

# Metallic nanoparticle production and consumption in China between 2000 and 2010 and associative aquatic environmental risk assessment

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**Abstract** With rapid advances in nanotechnology and nanomaterials, metallic nanoparticles (MNPs) have become widely used in many different products and industrial processes. Water is an important medium in the transfer and fate of MNPs. Accordingly, the potential for the inadvertent and incidental release of MNPs into aquatic environments through direct release and waste disposal has increased considerably in China in recent years. Environmental health and human safety are two of the greatest challenges facing the expanding nanomaterial field. However, existing knowledge on MNP toxicity is currently insufficient to carry out a comprehensive risk assessment due to a general lack of data related to the

environmental distribution of MNPs within aquatic environments. This study provides a summary of MNP production and consumption trends in China by means of statistical changes in MNP discharge and deposition between 2000 and 2010. China was used as a model for aquatic environmental risks associated with MNP consumption and production. MNP pollution of aquatic environments is discussed as well as the challenges that China will face in the future with increasing nanomaterial consumption and pollution. The study concludes with a discussion on managing MNP exposure of aquatic environments in China and its subsequent risks, if any, which may require greater attention.

**Keywords** Metallic nanoparticle · Nanomaterial · Nano risk · Aquatic environment · China

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## Introduction

The development of nanotechnology and nanomaterials in the twenty-first century has undergone unprecedented expansion. Metallic nanoparticles (MNPs) have wide-ranging functionality and utility for numerous industrial and consumer applications, including water treatment, energy production, novel therapeutic drug delivery systems, home appliances, consumer electronics, dietary supplements, and sports equipment (Hansen 2010). As nanomaterials (i.e., <100 nm) are integrated into more industrial and consumer products,

the potential for unintentional and incidental MNP discharge into the environment through direct release and waste disposal has significantly increased in recent years (Thomas et al. 2011; Wang et al. 2011). The environmental behavior and fate of MNPs with regards to adsorption, accumulation, persistence, aggregation, and mobility in different environmental media have an obvious effect on the quantity of MNPs in the environment, increasing the potential for environmental and human exposure. Since there is insufficient knowledge related to MNP toxicity, fate, and behavior at the present time, determining the severity of nanomaterial impacts on environmental and human health is impossible. Therefore, nanopollution can be considered an “invisible pollution” and may consequently be one of the most difficult types of pollution to manage and control.

Water is known to be an important medium in the transfer and fate of MNPs, which can enter aquatic environments alongside co-occurring chemicals by means of landfill leachates, road runoff, wastewater, etc. (Bernhardt et al. 2010). For these reasons, water safety is closely related to human health, and the potential human impact of MNP leaching into aquatic environments is starting to attract more attention. However, since only limited data are currently available, actual conditions of MNP discharge and distribution into aquatic environments remain obscure.

With rapid advances in nanotechnology, many products containing engineered MNPs have been marketed, such as scratch-free paint, sports equipment, electronic components, sunscreens, wrinkle, and stain resistant fabrics, medical products, etc. (Thomas et al. 2011; Wang et al. 2011). MNPs are also used in sunscreens and cosmetics due to their transparency and enhanced efficacy. They are further used in the manufacturing of tennis rackets and baseball bats to improve strength and reduce weight. The textile industry uses nanotechnology to produce stain, wrinkle, and water resistant clothing (Dhawan and Sharma 2010). Inorganic metal or metallic oxide nanomaterials concentrate on three product categories: paints, food packaging, and fuel additives (O'Brien and Cummins 2011). For example, silver (Ag) nanoparticles are used as anti-bacterial agents in washing machines; cadmium (Cd) nanoparticles are being explored in the development of efficient, low-cost solar panels; nanoscale iron (Fe) is used in water treatment applications to remove contaminants from

wastewater; spherical titanium (Ti) and carbon (C) nanomaterials are being evaluated for their potential to function as novel drug delivery systems based on their affinity to specific cellular organelles; and zinc oxide (ZnO) nanoparticles are used in food packaging as well as in common appliances, such as washing machines and water purifiers (O'Brien and Cummins 2011). For these reasons, the likelihood of environmental and human exposure to MNPs is growing.

China has become the largest nanomaterial market in the Asia-Pacific owing to the rapid economic development occurring in the region (MIIT 2011). Consequently, there is an increased potential risk in MNP contamination of China's aquatic environments (Piccinno et al. 2012). This study reviewed available data related to (1) Chinese MNP production and consumption, (2) statistical changes found in MNP discharge and deposition into aquatic environments in China between 2000 and 2010, and (3) MNP lifecycle processes in aquatic environments. Findings could provide insights into the emergent managerial practices related to the environmental risks of nanomaterials in China (and elsewhere) as a result of the growth in usage and application of nanotechnology.

## Methods

Data related to nanomaterial production and consumption in China between 2000 and 2010 were taken from the Chinese Nanomaterials Industry Market and Investment Forecast Report (2011–2017), papers published in domestic Chinese journals, conferences, and international journals as well as from patents granted by the United States Patent and Trademark Office (USPTO, <http://patft.uspto.gov>). Publication data were downloaded from the Science Citation Index Expanded (SCI-E) service and the Chinese National Knowledge Infrastructure (CNKI). Indicators based on publications provide important evidence of a country's progress in nanotechnology. Developing nanotechnological-based applications are contingent upon advanced scientific research. Patents were used to help judge inventive capacity. They were also used as a possible indicator of novel product/process development. Moreover, industry reports published by certain economic consultant companies can provide detailed developmental insights.

Data on wastewater discharge and dust deposition were obtained from Chinese environmental bulletins (2000–2010). In predicting MNP discharge values for particular surface water constitutions in China, resulting MNP concentrations were compared to ranking categories related to different orders of aquatic contamination, which are as follows: Rank 1 (0–10 ng/L), Rank 2 (10–100 ng/L), Rank 3 (0.1–1 µg/L), and Rank 4 (1–10 µg/L) (O'Brien and Cummins 2011). According to the Chinese environmental bulletins consulted, the Ministry of Environmental Protection defines industrial wastewater belonging to Rank 3 and residential wastewater belonging to Rank 2. Thus, this study obtained industrial and residential MNP wastewater discharge by multiplying wastewater discharge by 1 µg/L (the upper limit of Rank 3) and 100 ng/L (the upper limit of Rank 2), respectively. The mean value of dust containing nanoparticles was determined by monitoring dust reported in the Chinese environmental bulletins for 2000–2010. Thus, total MNP dust deposition can be calculated by total dust deposition multiplied by the mean coefficient of the dust containing nanoparticles.

Nanomaterial consumption data were taken from the Chinese Nanomaterial Industry Market and Investment Forecast Report (2011) and the Ministry of Industry and Information Technology of the People's Republic of China (MIIT) (2011). To understand relationships between increases in nanomaterial consumption and MNP pollution, a polynomial regression analysis was carried out between nanomaterial consumption and MNP pollution. All polynomial regression analyses were performed using the SPSS software package, version 12.0 (SPSS Inc., Chicago, IL, United States of America).

## Results

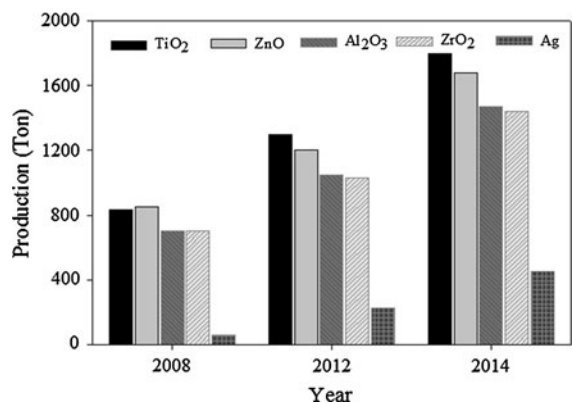
### MNP production and consumption in China

Nanotechnology is a highly intensive field wherein technological development critically depends on scientific research. China has already attained a leading position in terms of publications, paper citations, and patent applications after >20 years hard work, starting in the early 1990s (Bhattacharya et al. 2012). For example, China has emerged as the leader in nanotechnology literature, accounting for 23 % compared to the United States of America's 21 % of the relevant

literature published in 2009. It stands to reason that the top 1 % of all the cited literature on nanotechnology has the greatest international impact. Within this top 1 %, China has 132 while the United States of America has 257. Although a smaller number, it is still impressive, nevertheless. Similarly, nanotechnology patents granted by USPTO came in fourth with 34 between 2007 and 2009 (as shown in the Thomson Innovation Patent Database 2007). Subsequently, nanotechnological applications appear promising, especially when considering the substantial quantity of nanoproducts and nanomaterials produced and consumed (Zhang 2005).

In general, with increasing developments in Chinese nanotechnology, the management of MNP exposure of China's aquatic environments and its subsequent risks, if any, should be focused on in the eastern region of the country since almost all factories and enterprises related to nanotechnology are located in eastern China. Currently, the number of nanotechnology manufacturing factories and enterprises with a total investment of 10 billion United States dollars exceeds 800, mostly distributed in eastern China (Chinese Nanomaterial Industry Market and Investment Forecast Report, 2012–2017). However, the number of nanotechnology factories and enterprises with a total investment of \$80 million United States dollars was far lower, just over 100.

Typical Chinese MNP production estimates were based on the Chinese Nanomaterial Industry Market and Investment Forecast Report (2012–2017) (Fig. 1). Greater than 1,000 tons of metallic nanopowders have been produced since 2012, including Ti, ZnO, alumina



**Fig. 1** Typical MNP production in China between 2008 and 2014

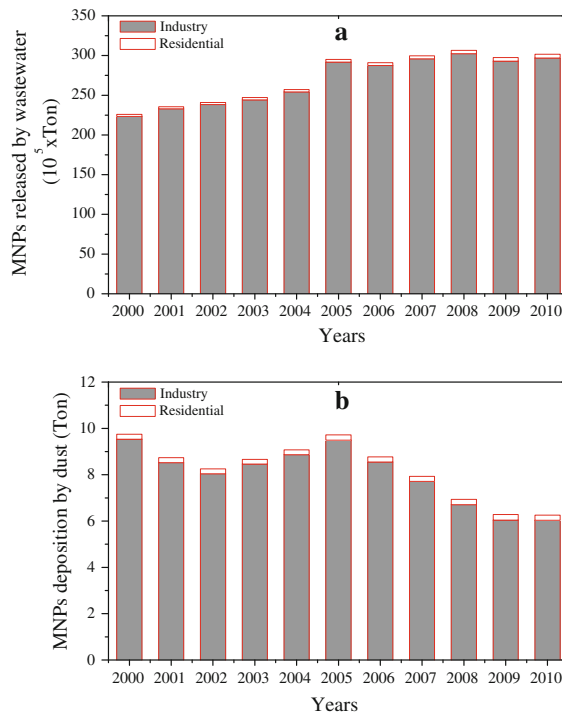
(Al<sub>2</sub>O<sub>3</sub>), and zirconia (ZrO<sub>2</sub>). Similarly, the market demand for Ag nanoparticles as an anti-bacterial agent was estimated to increase up to 366 tons by 2014 compared to 45 tons in 2008. This study estimated the amount of Ti nanopowders used as uvioreistant agents to be ~800 tons per year (since 2010). Furthermore, when a complete adjustment of economic structures is taken into account (i.e., the considerable needs related to environmental protection and energy saving measures), developmental steps in nanotechnology would increase rapidly, leading to an increase in financial commitment by more than 100 billion United States dollars by 2017 (Chinese Nanomaterial Industry Market and Investment Forecast Report, 2012–2017) due to the comparative market of MNP production and consumption in China. In particular, Ti, Ag, and ZnO nanomaterials should be emphasized because of their substantial production and consumption as well as their potential impacts on aquatic environments (O'Brien and Cummins 2011).

### Changes in MNP emissions in China over the past 11 years

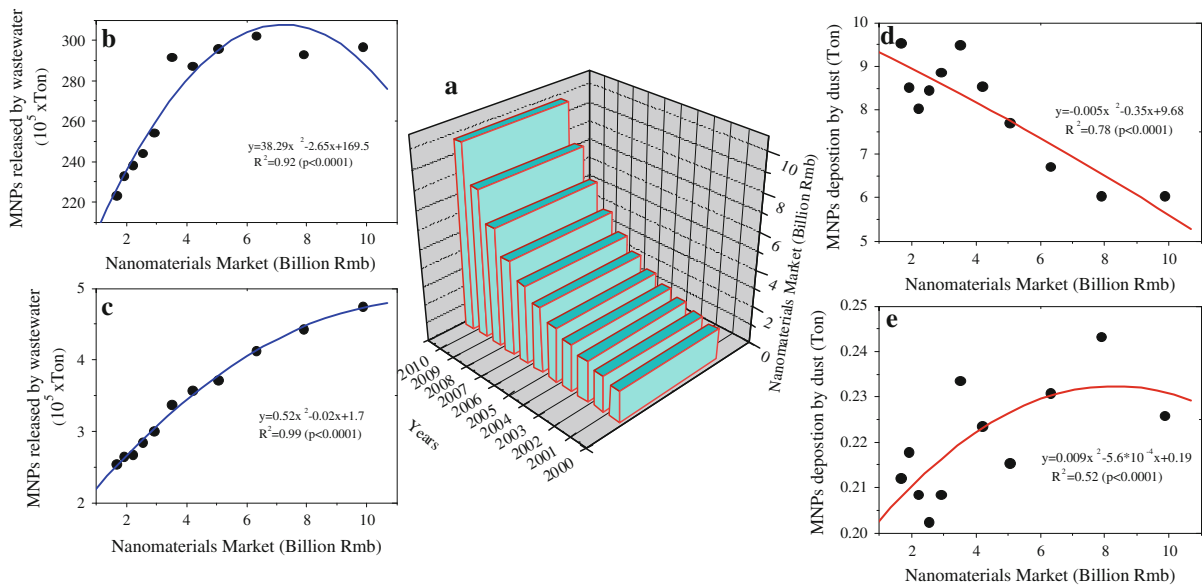
Facing a large nanomaterial market and strong consumer demand, it is necessary to evaluate MNP pollution and to characterize MNP distribution in aquatic environments to help control and ultimately prevent nanopollution from occurring. In China, less attention has been paid to MNP pollution. Only Luo et al. (2010) reported on distribution characteristics of TiO<sub>2</sub> in Xiamen Bay, Fujian Province. It stands to reason that an increase in nanomaterial development will accelerate MNP pollution into aquatic environments. To demonstrate the rapid development of nanomaterial integration into consumer markets in China, an evaluation of MNP discharge into aquatic environments over the past 11 years was carried out. Figure 2a shows the total amount of MNP discharged through wastewater between 2000 and 2010, with total emissions exceeding  $225 \times 10^5$  tons each year. It was determined that the primary source of MNP pollution came from industrial wastewater discharge, accounting for over 90 % of total MNP pollution. MNP from residential discharge was not a primary source of pollution. This study also estimated the effect of MNP dust deposition on aquatic environments (Fig. 2b). It

was found that gross MNP dust deposition significantly decreased in recent years but remains an important source of aquatic environment MNP pollution. Industrial sources were a primary source of MNP dust deposition while residential sources were not primary contributors. As has been observed with naturally occurring MNP dispersal, Lowry et al. (2010) reported that dispersal from manmade sources is closely related to the biogeochemical cycling of sulfur (S), C, phosphorus (P), and nitrogen (N) in an environment.

The World Market for Nanotechnology and Nanomaterials in Consumer Products (2010) reported revenue of approximately 1,545 million United States dollars from nanotechnology and nanomaterials in consumer products in 2009. This is expected to increase to 5,335 million United States dollars by 2015, an increase largely driven by the demand for consumer electronics and household cleaning products. The nanomaterial consumer market in China exhibited rapid growth over the past 10 years, close to ten billion RMB (1 USD = 6.47 RMB) in 2010 (Fig. 3a), where nanoparticles accounted for 93.8 %



**Fig. 2** MNP discharged via wastewater **a** and dust deposition **b** into aquatic environments in China between 2000 and 2010



**Fig. 3** Nanomaterial consumption in China between 2000 and 2011 **(a)**. **b** MNPs discharged from industrial sources via wastewater; **c** MNPs discharged from residential sources via

wastewater; **d** MNPs discharged from industrial sources via dust; and **e** MNP discharged from residential sources via dust

and composite nanomaterials accounted for 6.8 % of the nanomaterial market. Being a country in the midst of a rapidly developing nanotechnology industry, the nanomaterial market in China is expected to exceed 150 billion RMB within the period of the “Twelfth Five-Year Plan” (MIIT 2011), from 2011 to 2015.

Using statistical correlation analysis, this study found that MNP pollution produced by wastewater discharge significantly correlated to nanomaterial consumption (Fig. 3b–e), especially for MNP discharged from residential sources ( $R^2 = 0.99$ ,  $P < 0.0001$ ), which is much higher than that discharged from industrial sources. Nanomaterials consumed by residential sources are mostly discharged directly through water without pretreatment while industrial sources must follow strict pretreatment guidelines before discharging MNPs into aquatic environments. The overall impact of the nanomaterial consumer market on MNP dust deposition is relatively small. This is explained by the Chinese government’s gradual strengthening of management practices related to dust emissions. Moreover, technology related to dust emission pretreatment is more established and easier to implement. Although a national standard in China is required, many enterprises and industries still stealthily discharge wastewater to

reduce discharge fees. This makes it difficult to manage MNPs discharged by industrial wastewater, and is also why MNP pollution in aquatic environments has gradually increased over the past decade.

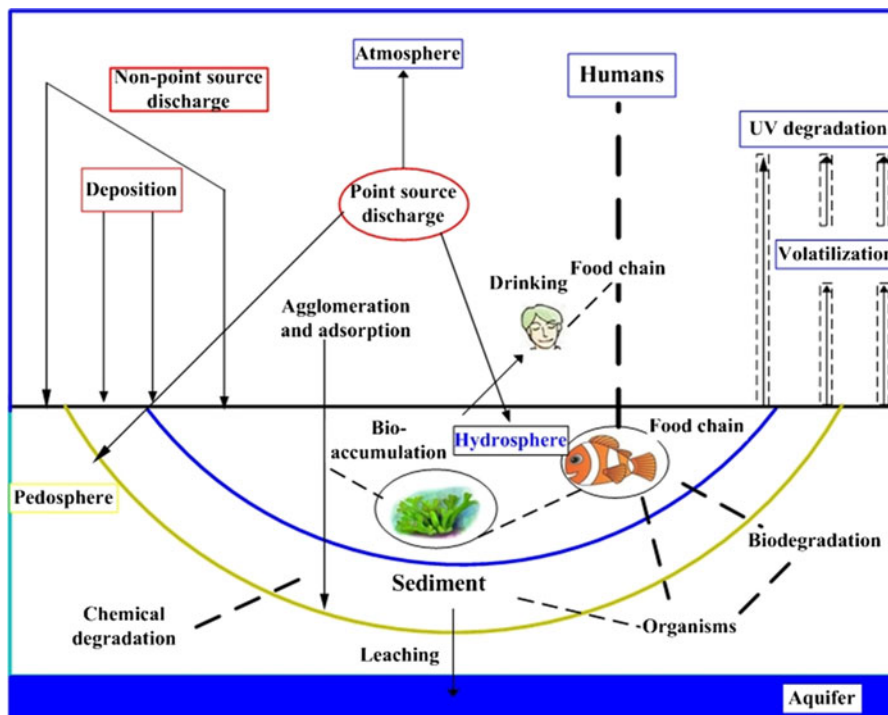
### Discussion

#### MNP lifecycle process risks on aquatic environments

MNPs integrated into products can be released during utilization, wear, recycling, and disposal. MNP populace exposure can occur through ingestion of contaminated drinking water and through food chain transfer (Fig. 4). The routes by which MNPs are released into aquatic environments originate from either point sources or nonpoint sources. MNP point sources include nanomaterial production or transport processes from manufacturing facilities and landfill wastewater treatment processes or stormwater runoff from manufacturing facilities or cities (Miao et al. 2010, 2011; Cao 2008). Most MNPs released into water are transported with other pollutants from nonpoint sources, such as from wearing or washing textiles containing MNPs, from using products that



**Fig. 4** MNP lifecycle processes in aquatic environments



contain nanomaterials (i.e., sunscreen, cosmetics, and paints), or from agrochemicals that leach or drain from landfills, rain runoff, etc., (Gottschalk and Nowack 2011; Gao et al. 2011, 2012). Many MNPs are inadvertently released into wastewater therefore making wastewater treatment plants an important point source for MNP discharge. These major pathways carry the greatest potential risk for MNP exposure from consumer products.

Adsorption, aggregation, and sedimentation are the primary behavioral characteristics of MNPs in water. Lin et al. (2010) concluded that most MNPs present in aquatic environments are aggregates rather than individual particles. Agglomeration and sedimentation are common mechanisms by which MNPs enter aquatic environments, affecting solubility. Water with elevated MNP concentrations can be expected to exhibit significant agglomeration within a couple of hours. Certain MNPs tend to form agglomerates when they come into contact with aquatic systems, distributing over time. However, smaller MNPs remain as colloids dispersed within solutions (Brar et al. 2010). Furthermore, certain MNPs are covered by surfactants during manufacturing processes, affecting the manner by which they dissolve due to the dissimilarity in agglomeration or aggregation in aqueous media

(Thomas et al. 2011). MNPs in fluids can in most cases be quickly and completely released during application whereas MNPs adsorbed within solid matrices are gradually or only partially released (Koehler et al. 2008). MNP dissolution is predicted to be higher in aquatic environments due to their smaller particle sizes and greater surface areas, which results in a greater available reactive area relative to mass (Yang and Xie 2006; Mihranyan and Stromme 2006). In general, the rate of MNP aggregation in a solution depends on MNP concentration, surface area, and forces involved in collisions (Miao et al. 2009).

MNP stability in aquatic systems is highly influenced by its solubility and dispersibility whereas MNP properties in water tend to change with time and the surrounding environment. Ionic strength, biochemical oxygen demand, hardness, alkalinity, pH, presence of organic matter, and composition are some hydrological parameters that modify MNP behavior in aquatic environments (Peralta-Videa et al. 2011). Moreover, dissolved ionic solutes and pH play crucial roles in MNP aggregation (Hotze et al. 2010). Differences in pH, ionic strength, quantity of natural organic compounds, and surfactants present in sea, fresh, and marine waters will have a significant influence on MNP performance and aggregation.

MNP surface area is an important factor in MNP toxicity because nanoparticle interaction with biological systems takes place on MNP surfaces (McNeil 2005). Furthermore, MNP transferability is typically proficient in and across epithelial cells. They can also be distributed to other body compartments, likely a function of size and surface properties (Tsuji et al. 2006; Thomas et al. 2006). MNPs come into contact with different biomolecules when absorbed into bodies, especially proteins (Bihari et al. 2008; Lynch and Dawson 2008), which alters MNP properties and affects MNP biodistribution and interactions with biostructures and cells. Moreover, once MNPs are inside a body, different salt concentrations and variable pH values can lead to MNP agglomeration suspension. In addition, current data related to the fate and behavior of many MNPs in environmental media that have been integrated into consumer and industrial products and processes are insufficient to draw comprehensive and definitive conclusions when applying a traditional quantitative risk assessment framework (Gottschalk and Nowack 2011; Christine et al. 2013).

Future challenges in MNP pollution of aquatic environments

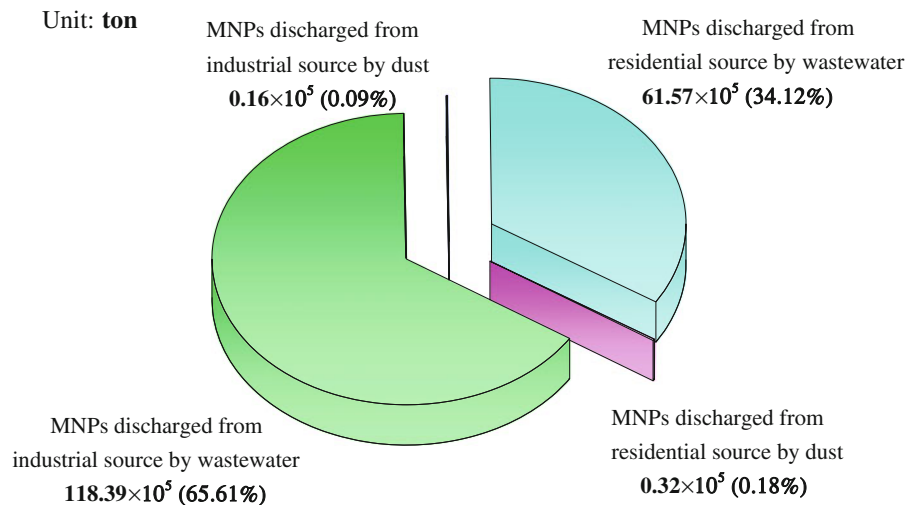
As Fig. 5 illustrates, MNPs discharged from industrial wastewater in China will progressively increase alongside increasing nanomaterial consumption, predicted at 100 billion United States dollars by 2017 and reaching and output of  $118.39 \times 10^5$  tons. Wastewater will also continue to be the primary pathway for

MNP distribution. Industrial source discharge will account for 65.16 % of total MNP discharge by 2017, a significant decrease compared to 2010 levels. Moreover, the ratio between MNPs discharged from residential sources via wastewater to total MNP discharge will significantly increase compared to 2010 levels (by nearly 40 %). MNP discharge from industrial and residential sources via dust will remain low when compared to total future MNP discharge, although this is expected to increase with increasing nanomaterial consumption.

Air  $PM_{2.5}$  (particulate matter with an aerodynamic diameter  $\leq 2.5 \mu m$ ) pollution has recently aroused concern with the Chinese government. Regional haze affecting cities in southern China has also recently been considered a weather event of great concern. The Chinese government has come to recognize the importance of protecting the environment while developing the economy. As such, it approved the first national environmental standard for limiting  $PM_{2.5}$  (Zhang et al. 2012). In light of this, it seems the Chinese government is currently committed to paying greater attention to  $PM_{2.5}$  pollution. MNP dust discharge will more likely be addressed in future initiatives in China.

For this study, China was used as a model where risks to aquatic environments by MNP exposure were directly associated with consumer products. Potential MNP exposure was calculated through wastewater and dust. Because precise MNP classifications and distributions could not be obtained for aquatic environments in China due to a general lack of data, a summary of the available data was applied instead.

**Fig. 5** MNPs discharged by wastewater and dust deposition into China’s aquatic environments in 2017, calculated by means of relationships ascertained between nanomaterial consumption increases and MNP pollution (as it relates to nanomaterial consumption in 2017)



Managing MNP release and exposure can be achieved through different exposure-reduction strategies, such as restricting its use, applying specialized disposal methods, using an ultrafiltration approach, and monitoring wastewater treatment processes as well the sludge applied to soils. The challenges China will be faced in the future are as follows: (1) how to define MNP discharged by industrial and residential sources; (2) how to address risk assessments and responsibility when MNPs impact human health; (3) whether to amend existing legislation or develop a new regulatory framework by which to determine MNP pollution risk; and (4) how to control MNP discharge and determine subsequent distribution to aquatic environments.

## Conclusion

Although attempts have been made to explicate behavioral and toxicological conditions on a number of MNPs contaminating aquatic environments, insufficient understanding related to the release, transport, transformation, and fate of MNPs in such environments remains unknown. This is largely due to a general lack of relevant data. Specifically, as nanotechnology promotes emerging MNP nanomaterials and they increase in number, understanding their behavior and effects on aquatic environments will remain challenging. Emerging MNPs are handled and disposed of as hazardous waste in the vast majority of research and industrial processes today. However, very little relevant data regarding the containment, commercial product usage, or disposal of MNPs are currently available. To assess and ultimately reduce the apparent risks posed by emerging MNPs on the environment at large and human health in general, potential exposure of these materials must be taken into account. Future methodological development includes refining behavioral relationships and applying a quantitative basis to transport and persistence ranking as well as employing relevant toxicological and regulatory limits on MNP concentrations in surface water.

Current data on the fate and behavior of many MNPs existing in environmental media integrated into consumer and industrial products and processes are insufficient to draw comprehensive and definitive conclusions when applying a traditional quantitative

risk assessment framework. Modeling techniques applying experimental fate, transport, and bioavailability data could be used to determine potential environmental MNP exposure and its short-term and long-term risks on aquatic environments. As MNP development progresses, it is hoped that at least the majority of MNPs will have no adverse effects on environments and human health under realistic exposure scenarios.

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