



## Is there evidence for man-made nanoparticles in the Dutch environment?



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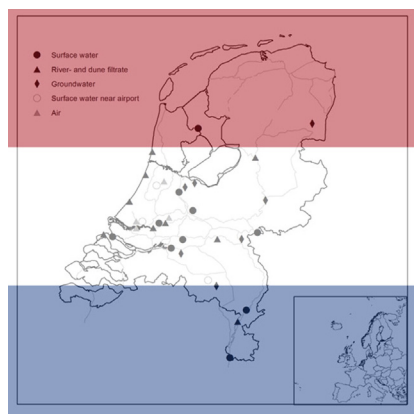
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### HIGHLIGHTS

- Different types of water were screened for *n*Ag, *n*Au, *n*TiO<sub>2</sub> and fullerenes.
- Air samples were analysed for the presence of inorganic nanoparticles.
- C<sub>60</sub> was detected in air, STP influents, effluents and sludge, but in no other aqueous samples.
- In STP sludge and influent evidence for *n*Ag and *n*Au was found.
- 1 to 5% of the total mass in the air samples were from particles smaller than 100 nm.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Only very limited information is available on measured environmental concentrations of nanoparticles. In this study, several environmental compartments in The Netherlands were probed for the presence of nanoparticles. Different types of water were screened for the presence of inorganic (Ag, Au, TiO<sub>2</sub>) and organic nanoparticles (C<sub>60</sub>, C<sub>70</sub>, [6,6]-phenyl-C<sub>61</sub>-butyric acid octyl ester, [6,6]-phenyl-C<sub>61</sub>-butyric acid butyl ester, [6,6]-phenyl-C<sub>61</sub>-butyric acid methyl ester, [6,6]-bis-phenyl-C<sub>61</sub>-butyric acid methyl ester, [6,6]-phenyl-C<sub>71</sub>-butyric acid methyl ester, [6,6]-thienyl-C<sub>61</sub>-butyric acid methyl ester). Air samples were analysed for the presence of nanoparticulate Mo, Ag, Ce, W, Pd, Pt, Rh, Zn, Ti, Si, B as well as Fe and Cu. ICP-MS, Orbitrap-HRMS, SEM and EDX were used for this survey. Water samples included dune and bank filtrates, surface waters and ground waters as well as influents, effluents and sludge of sewage treatment plants (STPs), and surface waters collected near airports and harbours. Air samples included both urban and rural samples.

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Water  
Air  
Field survey

C<sub>60</sub> was detected in air, sewage treatment plants, influents, effluents and sludge, but in no other aqueous samples despite the low detection limit of 0.1 ng/L. C<sub>70</sub> and functionalised fullerenes were not detected at all. In STP sludge and influent the occurrence of Ag and Au nanoparticles was verified by SEM/EDX and ICP-MS. In air up to about 25 m% of certain metals was found in the nanosize fraction. Overall, between 1 and 6% of the total mass from metals in the air samples was found in the size fraction <100 nm.

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## 1. Introduction

Engineered nanomaterials (ENMs) can be found in many different consumer products, and are used for industrial and agricultural processes (Baalousha et al., 2016). The production of ENMs is rising substantially (Keller et al., 2013; Foss Hansen et al., 2016; Virji and Stefaniak, 2014), due to their unique properties. C<sub>60</sub> and C<sub>70</sub> are constituents in cosmetics (Benn et al., 2011), while functionalised fullerenes are good semiconductors in organic solar cells (Kolkman et al., 2013). Also, C<sub>60</sub> is a product of combustion of common fuels (Tiwari et al., 2016). Nanosized silver (*n*Ag) is applied in healthcare products and textiles (Foss Hansen et al., 2016; Benn and Westerhoff, 2008). *n*Au is an integral part of electronic equipment and is employed as catalyst (Toro et al., 2007). *n*TiO<sub>2</sub> is used in consumer products such as sunscreens, cosmetics, tooth paste, paintings and certain food products (chewing gum) (Peters et al., 2014; Helsper et al., 2016). In the automotive industry nanomaterials of metals such as Mo, Ag, Ce, W, Pd, Pt, Rh, Zn, Ti, Si, B as well as Fe and Cu are common constituents in brake pads and discs, and due to abrasion nanoparticles can be formed. PGE (platinum group elements) are used in catalytic converters and coated diesel particulate filters (Jarvis et al., 2001; Wiseman and Zereini, 2009). In most car tyres besides black carbon also *n*SiO<sub>2</sub> is used as reinforcing filler and *n*ZnO is always used as a vulcanisation activator (Semaan et al., 2002). In addition, more nano-additives, such as W<sub>2</sub>S, Mo<sub>2</sub>S, BN, CeO<sub>2</sub> and possibly C<sub>60</sub> are being used in engine oils and fuels to improve performance (Lahouij et al., 2012; Jung et al., 2005).

Concerns have risen that ENMs may be toxic or harmful to the environment (Peralta-Videa et al., 2011; Pettitt and Lead, 2013; Klaine et al., 2008; Noël et al., 2016; León-Silva et al., 2016). Some ENMs are applied because of their antimicrobial properties (e.g. *n*Ag) (Marambio-Jones and Hoek, 2010) and it can therefore be expected that they have adverse environmental effects. Due to their high surface to volume ratio, many ENMs are highly active catalysts (Bäuerlein et al., 2009) which can – for example in combination with UV-light – cause reactive oxygen species (ROS) to be formed that have the potential to cause cell damage (Lin et al., 2014; Kamat et al., 2000; Miller et al., 2012; Bennett and Keller, 2011; Pulskamp et al., 2007; Alkahtane, 2015). It has also been shown that nanoparticles have the potential to damage DNA (Auffan et al., 2009). Apart from the toxicological problems caused by nanoparticles, they also can physically disturb vital functions of a cell, for example by blocking cell pores (Elsaesser and Howard, 2012). A more detailed review of the toxicological effects can be found elsewhere (Sajid et al., 2015).

Owing to their increasing production, application and their potentially adverse effects, it is important that qualitative and quantitative analysis of nanoparticles in various environmental compartments is possible. Relatively little is known about actual environmental exposure to ENM. This type of information is, however, crucial for sound environmental risk assessments (Foss Hansen et al., 2016; Meesters et al., 2014). Still, there are knowledge gaps regarding the actual use of nanoparticles (Tiede et al., 2016). There are no official inventories on production and use of ENMs. However, Foss Hansen et al. have launched a webpage that keeps track of products claiming to contain nanoparticles (Foss Hansen et al., 2016). Additionally, some papers have been published that give estimated numbers on production volumes (Tiede et al., 2016; Piccinno et al., 2012; Markus et al., 2013). Emissions of

ENMs to air, water and soil during their life cycle are largely unclear, quantitative data are currently lacking (Hischier and Walser, 2012). Based on emission estimates and market research information, coupled to fate and transport models, both emissions (Keller et al., 2013, 2014) and environmental concentrations (Gottschalk and Nowack, 2011; Gottschalk et al., 2009, 2013a, b; Sun et al., 2014) have been estimated (Markus et al., 2016). Not only the pristine form of the ENM, but also transformation products formed during treatment and in the environment are of relevance (Nowack et al., 2012). By using probabilistic techniques and comparing modelled environmental concentrations to species sensitivity distributions, the environmental risk of *n*Ag, *n*TiO<sub>2</sub>, *n*ZnO, carbon nanotubes and fullerenes are expected to be generally low with the exception of *n*Ag and *n*TiO<sub>2</sub> levels in sewage effluents (Gottschalk et al., 2009, 2013b).

Measured field data are essential in order to validate the expected concentrations and allow sound risk assessment (Paterson et al., 2011; Isaacson et al., 2009; Hassellöv and Kaegi, 2009).

Several methods have been developed to analyse inorganic and organic nanoparticles. These methods should comply with the definition of the European Commission on 'nanomaterial', i.e. they should be able to determine the particle size distribution in the 1–100 nm size range of single particles and aggregates (Calzolai et al., 2012) as well as the mass or the number of particles per unit of volume or mass. The most common analytical methods for the determination of nanoparticles are single particle inductively coupled plasma (SP-ICP-MS) (Pace et al., 2012; Mitrano, 2012; Borovinskaya et al., 2014; Lee et al., 2014), field flow fractionation (FFF) with and without ICP (Hagendorfer et al., 2012; Krystek et al., 2015; Astefanei et al., 2015), transmission electron microscopy (Kaegi et al., 2010), HPLC - high resolution mass spectrometry (HRMS) (Kolkman et al., 2013; Emke et al., 2015), UHPLC-HRMS (Carboni et al., 2016), LC-UV (Xiao and Wiesner, 2013; Carboni et al., 2014), and FFF coupled to HRMS (Herrero et al., 2015, 2014), multi angle light scattering and dynamic light scattering (Astefanei et al., 2015; Brar and Verma, 2011).

So far, most work has been focused on the development of analytical techniques, but the actual application of these methods has not been reported frequently. Relatively few groups collected environmental samples in order to determine the presence of nanoparticles, whether inorganic or organic. Fullerenes and functionalised fullerenes (C<sub>60</sub>, C<sub>70</sub> and *N*-methylfulleropyrrolidine C<sub>60</sub>) were measured in suspended material of wastewater effluents in Spain in concentrations up to the microgram per litre range (Farré et al., 2010). These high concentrations are, however, disputed (Wang et al., 2010).

Applying an UPLC-MS method around Barcelona, C<sub>60</sub> was detected up to 1.1 ng/kg and 25 pg/L levels in sediments and surface waters, respectively. C<sub>70</sub> was detected up to 330 pg/L in surface water and up to 7.2 ng/kg in sediments. Higher fullerenes, C<sub>76</sub>, C<sub>78</sub> and C<sub>84</sub> were not detected (Astefanei et al., 2014).

Using LC-MS, concentrations of aqueous *n*C<sub>60</sub> aggregates were found up to 130 ng/L in an industrial effluent and 98 ng/L in surface waters in Taiwan. *n*C<sub>70</sub> was found in industrial and municipal effluents in concentrations up to 37 ng/L (Chen and Ding, 2012). Emke et al. reported on C<sub>60</sub> concentrations between 2 and 19 ng/L in sewage treatment plants (STP) influents. The effluents did not contain any fullerenes (LOD < 0.1–0.2 ng/L) (Emke et al., 2015). Concentrations of functionalised fullerenes in influents and effluents were below the LOD (<0.1 ng/L).

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