



Survey of food-grade silica dioxide nanomaterial occurrence, characterization, human gut impacts and fate across its lifecycle



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HIGHLIGHTS

- We quantitatively assessed amorphous SiO₂ across its lifecycle.
- Ten of 14 foods contained amorphous nano-SiO₂ from 2 to 200 mg Si per serving.
- Food-grade SiO₂ showed adverse impacts on human microvilli at relevant levels.
- Amorphous nano-scale SiO₂ was found in raw sewage and effluent from WWTPs.

GRAPHICAL ABSTRACT

A Survey of Food-grade Nano SiO₂ across Its Lifecycle



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ABSTRACT

There is increasing recognition of the importance of transformations in nanomaterials across their lifecycle, yet few quantitative examples exist. We examined food-grade silicon dioxide (SiO₂) nanomaterials from its source (bulk material providers), occurrence in food products, impacts on human gastrointestinal tract during consumption, and fate at wastewater treatment plants. Based upon XRD, XPS and TEM analysis, pure SiO₂ present in multiple food-grade stock SiO₂ exhibited consistent morphologies as agglomerates, ranging in size from below 100 nm to >500 nm, with all primary particle size in the range of 9–26 nm and were most likely amorphous SiO₂ based upon high resolution TEM. Ten of 14 targeted foods purchased in the USA contained SiO₂ of the same morphology and size as the pristine bulk food-grade SiO₂, at levels of 2 to 200 mg Si per serving size. A dissolution study of pristine SiO₂ showed up to 7% of the dissolution of the silica, but the un-dissolved SiO₂ maintained the same morphology as the pristine SiO₂. Across a realistic exposure range, pristine SiO₂ exhibited adverse dose–response relationships on a cell model (microvilli) of the human gastro-intestinal tract, association onto microvilli and evidence that SiO₂ lead to production of reactive oxygen species (ROS). We also observed accumulation of amorphous nano-SiO₂ on bioflocs in tests using lab-cultured activated sludge and sewage sludges from a full-scale wastewater treatment plant (WWTP). Nano-scale SiO₂ of the same size and morphology as pristine food-grade SiO₂ was observed in raw sewage at a WWTP, but we identified non-agglomerated individual SiO₂ particles with an average diameter of 21.5 ± 4.7 nm in treated effluent from the WWTP. This study demonstrates an approach to track nanomaterials from source-to-sink and establishes a baseline occurrence of nano-scale SiO₂ in foods and WWTPs.

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1. Introduction

Research gaps exist related to the occurrence, transformations and safety of engineered nanomaterials across various components of their life cycle (i.e., source-to-sink) from synthesis, through use, and to end-of-life (NNI, 2014). Viewing nanomaterials with this life cycle perspective differs from a life cycle analysis, the latter of which is a tool for evaluating a product's environmental impacts through all stages of its life cycle. Food additives have emerged as a major use of engineered nanomaterials, yet recent reviews indicate we know little about their effects, their ability to undergo transformations, or the ability to characterize them in complex matrices (Canady and Tsytsikova, 2013; Singh et al., 2014; Szakal et al., 2014; Yada et al., 2014). Silicon dioxide (i.e., SiO₂) has the highest production volume of all engineered nanomaterials, and its use as a food additive is a major sector (Keller and Lazareva, 2014; Keller et al., 2013b; Lazareva and Keller, 2014). SiO₂ is widely used in foods and other products as a defoaming agent/stabilizer, adsorbent, carrier, conditioning agent, chill-proofing agent, filter aid, emulsifying agent, viscosity control agent, anti-settling agent, and anti-caking agent (Lewis & Harrison and Washington, 2010; OECD, 2004). The United States Food and Drug Administration (USFDA) and the Code of Federal Regulations Title 21 permit the use of SiO₂ in foods not to exceed 2% by weight (U.S.FDA, 2014). The United Kingdom Food Standards Agency permits the use of SiO₂ in foods as anti-caking agent (E551) (FSA, 2010), and it can be added to a broad variety of foods up to 5% by weight in European markets (Lewis & Harrison and Washington, 2010). Commonly used food-grade SiO₂ products include synthetic amorphous silica (SAS), sodium aluminum silicates, and calcium silicates (Lewis & Harrison and Washington, 2010; OECD, 2004). Food-grade SiO₂ is produced either thermally (e.g., fumed SiO₂) or through wet chemistry (e.g., SiO₂ gel and precipitated SiO₂) (OECD, 2004). The worldwide production of SiO₂ in 2000 was 432,900 tons, with ~90% reported to be SAS for food industry and other sectors (OECD, 2004). The production, use, and end-of-life disposal of food-grade SiO₂ can lead to direct and indirect exposure of humans and the environment (Hansen et al., 1999; van Kesteren et al., 2014), and there are emerging concerns about its potential risk to humans and ecosystems (Maynard, 2014).

The toxicity of food-grade SiO₂ has been evaluated in vitro and in vivo (ECETOC, 2006; OECD, 2004). The European Center for Ecotoxicology and Toxicology of Chemicals and the Organization for Economic Cooperation and Development found no acute toxicity of amorphous SiO₂ after conducting a toxicity study on rats through oral and dermal exposure, with the highest exposure concentration of 20 g SiO₂/kg body weight (ECETOC, 2006; OECD, 2004). No significant intestinal absorption of amorphous SiO₂ was observed in animals or humans (OECD, 2004). However, a recent study showed that fumed SiO₂, a form of SAS, produced reactive oxygen species (ROS) and caused red blood cell hemolysis (heparinized mouse cell) (Zhang et al., 2012). Other work suggests the impact of amorphous SiO₂ on humans, especially on the gastrointestinal tract, should be re-evaluated because there are significant differences in physiology and nutrient uptake of the gastrointestinal tract between humans and rats (Faust et al., 2014a; Maynard, 2014). Additionally, nano-sized SiO₂ (i.e., nano-SiO₂) particles can exist in intestine digestion stage even though they would agglomerate under acidic condition during the saliva and gastric digestion stages (Peters et al., 2012). Thus it is necessary to explore the impact of food-grade SiO₂ on microvilli, as the brush border cytoskeleton is more fragile in humans than rodents (Carboni et al., 1987).

Various approaches assess the release of SiO₂ into the environment at the end of its value chain (Keller et al., 2013a; Piccinno et al., 2012; Som et al., 2011). Material flow modeling estimates that approximately 7000 tons/year of nano-SiO₂ (including food-grade nano-SiO₂) enters wastewater treatment plants (WWTPs) (Keller et al., 2013a). Slight removal of uncoated nano-SiO₂ particles occurs in WWTPs, either by coagulation using poly aluminum chloride or by the activated sludge

process (Chang et al., 2007; Jarvie et al., 2009). These previous studies focus on well-dispersed spherical nano-SiO₂ particles (Chang et al., 2007; Jarvie et al., 2009), but the removal of food-grade SiO₂ nanomaterial (e.g., fumed nano-SiO₂) in the activated sludge process has not yet been fully explored. Experimental data on removal of food-grade SiO₂ is needed to understand its fate across the food-system and discharge into the environment (i.e., a life-cycle perspective).

The objective of this research was to understand occurrence, characterization and transformations of food-grade nano-SiO₂, a high volume engineered nanomaterial with direct human and environmental contact. First we characterized six food-grade SiO₂ bulk material powders used as food ingredients, aiming to provide information about: morphology (using transmission electron microscopy [TEM]), hydrodynamic diameter and zeta (ζ) potential (using Phase Analysis Light Scattering [PALS]), crystal structure (using X-ray diffraction [XRD]), and surface functionality (using X-ray photoelectron spectroscopy [XPS]). Second, we quantified the occurrence and characteristics the structure of SiO₂ incorporated into food and personal care products by purchasing 14 products in the USA with SiO₂ or anti-caking agents listed as ingredients. Third, as an indicator of a process leading to transformation of food-grade nano-SiO₂, we tested dissolution potential of bulk food-grade SiO₂ in ultrapure water, tap water, 1 mM of sodium bicarbonate (NaHCO₃), and a culture medium. Fourth, we quantified the impact of food-grade SiO₂ in culture media on microvilli using a Caco-2BBE1 cell system, which is an accepted model of the human intestine (Peterson et al., 1993; Peterson and Mooseker, 1993), and we hypothesized that the amorphous food-grade nano-SiO₂ could lead to the disruption of microvilli surface structure. Finally, we investigated occurrence and structural properties of bulk food-grade SiO₂ in lab-scale tests and prospected the presence of nano-SiO₂ in sludge samples from a local WWTP to determine whether SiO₂ is present in treated effluent that is discharged to surface waters or in sewage solids that are discharged to landfills, incinerators, or agricultural lands.

2. Materials and methods

2.1. Sample procurement and characterization

Six samples of food-grade SiO₂ (fumed, pyrogenic, not labeled as “nano”) were obtained from commercial vendors in the USA and China. All samples presented as white powders were sold specifically as food additives, and were considered as “pristine” SiO₂ because they had not yet been mixed or reacted with food matrices. Fourteen food products including personal care products were purchased in supermarkets in Scottsdale, Arizona (USA) in the fall of 2014 (see Table S1).

Details for sample preparation and analytical methods are provided in Supporting Information (SI), and briefly overviewed here. Hydrodynamic diameter and ζ potential were analyzed by PALS (ZetaPALS, Brookhaven Instruments Corp., NY, USA) in a 10 mM potassium nitrate solution from pH 2.5 to 9.5. Scanning transmission electron microscopy (STEM/TEM, Phillips CM-200, USA) or scanning electron microscopy (SEM) equipped with an electron dispersive X-ray (EDX) microanalysis system (Philips XL30-EDAX, USA) was used to acquire 10 to 15 images per sample. SiO₂ or food samples containing SiO₂ were dispersed in nanopure water before transferring small aliquots of supernatant liquid onto the surface of a copper grid (TedPella, Inc., CA, USA) or aluminum SEM stub (see SI for details). The primary particle size (i.e., not agglomerates) in TEM images was analyzed using the Image J software by averaging on 250–260 primary particles in each sample (see SI). Food-grade SiO₂ was dried at 60 °C for 24 h before XRD (Siemens D5000, TX, USA) and XPS (ESCALAB 220i-XL, Vacuum Generators, USA) analysis. Elemental composition of samples was determined using inductively coupled plasma mass spectrometry (ICP-MS, X-Series-2, Thermo Scientific, USA) after microwave digestion in the presence of

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