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# Characteristics of airborne gold aggregates generated by spark discharge and high temperature evaporation furnace: Mass–mobility relationship and surface area

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## ABSTRACT

The properties of gas-borne aggregates are important in nano-technology and for potential health effects.

Gold aggregates from three generators (one commercial and one custom built spark discharge generator and one high-temperature furnace) have been characterized. The aggregate surface areas were determined using five approaches – based on aggregation theory and/or measured aggregate properties. The characterization included mass-mobility relationships, effective densities (assessed by an Aerosol Particles Mass analyzer), primary particle analysis (based on Transmission Electron Microscopy), as well as total mass and number concentration outputs.

The relationships between mass and mobility are well described by power-law functions with exponents of 2.18–2.35. For all generators, the primary particles of the aggregates were fused together by a bridge with a diameter typically ~60–70% of the primary particle diameter (5–10 nm). The total mass outputs were 6.1–48.1 mg/m3 and the predicted surface area outputs in the range $0.9 \times 10^{-3}$–$17 \times 10^{-3}$ $\mathrm{cm^2/cm^3}$.

The aggregate effective densities differed considerably between generators. The difference could partly be explained by the differences in primary particle diameter, but not fully. This in turn may be explained either by a varying primary particle size with aggregate size, or by that there are slight differences in the morphology of the aggregates from the generators.

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*Abbreviations:* APM, aerosol particle mass analyzer; BET, Brauner, Emmett, Teller surface area; $b_r$, bridging; $C_i$, Cunningham factor; CMD, count median diameter; CPC, condensation particle counter; $D_\alpha$, exponent of Eggersdorfer et al. (2012) model; $d_{APM}$, primary particle diameter, APM; $d_b$, bridging diameter; DLCA, diffusion limited cluster aggregation; DMA, differential mobility analyzer; $c^*$, dimensionless drag coefficient; $d_{me}$, mobility diameter; $D_{mm}$, mass mobility exponent; DMPS, differential mobility particle sizer; $d_{pp}$, primary particle diameter, TEM; $d_{va\text{-}i}$, primary particle diameter, Sauter (APM and TEM); $\rho_{effd}$, effective density; ESP, electrostatic precipitator; $f_i$, friction factor; $F_i$, Stokes drag; GSTD, geometric standard deviation; HT, high temperature evaporation furnace; $K$, prefactor; $K_\alpha$, prefactor of Eggersdorfer et al. (2012) model; $Kn$, Knudsen number; $m_i$, Mass; $N$, number of primary particle particles in an aggregate; $N_{pp}$, number of primary particle number analyzed in the TEM image analysis; $Q$, elemental charge; $r_i$, radius; $SA_i$, surface area; SDG$_C$, spark discharge generator, custom; SDG$_P$, spark discharge generator, commercial; $SSA_i$, specific surface area; TEM, transmission electron microscopy; $N$, volume; $V$, voltage; $\Omega$, angular velocity; $X$, shape factor; Index $i$, refer to there being more than one variation of the parameter

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## 1. Introduction

The properties of airborne agglomerates and aggregates are important for many reasons. It is known that exposure to airborne particulates, which often includes agglomerates/aggregates, can have adverse health effects (Barath et al., 2010; Dockery et al., 1993). To understand the toxicological effect of inhaling aggregates it is important to (I) have robust but variable methods available for controlled generation of airborne aggregates (for controlled toxicological tests) and (II) methods assessing the critical properties of the aggregated particles. Studies have shown that for various particle types the surface areas of the particles produce coherent dose-response relations, suggesting that the promoter of toxicity is the surface area rather than the mass or particle number inhaled (Aitken, Chaudhry, Boxall, & Hull, 2006; Donaldson et al., 2008; Waters et al., 2009). Hence it is of great importance to have readily available tools and models for calculating the surface area dose for aggregated particles, be it in a lab during toxicological studies, occupational exposure setting, or for other applications.

Apart from surface area of the aggregates/agglomerates, produced by for example flame spray pyrolysis, high temperature evaporation condensation or spark discharge, there are several other aggregate properties that needs to be thoroughly characterized such as the mass–mobility relationships, number/mass output, primary particle size (Dillon, Copley, Koos, Bishop, & Grobert, 2013; Heurlin et al., 2012; Messing, Dick, Wallenberg, & Deppert, 2009; Shin et al., 2009).

In this study, the term "agglomerates" is used when primary particles are held together by weak van der Waals forces, and "aggregates" is use for primary particles more strongly bound together by partial melting (DFG, 2013; Lövestam et al., 2010). We here let aggregate include agglomerates if we do not know which nomenclature if most descriptive. Note that the nomenclature of the structures is not used unambiguously in the literature and the two terms can be used interchangeably depending on the research field.

Assessing the surface area of non-spherical nanoparticles is not straightforward, and as of today there few ways of performing direct measurements. For most methods used the measured surface area will be intimately related to the method by which it was determined. Although methods that measure surface area on-line are available (Asbach, Fissan, Stahlmecke, Kuhlbusch, & Pui, 2009; Fierz, Houle, Steigmeier, & Burtscher, 2011; Ntziachristos, Giechaskiel, Ristimäki, & Keskinen, 2004; Wang et al., 2010), these were typically not developed for non-spherical and porous particles, and thus do not work in an optimal way for all types of aggregates (LeBouf et al., 2011). Also there is data suggesting that the techniques based on unipolar diffusion charging does not relate directly to the surface area of the aerosol particles as suggested (Gopalakrishnan, Thajudeen, Ouyang, & Hogan, 2013; Ku & Kulkarni, 2012; Ku & Maynard, 2005). Transmission electron microscopy (TEM) imagery is often used to characterize primary particle size ($d_{pp}$) of aggregates. On-line techniques such as the aerosol particle mass analyzer (APM) that coupled in a series after a differential mobility analyzer (DMA) determines the mass–mobility relationship, as well as particle effective density ($\rho_{effd}$), with a relatively high resolution in both time and size (Ehara, Hagwood, & Coakley, 1996; Olfert, Symonds, & Collings, 2007). This technique also allows one to distinguish between externally mixed particles of different effective densities (McMurry, Wang, Park, & Ehara, 2002; Rissler et al., 2014). Various techniques have been used for specific surface area characterization, whereof nitrogen adsorption, or Brunauer Emmett Teller (BET) is the most common (Brunauer, Emmett, & Teller, 1938). BET has been successfully used for nanoparticle surface area characterization (Eggersdorfer, Groehn, Sorensen, McMurry, & Pratsinis, 2012; Ku & Kulkarni, 2012). However, the technique can prove challenging for many applications and nanoparticle sources since it is offline and a relatively large amount of material is needed (min. ~10 mg). Furthermore, the whole particle population collected over a long period of time is characterized, as a whole.

TEM imagery has also been compared to BET measurements and suggested as a stand-alone off-line method for determining specific surface area (Bau, Witschger, Gensdarmes, Rastoix, & Thomas, 2010). A combination of on-line and off-line techniques have also been suggested for the determination of surface area of aggregates, such as that of combining aerosol particle mass measurements (DMA–APM) and basic TEM imagery (Rissler et al., 2012, 2013; Thajudeen, Jeon, & Hogan, 2015).

Another approach to assess surface area is from a theoretical point of view, combined with on-line or off-line techniques. An extensive effort have been made in the area of modeling properties of fractal structures formed by diffusion limited cluster aggregation (DLCA), mainly focusing on number of primary particles in aggregates in relation to the aggregate radius of gyration or mobility size (Chan & Dahneke, 1981; Eggersdorfer, Groehn, Sorensen, McMurry, & Pratsinis, 2012; Meakin, Donn, & Mulholland, 1989; Sorensen, 2011). Furthermore, effort has also been devoted to linking the mobility of aggregates to results from image analysis using TEM (Thajudeen et al., 2015). Eggersdorfer, Groehn, Sorensen, McMurry, and Pratsinis (2012) suggest predicting surface area from DMA–APM measurements solely – for aggregates formed by DLCA. There is also considerable work put into describing the properties of aggregates by using a purely theoretical approach (Chan & Dahneke, 1981; Dahneke, 1982).

The aim of this study is to compare number, mass and surface area output concentrations of gold aggregates generated by three methods. These methods were a high temperature furnace (HT), a commercial spark discharge generator ($SDG_P$), and a novel in-house constructed spark discharge generator ($SDG_C$). This was accomplished by a detailed characterization regime in combination with theory and semi-empirical models. The instruments used for characterization were: a transmission electron microscopy (TEM), a Differential mobility analyzer – aerosol particle mass analyzer (DMA–APM), and a differential mobility particle sizer (DMPS).