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Title: Potential Exposure and Treatment Efficiency of Nanoparticles in Water Supplies based on Wastewater Reclamation

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Highlights

- We present current knowledge on nanoparticle fate in wastewater reclamation systems for potable reuse

Nano impact (120 words)

In this article we present the first ever evaluation of the removal of nanoparticles in waste water and water treatment processes in order to estimate concentrations of NPs in reclaimed wastewater for potable reuse. Based on mass flow analysis of two specific water reclamation cases (i.e. Orange County and Berlin) and a literature review, we find that silver (Ag), titanium dioxide (TiO_2), and zinc oxide (ZnO) nanoparticles may occur in concentrations up to 3 µg/L (Ag), 147 µg/L (TiO_2), and 0.3 µg/L (ZnO). Critical research needs evolve around understanding the fate of nanoparticles treated by reverse osmosis, UV and disinfection processes and understanding which kinds of nanoparticles in various types of products end up in our water supply.

Abstract

Water scarcity brings an increased focus on wastewater reclamation for drinking water supply. Meanwhile, the production volume of nanoparticles (NPs) is rapidly increasing, but to date there has been little attention to the fate of NPs in water systems based on wastewater reclamation. We have investigated the possible concentrations of silver (Ag), titanium dioxide (TiO_2), and zinc oxide (ZnO) nanoparticles in tap water for water supplies based on reclaimed wastewater. Tap water concentrations of the NPs were assessed by mass flow analyses of two typical wastewater reclamation concepts: 1) advanced membrane treatment and 2) bank infiltration, similar to systems established in Orange County, CA, USA and Berlin, Germany. The mass flow analyses are based on a literature review of known wastewater concentrations of NPs and removal efficiencies for the implemented treatment stages in two case systems. Few studies are available on the removal efficiencies of NPs by advanced water treatment processes with a majority of identified studies focusing on removal efficiencies in wastewater treatment plants and fate in surface waters. The NP removal efficiency of several treatment processes is unknown at this stage. We found worst case removal efficiencies for the two cases to be 97-99.97% for Ag-NPs, 91-99.2% for TiO_2-NPs, and 92-93% for ZnO-NPs. Corresponding worst case concentrations in the tap water for the advanced membrane treatment were 0.04 µg/L (Ag), 147 µg/L (TiO_2), and 0.28 µg/L (ZnO). Concentration for ZnO-NPs also includes zinc ions and the concentration of ZnO-NPs is likely to be lower than indicated here. Worst case removal by the wastewater reclamation bank infiltration system was predicted to lead to tap water concentrations up to 3.3 µg/L (Ag), 13 µg/L (TiO_2), and 0.25 µg/L (ZnO). Overall, it is found that the primary removal mechanisms of NPs are aggregation, sedimentation, coagulation, and biosorption and this supports conventional biological treatment processes as likely effective barriers against NPs. Advanced treatment methods such as microfiltration and ultrafiltration can exhibit very low removal of ZnO-NPs or zinc ions caused by dissolution of ZnO-NPs. There are marked knowledge gaps, and further research in NP fate in water treatment is encouraged.

1 Introduction

Engineered nanoparticles (NPs) are used to an ever increasing extend e.g. in consumer products, but we know very little about how they are used and where they end up. According to The Nanodatabase (www.nanodb.dk), which is a database maintained by the Danish Ecological Council, the Danish Consumer Council and Department of Environmental Engineering at the Technical University of Denmark, more than 1200 products claimed to be based on nanotechnology or containing nanomaterials are now available to the European consumers on-line. 52% of these products entail nanoparticles suspended in liquids e.g. personal care products such as cosmetics and only about 16% and 7% have surface bound NPs or NPs suspended in solids and hence it is reasonable to assume that most of these compounds will ultimately end up in our wastewater treatment systems (Figure 1).

Treated wastewater ends up in recipients such as rivers, lakes, and oceans, where it may planned or unplanned (de facto) become the indirect source for drinking water supplies [1,2]. Although few large scale reclamation plants for potable reuse are operational, it has been suggested that direct potable reuse can play a much larger role in future solutions to water scarcity [3]. With occurrence of nanoparticles in wastewater it is therefore relevant to investigate their potential presence and effective treatment in drinking water.

In this paper, we estimate the concentrations of silver (Ag), titanium dioxide (TiO_2), and zinc oxide (ZnO) NPs that can be expected to end up in the water supply as a consequence of wastewater reclamation. NanoAg, nanoTiO_2 and nanoZnO was chosen as they are the most commonly used NPs in consumer products (Figure 2) [4,5]. These three NPs are often suspended in liquids when used in consumer products and hence can be expected to end up in the wastewater. Other nanoparticles which are produced in large quantities annually are carbon black and silicon dioxide[6]. However, carbon black nanoparticles are predominantly used in products (e.g. tires) which are disposed at landfills [7,8] while the silicon dioxide is predominately used in food products and is not viewed as a health hazard even in concentrations of 1500 mg/L [6]. Therefore, these nanoparticles are not assessed in this study.

2 Method

The exposure and fate of nanoparticles in water treatment was investigated by mass flow analysis of two typical wastewater reclamation concepts based on a review of the current knowledge on nanoparticle fate in water treatment systems. A literature review was conducted to find the current knowledge on typical NP concentrations in wastewater, and their fate and transformation in wastewater treatment processes, advanced wastewater treatment, surface water, drinking water treatment, and natural filtration through the soil column.

2.1 Two typical concepts for wastewater reclamation

We estimated the potential NP concentration in drinking water for two existing water reuse systems in Orange County, California, USA [3] and Berlin [1]. Orange County is situated in a water scarce region and relies partly on import of water from outside the area. The Municipal Water District of Orange County (MWDOC) base its water supply on 62% groundwater, 34% imported water, and 4% surface water [9]. Since the 1970’s the groundwater has been replenished by reclaimed wastewater. The wastewater reclamation system of Orange County (Figure 3) consists of a conventional wastewater treatment plant (Plant no. 1) which discharges its effluent to the Advanced Water Treatment Facility that is part of the Groundwater Replenishment System (GWRS) for the Orange County area. The Advanced Water Treatment Facility includes treatment methods such as disinfection, UV, microfiltration, and reverse osmosis. The effluent from the Advanced Water Treatment Facility is suitable for drinking and is subsequently delivered to the Santa Ana Groundwater Basin. After abstraction, the groundwater is disinfected before distribution to the public water supply [10].

Berlin’s water supply is less technology intensive than in Orange County. In Berlin, local groundwater is abstracted from local aquifers and then treated by aeration and sand filtration before distribution to the city (Figure 3B). The aquifers are recharged with water from the local rivers and lakes [11]. It is a “de facto” reclamation scheme because the same rivers and lakes also receives effluent from the local conventional wastewater treatment plants [12]. Because of the recognized link between wastewater effluents and the drinking water [13,14], Berlin has had a high awareness on keeping the state of the lakes and rivers healthy. The groundwater abstraction mainly occurs from soil layers dominated by sand and gravel in a depth of 30 to 50m below surface [12]. In the central area of Berlin, the sewage system is combined, e.g. rain water and wastewater are collected by the same pipes. In the event of an overflow in this system, some of the sewage water is discharged to the rivers untreated because of WWTP capacity overload. In the aftermath of such event, increased concentrations of usually well removed contaminants have been detected in the rivers while contaminants usually difficult to remove are found in lower concentrations in the rivers due to dilution [15].

2.2 Mass flow analysis

The removal efficiency by the two systems in Berlin and Orange County was assessed using mass flow analysis following the basic principles presented by Brunner [16]. The considered mass flow analysis is a simple model which is based on removal efficiencies identified for each of the treatment processes used in the investigated waste water reclamation systems. The setup of the model is that escaping nanoparticles from a previous treatment step is removed by the identified removal efficiency for a proceeding treatment process. Therefore, the model does not take concentration dependency of the identified removal processes into account e.g. It is well-known that removal by aggregation is concentration dependent.

In order to properly evaluate the NP fate in the reclamation concepts, the removal efficiencies of the three NPs by the treatment stages are assessed in two scenarios representing the worst and best case evaluations of the assumed removal efficiencies. The lowest removal efficiency in each range is assumed to represent the worst case removal scenario while the corresponding highest removal efficiency is used for the best case removal scenario. Where only a single value for the NP removal efficiency in a given treatment stage has been identified, this single value is assumed to be the removal efficiency in both the minimum and maximum removal scenarios. Where there is no documentation for the removal efficiency of a given NP by a given treatment stage, minimum removal is assumed to be 0% and maximum removal to be 100%.

3 Results

3.1 NP concentrations in wastewater

The estimation of potential end concentration of NPs in drinking water starts with the estimation of concentrations in the wastewater influent. Gottschalk et al. [17] modelled concentrations of Ag-NPs, TiO_2-NPs, and ZnO in WWTP effluent for US, EU, and Switzerland. Tiede et al. [8] used different forms of modeling to calculate concentrations for TiO_2-NPs and Ag-NPs in the WWTP effluent which again are based on the use, product concentration, and fate estimations reported by Boxall et al. [18], Mueller & Nowack [19], and Gottschalk et al. [17]. From their results we assumed WWTP influent concentrations of 107.2 µg/L for Ag-NPs, 1,636.4 µg/L for TiO_2-NPs, and 3.6 µg/L for ZnO-NPs.

3.2 Fate of NPs in treatment processes

When it comes to understanding and mapping what happens with the NPs in treatment processes, the specific fate and transformation process in the sewer, the WWTP, microfiltration, etc. is of vital importance. Out of 71 studies identified for the period 2008-2013 (Figure 4) wastewater treatment plants (27) and surface water (17) has received most attention. Then come microfiltration (7), drinking water treatment plants (7), ultrafiltration (6), and soil and groundwater (6). Sewers, reverse osmosis, UV and disinfection have been covered by 0-1 study each.

The estimated removal efficiencies of Ag-NPs, TiO_2-NPs, and ZnO-NPs by each treatment stage identified in the literature are presented in table 1 and will be discussed in the following in the light of Orange County and Berlin.

3.2.1 Sewer

Kaegi et al. [20] observed that due to the much larger surface areas, the Ag-NPs will tend to attach to the larger surface of suspended solids rather than the biofilms in the sewage pipes. Therefore, it is likely that about 0% of Ag-NPs will be retained in the sewers although a portion of the Ag-NPs might have become sulfidized upon arrival at the WWTPs. No identified studies have investigated the fate of TiO_2-NPs and ZnO-NPs in the sewers.

3.2.2 WWTP

The removal efficiency of Ag-NPs by WWTPs has been found to be in the range 39-99.9% for varying conditions related to NP surface coating and concentration of TSS [17,19–24]. However, peak loadings of Ag-NPs to the WWTP, may cause a large fraction of the Ag-NPs to end up in the effluent due to constraints in sorption kinetics and capacity of the biosolids to which the Ag-NPs could be attached/absorbed [22]. Musee et al.[25] found that Ag-NPs have a high attraction to the sludge and low solubility regardless of the pH-level of the wastewater. In general, activated sludge (e.g. heteroaggregation) is likely to be the main driver for retention of Ag-NPs [20–23,26,27].

Similar to the removal efficiency of Ag-NPs, increasing concentration of TSS is likely to result in increasing removal of the influent TiO_2-NPs[24,27]. Removal efficiencies of TiO_2-NPs at 23-97% depending on the type of surface coating and concentration of TSS have been observed [24,27]. Due to the very low solubility of TiO_2-NPs, the presence of ionic Ti is not expected [24]. The results presented by Johnson et al. [26] indicate that the primary treatment can remove about 13% and the secondary treatment (activated sludge) removes further 88%. Overall, Johnson et al. found the removal efficiency of TiO_2-NPs by WWTPs to be 90% [26].

Most of the ZnO-NPs are estimated to agglomerate and aggregate in WWTPs [28,29]. Overall, the removal efficiency of ZnO-NPs in WWTPs is likely to be 88-100% [17,28].

It should be noted that all three NPs are found to have a potential inhibitive effect on the microbial community in the WWTPs, at least in the short term until the microbes have adapted to the new compounds[21,24,25,28,30–34].

3.2.3 Microfiltration and Ultrafiltration

Micro- and ultrafiltration are key treatment stages in many wastewater reclamation schemes [2]. Although microfiltration and ultrafiltration membranes are likely to retain a significant large portion of NPs, a small fraction is still expected to break through. Abbott Chalew et al. found that 55-99% of Ag-NPs and 56-100% of TiO_2-NPs are likely to be retained by microfiltration membranes and that 98-100% of Ag-NPs and 96-100% of the TiO_2-NPs will be retained by ultrafiltration membranes [35]. However, the expected dissolution of ZnO-NPs to zinc ions at neutral pH-values are expected to cause a much lower removal efficiency of ZnO-NPs of 17-64% by microfiltration and 4-98% by ultrafiltration membranes although it is unclear how large a fraction is transformed to zinc ions [35] (Table 1). Ladner et al. found that the removal efficiency towards NPs of a membrane largely depends on the properties of the NP as well as membrane surface functionality [36]. Especially, the surface charge of the nanoparticles is essential as electrostatic repulsion/attraction with the typically negatively charged membranes is an important aspect in the overall removal efficiency. Moreover, the molecular weight cut-off (MWCO) of the membrane is an important parameter with higher MWCO providing an increasing risk for breakthrough of especially negatively charged nanoparticles. Overall, 0-10% of negatively charged Ag-NPs and TiO_2-NPs and about 100% of positively charged TiO_2-NPs were observed to be removed by microfiltration membranes (pore size 0.1-10 µm [37]). For ultrafiltration membranes (pore size 1-100 nm [37]), 60-90% of negatively charged Ag-NPs (including the 14% of Ag-NPs(-) which was dissolved), 95-100% of TiO_2-NPs(-) and about 100% of TiO_2-NPs(+) was observed to be removed [36]. The removal efficiencies found by Abbott Chalew et al. [35] are valid for pH=7-8 of the solution while the results by Ladner et al. [36] is for pH = 8.2-9.6. This indicates that the pH of the solution also has an effect on the removal efficiency of NPs by a given membrane.

In Orange County, the water is treated by microfiltration after the disinfection stage in the Advanced Water Treatment Facility. The pH-value of the microfiltration influent and effluent is 7.3 and 7.5, respectively [10]. Therefore, we assume that the removal efficiencies by Abbott Chalew et al. [35] should be applicable to the microfiltration units in the ATWF. It is noted that the removal efficiency in microfiltration/ultrafiltration membranes possibly can increase for ZnO-NPs by adjusting for higher pH as some results indicate [35].

3.2.4 Disinfection

When the effluent from the WWTP arrives at the Advanced Water Treatment Facility (AWTF) in Orange County, the first stage of treatment is disinfection by sodium hypochlorite. Yuan et al. found that by adding sodium hypochlorite to the water about 95% of Ag-NPs was removed irrespective of pH-level in the water [38]. Hydrogen peroxide and other disinfectants are also used in the AWTF, which might contribute further to the overall removal of NPs. Additionally, no studies, which investigated the corresponding removal efficiency of TiO_2-NPs and ZnO-NPs, were found.

In Orange County, the groundwater is typically disinfected prior distribution to the consumer. 14% of the extracted groundwater is treated by ultrafiltration and reverse osmosis among others before distribution [39]. However, for this mass flow analysis, these water treatment processes are disregarded at this stage due to their small share. In Anaheim, Orange County, sodium hypochlorite (12.5%) is used as the only treatment of groundwater prior to distribution (Personal communication) while the water utility in Irvine Ranch adds chloramines at about 3 ppm in order to keep a ratio of 4.5:1 chloramine to ammonia (Personal communication). The pH-level of the groundwater in Anaheim and Irvine Ranch is 7.9 and 8.2, respectively (Personal communication). However, Yuan et al. found that changes in pH-level would have little effect on the removal efficiency of Ag-NPs [38]. Therefore, the removal efficiency of the remaining Ag-NPs in the treatment of groundwater is estimated to be approximately 95%. The literature does not provide corresponding values for TiO_2-NPs and ZnO-NPs and therefore, their removal efficiencies are assumed to be 0% and 100%, respectively in the mass flow analysis.

3.2.5 Reverse Osmosis

To the best of our knowledge, no studies have been published on the effectiveness of RO when it comes to NPs. Due to its smaller membrane pore sizes, the removal efficiency by reverse osmosis can be expected to be better than the removal efficiencies for ultrafiltration. Studies of micropollutants have shown that molecules larger than the membrane pore size can pass a reverse osmosis membranes. It was found that the rejection rates of micropollutants by reverse osmosis could be influenced by other parameters than size exclusion, including feed water quality, fouling and membrane materials [40,41]. Therefore, the potential of reverse osmosis in retaining NPs might be less than one can expect from size exclusion alone. To account for this knowledge gap, the reverse osmosis removal rates for all three NPs were assumed between 0% and 100% in the mass flow analysis.

3.2.6 UV

After the reverse osmosis treatment process, the permeate water is treated by UV irradiation. At this stage, the pH of the water is at 5.7 [10]. Yuan et al. observed that at pH=5.2, UV radiation removes about 60% of the Ag-NPs in the water, but found no effect of the UV treatment on NPs at pH=7.5 [38]. We assume that the removal efficiency of Ag-NPs by the Advanced Treatment Water Facility’s UV treatment is likely to be up to 60%. Corresponding removal efficiencies were not found for TiO_2-NPs and ZnO-NPs.

3.2.7 Surface Water

The effluent from the WWTPs in Berlin is discharged to the surrounding rivers and lakes. Here, the NPs are either transported away from the city or transported through the soil down to the groundwater at the sites where bank filtration and artificial recharge is situated.

The effluent from the Advanced Water Treatment Facility in Orange County is pumped to the Kraemer, Miller, and Miraloma Basins or to the Talbert Seawater Intrusion Barrier at the coast. At the Kraemer, Miller, and Miraloma Basins, the treated water percolates to the groundwater while the treated water directed to the intrusion barrier is pumped into the groundwater reservoir. In both cases, the water is mixed with the existing groundwater. Due to the assumed short residence time in the basins, no interactions or transformation of NPs are expected.

Several studies have been made on the fate and transformation of Ag-NPs in surface waters. The potential retention of Ag-NPs in surface water has been predicted by Monte Carlo simulations to be around 50% [17,19]. The possible retention of TiO_2-NPs and ZnO-NPs in surface water has been assessed by experimental results and Monte Carlo simulations which estimate that 53-100% of TiO_2-NPs are likely to be retained while ZnO-NPs will be removed 24-75% [17,19,42,43]. The retention of Ag-NPs, TiO_2-NPs, and ZnO-NPs in surface water are likely to be determined by several factors such as the surface coating of the NPs, flow rate, and pH [17,19,42,43].The found percentage ranges for retention are estimated to be the best guess for Berlin while no retention is assumed in the basins in Orange County.

3.2.8 Soil and Groundwater

The unsaturated layers in Berlin and Orange County are predominantly sand and gravel, especially in the top layers [10,12,44]. However, silts and clays are also present in large quantities in certain areas of Orange County. The proportion mechanisms of sand, silt, and clay and particle route in the Santa Ana groundwater basin in Orange County is unknown. The removal efficiency by the unsaturated zone in both cases are estimated to be 26-71% for Ag-NPs and 5-99% for TiO_2-NPs [45–47]. Based on laboratory tests, the retention in the natural aquifers are expected to be governed by ionic strength, NOM, residual chloride, low grain sizes, and pH [43,45–48]. Based on the results produced by Keller et al., it is estimated that 72.7% of TiO_2-NPs and 21.5% of ZnO-NPs will sediment in groundwater media [43]. However, no studies were found on the potential effect by the interaction between groundwater and soil which could provide a different removal efficiency altogether. Nonetheless, it is assumed that sedimentation and sorption in soil is likely to act as another barrier for the transport of NPs through the soil layers.

Due to the sandy soil in the two areas[12,49], the removal efficiency for both Berlin and Orange County is estimated to be 50-71% for Ag-NPs in the unsaturated zone, 5-99% for TiO_2-NPs in unsaturated soil and 72% in saturated soil layers, and about 21.5% for ZnO-NPs in saturated soil layers[43,45–47].

3.2.9 Water Treatment Plants

A handful of studies have investigated the possible removal of NPs by conventional drinking water treatment. Z. Li et al. observed that a sand filter retained about 40% of Ag-Citrate-NPs, about 25% of ZnO-PVA-NPs, and 0% of Ag-PVPNPs [50]. However, all the uncoated NPs examined, namely TiO_2-NPs, ZnO-NPs, and CeO2-NPs, were almost 100% retained by the sand filter. Moreover, the removal efficiency of each investigated type of NP will most likely be constant once a given pore volume has been reached but at various C/C_0-values (C = effluent concentration, C_0 = influent concentration) [50]. In addition, Z. Li et al. developed a model based on the experimental results for filter sand and ran simulations for two cases of NP-influent: single spiked input and continuous flow input [50]. The sand filter was observed to retain a large amount of the NPs in the event of a single spiked input (influent concentration of NPs of 50-500 ppm) but if a continuous flow (influent concentration of NPs of 5-50 ppm) was simulated, almost a 100% breakthrough rate was observed. Z. Li et al. explains this phenomenon as being caused by steric repulsion of the coated NPs [50].

Abbott Chalew et al. found the average removal efficiency of Ag-NPs by traditional water treatment plants to be around 80-98% [35]. For TiO_2-NPs, the average removal efficiency was found to be in the region of 92-97% for the simulated traditional water treatment, while for ZnO-NPs it was found to be 1-52%. The removal efficiencies for ZnO-NPs includes zinc ions, which indicate that the removal of ZnO-NPs is likely to be greater than presented. By comparing the observed removal efficiencies by Z. Li et al. [50] and Abbott Chalew et al. [35], aeration should remove 50% or more of the NPs in the influent to the water treatment plant.

The six drinking water treatment plants in Berlin treat the extracted groundwater by conventional processes such as sand filter, primary treatment, and secondary treatment. Li et al. found that less than 50% of surface coated NPs will be retained by sand filters [50]. As many engineered NPs are surface coated, this is likely to be the removal efficiency of sand filters in most cases. Abbott Chalew et al. found removal efficiencies for conventional drinking water treatment processes of Ag-NPs, TiO_2-NPs, and ZnO-NPs to be 80-98%, 92-97%, and 1-52%, respectively [35]. In Orange County, the extracted groundwater is treated by disinfection which is covered in section 3.2.6.

3.3 Mass Flow Analysis

3.3.1 Orange County

Our results show that Ag-NPs are almost completely removed (99.97%) by the wastewater reclamation system in Orange County primarily due to the effectiveness of the processes in the conventional wastewater treatment plant and the disinfection stage in the Advanced Water Treatment Facility (figure 5). The worst case removal scenario predicts that about 9% of TiO_2-NPs and 8% of ZnO-NPs will pass through the treatment processes and may end up in the tap water.

Figure 6 depicts the concentrations after treatment by each treatment stage in Orange County. The estimated worst case concentration of TiO_2-NPs in the influent to the WWTP is predicted to be 147 µg/L while ZnO-NPs and Ag-NPs were predicted in concentrations of 280 ng/L and 37 ng/L, respectively.

3.3.2 Berlin

The overall removal of NPs in Berlin was estimated based on the assumed removal efficiencies for the treatment stages in Table 1. The results show that the Berlin reclamation sequence is most efficient in retaining TiO_2-NPs. The system is also estimated to retain almost all of the Ag-NPs arriving at the WWTPs.

ZnO-NPs seem to be the most difficult NP of the three to remove (figure 7). Especially, the usage of conventional wastewater treatment seems to be crucial in order to ensure high removal efficiency.

The estimated concentrations of the three NPs in the tap water to the consumers in Berlin indicate that TiO_2-NPs can be found in the largest concentrations followed by Ag-NPs. The worst case scenario predicts that all three NPs may end up in the tap water in concentrations between 0.25-13 µg/L (figure 7). In addition, the system in Berlin utilizes surface water treatment plants (SWTPs) which treats the river water in order to minimize the concentration of phosphorous among others in the local surface water. In these plants flocculation, sedimentation, and filtration occurs [12]. These processes are likely to contribute to the overall NP removal efficiency of the system in Berlin. However, no information regarding the treated volume could be found. The SWTPs are therefore not included in the mass flow analyses.

4 Discussion

4.1 Evaluation of the Wastewater Reclamation Systems in Orange County and Berlin

The mass flow analyses in the previous sections of the wastewater reclamation systems in Orange County and Berlin indicate that considering a worst case scenario, the advanced treatment currently in operation in Orange County is likely to be more efficient towards Ag-NPs than the corresponding system in Berlin while the opposite is the case for TiO_2-NPs and ZnO-NPs (Table 2)

The concentration of TiO_2-NPs in the tap water in the worst case scenario for Berlin is less than 10% of the corresponding concentration in Orange County. On the other hand, the system in Orange County has a worst case overall treatment efficiency of 99.97% for Ag-NPs, whereas Berlin is predicted to remove just 97% of the Ag-NPs. For ZnO-NPs, the found concentrations in the worst case scenario are almost equivalent for both systems.

The two disinfection stages (sodium hypochlorite) in the wastewater reclamation system in Orange County are the main cause for the higher worst case removal efficiency of Ag-NPs compared to the system in Berlin. For Ag-NPs, the difference in worst case removal efficiencies for the two systems are mainly a 95% removal efficiency of the two disinfection stages and 55% removal efficiency of the microfiltration treatment in Orange County versus a 50% and 80% removal by the surface water and WTP respectively, in the Berlin system. Therefore, size exclusion by microfiltration [35]and especially oxidative dissolution and aggregation of Ag-NPs by disinfectants [38] seem to be a more effective barrier than ones provided by the surface water (aggregation) and drinking water treatment plants (coagulation and bioadsorption) in Berlin [35,42,43,50,51].

The primary reason for the difference in the worst case removal efficiency of TiO_2-NPs by the two wastewater reclamation systems are due to the processes in surface waters and water treatment plants in Berlin. The found worst case removal efficiencies of TiO_2–NPs by the advanced treatment in Orange County are down to the lower minimum removal efficiency by microfiltration. The different removal mechanisms provide the system in Berlin with the advantage as no removal efficiency of TiO_2-NPs by disinfection, reverse osmosis, or UV could be identified. The differences between microfiltration treatment used in Orange County and the processes occurring in water treatment plants and rivers are predominantly due to the removal mechanism in microfiltration being reliable on size exclusion [35] while the natural attraction of TiO_2-NPs to the particles and matter in surface water (including mutual attraction between the TiO_2-NPs) [42,43,51] as well as the coagulation [35] and bioadsorption [50] in conventional water treatment are observed to more effective.

The difference between the found worst case removal efficiencies of ZnO-NPs for Berlin and Orange County is the smallest compared to the corresponding for Ag-NPs and TiO_2-NPs. However, the WWRS in Berlin is still estimated to be slightly more effective than the corresponding in Orange County due to slightly higher overall estimated worst case removal efficiency of ZnO-NPs by surface waters and WTPs than the corresponding by microfiltration.

4.2 The known barriers against NPs

The literature search results indicate that ultrafiltration provides the best removal efficiency of Ag-NPs (98-100% removed) due to size exclusion [35]. Moreover, the heteroaggregation occurring in treatment processes in WWTPs and drinking water treatment plants are likely to provide a significant retention of Ag-NPs. The dominant removal mechanism in WWTPs suggested by the identified studies is the natural attraction of Ag-NPs to the total suspended solids in the medium and the bacteria in the activated sludge [20,30]. Disinfection and microfiltration are also likely to retain a large ratio of Ag-NPs. UV disinfection was effective predominantly due to dissolution and aggregation of the Ag-NPs [38].

By similar comparison, ultrafiltration is likely to be the most efficient barrier towards TiO_2-NPs (96-100% removed) due to size exclusion [35]. However, the coagulation, sedimentation, and flocculation in the water treatment result in a high removal efficiency of TiO_2-NPs (92-97%). Furthermore, high single removal efficiency of TiO_2-NPs were documented for WWTPs, surface waters, soil, and microfiltration which highest estimated removal efficiency of TiO_2-NPs were found to be 97%, 99.9%, 99%, and 100%, respectively. The aggregation and biosorption of TiO_2-NPs in WWTPs were observed to be the primary removal mechanism [24,27]. The removal efficiency of TiO_2-NPs in (sandy) soil was very much dependent on the pH-value of the soil which could lead to almost negligible removal efficiency (5%). Moreover, if the soil has a high dissolved organic carbon content a high removal efficiency (>95%) can be expected due to the composition of the clay. Size exclusion of aggregated TiO_2-NPs due to the pore size of the microfiltration membrane was the main reason for its high removal efficiency [35]. In surface waters with acidic conditions and low flow rate (3.2 L/s), the sedimentation of TiO_2-NPs is found to be almost 100% regardless of surface coating. However, by increasing the pH and flow rate, the TiO_2-NPs are more difficult to settle [42].

The processes in the WWTP is likely to be the most efficient barrier against the breakthrough of ZnO-NPs. Hou et al. found that about 70% of ZnO-NPs (no surface coating) are likely to rapidly settle in the primary clarification tank and the remaining ZnO-NPs are suggested to be completely removed by processes in the aeration and activated sludge treatment stage [28]. The primary removal mechanism was found to be biosorption. The rapid settling is also observed in surface water media where 8 hours of flocculation and 1 hour of sedimentation of ZnO-NPs is likely to result in 75% removal of ZnO-NPs [51]. However, a removal efficiency of ZnO-NPs of 24% in river water media indicates that the flow rate are the primary parameter for the removal of ZnO-NPs in surface water [43]. Ultrafiltration and microfiltration can also exhibit a high removal efficiency of ZnO-NPs (64% and 98%, respectively) but due to the dissolution of ZnO-NPs into zinc ions, these barriers are also observed to exhibit very low removal efficiencies (17% and 4%, respectively) [35].

In general, the identified studies highlights aggregation – and especially heteroaggregation – and size exclusion as essential removal mechanisms in waste water and water treatment [20,52].

4.3 Knowledge gaps

In our attempt to complete a mass flow analysis for potable water reclamation in Orange County and Berlin, a marked lack of observations from several treatment stages and compartments is evident. This prevents an accurate estimation of the fate of the investigated NPs. Of the three NPs in question, ZnO-NPs have received least attention. Only single observations were found for several combinations of NP type and treatment stage, for example ZnO-NPs in WWTP, and groundwater (Table 1). Ranges of removal efficiency of Ag-NPs and TiO_2-NPs were found for wastewater treatment, microfiltration, and soil layers whereas only single observations for the removal efficiency of Ag-NPs and TiO_2-NPs could be found for disinfection and groundwater, respectively.

No studies which examined the removal efficiency of NPs by reverse osmosis were identified. The pore size of the reverse osmosis membranes is below 0.1 nm and it is justified to assume that the reverse osmosis membranes will have a removal efficiency at least equal to ultrafiltration membranes or better. But as

Bellona et al. [40] found, rejection at the membrane might not be as straightforward, because size exclusion of compounds may not be the only parameter controlling rejection. In addition, primarily due to their tendency to dissolve into zinc ions, ZnO-NPs have been observed to be difficult to remove by size exclusion[35] and further studies are needed in order to determine the exact concentrations of ZnO-NPs which are able to breakthrough the barriers in a given system. In the review of the identified studies, the WWTPs were found to exhibit the highest removal efficiency of ZnO-NPs. Moreover, WWTPs are also likely to exhibit a relative high removal efficiency of Ag-NPs and TiO_2-NPs which indicate that the biological processes in the WWTPs at the current knowledge level appears as the most efficient NP barrier in wastewater reclamation systems.

In general, further studies are needed in order to attain more knowledge of the fate of NPs in the various treatment stages. Only the studies of the removal efficiency of the three NPs by WWTPs and surface waters can be regarded as providing a minimum level of nuanced understanding of the fate and behavior of NPs. This indicates that the primary focus from the research community has been on the release to and fate in the environment and to lesser extent the risk of exposure to humans through drinking water. Therefore, knowledge is lacking on the removal efficiency of more advanced treatment processes, with no observations reported for reverse osmosis. Furthermore, the studies evaluated in this study have primarily been conducted in laboratory settings or by modelling and simulation, which might provide a distorted perception of the transformation of NPs in real environments which is also highlighted in the work by Garner and Keller[52].

Finally, an important knowledge gap lies in the estimation of the influent concentration. The influent concentration of each NP in the mass flow analyses are based on results found by Tiede et al. [8] and Gottschalk et al. [17] and are based primarily on the observed behavior of NPs in the various treatment processes and previous results from similar studies as well as assumptions on the production volume and WWTP influent concentration of NPs. Although, we do believe that these concentrations represent the best known emission concentrations of NPs, it should be acknowledge that different estimations are available and that influent and effluent estimations will of course depend on fundamental assumptions about, for instance, production volumes[53].

5 Conclusions

Based on a mass flow analysis of possible nanoparticle fate and treatment in two typical potable reuse systems, we have found that:

- Considering a worst case scenario, nanoparticles may reach the drinking water supply in ng/L to µg/L concentrations after both advanced membrane based reclamation and simpler conventional water treatment.
- There are marked knowledge gaps and actual removal efficiencies for several combinations of nanoparticle and treatment stages are largely unknown.
- Observations reported so far support biological treatment processes as the most efficient engineered barriers against nanoparticle in wastewater reclamation systems for potable reuse.

Whether the estimated concentrations pose a risk for humans is yet to be determined.

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Figure text

Figure 1. Number of products with registered location of nanoparticles (nanodb.dk).

Figure 2. Number of registered nanoparticles grouped by product category (nanodb.dk).

Figure 3. Conceptual diagram of the wastewater reclamation systems in A Orange County and B) Berlin [1,2,54].

Figure 4. Number of published studies dealing with NP fate in water treatment processes grouped by year. Retrieved from the International Council on Nano Technology ICON database and Web of Science. 'Other' refers to publications dealing with NPs different from Ag-, TiO_2-, and Zn-NPs.

Figure 5. Estimated accumulated percentage of removed NPs after each treatment stage in the Orange County wastewater reclamation system for the removal scenarios: Minimum and maximum. Where no removal efficiency has been found in the literature it is assumed that 0% was removed for the minimum scenario and 100% removed for the maximum scenario. It is assumed that no NPs are removed in the sewers.

Figure 6. Concentration of NPs (μg/L) after each treatment stage in the Orange County wastewater reclamation system for the removal scenarios: Minimum, and maximum. The estimated concentrations of Ag-NP and TiO_2 in the influent to the WWTP are based on the study by Tiede et al. [8] while the corresponding concentration of ZnO-NPs is based on the study by Gottschalk et al. [17].

Figure 7. Estimated accumulated percentage of removed NPs after each treatment stage in the Berlin wastewater reclamation system for the minimum and maximum removal scenarios. Where no removal efficiency has been found in the literature it is assumed that 0% was removed for the minimum scenario and 100% removed for the maximum scenario. It is assumed that no NPs are removed in the sewers. WTP: Conventional drinking water treatment.

Figure 8. Concentration of NPs (µg/L) after each treatment stage in the Berlin wastewater reclamation system for the removal scenarios: Minimum, and maximum. The estimated concentrations of Ag-NP and TiO_2 in the influent to the WWTP are based on the study by Tiede et al. [8] while the corresponding concentration of ZnO-NPs is based on the study by Gottschalk et al. [17]. WTP: Conventional drinking water treatment.

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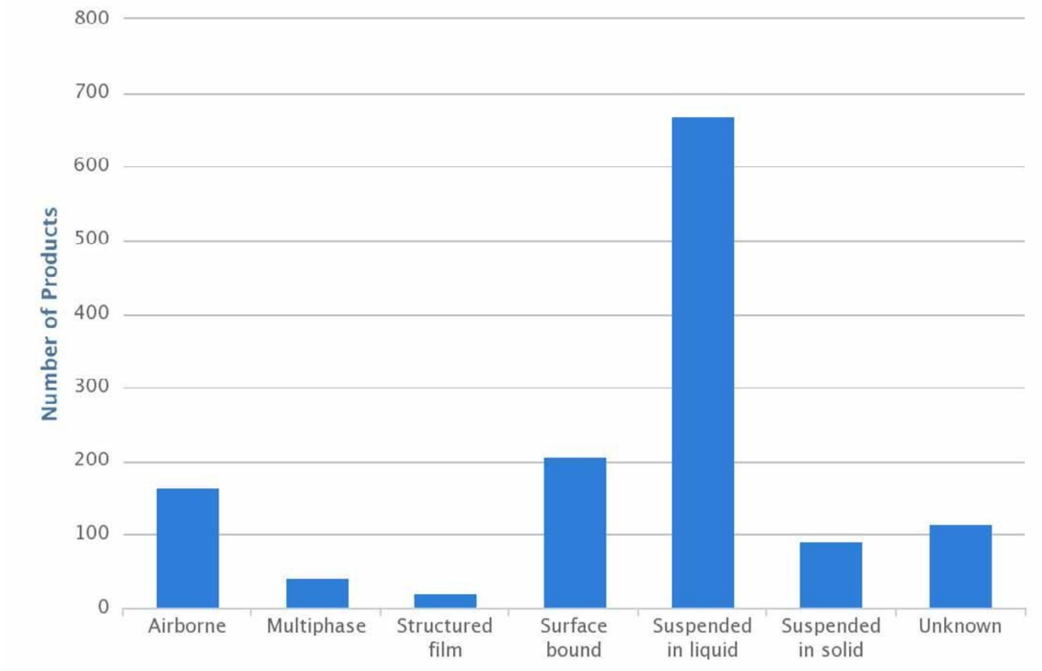
Table 2. Summary of estimated removal efficiencies and worst case concentrations of each NP by the wastewater reclamation system in Orange County and Berlin.



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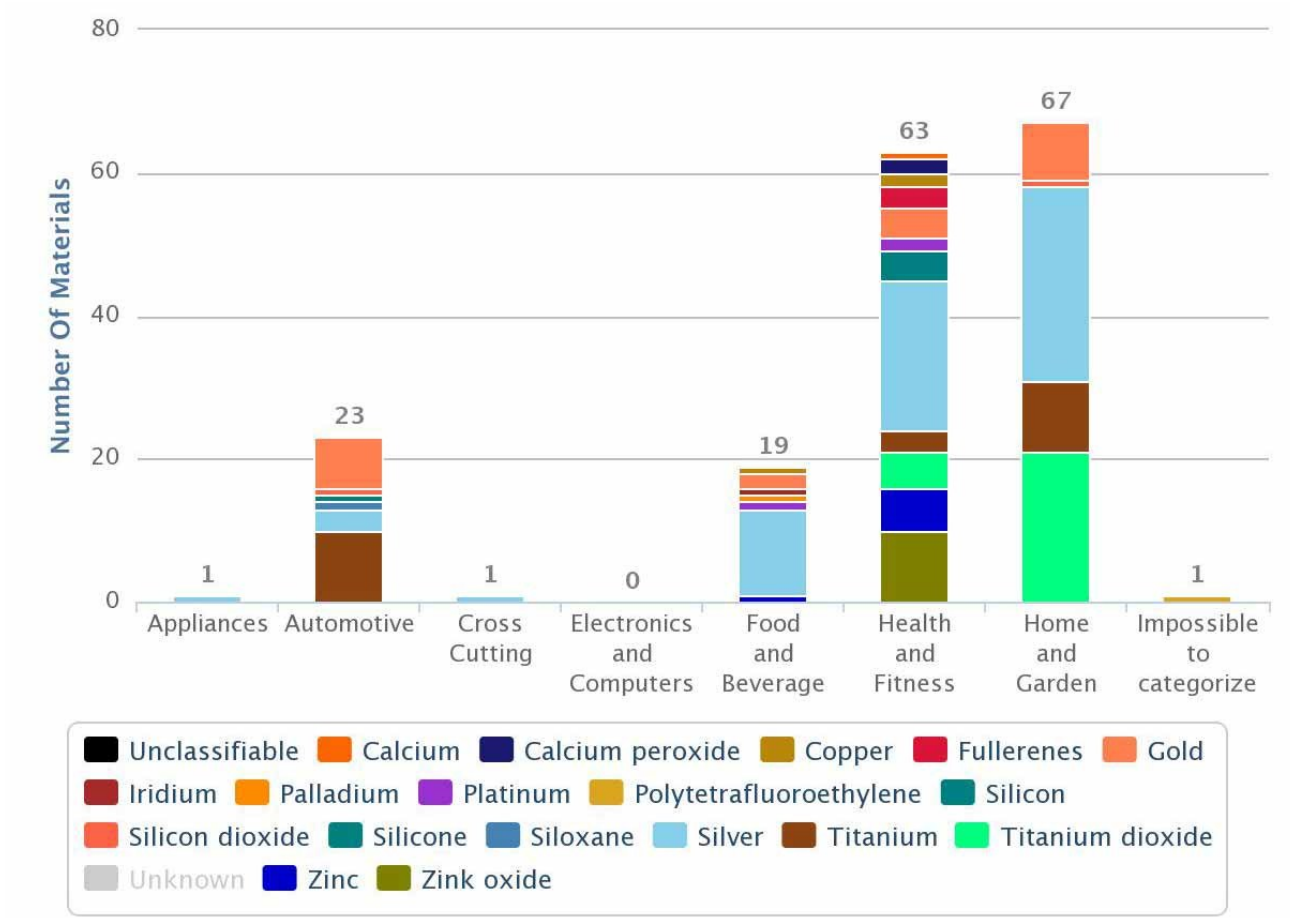

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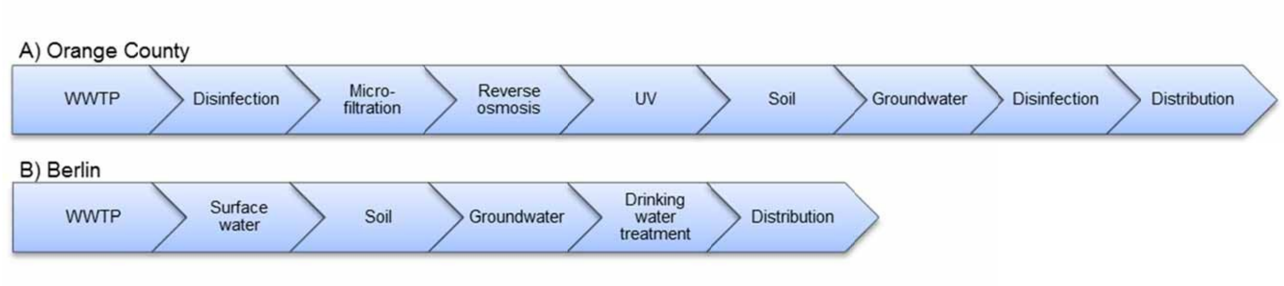

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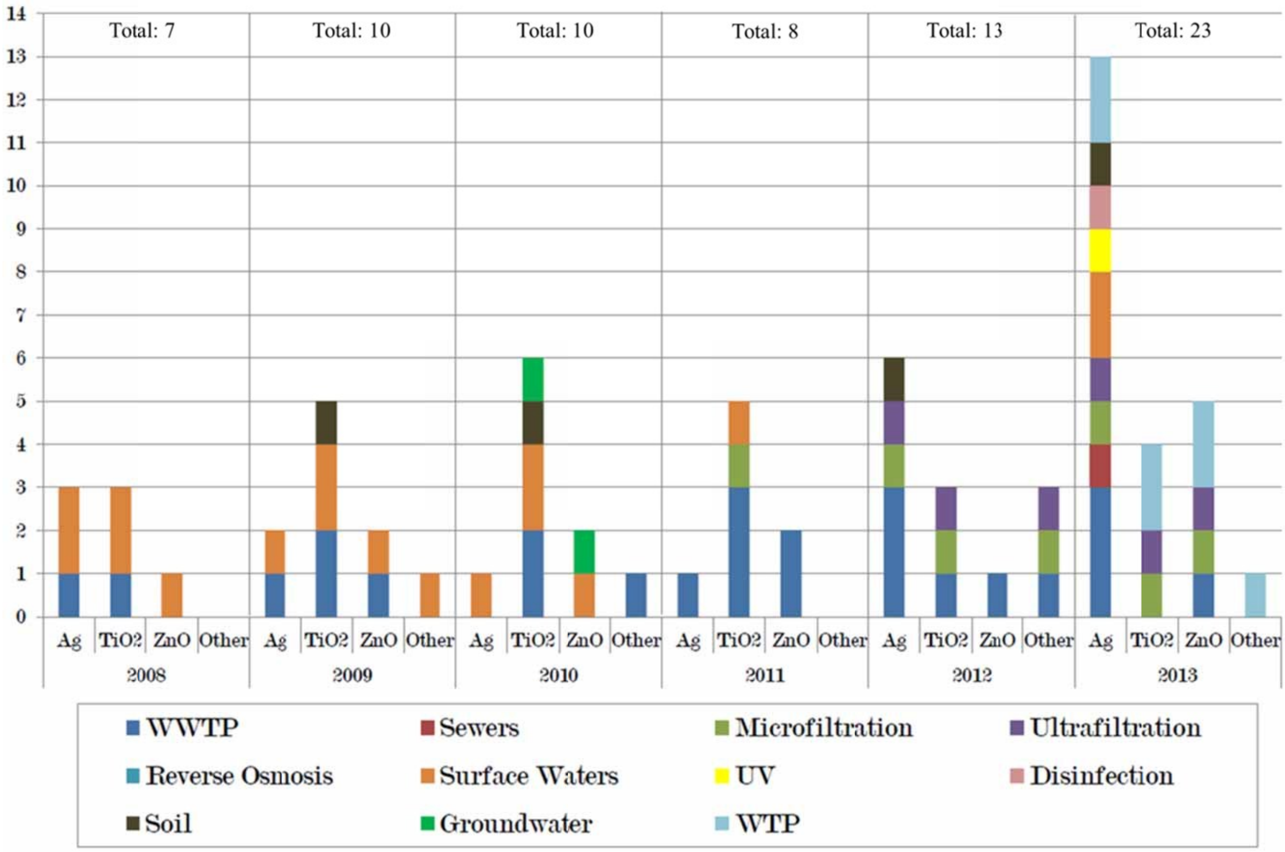

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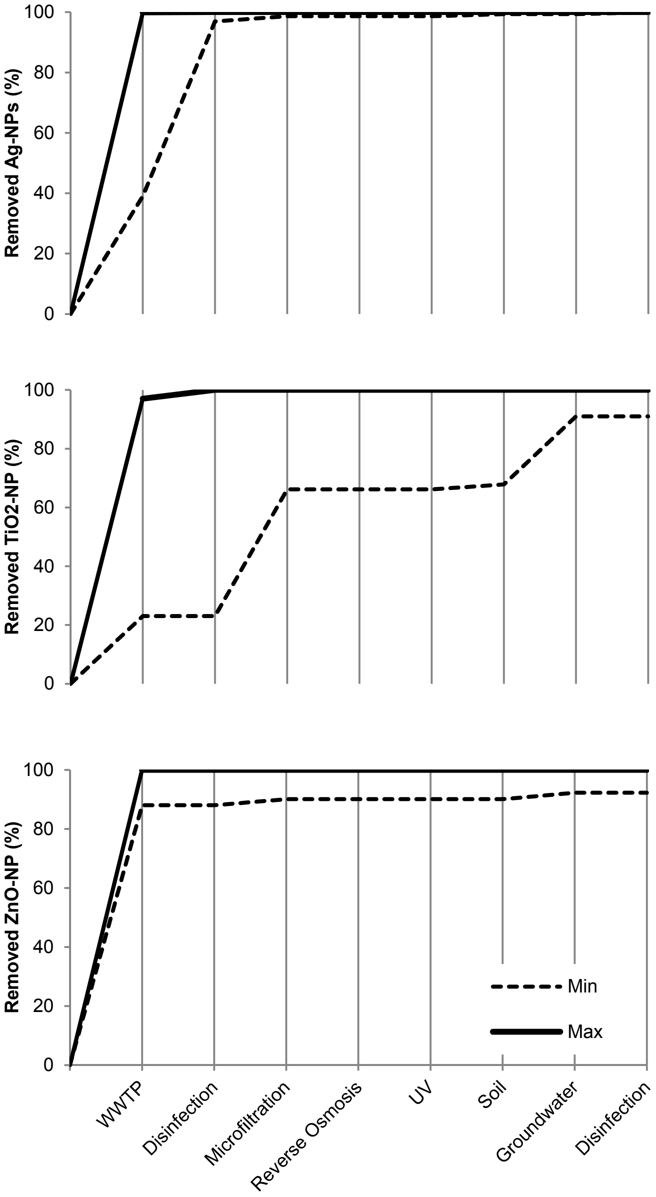

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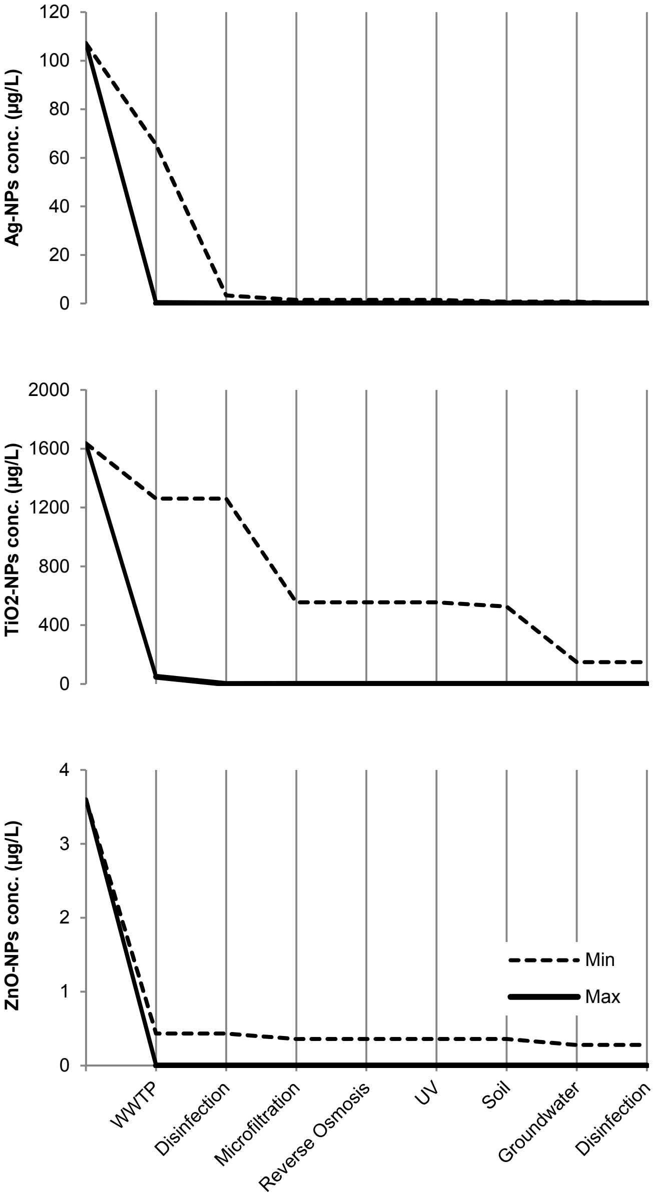

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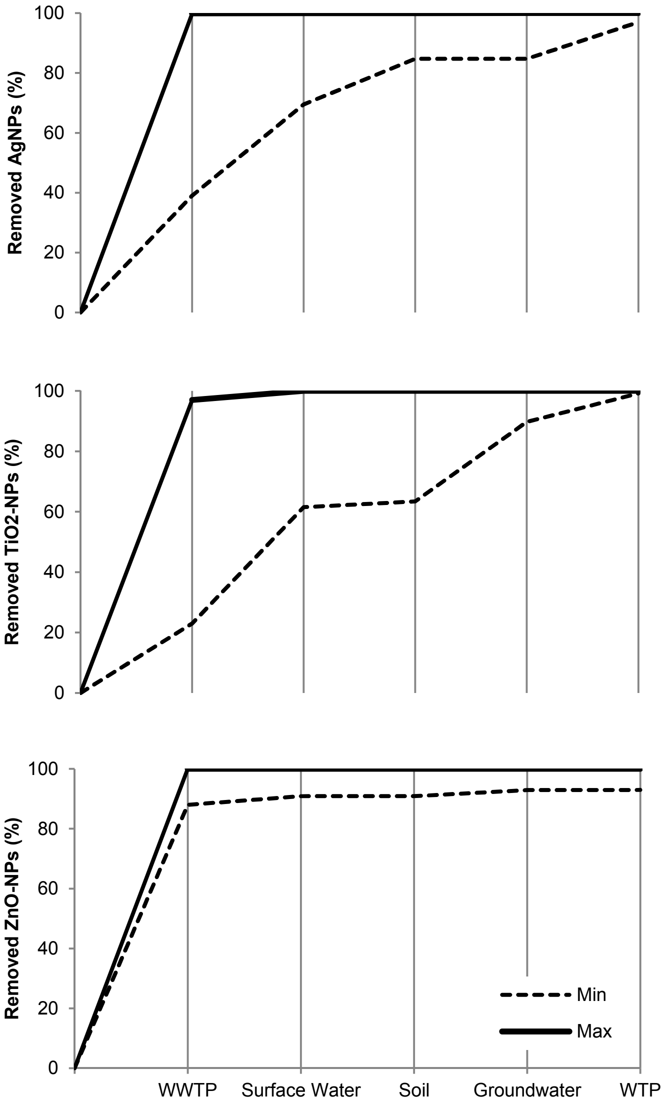

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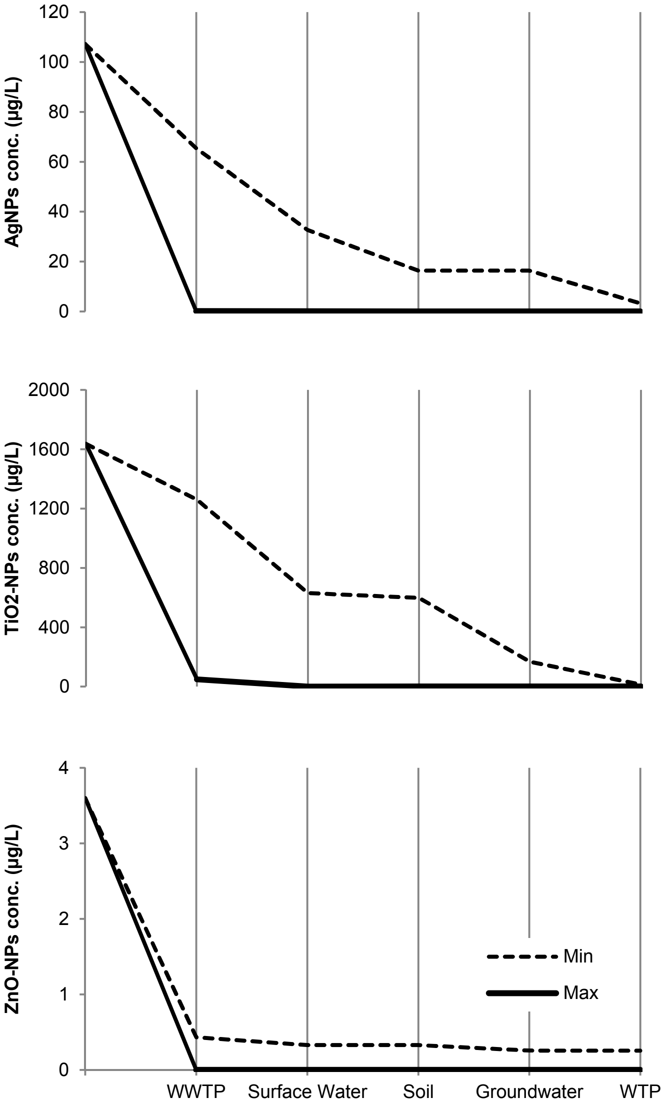

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Table 1. Overview of removal efficiencies for Ag-NPs, TiO_2-NPs, and ZnO-NPs in water treatment reported from field studies (field), laboratory experiments (lab), and computer simulations (sim).

Stage	Reference	Comments	Ag	TiO_2	ZnO	Type of study	Considered in the mass flow analysis
Sewers	(Kaegi et al., 2013)	The Ag-NPs are observed to be stabilized by adsorptions to sulfides and suspended solids.	0	-	-	Lab.+ field	Yes
Wastewater Treatment Plants	(Kaegi et al., 2013)	Overall, sedimentation, aggregation and adsorption are identified as the primary mechanisms for the removal of NPs. Increasing concentration of Total Suspended Solids (TSS) is observed to increase the removal of NPs (M. a Kiser et al., 2010; Wang et al., 2012). In addition, Li et al (L. Li et al., 2013) found the removal efficiency of mechanical treatment and biological treatment to be 35% and 72%, respectively.	98.9-99.9	-	-	Lab.+ field	Yes
	(Jeong et al., 2012)		70-90	-	-	Lab.	Yes
	(L. Li et al., 2013)		95	-	-	Lab.	Yes
	(Wang et al., 2012)		39-59 84-92	65-98	-	Lab.	Yes
	(M. a Kiser et al., 2010)		-	23-88	-	Lab.	Yes
	(Mueller and Nowack, 2008)		81-92*	81-87.6*	-	Sim.	Yes
	(Gottschalk et al., 2009)		~76*	~76*	~88*	Sim.	Yes
	(Gottschalk et al., 2010)		-	~86.6*	-	Sim.	Yes
	(Johnson et al., 2011)		-	~89.5	-	Field	Yes
	(Hou et al., 2013)		-	-	~100	Lab.	Yes
Microfiltration	(Abbott Chalew et al., 2013)	Abbott Chalew et al. reports for pH 7-8 and Ladner et al. reports for pH 8.2-9.6.	55-99	56-100	17-64	Lab.	Yes
	(Ladner et al., 2012)		0-10	0-10 ~100	-	Lab.	No$^{(1)}$
Ultrafiltration	(Abbott Chalew et al., 2013)	Abbott Chalew et al. reports for pH 7-8 and Ladner et al. reports for pH 8.2-9.6.	98-100	96-100	4-98	Lab.	Yes
	(Ladner et al., 2012)		60-90	95-100	-	Lab.	No$^{(1)}$
Reverse Osmosis	N/A	It is expected that that reverse osmosis should have removal efficiency better than ultrafiltration (Abbott Chalew et al., 2013; Ganzleben et al., 2011) actual removal efficiency has not been investigated.	N/A	N/A	N/A	N/A	
UV	(Yuan et al., 2013)	Oxidative dissolution was found to be the dominant reaction when exposing Ag-NPs to disinfectants. In addition, the level of pH, sodium nitrate, humic acid and the type of disinfectant is important for predicting the removal of NPs by disinfection.	17-67*	-	-	Lab.	Yes

Stage	Reference	Comments	Ag	TiO_2	ZnO	Type of study	Considered in the mass flow analysis
Disinfection	(Yuan et al., 2013)		95*	-	-	Lab.	Yes
Surface Waters	(Gottschalk et al., 2009)	The dominant removal mechanisms in surface waters seem to be low pH and flow rate as well as increased flocculation, sedimentation, aggregation, and electrostatic attraction.	~50	~99	~70	Sim.	Yes
	(Gottschalk et al., 2010)		-	~50	-	Sim.	Yes
	(Ticiana Boncagni et al., 2009)		-	~100	-	Lab.	Yes
	(Zhang et al., 2008)		-	53-75*	~75*	Lab.	Yes
	(Keller et al., 2010)		-	~77*	~24*	Lab.	Yes
Soil	(Sagee et al., 2012)	In general, the studies observed that low grain sizes, low concentration of humic acid, low flow conditions, low pH-value, and high ionic strength of the soil promotes electrostatic attraction and mechanical straining of NPs.	22-80*	-	-	Lab.	Yes
	(Fang et al., 2009)		-	17-99.8	-	Lab.	Yes
	(Solovitch et al., 2010)		-	5-99	-	Lab.	Yes
Groundwater	(Keller et al., 2010)	Increasing ionic strength and concentration of Total Organic Carbon (TOC) promote retention of NPs.	-	72*	21,5*	Lab.	Yes
Water Treatment Plants	(Abbott Chalew et al., 2013)	Sand filtration is observed to remove 100% of uncoated NPs while below 40% of coated NP (depending on type and surface coating) can be removed by sand filters. Moreover, sand filtration seems to be susceptible towards a continuous influent seems resulting in a larger breakthrough of NPs than in a peak flow (Z. Li et al., 2013).	80-98	92-97	1-52	Lab.	Yes(2)

*The removal efficiencies are read from graphs, tables or data. (1) Not considered since the microfiltration influent and effluent in the ATWF of Orange County has pH 7.3 and 7.5 (GWRS, 2013). (2) The conventional water treatment simulated by Abbott Chalew et al. (Abbott Chalew et al., 2013) is based upon coagulation and the removal efficiencies is therefore not necessarily the correct removal efficiencies for the drinking water treatment plants in Berlin.

Table 2. Summary of estimated removal efficiencies and worst case concentrations of each NP by the wastewater reclamation system in Orange County and Berlin.

		Ag	TiO_2	ZnO
Removal Efficiency Range (%)	Orange County	99.97-100	91-100	92-100
	Berlin	97-100	99.2-100	93-100
Worst Case Concentrations in Tap Water (μg/L)	Orange County	0.04	147	0.28
	Berlin	3.3	13	0.25