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ACCEPTED MANUSCRIPT

Stochastic fate analysis of engineered nanoparticles in incineration plants

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9 Abstract

10 Worldwide waste is increasing and can contain engineered nanoparticles (ENP) made from almost the entire spectrum of the periodic table. An increasing amount of waste (including nanowaste) is incinerated. A full-scale experiment in a modern 11 12 waste incineration plant showed that even inert nanoparticles (nano-CeO₂) are successfully removed from the flue gas and transferred to the solid incineration residues. Predicting the fate of nanomaterials in incineration plants with models based 13 14 on real measurements would reduce the immense efforts (time and resources) for real-scale experiments. Here, we develop a model for the ENP fate in incineration plants, based on the data of the nanoCeO2-experiment. We investigated all 15 16 possible transfers and sinks of ENP throughout the incineration by linking ENP concentration measurements to the 17 nanomaterial flows and retention times. The model also delivers information on the associated uncertainties and how they 18 propagate through the incineration system by using a fully probabilistic material flow analysis. The model can be generalized to other ENP and also to other incineration plants. We show that the output of the measurements was 19 20 consistent albeit relying on multiple measurement methods, and that a one day sampling period is sufficient to obtain an overview on the fate of nanoparticles in incineration plants. In addition to the dynamic results, a generalized steady state 21 22 mass flow with transfer factors is provided and can be used for modeling purposes of CeO₂ or other nano sized metals with 23 similar physic-chemical properties.

24 **1. Introduction**

25 1.1. Sustainable treatment of nanowaste is increasingly important

26 Nanoparticles from natural sources such as sea salt, soil dust or pollen lead to ambient concentrations of a few thousand 27 particles per cm³ (Shi et al., 2001). Anthropogenic sources such as traffic or heating further increase human exposure to 28 nanoparticles. A new emission source is arising from the growing production and application of engineered nanoparticles 29 (ENP) in industrial and consumer products (Gottschalk et al., 2010), which already represents a multiple billion USD industry 30 (Becker, 2013). Various emission hotspots of ENP along their lifecycle in nanoenabled products have already been identified 31 (Koehler et al., 2008), such as indoor emissions from nanopowder production and handling (Savolainen et al., 2010) or 32 nanosilver release from textiles into wastewater (Lorenz et al., 2012). A further potential hotspot of accumulated and 33 potentially re-emitted ENP are waste treatment systems (Musee, 2011), which have to deal with an increased amount of 34 products containing ENP that have reached the end of their use. So-called nanowaste, i.e. discarded products that contain 35 ENP, needs to be carefully evaluated with respect to sustainable waste management options. Increasingly important 36 worldwide is incineration, which is a popular disposal option for conventional as well as for hazardous waste. 37 Unfortunately, the amount and fate of nanoparticles in incineration plants is poorly understood because of virtually no 38 experimental study and very simplistic modeling (Asmatulu et al., 2012; Bystrzejewska-Piotrowska et al., 2009; Mueller et 39 al., 2013; Musee et al., 2011; Walser et al., 2012). Complicating aspects of nanoparticles are that they combine characteristics from gases (e.g. no sedimentation) and from larger particles (e.g. carrier for other compounds). Moreover, 40 41 the complex chemical and physical conditions in real incineration plants make mechanistic fate modeling of ENP even more 42 unrealistic. However, a real-scale experiment to test whether incineration plants are a favorable solution for the disposal of nanowaste is a theoretical and practical challenge. Such studies have to deal with (i) the development of cutting-edge 43 44 measurement capabilities, (ii) the extremely low nanoparticle concentrations on top of a commonly high elemental 45 background concentration, and (iii) a complex large scale experimental setting. A recent study (Walser et al. 2012) showed that modern incineration plants can remove inert nanoparticles from the flue gas and deposit them in the solid incineration 46 47 residues. This experimental study also showed that location, residence time and residence form of ENP during the waste 48 treatment process cannot be modeled in detail without adding dynamics to the temporally and spatially resolved ENP fate 49 parameters.

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