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Workplace Exposure to Submicron Particle Mass and Number Concentrations From Manual Arc Welding of Carbon Steel

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APPLIED STUDIES

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Particle emissions from manual shielded metal arc welding of carbon steel were sampled in a typical industrial maintenance and metal fabrication workplace environment. Particle number measurements over the size range from 14 nm to 10 μm using a scanning mobility particle sizer and an optical particle counter showed that welding produced an approximately lognormal particle mode with a 120 nm count median and a geometric standard deviation of 2.07. This study produced welding particle number concentrations on the order of 2×10^5/cm^3 in the building air 8.5 m away from the welding. Workplace exposure samples were below the current 8-hour American Conference of Governmental Industrial Hygienists mass concentration threshold limit value of 5 mg/m^3. Submicron particles comprised 80% of the total aerosol mass collected by a cascade impactor during welding. The concentration of larger particles was indistinguishable from indoor background. Microscopy showed that the welding emissions are dominated by clusters formed from <0.1 μm primary spheres. These data on the particles resulting from aerosol transformation by natural dilution inside an industrial building can be compared with laboratory-scale studies of welding particulate. The particle number characteristics observed in this study are significant because toxicological hypotheses suggest that number or surface area may be a better metric than mass when evaluating the health effects of fine particles.

Keywords: electron microscopy, occupational exposure, particle number, SMAW, welding

Inhalation exposure to submicron particles is a concern for both environmental and occupational health research. Epidemiology has shown that adverse acute health effects are associated with small incremental changes in the concentration of ambient particulate matter (PM) even at concentrations previously considered to be safe.[1–3] Current toxicological hypotheses involve particle surface area; particle number; ultrafine particles (<50 nm); bioavailable transition metals; acid aerosols; secondary sulfate and nitrate particles; peroxides; organic compounds; soot; and interactions between copollutants.[4–6] Occupational exposure limits (OELs) are based on the responses of healthy working-age individuals and allow PM exposures that are one to two orders of magnitude higher than ambient air quality standards, which are written to protect sensitive individuals with an adequate margin of safety.[7,8] Research is needed on occupational PM characteristics beyond the mass-based measurements routinely made for regulatory compliance monitoring.

Inhaled particles provide a vehicle for metals to enter the body in inappropriate doses. Larger particles that are deposited in the upper airways are rapidly cleared from the respiratory system into the pharynx, where they are swallowed. For these particles the dose-response relationships are based on the ingestion pathway.[9] However, submicron particles are deposited deep in the lung and ultrafine particles can pass from the lung and be circulated to other parts of the body.[10] Further, recent laboratory studies have shown that ultrafine particles (<50 nm) of metal oxides and

carbon black have greater biological effects than an equal mass dose of fine (100–300 nm) particles.[11,12]

The establishment of OELs for airborne contaminants is based on the amount of mass inhaled by a worker in a typical 8-hour workday. An 8-hour time-weighted average (TWA) threshold limit value (TLV®) of 5 mg/m³ for welding fumes not otherwise specified (NOS) has been established by the American Conference of Governmental Industrial Hygienists (ACGIH).[7] This limit is based on metal arc or oxyacetylene welding of iron, mild steel, or aluminum. The TLV applies only to unspecified contaminants in the welding fume (e.g., the total fume and not its individual components). The literature used to establish the TLV has been documented.[13] Evaluation of occupational exposure to welding fume NOS is traditionally performed by the collection of "total PM" using a collection device having the ability to capture particles that may be hazardous when deposited anywhere in the respiratory tract. The term "total PM" is defined by ACGIH as airborne material sampled with a 37-mm closed-faced cassette. Occupational sampling for comparison of worker exposure to mass-based guidelines involves the capture of PM in size ranges from ultrafine to extremely course.

Limited data are available on submicron particle mass and number concentration, especially for ultrafine particles in occupational settings. An arc welding emission study using a microorifice cascade impactor showed particle size measurements having mass geometric means ranging from 0.46 to 0.59 μm.[14] The smallest fraction resolved by the impactor was d<0.071 μm. An animal inhalation study of stainless steel welding reported that diameters of fume particles were lognormally distributed and varied in size from 0.02 to 0.81 μm.[15] Published studies of welding emissions by microscopy have shown individual particles on the order of 0.1 μm, clusters of particles, and fine particles in chain aggregates.[16,17] Studies of rat lungs after exposure to stainless steel welding fumes have shown a major population of 0.1–0.25 μm particles and a minor population of 5 to 100 nm particles.[18] Occupational exposures below the mass concentration threshold identified in the published welding fume ACGIH TLV may be biologically important because large number counts of ultrafine particles enhance aerosol surface area. Animal studies evaluating particle dissolution and cytokine induction on exposure of rat lungs to welding fumes suggest the importance of particle size because the dissolution of low-solubility materials is controlled by surface area.[19–21]

Aerosols formed by metal vaporization evolve rapidly due to condensation and coagulation as the hot vapor cools and is diluted by mixing. This study complements recent laboratory-scale research on welding emissions by providing field data on the welding aerosol that occurs under well-mixed conditions in a typical industrial building.[22]

METHODS

The goal of this study was to characterize the aerosol produced by natural mixing of welding fumes with the air in a typical industrial building. An experimental design was selected to produce mass concentrations much higher than ambient air but below the current welding fume 8-hour TWA TLV. All welding emission measurements were made in a university-operated research facility located in a converted equipment manufacturing shop. No operations other than the test welding were taking place in the building during the sampling. The site, located in an industrial zone 150 m from an interstate highway, is typical of locations where welding occurs in industrial maintenance and steel fabrication occupations. The building is a metal structure 18 m wide by 52 m long by 11 m high with a 10-ton overhead bridge crane. Rollup overhead doors, wall louvers, and three 1.5 m diameter wall fans provide ventilation. This study was performed in the winter with the fans off, resulting in three to four building air changes per hour from infiltration and natural convection. Natural gas fired radiant heaters maintained an inside air temperature of 10–15°C. Table I summarizes the outdoor ambient conditions.

TABLE I. Ambient Conditions

Sampling Date	December 16, 2000	December 20, 2000
Outdoor temperature	0°C	3°C
Wind	9 mph	10 mph
$PM_{2.5}$	10 μg/m³	23 μg/m³
PM_{10}	35 μg/m³	68 μg/m³

The welding particle emission source consisted of manual shielded metal electrode electric arc welding (SMAW) of carbon steel. Welding took place on a carbon steel pipe with the arc 15–30 cm above the concrete floor. The electrode was type E6011, 0.32 cm diameter, Lincoln (Cleveland, Ohio) Fleetweld 180. Welding current was 115 amps and an electrode was consumed every 2 min during the sampling period.

Figure 1 shows the configuration of the welding operation and the sampling instruments. The monitoring locations were selected to approximate the particle exposure received by workers in the

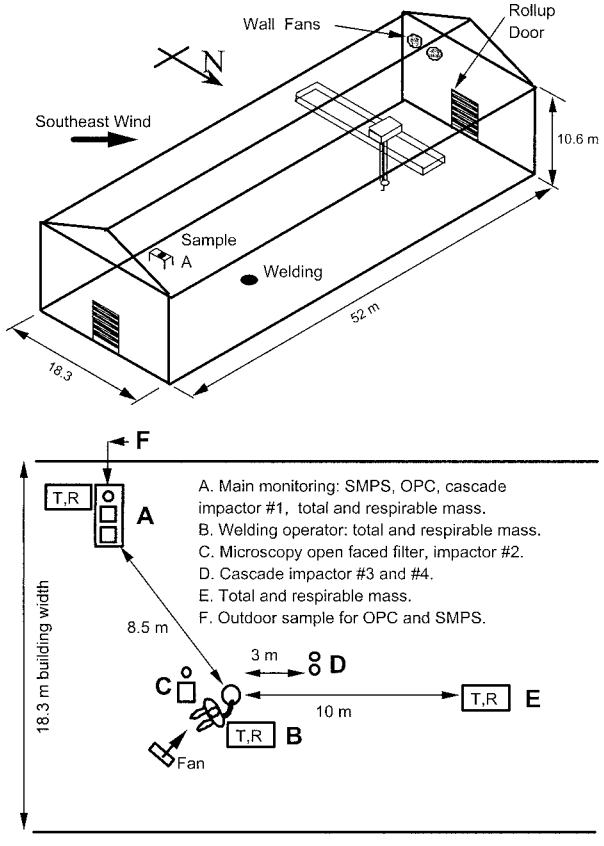

FIGURE 1. Schematic of indoor air sampling intended to simulate general workplace exposure in a typical building used for light fabrication or industrial maintenance welding

TABLE II. Particle Mass Loading

Sample Location	Total Aerosol (mg/m³)[A] 160 Min TWA[D]	Total Aerosol (mg/m³)[A] 8-Hour TWA	Respirable Aerosol (mg/m³)[B,C] 160 Min TWA	Respirable Aerosol (mg/m³)[B,C] 8-Hour TWA
Monitoring station, location A	1.74	0.58	1.13	0.377
Welder breathing zone, location B	3.14	1.04	1.38	0.460
Downwind exposure, location E	4.28	1.43	1.55	0.517

[A]50% cut point of approximately 25 μm.
[B]50% cut point of 3.5 μm.
[C]TLV is a threshold exposure limit for total aerosol. A TLV for the respirable fraction of welding aerosols has not been established.
[D]TWA exposure for the time period shown.

general area who were not actually performing the welding. A fan was placed behind the welder to purge the breathing zone and allow the emissions to mix with the room air by convection. The main particle monitoring station ("A" in Figure 1) was located at tabletop height 8.5 m (28 ft) from the welding. A TSI, Inc. (St. Paul, Minn.) model 3080 scanning mobility particle sizer (SMPS) consisting of a model 3081 differential mobility analyzer (DMA), a model 3022 condensation particle counter (CPC), and Aerosol Instrument Manager Version 4.0 software was used to measure particle size and number concentration from 14 to 600 nm. The data were collected using a sheath flow of 3 L/min and a sample flow of 0.3 L/min. A Lasair model 310 (Particle Measuring Systems, Boulder, Colo.) optical particle counter (OPC) was used to measure particle number concentration from 0.3 to 10 μm. The OPC sample was diluted approximately 600:1 with HEPA-filtered air because the concentration during welding was greater than the instrument range. Calibration runs showed a linear relationship between dilution ratio and measured concentration over the particle size range of interest.

Particle mass and size were measured using an Andersen 1-acfm cascade impactor with a preseparator, eight impaction stages coated with Dow 316 silicone oil, and a Whatmann 910AH glass fiber final filter. Cascade impactor samples were taken at tabletop height at location A and at 3 m above the floor at locations C and D in Figure 1.

Particle samples for electron microscopy were collected on polycarbonate membranes (Millipore, type GTTP) using an open-faced filter holder 1 m horizontally and 0.75 m above the welding site (C in Figure 1). Sampling at 1 ft³/min for 30 sec provided an appropriate density of particles for imaging.

Occupational exposure monitoring of airborne mass concentration was conducted. One total and one respirable mass sampler were placed within 1 m of the SMPS and OPC, location A. One total and one respirable mass sampler monitored the breathing zone of the welder, location B. A final set of total and respirable mass samplers was placed 10.7 m (35 ft.) from the welding, location E.

Total and respirable aerosol mass were captured on Teflon®

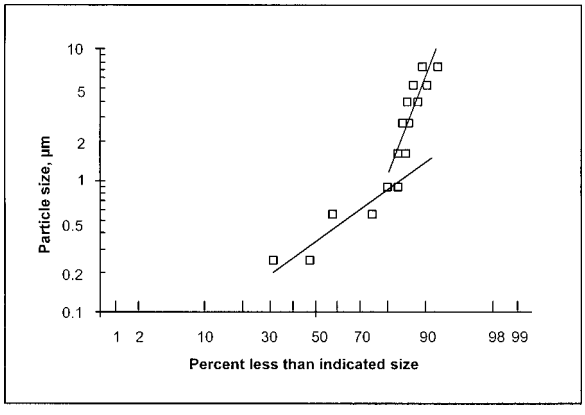

FIGURE 2. Log-probability mass distribution of indoor PM measured during SMAW welding. Duplicate cascade impactor measurements were made 3 m from the welding operation. Submicron particles comprise over 80% of the particle mass.

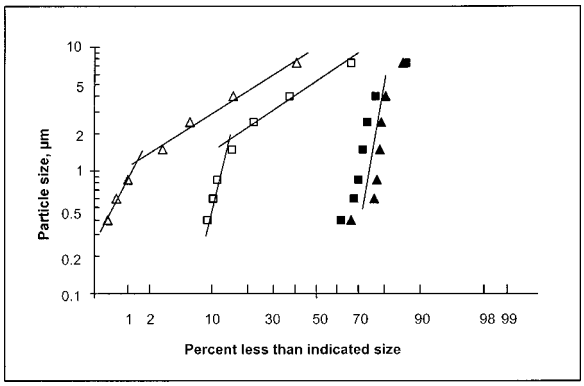

FIGURE 3. Log-probability particle volume calculated from OPC size and number measurements before (open symbols) and during (black symbols) welding on December 16 (triangles) and December 22 (squares). Points are the average of measurements made at 6-min intervals.

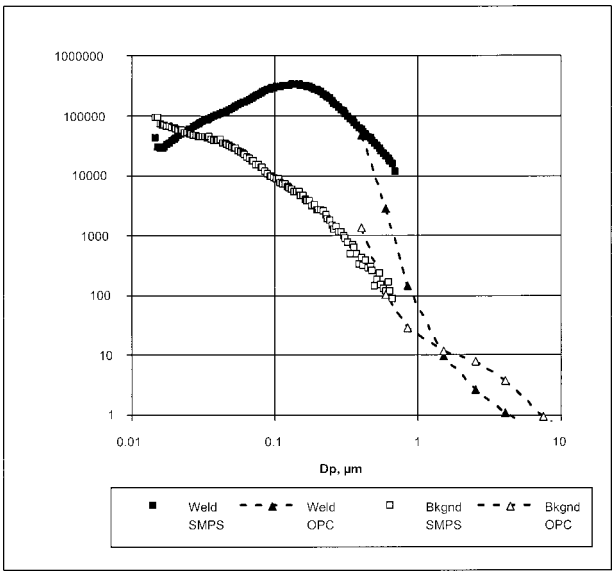

FIGURE 4. Particle number at the monitoring station 8.5 m horizontally and 1 m above the arc welding operation using a combination of an SMPS (squares) and OPC (triangles). For particles larger than 1 μm the concentration during welding operation (black symbols) was approximately the same as in room air prior to the start of welding (open symbols).

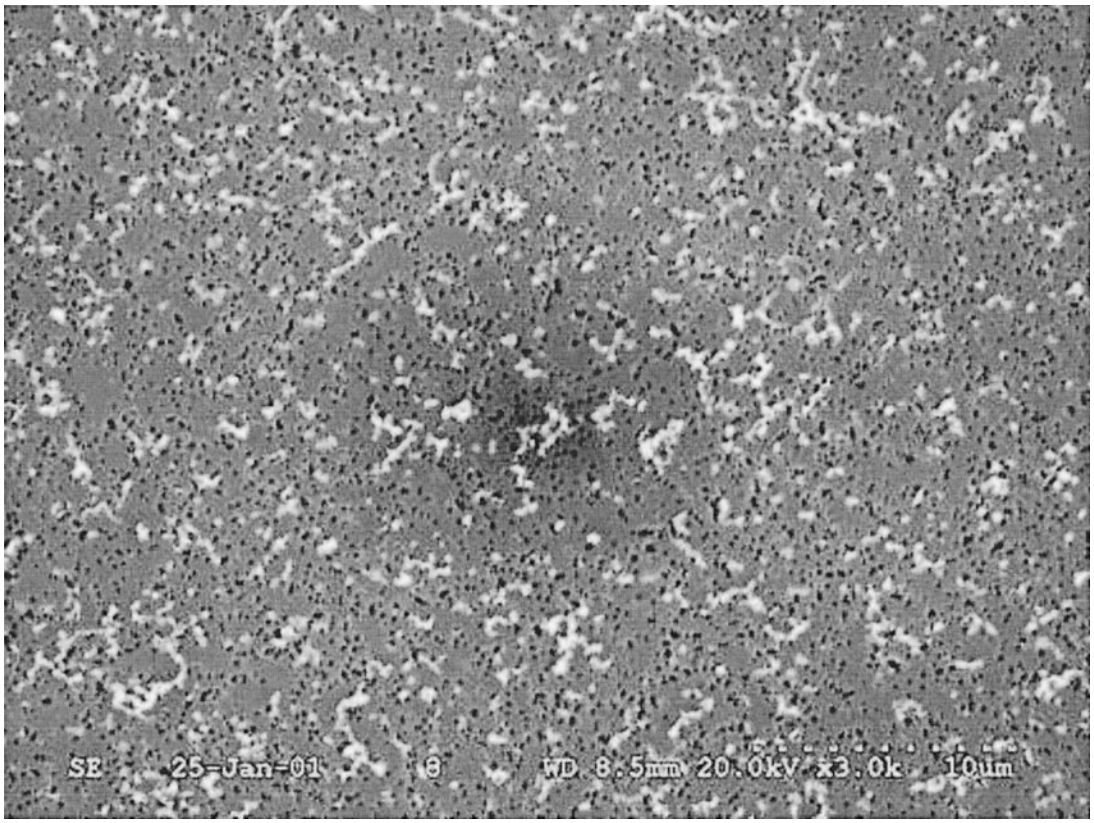

FIGURE 5. Open-face filter sample 1 m from the welding operation viewed at 3000×. The black circles are filter pores. The individual particles are clusters and long chains, not compact spheres.

preweighed, matched weight mixed cellulose ester membrane filters (0.8 μm pore size) housed in a two-piece, closed-face, 37-mm, polystyrene SKC cassette holder. For the collection of total aerosol mass the cassette holder was connected directly to a sampling pump allowing for the collection of particulate mass under the influence of negative pressure. For the collection of respirable mass the cassette holder was placed in a 10-mm nylon Dorr-Oliver cyclone separator (Mine Safety Appliances, Pittsburgh, Pa.) connected to a sampling pump. All connections were made using Tygon® laboratory tubing. Negative pressure was provided by SKC Aircheck high-flow sampling pumps (model 22451; SKC, Eighty Four, Pa.). The configuration and calibrated flow rate of the 37-mm cassette and of the cyclone separator allowed for collection of total and respirable particulate mass at 50% cut points of 25 and 3.5 μm, respectively. Both the total and respirable aerosol samples were gravimetrically analyzed by an American Industrial Hygiene Association-accredited laboratory using National Institute for Occupational Safety and Health Method 0500 for total particulate.

Quality assurance methods were employed to verify all instrument airflows and to check data for consistency between different instruments and between multiple runs. The time series data and the data obtained on 2 separate days provide a measure of reproducibility. A Gilibrator bubble tester (Gilian Instrument Corp.) was used to calibrate the total and respirable mass samplers at flow rates of 2.0 and 1.7 L/min, respectively. The Gilibrator was also used to verify the inlet flow rate on the SMPS and to verify the dilution ratio on the OPC. Aerosol dynamics simulations were run using MAEROS (Multicomponent Aerosol Time Evolution) to evaluate the expected effect of coagulation and deposition on the observed particle distribution.[23]

RESULTS

Table II shows the mass concentrations measured in the workplace air at the monitoring station, in the welder's breathing zone, and 10 m downwind of the welding. Airborne 8-hour TWA exposures to total PM at indoor locations, selected for their expected high fume concentration, were below the ACGIH TLV of 5 mg/m³ for welding fumes NOS. Both the direct measurement of particle mass distribution using the cascade impactor and the calculation of particle mass from the OPC size data show that SMAW emissions consist almost entirely of particles smaller than 1 μm. The cascade impactor data, plotted on log-probability axes in Figure 2, shows that over 80% of the mass during welding was in particles smaller than 1 μm. Figure 3 shows the PM size distribution both before and during welding calculated from OPC particle number measurements converted to volume assuming the particles are spheres. OPC data indicate that 70–80% of the PM volume during welding was in submicron particles.

The particle number distribution over the entire size range from 14 nm to 10 μm is shown on a log-log graph in Figure 4. The particle number increased by one to two orders of magnitude in the 0.05–0.5 μm size range during welding, whereas the background number of large particles remained approximately constant. The welding PM size distribution was approximately log-normal with a count median diameter of 120 nm and a geometric

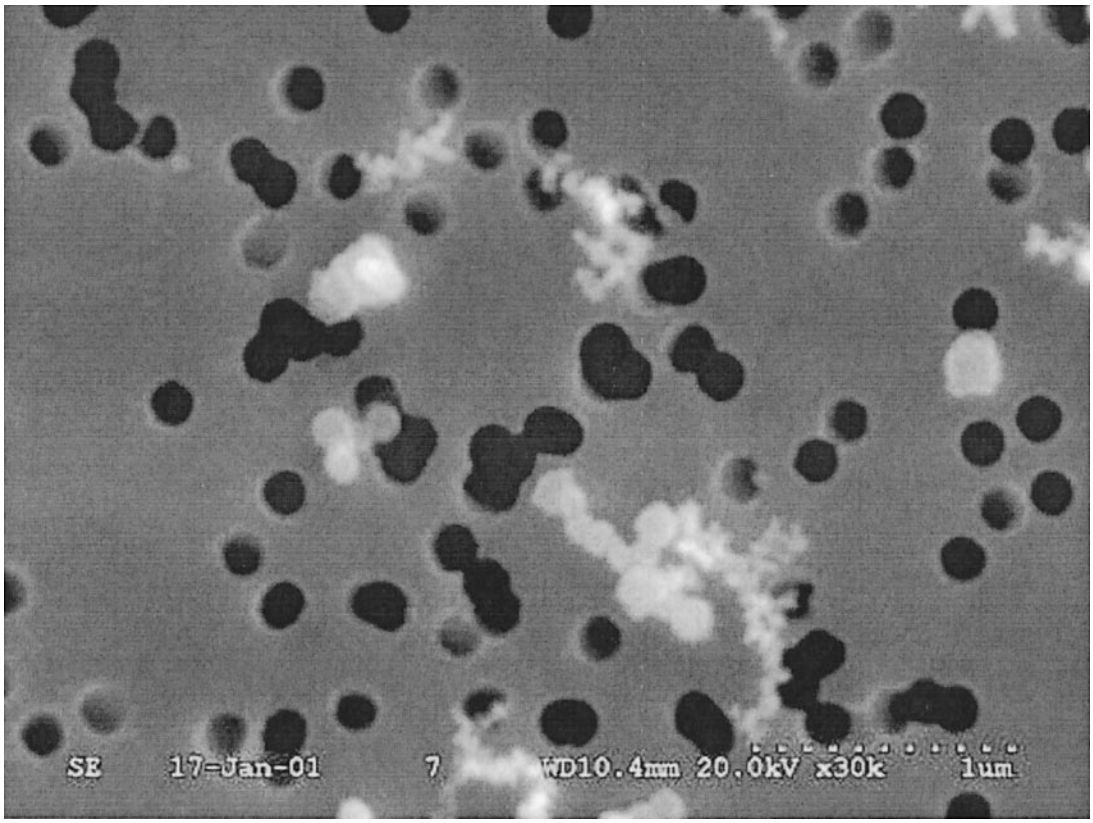

FIGURE 6. Detail of the welding particles at 30,000× shows that the aggregates are formed from primary particles less than 0.1 μm diameter. The spherical primary particles within a single aggregate have similar diameters but different aggregates contain different size primary particles.

standard deviation of 2.07. At monitoring location A the average total particle number in the range of the SMPS during welding was 250,000/cm³.

There is reasonable agreement between the SMPS and OPC measurements in the size range of overlap between the instruments considering that these instruments determine particle size by different physical principles. The OPC determines size by the intensity of the scattered light, which depends on particle shape and index of refraction. The SMPS determines size by electrical mobility, which depends on charge and aerodynamic drag, which is roughly proportional to surface area for fractal aggregates. The fundamentals of these techniques, including discussion of nonspherical particles, were reviewed by Baron and Willeke.[24]

Electron microscopy results showed that the welding particles were clusters and chain aggregates formed from much smaller primary particles. Figure 5 shows the open-faced filter deposit at 3000×, and Figure 6 shows typical aggregates at 30,000×. The sizes of the primary particles within a single cluster were uniform, but there were large differences in the primary particle sizes between different clusters.

DISCUSSION

The count median diameter of 120 nm and the particle size range reported by this study are consistent with results from earlier studies investigating welding particle size. These measurements of welding fumes in the mixed air of a typical industrial building are useful for comparison with welding fumes sampled in small laboratory enclosures, as in the studies by Hewett and Zimmer.[14,22] This study provides simultaneous mass and number measurements over the full particle size range from ultrafine to extremely coarse. Using the Hatch-Choate equations for a lognormal distribution, the count median diameter measured in this study converts to a mass median diameter of 590 nm.[25] This agrees well with the 500–700 nm mass median measured by Hewett using a cascade impactor.[14]

Coagulation simulations using MAEROS indicated that the size distribution of an aerosol with the mean size and number concentration reported by this study is relatively stable on the scale of minutes and that even 1 hour of aging would decrease the particle number by only about two-thirds, resulting in a 44% increase in the size distribution mode diameter. Because the building air exchange time was 15–20 min, the measurements were considered to represent a typical quasi-steady state aerosol size distribution for welding inside a large industrial building. The aerosol in the building air was slightly larger than the aerosol measured in an enclosure by Zimmer.[22] This is consistent with the extra time available for particle growth by coagulation as the welding fumes circulated in the building.

The high concentration of ultrafine particles in the indoor air prior to the start of welding (open squares on Figure 4) is suspected to be a result of the presence of the unvented, natural gas-fired, radiant space heaters because these particles were not found in the outdoor air sampled at the start and end of the session. The decrease in the ultrafine particle number after welding started was likely due to scavenging of ultrafines by coagulation with the increased number of larger particles from welding.

The difference observed in this study between weighed mass from the impactor and filters and calculated mass from particle number measurements is common in studies of this type. The relationship between number and mass is very sensitive to the details of the particle size distribution and requires making assumptions about particle shape and density.

The SEM data indicate that the particles measured in this study were aggregates, not spheres. The particle morphology observed in this study is also important for estimating the PM surface area. The clusters may behave as a single unit for aerodynamic deposition in the lung, but the primary particles have much more surface area than would be present if the same aggregate particle mass was in the form of a single sphere.

CONCLUSIONS AND IMPLICATIONS

Recent studies have shown that ultrafine particles may have greater biological effects than an equivalent mass of the same substance as larger particles.[10–12,21] This study reports the particle number concentration that was observed in a typical industrial maintenance or fabrication shop setting when the mass concentration of total aerosol was high but within current occupational guidelines for welding fumes NOS. Additional studies of this type are needed to build a database that can be used to estimate workplace ultrafine particle exposure for situations, such as retrospective occupational health studies, in which particle number measurements are unavailable. These data also are useful for estimating expected conditions when designing human exposure studies related to occupational exposure to ultrafine PM.

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