



Origin and fate of nanoparticles in marine water – Preliminary results

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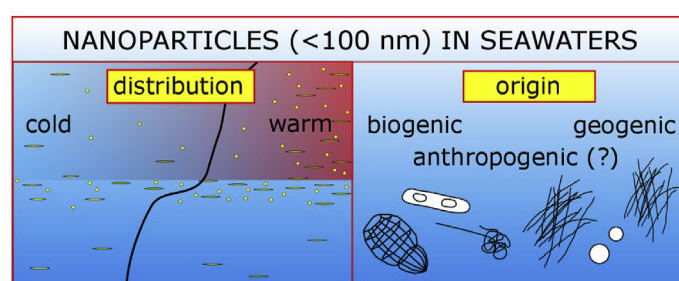
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HIGHLIGHTS

- The number of nanoparticles in seawater vary from not detected to $380 \times 10^2 \text{ cm}^{-3}$.
- The number of nanoparticles increases during intense primary production.
- Thermal stratification favors accumulation of nanofibers.
- Pyrite and biogenic silica and manganese/iron oxides nanoparticles were identified.
- Probably only asbestos nanofibers were of anthropogenic origin.

GRAPHICAL ABSTRACT



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ABSTRACT

The number, morphology and elemental composition of nanoparticles (<100 nm) in marine water was investigated using Variable Pressure Scanning Electron Microscopy (VP-SEM) and Energy-dispersive X-ray spectroscopy (EDS). Preliminary research conducted in the Baltic Sea showed that the number of nanoparticles in seawater varied from undetectable to $380 \times 10^2 \text{ cm}^{-3}$. Wind mixing and density barriers (thermocline) had a significant impact on the abundance and distribution of nanoparticles in water. Many more nanoparticles (mainly nanofibers) were detected in periods of intensive primary production and thermal stratification of water than at the end of the growing season and during periods of strong wind mixing. Temporal and spatial variability of nanoparticles as well as air mass trajectories indicated that the analysed nanofibers were both autochthonous and allochthonous (atmospheric), while the nanospheres were mainly autochthonous. Chemical composition of most of analysed nanoparticles indicates their autochthonous, natural (biogenic/geogenic) origin. Silica nanofibers (probably the remains of flagellates), nanofibers composed of manganese and iron oxides (probably of microbial origin), and pyrite nanospheres (probable formed in anoxic sediments), were all identified in the samples. Only asbestos nanofibers, which were also detected, are probably allochthonous and anthropogenic.

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1. Introduction

The term “nanoparticles” refers to particles with dimensions in the range of nanometers. In this paper we follow the definition of

Maurice and Hochella (2008), who describe them as particles of which at least one dimension is smaller than 100 nm. This can include spherical, tubular or irregularly shaped particles existing in fused, aggregated or agglomerated forms. Properties of nanoparticles (NPs) are different from their bulk counterparts. They are characterized by extremely high surface area to volume ratios and unique physical and chemical properties (e.g. ease of suspension in liquids, increased strength/weight ratios, enhanced conductivity, and improved optical or magnetic properties). It is commonly assumed that their adverse effects cannot be predicted from toxicity estimates based on studies of the same material in macroscopic size ranges, which obey the laws of classical physics (Hochella et al., 2008; NIOSH, 2013; SCENHIR, 2006). They are ubiquitous in aquatic environments and possess the ability to influence water chemistry and processes in a very different way to macro-sized counterparts as they are generally more reactive (Ju-Nam and Lead, 2008; Wigginton et al., 2007). They can be either organic, inorganic or a combination, of natural origin (e.g.: from living organisms such as pollen, sea salt, dust, erosion particles, volcanic explosions), incidental origin (e.g.: industrial processes, diesel engine emissions) or engineered (e.g.: fullerenes, metal oxides, nanotubes) (Hassellöv and Kaegi, 2009; Nowack and Bucheli, 2007; Peralta-Videa et al., 2011).

The specific properties of NPs makes them valuable materials. In general, the properties of NPs mean that they have great economic potential and the application of nanotechnology is beneficial to individuals and organisms (NIOSH, 2013; SCENHIR, 2006). Roco (2011) anticipated that products of nanotechnology would be worth \$1 trillion in 2015 whilst products that incorporate nanotechnology are expected to bring about \$3 trillion by 2020 (Roco, 2011). There are, however, issues with such estimates as the amount of NPs produced (so-called “engineered” nanomaterials –ENMs) cannot be estimated reliably due to issues of data quality, problems related to the definition of particular nanomaterials and the rapidly evolving nature of the production processes (Hendren et al., 2011; Piccinno et al., 2012). According to the Project on Emerging Nanotechnologies (PEN, 2014), the number of nanotechnology consumer products registered in dedicated inventories increased by about 125% each year between 2008 and 2010 and it contained 1628 products or product lines in October 2013.

Growing interest in nanotechnology raises concerns about the effect of NPs on human health, safety and environmental impacts. Four possible impacts of NPs have been defined: (1) direct toxic effects, (2) changes in the bioavailability of nutrients and toxins, (3) indirect effects resulting from their interaction with natural organic compounds, and (4) changes in environmental microstructures (Klaine et al., 2008; Peralta-Videa et al., 2011; Simonet and Valcárcel, 2009). Recently, the number of studies on the toxicity, fate and transport of engineered nanomaterials has been growing rapidly (Handy et al., 2012a; Handy et al., 2012b; Ju-Nam and Lead, 2008; Nowack and Bucheli, 2007; OECD, 2014; Vejerano et al., 2014) but such critical issues as the magnitude of engineered nanoparticles (ENPs) that reach the environment have received less attention (Hendren et al., 2011; Piccinno et al., 2012; Sun et al., 2014). In recent years, there have been some studies on nanoscale particles in the atmosphere (Hussein et al., 2005; Longley et al., 2003; Murr and Garza, 2009; Noble et al., 2003; Young and Keeler, 2004), sewage (Eduok et al., 2005) and fresh water (Baalousha and Lead, 2007). Furthermore an attempt has been made to derive a global nanomaterial budget for Earth (Hochella et al., 2012). However, basic research on concentrations of ENPs and occurring naturally in the marine environment nanoscale particles that would provide estimates of background levels are missing. Without such information no preventive measures can be taken. Furthermore, the possibility of negative synergistic effects

occurring between NPs and other human related pressures (e.g. climate change, ocean acidification, chemical pollution, etc.) also have to be considered. Thus, basic knowledge on the number, distribution, origin and fate of engineered and naturally occurring NPs as well as incidental NPs in the aquatic environment is essential. This study characterizes nanoparticles in marine water, using Variable Pressure Scanning Electron Microscope (VP-SEM) and Energy-dispersive X-ray spectroscopy (EDS). These techniques do not require the studied materials to be conductive and do not destroy widely produced engineered materials. The research was conducted in the southern Baltic, which is a shelf sea with long water exchange time (approximately 25 years: HELCOM, 2009), and exposed to pollution from several European countries. Morphology, chemical composition and regional variability of NPs numbers were studied and 48-h air-mass backward trajectories were calculated in order to determine origin of nanoparticles in marine water. It is assumed that biogenic and geogenic NPs are present in marine water and that they originate from both autochthonous and allochthonous sources. The influence of season on number of NPs in seawater was also studied. Our hypothesis is that primary production and water stratification, which strongly depend on season, influence the distribution of NPs in the water column. To our knowledge, this is the first study addressing sampling and examination of NPs in marine waters.

2. Materials and methods

2.1. Field work

The analysis were conducted on samples taken in June and November 2014 at stations with different levels of human impact, located in the southern Baltic Sea area (Fig. 1). The selected sampling stations corresponded to the national monitoring stations within the Polish Economic Zone. Water samples were collected along two transects. The first station of transect no. 1 was Zn2 (Vistula River mouth) in the Gulf of Gdansk and the last station of this transect was P1 (Gdansk Deep), in an area of open sea. The second transect was located entirely in the open sea area. It started in the middle of the Polish coast (P5) and ended in the Bornholm Deep area (P5). In addition, water samples were collected in the open sea area at the slope of the Gotland Deep (P140), in a sheltered part of the Gulf of Gdansk located to the east of the mouth of the Vistula River (KO), and in the Pomeranian Bay (B13). The Gulf of Gdansk and Pomeranian Bay are both subject to strong human pressure. The Gulf of Gdansk receives water from the Vistula, the second largest river that flows into the Baltic Sea. The Pomeranian Bay, in turn, is the estuary of one of the major rivers in Poland – the Oder river. In November 2014, water samples were additionally collected from the Vistula River (about 30 km south from the mouth). At all stations, samples were collected from the surface and from the depth of the thermocline and/or halocline, depending on water depth in the area of sampling. A total of 35 water samples were taken. Water salinity at the stations varied from 6.8 PSU (Practical Salinity Units) to 7.9 PSU at the surface, and from 7.1 PSU to 16.5 PSU at the bottom. Each sample was taken with a ten Niskin bottles (10 dm³ each). Then a volume of 100 cm³ was transferred from each Niskin bottle (to ensure the representativeness of a sample) into one 1 dm³ dark glass bottle and stored in a cool and dark place until laboratory treatment. All bottles were cleaned beforehand, following the procedure described in the next section (Laboratory treatment and microscopic analysis).

2.2. Meteorological conditions

The 48-h air-mass backward trajectories were calculated at 6 h

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