Number Size Distribution of Fine and Ultrafine Fume Particles From Various Welding Processes

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Received 30 July 2012; in final form 15 August 2012; Advance Access publication 1 October 2012

Studies in the field of environmental epidemiology indicate that for the adverse effect of inhaled particles not only particle mass is crucial but also particle size is. Ultrafine particles with diameters below 100 nm are of special interest since these particles have high surface area to mass ratio and have properties which differ from those of larger particles. In this paper, particle size distributions of various welding and joining techniques were measured close to the welding process using a fast mobility particle sizer (FMPS). It turned out that welding processes with high mass emission rates (manual metal arc welding, metal active gas welding, metal inert gas welding, metal inert gas soldering, and laser welding) show mainly agglomerated particles with diameters above 100 nm and only few particles in the size range below 50 nm (10 to 15%). Welding processes with low mass emission rates (tungsten inert gas welding and resistance spot welding) emit predominantly ultrafine particles with diameters well below 100 nm. This finding can be explained by considerably faster agglomeration processes in welding processes with high mass emission rates. Although mass emission is low for tungsten inert gas welding and resistance spot welding, due to the low particle size of the fume, these processes cannot be labeled as toxicologically irrelevant and should be further investigated.

Keywords: nanoparticles; size distribution; ultrafines; welding

INTRODUCTION

About 20 years of studies in environmental epidemiology and toxicology suggest that in respect to the toxicological effect of aerosol particles not only the particle material and mass are crucial but also particle size and surface properties are (Ferin et al., 1990; Oberdörster et al., 1990; Oberdorster et al., 1994; Oberdörster et al., 1995; Donaldson et al., 1998; Oberdörster et al., 2000; Utell and Frampton, 2000; Donaldson et al., 2001; Oberdörster, 2001; Donaldson et al., 2002). Studies suggest that the respiratory and the cardiovascular effects of inhaled ambient particles are correlated with the total particle surface, rather than particle mass, present in the lungs (Brown et al., 2001). Therefore, ultrafine particles (particles with diameters below 100 nm) are considered as toxicologically more relevant than fine or even coarse particles. These particles not only have a high cumulative particle surface per unit mass but also have lung deposition of ultrafine particles that is considerably higher than that of fine and coarse particles (Brown et al., 2002; Kim and Jaques, 2005; Moller et al., 2008). Especially, coating of the surfaces of ultrafine particles with reactive substances like metals and organic substances like polycyclic aromatic hydrocarbon may be responsible of increased morbidity and mortality in

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environments with high concentrations of ultrafine particles. Particle surface seems to be important for the toxicological properties of inhaled particles, and aerosol particles with sizes below about 30–50 nm seem to have properties that differ from those of larger particles: such particles are poorly phagocytized by macrophages (Ferin, 1994; Brown et al., 2002) but may penetrate into various other cells like epithelium cells (Ferin et al., 1990; Ferin et al., 1992) and may therefore transport toxic or irritative materials in tissues that are normally not exposed to ambient aerosols (Limbach et al., 2009). Furthermore, nanometer-sized clusters of certain materials may have completely different chemical properties than the bulk material (Stark, 2011).

However, there seems to be a considerable difference between ultrafine particles in the urban environment and the workplace—especially, if it is considered that welding fumes consist of agglomerates of primary welding fume particles that are of ultrafine size. If mortality data derived for environmental studies [i.e., 1.5% per 10 µm m 3 increase in particle mass (Wichmann et al., 2000)] are transferred to workplaces with typical welding fume exposures [welding fume concentration up to above 10 mg m 3 (Pourtaghi et al., 2009)], this will result in unrealistically high values of mortality. There are different possible explanations for this apparent paradox: first, there may be a considerable ‘healthy worker effect’ (Baillargeon, 2001), which leads to a selection of subjects non-susceptible to ultrafine particles. Second, toxicity of environmental particles may be due to chemical components that are not present in welding fume particles. Third, the particle size distributions may be different between environment and welders’ workplace.

Since particle size seems to play a crucial role for characterizing the toxicological properties of inhaled particles, in this paper, particle number concentrations and size distributions were measured for seven different welding techniques, using a fast mobility particle sizer (FMPS).

METHODS

Welding

The following welding processes and process variants were investigated:

1. Manual metal arc welding (MMAW), base material: high-alloyed steel (EN10027-2: 1.4301), filler metal: high-alloyed basic covered electrode (Thermanit JE spezial, Böhler welding, Germany).
2. Metal active gas welding (MAG), base material: high-alloyed steel (EN10027-2: 1.4301), filler metal: high-alloyed welding wire (Thermanit X, Böhler welding, Germany), shielding gas: 97.5% Ar, 2.5% CO 2, short arc mode.
4. Metal inert gas soldering on zinc plated base material (MIG soldering), base material: hot-dip zinc coated steel sheet EN 10346: DX51D+Z275, filler metal: copper (96%), manganese (1%), silicium (3%) (CuSi3Mn; Bercoweld S3, Bedra, Germany), shielding gas: Argon.
5. Laser welding, base material: hot-dip zinc coated steel sheet (1 mm) EN 10346: DX56D+Z100, CO 2 Laser (6 kW), output power: 30% pulsed with 50 kHz, focal length: f = 175 mm, no cross-jet, coaxial gas nozzle, shielding gas: Helium, welding speed: 3 m min⁻¹, full penetration welds.
7. Resistance spot welding (RSW), medium-frequency inverter spot welding (1 KHz), base material: hot-dip zinc coated steel sheet (1 mm) EN 10346: DX51D+Z275, two steel sheets joined with 4.5 mm welding spots.

For processes MMAW, MAG, MIG, and TIG, bead-on-plate welding was performed manually by an experienced professional welder. Welding was performed in five welding episodes of 40 s. After each welding episode, a break of about 2–5 min followed to allow the particle sampling system to flush. For laser welding, automatic welding was performed for 14 s with a welding speed of 3 m min⁻¹. For RSW, a total of 25 single welding spots were analyzed.

Welding fume sampling

Above the welding process a funnel-shaped fume hood was placed, which aspirates the fume with a volume flow of 38 l s⁻¹. Within the aspiration flow, a welding fume sample was taken (10 l min⁻¹) under approximately isokinetic conditions. The distance between welding process and site of sampling was 60 cm (Fig. 1). This distance was chosen since it represents approximately the distance between the welding process and the breathing zone of welders. Very close to the sampling site, the aerosol sample was
diluted by a factor of 20, using a commercial device (dilution system type DIL 550, Topas, Germany).

**Particle size analysis**

Particle number size distribution measurements were performed using a FMPS (TSI Incorporated model 3091, USA; Jeonga and Evansa, 2009; Leskinen et al., 2012). This device classifies particles in the size range from 5.6 nm to 0.56 µm according to their electrical mobility. Number concentration is assessed by measuring the electrical current of charged aerosol particles (Tuch et al., 2000). Since particle number size distribution is measured simultaneously in 32-size channels with a time resolution of 1 s, this device is suitable for the measurement of fast changing, unstable aerosols.

**Data analysis**

Each measurement consisted of five welding episodes of 40 s (MMAW, MAG, MIG, and TIG), five welding episodes of 14 s (Laser), or one welding spot (RSW). Particle size distributions measured in 1-s intervals were averaged over the entire welding episode. The start of the welding episode was identified by the fast increase of total number concentration. From the averaged size distribution, the mean total number concentration of particles within the measuring range (5.6 nm to 0.56 µm), the maximum total number concentration, the mode (size channel with the highest number concentration), the fraction of particles with diameters below 100 nm ($F < 100$) (number of particles <100 nm/total number concentration), and the fraction of particles with diameters below 50 nm ($F < 50$) (number of particles <50 nm/total number concentration) were calculated.

**RESULTS**

In Figs 2–8, the average particle number size distribution of the five welding episodes is shown as a function of particle size. The average ‘mean concentration’, average ‘maximum concentration’, the mean fraction of particles with diameters smaller than 100 respectively 50 nm, and the modal diameter are summarized in Table 1.

As can be seen, particle size distributions for MMAW, MAG, MIG, and laser welding are quite similar. Although sampling of the welding particles was performed close to the welding arc (60 cm), in contrast to our expectation, the fraction of ultrafine particles with diameters below 50 nm was small (10–16%). The mode of the main peak of the distribution for all these welding techniques was about 110–140 nm. This peak may be attributed to particles...
that have already grown due to agglomeration processes. A small peak at or below about 10 nm presumably represents remaining primary particles or small agglomerates.

Size distributions of particles produced by TIG welding or RSW have a completely different structure. Particles produced by this technique are (at 60 cm from the welding site) nearly all smaller than 100 nm, and at least 90% are smaller than 50 nm. These particles are nearly exclusively ultrafine, indicating that agglomeration of primary particles is much slower.

**DISCUSSION**

The results of this study were rather surprising since it turned out that some welding techniques (MMAW, MAG, MIG, and laser welding) nearly exclusively consisted of particles in the size range between about 60–200 nm, and only a small fraction of particles with diameter below 50 nm was observed, which are supposed to be toxicologically of particular interest. In contrast to these welding processes, TIG welding and RSW produced solely particles in the ultrafine range below 50 nm—thus, particles that are toxicologically of particular interest. Aerosol particles in the size range considerably below about 100 nm increase in size due to agglomeration with decreasing initial particle size and increasing initial particle number concentration. A summary of mass emission rates of different welding techniques is given in the paper of Pohlmann et al. (2012). They found for similar welding techniques the following values: MMAW, 5.5 mg s⁻¹; MAG, 2.9 mg s⁻¹; MIG-Alu, 24 mg s⁻¹; MIG-soldering, 2 mg s⁻¹; and laser welding, 8 mg s⁻¹. On the hand, for the clean welding techniques, they found emission rates of 30 µg s⁻¹ (TIG) and 23 µg s⁻¹ (RSW). Since total particle number concentration of TIG and RSW was similar to that observed for the other welding processes, the rating that TIG welding and RSW are clean and toxicologically of minor importance (Spiegel-Ciobanu, 2005) should be questioned, despite the low mass emission rates of these techniques.

The reason for the different particle size distribution found for TIG welding and RSW in contrast to the other welding techniques under consideration is most probably due to the different coagulation dynamics after the primary particle formation (Preining, 1998; Limbach et al., 2009). Aerosol particles in the size range considerably below about 100 nm increase in size due to agglomeration with decreasing initial particle size and increasing initial particle number concentration. The data from Preining (Preining, 1998) indicate that half-life of a 1-nm particle with an initial particle mass concentration of 1 mg m⁻³ is about 2 ms. If particle concentration is lower, agglomeration slows down: for a 1 nm particle at 1 µg m⁻³, half-life is about 2 s. During arc welding, particles are mainly formed by condensation from extremely hot vapors during cooling. The initial particle number concentration is supposed to be extremely high, resulting in extremely short half-lives of the primary particles. Thus, particle size increases and particle concentration decreases fast. With increasing particle size and decreasing number concentration, this process becomes slower until it nearly stops at the minimum of intrinsic particle motion at a diameter of about 100–400 nm. Hence, the initially nanometer-sized primary particles emitted from arc welding processes with high mass emissions are nearly completely agglomerated to 100–200 nm particles when reaching the measurement site (and the respiratory zone of a welder) 60 cm from the arc.

### Table 1. Mean particle number concentration (Cmean), maximum particle number concentration (Cmax), fraction of particles smaller than 100 nm ($F < 100$), fraction of particles smaller than 50 nm ($F < 50$), and mode of the distribution for the 7 welding techniques. Shown is the average and standard deviation (Std. Dev) of the five measured emission episodes for each technique (25 spots for RSW).

<table>
<thead>
<tr>
<th></th>
<th>C mean average (cm⁻³)</th>
<th>Standard deviation (cm⁻³)</th>
<th>Cmax average (cm⁻³)</th>
<th>Standard deviation (cm⁻³)</th>
<th>$F &lt; 100$ average</th>
<th>Standard deviation</th>
<th>$F &lt; 50$ average</th>
<th>Standard deviation</th>
<th>Mode average (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MMAW</td>
<td>2.6E + 06</td>
<td>4.5E + 05</td>
<td>6.5E + 06</td>
<td>1.2E + 06</td>
<td>0.24</td>
<td>0.02</td>
<td>0.10</td>
<td>0.02</td>
<td>143</td>
</tr>
<tr>
<td>MAG</td>
<td>2.1E + 06</td>
<td>1.1E + 06</td>
<td>4.7E + 06</td>
<td>2.5E + 06</td>
<td>0.54</td>
<td>0.02</td>
<td>0.16</td>
<td>0.02</td>
<td>108</td>
</tr>
<tr>
<td>MIG alu</td>
<td>5.0E + 06</td>
<td>1.6E + 06</td>
<td>1.2E + 07</td>
<td>4.7E + 06</td>
<td>0.35</td>
<td>0.03</td>
<td>0.13</td>
<td>0.06</td>
<td>124</td>
</tr>
<tr>
<td>MIG solder</td>
<td>2.1E + 06</td>
<td>1.6E + 06</td>
<td>5.1E + 06</td>
<td>3.9E + 06</td>
<td>0.44</td>
<td>0.00</td>
<td>0.15</td>
<td>0.02</td>
<td>124</td>
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<tr>
<td>Laser</td>
<td>5.0E + 07</td>
<td>3.8E + 06</td>
<td>1.2E + 08</td>
<td>8.6E + 06</td>
<td>0.42</td>
<td>0.02</td>
<td>0.12</td>
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<td>TIG</td>
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<td>4.5E + 05</td>
<td>3.3E + 06</td>
<td>4.9E + 05</td>
<td>0.99</td>
<td>0.01</td>
<td>0.90</td>
<td>0.05</td>
<td>16</td>
</tr>
<tr>
<td>RSW</td>
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<td>9.3E + 05</td>
<td>4.7E + 06</td>
<td>2.2E + 06</td>
<td>1.00</td>
<td>0.00</td>
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<td>0.01</td>
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</table>
Fig. 2. Average particle number concentration as a function of particle size for the five measured emission episodes for MMAW.

Fig. 3. Average particle number concentration as a function of particle size for the five measured emission episodes for MAG.
Particle mass emission of TIG welding and RSW is considerably lower (0.01–0.05 mg s⁻¹). Due to this low mass emission, the initial particle number concentration is supposed to be lower for these techniques, resulting in a very slow agglomeration speed. At the site of measurement, these particles are still in the size range of about 10 nm, and this fraction of nanoscaled particles is stable for at least some minutes since number concentration is relatively low.

Particle sources in ambient air also have relative small emission rates (vehicle exhaust for example), resulting in a slow agglomeration speed and a relatively stable fraction of 10–20 nm particles (nucleation mode), which exists in addition to the fraction of aged, agglomerated particles (accumulation mode) (Kelkar and Joshi, 1977; Lippmann, 1980). Hence, the urban ambient aerosol and welding fume emission from welding techniques with low mass emission rates (TIG and RSW) are in respect to particle size distribution quite similar.

If it is considered that accumulated particles of 100 or 200 nm also consist of about 10 nm primary particles that are more or less loosely bound to each other, the crucial question for assessing possible biological effects of these particles upon inhalation is, if the agglomerates remain as agglomerates or if the agglomerates break down to smaller aggregates or even primary particles in contact with the lung surface—which may significantly influence their toxicological properties. However, a recent experimental study, in vitro and in vivo in an animal model, suggest that break down of agglomerates on the lung epithelium is unlikely (Schaudien et al., 2011). Therefore, particle sizes measured air borne in the respiratory zone of the subject seem to be a reasonable estimate of the size related properties of particles in the lungs.

CONCLUSIONS

Whereas welding techniques with high mass emission rates (MMAW, MAG, MIG, and Laser) show predominantly particles with diameters about 100–200 nm and few nanoscaled particles, so-called ‘clean’ techniques with low mass emission rates (TIG, and RSW) emit nearly exclusively particles smaller than 50 nm with number concentrations similar to other welding techniques. Such particles are in respect to their potential toxicological properties of particular interest. Therefore, the health effect of exposure to welding fumes with low mass emission rates but consisting of nanoscaled particles should be further investigated.
Fig. 5. Average particle number concentration as a function of particle size for the five measured emission episodes for MIG soldering.

Fig. 6. Average particle number concentration as a function of particle size for the five measured emission episodes for laser welding.
Fig. 7. Average particle number concentration as a function of particle size for the 5 measured emission episodes for TIG.

Fig. 8. Average particle number concentration as a function of particle size for the 25 measured emission episodes for RSW.
REFERENCES