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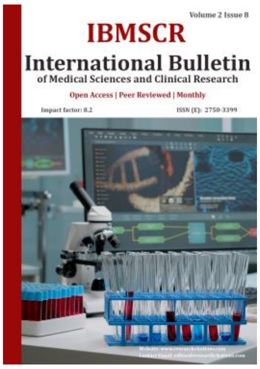

THE IMPORTANCE OF E171 DYE IN THE FOOD INDUSTRY AND ITS EFFECT ON THE BODY

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Abstract

TiO2 (E 171) is a pigment used to alter the visual properties of food and beverages. It is also used in cosmetics, pharmaceuticals, and toothpastes, which means that we may also come in contact with the substance while taking medicines or using personal hygiene products such as toothpastes or mouthwashes. Titanium dioxide (TiO2) nanoparticles (NPs) have been widely applied in various industrial fields, such as electronics, packaging, food, and cosmetics. Titanium dioxide (TiO2) is commonly applied to enhance the white colour and brightness of food products. TiO2 is also used as white pigment in other products such as toothpaste. A small fraction of the pigment is known to be present as nanoparticles (NPs). Submicron-sized TiO2 particles, in Europe listed as E 171, are widely used as a food additive although the relevant risk assessment has never been satisfactorily completed. For example, it is not possible to derive a safe daily intake of TiO2 from the available long-term feeding studies in rodents. Also, the use of TiO2 particles in the food sector leads to highest exposures in children, but only few studies address the vulnerability of this particular age group. Extrapolation of animal studies to humans is also problematic due to knowledge gaps as to local gastrointestinal effects of TiO2 particles, primarily on the mucosa and the gut-associated lymphoid system. Tissue distributions after oral administration of TiO 2 differ from other exposure routes, thus limiting the relevance of data obtained from inhalation or parenteral injections. Such difficulties and uncertainties emerging in the retrospective assessment of TiO2 particles exemplify the need for a ft-to-purpose data requirement for the future evaluation of novel nano-sized or submicron-sized particles added deliberately to food.

Keywords: E171, Titanium dioxide, Nanoparticles

Introduction

Titanium dioxide (TiO2) is a naturally occurring metal oxide and is one of the fie engineered nanomaterials most commonly used in daily consumer products, including food1. The TiO2 food additive, referred to as E171 in the European Union (EU), is commonly used as a whitening and brightening agent in confectionary (candies and chewing gum), white sauces and icing1–3. The Food and Drug Administration approved the use of food-grade TiO2 in 1966 with the stipulation that TiO2 levels must not exceed 1% of the food weight4. In Europe, the current EU Directive 94/36/EC authorizes the use of E171 in foodstuff without establishing an acceptable daily intake level by the Joint FAO/WHO Expert Committee on Food Additives, based on TiO2 absorption considered to be very low5. Nevertheless, the common use of E171 leads to signifiant levels of daily dietary intake of nanoparticulate matter among humans1. Indeed, E171 batches show broad size distributions of TiO2 primary particles [1].

Titanium dioxide is a common additive in many food, personal care, and other consumer products used by people, which after use can enter the sewage system and,

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subsequently, enter the environment as treated effluent discharged to surface waters or biosolids applied to agricultural land, incinerated wastes, or landfill solids. This study quantifies the amount of titanium in common food products, derives estimates of human exposure to dietary (nano-) TiO2, and discusses the impact of the nanoscale fraction of TiO2 entering the environment. The foods with the highest content of TiO2 included candies, sweets, and chewing gums. Among personal care products, toothpastes and select sunscreens contained 1% to >10% titanium by weight. While some other cremes contained`titanium, despite being colored white, most shampoos, deodorants, and shaving creams contained the lowest levels of titanium (<0.01 μg/mg). For several high-consumption pharmaceuticals, the titanium content ranged from below the instrument detection limit (0.0001 μg Ti/mg) to a high of 0.014 μg Ti/mg. Electron microscopy and stability testing of food-grade TiO2 (E171) suggests that approximately 36% of the particles are less than 100 nm in at least one dimension and that it readily disperses in water as fairly stable colloids. However, filtration of water solubilized consumer products and personal care products indicated that less than 5% of the titanium was able to pass through 0.45 or 0.7 μm pores. Two white paints contained 110 μg Ti/mg while three sealants (i.e., prime coat paint) contained less titanium (25 to 40 μg Ti/mg). This research showed that, while many white-colored products contained titanium, it was not a prerequisite. Although several of these product classes contained low amounts of titanium, their widespread use and disposal down the drain and eventually to wastewater treatment plants (WWTPs) deserves attention. A Monte Carlo human exposure analysis to TiO2 through foods identified children as having the highest exposures because TiO2 content of sweets is higher than other food products and that a typical exposure for a US adult may be on the order of 1 mg Ti per kilogram body weight per day. Thus, because of the millions of tons of titanium-based white pigment used annually, testing should focus on food-grade TiO2 (E171) rather than that adopted in many environmental health and safety tests (i.e., P25), which is used in much lower amounts in products less likely to enter the environment (e.g., catalyst supports, photocatalytic coatings) [2].

As a bulk material, titanium dioxide (TiO2) is primarily used as a pigment because of its brightness, high refractive index, and resistance to discoloration. The global production of TiO2 for all uses is in the millions of tons per year. Nearly 70% of all TiO2 produced is used as a pigment in paints, but it is also used as a pigment in glazes, enamels, plastics, paper, fibers, foods, pharmaceuticals, cosmetics, and toothpastes.1 Other TiO2 uses include antimicrobial applications, catalysts for air and water purification, medical applications, and energy storage. Recently, more attention has been given to the use of TiO2 as a nanomaterial. In 2005, the global production of nanoscale TiO2 was estimated to be 2000 t worth $70 million;2 approximately, 1300 t was used in personal care products (PCPs) such as topical sunscreens and cosmetics. By 2010, the production had increased to 5000 t, and it is expected to continue to increase until at least 2025 with greater reliance upon nanosize TiO2. Consequentially, many sources of nanoscale TiO2 could result in human exposure and entrance of this material into the environment (air, water, or soil compartments) [3,4].

TiO2-containing materials are produced in a range of primary particle sizes. Many applications of TiO2 would benefit from smaller primary particle sizes, and the percentage of TiO2 that is produced in or near the nano range is expected to increase exponentially. TiO2 nanoparticles are generally synthesized with a crystalline structure (anatase, rutile, or brookite, each of which has unique properties). The most common procedure for synthesis of

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TiO_2 nanoparticles utilizes the hydrolysis of titanium (Ti) salts in an acidic solution. Use of chemical vapor condensation or nucleation from sol−gel can control the structure, size, and shape of the TiO_2 nanoparticles.8,9 To increase photostability and prevent aggregation, TiO_2 nanomaterials (particles, tubes, wires, etc.) are commonly coated with aluminum, silicon, or polymers[4,5].

TiO_2 nanomaterials in foods, consumer products, and household products are discharged as feces/urine, washed off of surfaces, or disposed of to sewage that enters wastewater treatment plants (WWTPs). Although WWTPs are capable of removing the majority of nanoscale and larger-sized TiO_2 from influent sewage, TiO_2 particles measuring between 4 and 30 nm were still found in the treated effluent. These nanomaterials are then released to surface waters, where they can interact with living organisms. One study monitoring TiO_2 nanomaterials found the highest concentrations in river water to be directly downstream of a WWTP.14 TiO_2 nanomaterials removed from sewage through association with bacteria may still end up in the environment if the biomass is land applied [6,7].

Although the release of TiO_2 nanomaterials to the environment has been shown qualitatively, quantification of how much is released is difficult. It is impossible to determine all sources or measure the amount of released TiO_2 nanomaterials, which is why emissions are often modeled to better predict the impact of TiO_2 nanomaterials on the environment. The same difficulty applies to the assessment of human exposure, as estimated uptake rates of different types of nanoparticles range from 0% to 8.5% depending on type, size, and shape of the nanoparticles [8,9,10].

Toxicity studies mainly report a risk from nanoparticular TiO_2 due to inhalation (inflammation and possible link to asthma), but titania has also been linked to Crohn's disease from gastrointestinal intake and it has been classified as a possibly carcinogen.18−21 However, a risk assessment has not been published yet and care has to be taken when comparing exposure to effect. Not only have modifications been reported to have diverging toxicological properties (anatase is 100 times more toxic than rutile in the nanoparticular form) but also coatings, size, and shape modify the toxicity of nanoparticles, and only a small number of them have been tested. Once in the environment, even less is known about how TiO_2 nanomaterials affect organisms, although nanosized TiO_2 has been shown to inhibit growth of algae and bioaccumulate in Daphnia magna. However, several studies have indicated that TiO_2 tends to be less hazardous to organisms than other nanomaterials such as multiwall carbon nanotubes, nanocerium oxide, and nanozinc oxide. Previously, primary particle size was generally accepted as a large factor in toxicity, with smaller particles tending to be more toxic. However, recent studies have shown that particle size is only a single (and perhaps minor) factor influencing the toxicity of nanoparticles. Risk assessment of certain nanomaterials is still quite difficult because nanotoxicology studies rarely have enough reliable information on the physicochemical characteristics of the nanoparticles tested [10,11].

Many fate and transport as well as toxicity studies have used a readily available TiO_2 nanomaterial (Evonik Degussa P25) because the primary crystals are <50 nm in size and uncoated. P25 is advertised as "titanium dioxide without pigment properties". On the basis of information on the manufacturers Web site, P25 is used primarily as a photocatalyst, catalyst carrier, and heat stabilizer for silicon rubber. This material is agglomerated in the dry powder state and readily aggregates to several hundred nanometers in water. However, usage of TiO_2

in the food, beverage, and paint markets dwarfs the usage of P25. For example, food-grade TiO_2 (referred to as E171) is purchased by the ton and is available as synthetic forms of anatase, rutile, and others. Only one study reports the titanium content of a few commercial products; we know very little about size or surface properties of E171 forms of these TiO_2s in comparison with the vast amount of data on P25 even though E171 and other commercially used whiteners represent the majority of TiO_2-containing materials that enter the ecosystem today [9,12].

This paper aims to begin filling the large knowledge gaps that exist regarding commonly used sources of TiO_2 materials. We obtained a broad spectrum of commercial products that either listed titanium dioxide on the label or had a “white” color and quantified the titanium content. Selected products were further characterized by electron microscopy. Using this new and already existing TiO_2 data, a human exposure analysis was conducted that indicates children may be disproportionately exposed to higher levels of all sizes of TiO_2. Finally, characteristics of E171 were compared against those of the titanium observed in food products and against those of P25 in an attempt to argue that greater efforts to elucidate fate and transport are needed for materials containing E171[8,11,12].

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