, AgCl-NPs, Ag_2S-NPs, Ag-oxides, Ag-phosphates, Ag-sulfates, Ag-zeolites). After washing the textiles, more stable Ag species were formed, including AgCl and Ag_2S. AgNP concentrations depend greatly on how they are associated/integrated with fabric. During initial washing cycles, Quadros et al., reported that AgNPs embedded on textile surfaces do not release further once they are depleted [41]. Long-term washing of textiles should reduce their antibacterial properties; however, this reduction has not been experimentally proven.

The impact of several factors on AgNP transformations were assessed, primarily focused on the production/manufacturing phase. The potential transformations that may occur during the usage and disposal scenarios were not accounted for in the life-cycle assessment.

4.1.1.2. CP utilization – release into water from products other than textiles

Although many AgNP-CPs are not textiles, few studies have been conducted on their fate. One study used 13 different AgNP-containing baby products, including plush toys, baby blankets, sippy cups, baby wipes, breast milk storage bags, humidifiers, and disinfecting sprays [41]. They simulated realistic usage scenarios by analyzing the products in a leaching study using milk, orange juice, saliva, urine, and sweat. Since Cl^- salts are present in urine and sweat, the CPs release the most AgNPs regardless of the product. They quantified the concentrations of AgNPs released in simulated scenarios, but little attention was given to characterize of these AgNPs, especially potential speciation alterations. In addition, washing machines that promote antibacterial activity with AgNPs may release AgNPs into the water source. The study by Farkas et al., investigated the simulation of common clothes washing using nano-Ag washing machines [43]. In the study, detergents were used and deactivated and activated modes of the "nano-silver wash" function were used. 11µg/L of Ag was detected in the wash water from the “nano-silver wash” mode. The AgNPs discharged mainly consisted of particles < 20 nm. The AgNPs were not characterized in detail once again. However, further investigation was conducted on the effect of the discharged water (containing AgNPs) on bacterial growth. By using flow cytometry, they measured the impact of the washing machine discharge on natural bacterial-plankton communities after 7 days. After seven days of AgNP exposure, bacterial growth was significantly reduced (85% at 12.5 g/L, 62% at 2.5 g/L), demonstrating the ecotoxicity of the released AgNPs.

The presence of liquid CPs-AgNPs in various water sources may also be associated with solid CPs-AgNPs. A common application of liquid AgNP-CPs is paint. Paints containing AgNP can potentially release AgNPs into water sources through exposure to rainwater and washing tools (paint brushes, containers, and gloves). Water run-off from facade panels painted with AgNPs-paints has also been assessed to further assess this scenario. There was a mixture of individual AgNPs as well as AgNPs bound to organic binders in the runoff. Analysis of SEM/EDX showed that Ag(0) is transformed into Ag_2S when it interacts with dissolved H_2S in rain water [35].

4.1.1.3. Wastewater treatment scenarios

Wastewater treatment plants (WWTP) use a physical and chemical processes, including aeration, activated sludge treatment, separation, filtration, oxygen treatment, UV exposure, and chlorine treatment. It is essential to understand how AgNPs transform in wastewater when exposed to different processes and conditions. Under simulated wastewater treatment conditions using pristine AgNPs, several studies have investigated the transformation of AgNPs [44-47]. In most of these studies, Ag_2S was confirmed as a more stable/nontoxic form of Ag in sludge and effluent waters. Under certain conditions, Ag_2S, however, can undergo further transformations, resulting in a more toxic form [48]. In this study, they found that ozone treatment produces more toxic forms of silver from WWTP effluent due to the oxidation of Ag_2S. In a quasi-equilibrium batch adsorption study, AgNPs released from socks were assessed by Benn et al., [24]. An adsorption capacity of 3.4 - 17 µg-Ag/g-biomass was calculated and consistent with a Freundlich isotherm model. They used the data to estimate Ag concentrations in WWTP effluent and sludge using a general fate model for sorption. In a similar study, metallic Ag dietary supplements were adsorbable under WWTP conditions [30]. With a biomass dose of 1000 mg/L, 80-90% of the Ag was removed but no analyses or model predictions were presented regarding the physicochemical properties of the AgNP. According to Kim et al., AgNPs were detected in real sewage sludge under anaerobic conditions from the reaction between reduced sulfur and AgNPs; these findings proved that Ag_2S is formed in situ during water treatment processes [49].

4.1.1.4. Natural surface water

A variety of environmental conditions and components may be exposed to AgNPs accumulated in natural bodies of surface water, including direct sunlight, temperature fluctuations, dissolved oxygen, and natural organic matter (NOM). Laboratory-synthesized AgNPs have been studied to determine the impact of some of these parameters on transformations. As part of these studies, AgNPs were transformed as a function of pH, ionic strength, and background electrolytes [50]; NOM transformed Ag^+ to AgNPs [51]; sunlight and different capping agents affected aggregation and dissolution [52], morphological transformations due to the influence of inorganic anions [53, 54] and a function of both NOM and light irradiation [55]. However, AgNPs released from CPs have received relatively few detailed investigations. Using mesocosm systems containing seawater, sediment, creatures from the sea (crabs, mussels), biofilm slides, and marsh grass, Cleveland et al., simulated exposure to estuarine conditions with AgNP-containing CPs (teddy bear, wound dressing, socks) [56-59]. Under realistic conditions, they aimed to measure AgNP bioaccumulation in aquatic organisms from CPs. In this study, no investigation was conducted into the transformation of AgNPs after exposure to the relevant environmental conditions. Overall, detailed experiments evaluating AgNP transformations in natural surface waters are lacking.

5. RESEARCH RECOMMENDATIONS AND GAPS

5.1 Consideration of All Possible Interactions in each Scenario

The possible interactions between AgNPs-CPs and relevant environmental factors must be taken into consideration according to the specific applications of the CPs (e.g., the potential transformations of AgNPs during the use of textiles due to body temperature, excess heat from ironing or drying clothes, exposure to sunlight (UV light), and the presence of other substances).

5.2 Effects of Capping Agents

Due to the instability of AgNPs caused by their high surface area to volume ratio, a wide variety of capping agents are used stabilize them. Citrate, Tween 20, Poly vinylpyrrolidone (PVP) and polyethlene glycol (PEG) are some of the most widely used capping agents for the synthesis of AgNPs. Although coated AgNPs are commonly incorporated into CPs, most of the studies cited in this report did not mention or report any details on the type, characteristics or potential effects of the coating agents incorporated into the products. However, except for one study, Radwan et al., conducted effects of particle size and capping agent of AgNPs in colloidal CPs [60].

5.3 Potential Transformations as a Function of Natural Environmental Factors

Physicochemical transformations of AgNPs are possible when they are exposed to natural environmental factors such as exposure to sunlight, oxygen, sulfides, and pH changes. Except for a few experiments on textile washing scenarios, the possible transformations of AgNPs released from different CPs under various environmental conditions have not been reported. Furthermore, the potential environmental impacts of the transformed AgNPs have been rarely studied. However, except for one study, Radwan et al., conducted the transformation of AgNPs after interactions with surfactant-based household surface cleaners [61].

5.4 Ecotoxicity Analyses

Many research studies have included ecotoxicity analyses on pristine laboratory synthesized AgNPs using different aquatic, bacterial, and plants species. However, except for one study, similar ecotoxicity analyses have not been performed with AgNP-containing CPs. Cleveland et al., conducted a mesocosm experiment by exposing AgNP-CPs (teddy bear, wound dressing, and socks) to estuarine water conditions [56]. Even in this study, a detailed toxicity assessment was not conducted using standard toxicity assays. Detailed ecotoxicity analyses on AgNP-CPs for different ecosystems (water, soil, and sediments) are essential. Previous studies that used laboratory-synthesized AgNPs, evaluated the potential toxicity and interactions of AgNPs on a wide range of different organisms and environmentally relevant bacterial species. Similarly, for AgNP-containing CPs, a detailed ecotoxicity assessment of on a variety of organisms should be completed.

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Table 1. Summary of past research using AgNP containing consumer products.

Consumer Product- characteristics			**Environmental Exposure**			**Human exposure**		**Reference**
			AgNPs behavior					
Product	**Form ofAgNP**	**Characterizing techniques**	**Scenario**	**Transformation**	**Toxicity For ecosystem**	**Exposure pathways**	**Toxicity assays**	
Kinetic go green™, Oso fresh (Plastic food storage containers),FreshLonger™ (plastic food storage bags)	Nano-Ag in LDPE (bags) or PP (containers)	ICP-MS, SEM/EDX,	Migration of AgNP fromthe packages to food 1. ethanol (50%) and acetic acid (3%) exposurefor 10 days (at 40°C) and 2 hr (at 70°C) Microwave heating	AgNP of different size and aggregates released from the food packages. Release of nano-plastics along with AgNPs in acidic conditions. AgNPs release is higher in microwave than oven.	Not studied	Not studied	Not studied	Vance etal., 2015, [5]
Kinetic go Green ™ , Always Fresh Containers™ (food storage box), FreshLonger ™, Miracle Food Storage ™	Nano Ag, Polyethylene(storage box), HDPE (bags)	ICP-MS, TEM/EDX,	Migration of AgNP fromthe packages to food, 1. Milli-Q water 2. 3% acetic acid 3. 10% ethanol At 40°C for 10 days	Highest release at acidic conditions and higher temperatures. Migration to water and 10% ethanol was below detection limit. Released particles are spherical shape, in agglomerates.	Not studied	Not studied	Not studied	Hansen etal., 2016, [12]
Textiles		ICP-MS, AAS,SEM/EDX, DLS	Extraction to artificial body fluids (sweat andsaliva)- effect of pH, temperature.	AgNPs release Ag^+ in sweat, $Ag^+ + Cl^- = AgCl$	Not studied	Not studied	Not studied	Stefaniak etal., 2014, [14]
Plastic bag Sunriver Industrial Co., China)	Nano Ag	SEM/EDX	Migration of AgNP fromthe packages to food 1. ultra-pure water 2. 4% acetic acid 3. 95% ethanol hexane	< 300 nm size Ag wasmigrated, Time and temperature dependent migration	Not studied	Not studied	Not studied	US.EPA, 2012, [19]
Socks	100-500 nm sized Agparticles	SEM, TEM, IES, ICP-AES	Washing with ultrapurewater and tap water without detergents	AgNP $\rightarrow$ Ag^+/colloidal Ag Colloidal Ag $\rightarrow$ Ag^+(after prolong exposure to water)	Model simulation was performed topredict the Ag leaching from biosolid under TCLP conditions	Not studied	Not studied	Benn et al., 2010, [20]
			Wastewater treatment (Adsorption to activatedsludge)	Sludge was not characterized for Ag speciation				
Socks	AgNP boundto fiber surface, incorporated in fibers, AgCl incorporated in binder on the fiber surface	ICP-OES, XFS,ISE	Washing (ISO method forwashing test) – effect of pH, surfactants, oxidizingagents	$Ag(0) \rightarrow Ag^+$ More Ag^+ release at pH 7 than at pH 10 $Ag + H_2O_2 \rightarrow$ $Ag^+ + OH\cdot$ $Ag + OH\cdot \rightarrow$ $Ag^+ + OH$	Not studied	Not studied	Not studied	Geranio et al., 2009, [21]

Socks, T-shirts, trouser	Ag-wires, Ag integrate in polyamide fiber, AgNPincorporated in cotton fibers, Ag(0), AgCl	Based on the information obtained from manufacturer and SEM/EDX,ICP-OES and XRF	Washing with tap waterand detergents	Release of AgCl, Ag_2S, Aggregations of metallic AgNPs	Not studied (antibacterial activity of the textile after washinghas been tested)	Not studied	Not studied	Lorenz et al.,2012, [22]
Socks	Metallic Ag in high concentration (14500mg/kg)	ICP-MS, STEM/EDX,	Washing with detergents	Metallic Ag particles of 20-30 nm. AgNPs in Si matrix or associate with S.	Not studied	Not studied	Not studied	Mitrano et al.,2014, [23]
Socks, T- shirt, medical masks, toothpaste, shampoos,		ICP-OES, SEM/EDX,	Washing with tap water	Release as particles or associated with particles > 100 nm (liquid phase products). Released as agglomerates of AgNP (< 20 nm) (textiles/fabric).	Not studied	Not studied	Not studied	Benn and Westerhoff, 2008, [24]
			Released into air duringuse (humidifiers)	Rate of release 0.08 – 0.11µg Ag hr^{-1}. Characterization was not successful due to the low concentration.				
			Landfill disposal (TCLPleaching test to simulate)	1.7 µg/g (medical mask), 54 µg/g (medical cloth), 0.2 µg/g (humidifier)				
Wound dressing, underwear, baby blanket, gloves, pants, wipes	Size of the AgNPs in each product is given in the original article	ICP-AES, SEM/EDX, TEM/EDX, UV-vis	Not studied	Not studied	Not studied	Potential exposures (ingestion) have estimated using mathematical formulae	Not studied	Tulve et al.,2015, [26]
Plush toy,sippy cup								
disinfectingspray, antibacterialspray, antiviral spray		ISE, DLS, TEM/EDX, UV-vis						
Textiles	Ag integrate in polyamidefiber, AgNP incorporated ncotton fibers, (Ag_2S-NP, Ag Zeolite, Ag_2S,$AgNO_3$, AgCl-NPs, Ag oxides)	XANES	Washing procedure (ISO105-IS washing procedure and machine washing). AgNPs transformationwithin fabric.	Increased the AgCl- NP andAg_2S-NP fraction. However, highly depend on the association mode of AgNPs in the original fabric.	Not studied	Inhalation	Not studied	Lombi et al.,2014, [27]
AgNP containingtextiles (Acticoat TM)	50 – 200 nm	ICP-AES, ICP-MS, SEM/EDX, AFM	Not studied	Not studied	Not studied	dermal	In vitro analysis skin analysis. Aggregations of AgCl on the skin layers	Bianco et al.,2015, [28]
NANO BeBe+ (Baby feeding bottle, Baby dream Co. Ltd, Korea) and foodbox	Nano Ag Polycarbonate (feeding bottle) Polypropylene (food box)	ICP-MS, SEM -EDX	Migration of AgNP fromthe packages to food 1. Milli-Q water 2. 3% acetic acid 3. 95% ethanol 4. 10% ethanol	Migration of AgNp dependon the nature of polymer inthe product Low release of AgNPs.	Not studied	Not studied	Not studied	Ramos et al.,2016, [29]
Cosmetic spray	AgNPs	TEM, ICP-MS, CPC, DMA	Spraying the antibacterialsolution inside a	Not studied	Not studied	Not studied	Not studied	Losert et al.,2015, [31]

			glove box					
Anti-odor spray, surface disinfectant, a throatspray	Ag particles > 450 nm, ionic Ag, nano Ag	ICP-MS, TEM,SEM/EDX, DLS, UV-vis,	Spraying inside a polyethylene chamber	Not studied	Not studied	Not studied	Not studied	Quadros andMarr, 2011, [33]
Antibacterialhydrogellotion, antibacterial nasal spray	Develop a separation method for AgNP and Ag^+ in solutions	UV-vis, SEM,TEM	Spraying the antibacterialsolution	Not studied	Not studied	Not studied	Not studied	Chao et al.,2011, [34]
Nano-Ag spray in two types of sprays (spray and pump spray)		ICP-MS, TEM/EDX, SMPS	Spraying the antibacterialsolution inside a glove box	Propellant gas sprays release nano-sized particles even NPsare not present in the spray solution	Not studied	Not studied	Not studied	Hagendorfer etal., 2010, [39]
Baby blanket, plush toy, breast milk storage bag,sippy cup (rubber ring), disinfecting spray, surface wipes, kitchen scrubber, humidifier accessory cube	Sizes of the AgNPs in each product is givenin the original article	SEM/EDX	Release into liquid media(tap water, synthetic sweat, saliva, urine, milkformula, orange juice)	Most product released Ag inionic form via dissolution. Synthetic sweat and urine extract the highest amount ofAg.	Not studied	Ingestion, dermal (NIOSH Method 9102)	Not studied	Quadros et al., 2013, [41]
		SMPS, optical particle counter	Aerosol release (in a simulated the product usein a room with carpeted floor, painted walls, a door and a window, two desks, two cushioned chairs, bookcase and wooden wardrobe)	very low emission rate	Not studied	Inhalation	Not studied	
WashingMachine	<20 nm sizeAgNPs	ICP-MS, SEM, STEM, Nano particle tracking analysis	Washing cycle	Release of AgNP (11 µg/L)	Affect on the growth of naturalbacterioplanktonwas tested	Not studied	Not studied	Farkas et al.,2011, [43]
Antiperspirant,plant strengthenng agent	8 nm – µm size range (solutions) nm-size aerosols	TEM/EDX, ICP-MS, SMPS	Spraying the antibacterialsolution in a glove box	Not studied	Not studied	Inhalation exposure	Not studied	Kaegi et al.,2011, [45]
Wound dressings,Catheters	Nano Ag andAg^+	SEM, TEM, ICP-MS, DLS,UV-Vis, (NTA)	Not studied	Not studied	Not studied	AgNPs extraction on to water, saline and human plasma	Not studied	Sussman et al., 2015, [50]

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