Characteristics of Lead Aerosols in Different Work Environments

Air samples were collected to examine physical and chemical characteristics of lead aerosols in different work environments in the airborne lead concentration range from 28 – 783 μg/m³. Size distribution of aerosols was measured using a Marple personal cascade impactor, and lead analysis was conducted using graphite atomic absorption spectrometry. Lead aerosol size distribution was found to be different from that of total particles. Such differences are not negligible and must be taken into account when estimating aerosol deposition within the human respiratory tract. Variation of MMAD (mass median aerodynamic diameter) and GSD (geometric standard deviation) of lead aerosols in the coarse mode with total airborne concentration was found to be small, and the assumption that lead aerosol distribution is constant is reasonable for establishing the relationship between airborne lead and blood lead concentrations. Respirable lead concentration is shown to be well correlated with the total airborne lead concentration in all work areas of the three plants except the charging area of the capacitor plant. Therefore, it is expected that either total airborne lead or respirable airborne lead concentration can be used to correlate blood lead concentration.

Keywords: air sampling, blood lead, lead aerosols

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formed due to nucleation, coagulation, and the condensation processes, and size distribution of lead aerosols depends on the temperature history of the system, chemical reaction rates, and surface tension of various species.

A survey of occupational lead exposure in Taiwan shows that lead powder (PbO and Pb₃O₄), lead-based stabilizing agent (lead stearate, tribasic lead sulphate, dibasic lead stearate), and lead-acid battery manufacturing are the three major industries that often have PbA levels over the current Taiwan time-weighted average PEL (TWA-PEL), 0.1 mg/m³, (10) Over 10% of the workers in these plants have PbB levels over 60 µg/dL. (12) Currently, the government is making serious efforts to reduce occupational lead exposure in these industries.

To assist the government in controlling occupational lead exposure, a lead-acid battery plant, a metallic film capacitor plant, and a lead powder plant were chosen to study lead and use for comparison of the physical and chemical characteristics of lead aerosols. The metallic film capacitor plant was chosen because it once had very high PbA concentrations, mainly contributed by the tin-lead metal sputtering process. Since the plant changed from tin-lead to tin-zinc, the PbA concentration has been reduced appreciably, but some areas of the plant still claim to have PbA concentration over the Taiwan TWA-PEL. It is suspected that lead contained in zinc as an impurity may be the cause of high PbA concentration, but the amount of lead in the zinc of the capacitor plant is not currently available.

The objectives of this study were to investigate the differences in lead particle morphology, possible variation of lead particle size distribution with total PbA concentration, content of lead in each particle size range, concentration of lead aerosols less than 1.0 µm in d₅₀, and concentration of respirable lead in total PbA in different areas of the three plants. The authors chose to study both particle and lead aerosol size distributions simultaneously because airborne particles usually contain elements other than lead, or intermix with lead oxides; hence, lead aerosol size distribution may be different from that of particles in the work areas. If particles contain elements other than lead, it will be more appropriate to estimate lead aerosol deposition in the human respiratory tract based on particle size distribution and lead content in each size range.

Area sampling was used to compare the physical and chemical characteristics of lead aerosols in different work environments. Subsequent work will focus on the relationship between the PbA determined from personal sampling and PbB in different work environments.

**Experimental Methods**

In this study airborne particles were sampled using Marple personal cascade impactors (model 298, Andersen Sampler, Inc.) at 1.5 m above the ground. For reference only, total PbA concentrations were determined by Millipore 37-mm filter cassettes. More detailed descriptions of work sites as well as the sampling and analysis procedure are described below.

**Work Sites**

The manufacturing processes of the lead-acid battery plant were similar to those described in Hodgkins et al. (9) The processes include Pb oxide production, plate casting, plate cutting, plate pasting, plate drying, negative plate encapsulation, plate stacking, cast-on-strap, cover placement, acid filling, charging, etc. Plate cutting, casting, and pasting areas were chosen for this study. They are all at the same ground levels. The plant has a very good ventilation system where clean air is constantly blowing from the top of the ceiling and venting from the doors, which are normally open.

The processes in the metallic film capacitor plant involved rolling metallic films into a cylindrical element; bundling 25–35 elements into a single honeycomb using paper tapes, metal sputtering (tin-zinc) both open ends (top and bottom) of the elements in a sputtering machine; removing paper tapes; encapsulating; charging; and assembly, etc. Sputtering, charging, and assembly areas were chosen for the study. No Impactor samples were taken for the assembly area because lead concentration was found to be too low for size distribution measurement. The metal sputtering and paper tape removal areas were in the same room, which was air conditioned. The charging area was in a separate room with its own air condition. Charging was an automatic process in which capacitors were automatically lined up to be charged one after another in a rotating device, using mechanical abrasion. Unlike in the battery plant, windows and doors were normally closed.

The main product of the lead powder plant was litharge (PbO). Lead powder was first generated by melt-blown devices and transported to furnaces by conveyors. In these melt-blown devices, lead ingots were first melted in crucibles; the melt then was blown through air nozzles to become lead powders. Lead powder was heated in furnaces to about 600°C for conversion into litharge. Particles were generated mainly during powder transportation, powder loading and unloading into furnaces, and lead powder packaging. The plant uses stand-alone electric fans for ventilation near suspected pollutant sources. It also counts on natural ventilation through doors and windows, which were kept open.

**Sampling and Analysis**

Typical sampling time ranged from 5 to 8 hours. The Marple personal cascade impactor was operated at 2 L/min, and the cut points (in d₅₀) of the eight stages were 0.52, 0.93, 1.55, 3.5, 6.0, 9.8, 14.8, and 21.3 µm. Fraction of internal particle loss, f, and inlet sampling efficiency, e, in a quiescent environment were determined by Rubow et al. (12) Overall sampling efficiency, E, of the impactor is calculated as E = e (1-f) and was found to be ≥90% for particles less than 7 µm in d₅₀, 84% for 10 µm particles, and 56% for 20 µm particles with an inlet visor. (12) In this study wind speeds in the workspaces were typically less than 1.0 m/sec, considered as nearly calm wind situations. Measured particle mass concentration at each size range was divided by E as shown in Figure 7 of Rubow et al. (12) to obtain a more accurate size distribution. A thin layer of silicon grease (No. 11025, Cling-Surface Co., Inc., Angola, N.Y.) was sprayed onto the substrate to prevent particle bounce following the procedure described in Marple et al. (13) Effectiveness of silicon grease to reduce particle bounce was studied previously (14).

MCE filters (mixed cellulose esters, GN-4, pore size: 0.8 µm, Gelman Sciences, Ann Arbor, Mich.) were used when the filter was to be analyzed only chemically and PTFE filters (Zeflour, pore size: 2.0 µm, Gelman Sciences) were used when the filter was to be analyzed both chemically and gravimetrically. To determine particle size distributions, PTFE filters were first weighed to within ± 1.0 µg accuracy before lead analysis. Weighing was performed in a chamber where relative humidity was controlled within 40 ± 5% and temperature was maintained from 15–22°C. Table 1 shows the precision of weighing in which the same blank filters were weighed once per day after 24 hours conditioning for 5 days consecutively. Average standard deviations for two blank MCE and two PTFE filters were shown to be 15.1 and 3.9 µg, respectively. Precision was hence greatly increased using PTFE filters, which did not absorb moisture as readily as MCE filters. For a typical sampling time of 6 hours at the flow rate of 2 L/min, the minimum

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particle concentration that can be determined accurately is about 6.8 μg/m³ by the PTFE filters. Compared with the minimum of about 10 μg/m³ found in the present study in the lower stages of the Marple personal cascade impactor, the precision of weighing was acceptable.

<table>
<thead>
<tr>
<th>TABLE I. Precision of Weighing for Blank Filters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Filter Material</td>
</tr>
<tr>
<td>Sample No.</td>
</tr>
<tr>
<td>Day 1</td>
</tr>
<tr>
<td>Day 2</td>
</tr>
<tr>
<td>Day 3</td>
</tr>
<tr>
<td>Day 4</td>
</tr>
<tr>
<td>Day 5</td>
</tr>
<tr>
<td>Average</td>
</tr>
<tr>
<td>SD</td>
</tr>
<tr>
<td>Average SD</td>
</tr>
</tbody>
</table>

Note: Measurements are in milligrams.

To determine the total PbA concentration or lead aerosol size distribution, the substrates were first treated by a microwave digestion system (model MDS 2000, CEM Corp., Matthews, NC). A graphite atomic absorption spectrometer (model Z 8100, Hitachi Corp., Tokyo, Japan) was then used to determine the lead aerosol concentration. The quality assurance procedure was similar to that described in Baldwin et al. The limit of detection was found to be 0.05 μg/m³, which is about one-sixth of the lowest lead concentrations found in the lower stages of the Marple personal cascade impactor.

Accuracy and precision of lead analysis is shown in Table II. The data shown are for reference samples taken with MCE filters, although PTFE filters show similar results. Two different standard materials, NIST SRM 3087 (metals on filter media) and NIST SRM 1648 (urban particle matter), were used to quantify the accuracy (or percentage of recovery) of lead analysis. In another test, silicon grease was sprayed onto the filter and NIST 1648 was added (NIST 1648 + grease) to determine if there was any interference with chemical analysis by the grease coating. Each test was repeated five times to determine the precision. The table shows for NIST 3087, NIST 1648, and NIST 1648 + grease, the average recovery plus or minus one standard deviation is 96.0 ± 7.2%, 93.8 ± 4.9%, and 104.2 ± 6.4%, respectively. The test results indicate that current lead analysis yields data of good quality.

To observe particle morphology using scanning electron microscopy, particles were collected on the filter cassette using polycarbonate filters.

**RESULTS AND DISCUSSION**

**Particle Morphology**

Particles collected at different work areas look different. Figures 1(a)–(c) show typical pictures of particle morphology at the plate cutting area of the battery plant, the sputtering area of the capacitor plant, and the furnace area of the lead powder plant, respectively. Particles generated by mechanical cutting are large and look irregular, as shown in Figure 1(a). Many submicron particles are generated in the high temperature sputtering process. Some particles further coagulate into chain-like aggregates as shown in Figure 1(b). Observation of individual particles shows that many fine particles are spherical. Since the paper tape removing area was in the same room, many large particles were also collected on the filter samples.

In the lead powder plant, since the furnace temperature of 600°C was below the melting point of litharge (888°C), it was expected that fewer submicron particles would be generated than in the metal sputtering area of the capacitor plant. Particles collected in the furnace area of the lead powder plant indeed look spherical and large as shown in Figure 1(c).

**Total PbA Concentration**

Total PbA concentrations determined by Marple cascade impactors are shown in Table III. Data obtained from the 87-mm cassette are included in Table III when impactor data are not available. Overall sampling efficiencies were used to correct original lead aerosol concentration measured at each stage and after filter of the Marple cascade impactor. Total PbA concentration was calculated as the sum of the corrected Pb concentrations at each stage and after filter, and it should represent the actual PbA concentration better.

Total PbA concentration varied by work site, and many samples were above the Taiwan PEL, 0.1 mg/m³. In the battery plant, average total PbA concentrations (plus or minus one standard deviation) were 164 ± 155 and 200 ± 81 μg/m³ for the cutting and pasting areas respectively, as determined by the Marple cascade impactor. Standard deviations were large because some samplers were located nearer the pollutant sources than others, and air circulation patterns might be different at different sampling locations. At the casting areas, all samples resulted in total PbA concentration below 0.1 mg/m³.

At the assembly area of the capacitor plant, total PbA concentration was well below 0.1 mg/m³, as shown in Table III. The sputtering and charging areas had much higher PbA concentrations, 183 ± 59 and 380 ± 33 μg/m³, respectively, as determined by the Marple cascade impactor. High total PbA concentration in the sputtering area was contributed mainly by the paper tape removing process in the area, in which many large particles were generated. Particle size distribution will be discussed later. Compared with the battery plant, the relative...
standard deviation, defined as the standard deviation divided by the average of the PbA, was smaller presumably because the working areas were more airtight and aerosol particles were more uniformly distributed in this plant.

### TABLE III. Total PbA Concentration in Three Work Environments

<table>
<thead>
<tr>
<th>Plant</th>
<th>Site</th>
<th>No. of Samples</th>
<th>Range $\mu$g/m$^3$</th>
<th>Avg. $\pm$ SD $\mu$g/m$^3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Battery</td>
<td>casting</td>
<td>4$^a$</td>
<td>28–80</td>
<td>57 $\pm$ 23</td>
</tr>
<tr>
<td></td>
<td>cutting</td>
<td>4</td>
<td>69–395</td>
<td>164 $\pm$ 155</td>
</tr>
<tr>
<td></td>
<td>pasting</td>
<td>4</td>
<td>123–300</td>
<td>200 $\pm$ 81</td>
</tr>
<tr>
<td></td>
<td>Overall average</td>
<td></td>
<td>140 $\pm$ 112 $\mu$g/m$^3$</td>
<td></td>
</tr>
<tr>
<td>Capacitor</td>
<td>sputtering</td>
<td>4</td>
<td>110–249</td>
<td>183 $\pm$ 59</td>
</tr>
<tr>
<td></td>
<td>charging</td>
<td>4</td>
<td>340–419</td>
<td>380 $\pm$ 33</td>
</tr>
<tr>
<td></td>
<td>assembly (4)$^b$</td>
<td></td>
<td>11–15</td>
<td>14.0 $\pm$ 2</td>
</tr>
<tr>
<td></td>
<td>Overall average</td>
<td></td>
<td>281 $\pm$ 114 $\mu$g/m$^3$</td>
<td></td>
</tr>
<tr>
<td>Lead powder</td>
<td>furnace</td>
<td>6</td>
<td>215–783</td>
<td>485 $\pm$ 245</td>
</tr>
<tr>
<td></td>
<td>melt-blown device</td>
<td>(4)</td>
<td>21–194</td>
<td>101 $\pm$ 87</td>
</tr>
<tr>
<td></td>
<td>Overall average</td>
<td></td>
<td>485 $\pm$ 245 $\mu$g/m$^3$</td>
<td></td>
</tr>
</tbody>
</table>

$^a$Data from Marple cascade impactor, corrected  
$^b$Data (in parentheses) from 37-mm filter cassette, not included in the overall average

At the lead powder plant, average total PbA concentration, $485 \pm 245$ $\mu$g/m$^3$, was very high at the furnace area as determined by the Marple cascade impactor. The melt-blown area had average total PbA concentration near the Taiwan PEL as determined by filter samples. The large relative standard deviation of total PbA concentration seen at the furnace area is again the indication of an open work site. The high total PbA concentration shows that the ventilation system is not efficient enough to reduce aerosol concentration generated from pollutant sources.

### Particle and Lead Size Distributions

Experimental results show that the lead aerosols were mainly in the coarse mode. Typical size distributions are shown in Figures 2(a)–(c) for the cutting area of the battery plant, the sputtering area of the capacitor plant, and the furnace area of the lead powder plant, respectively. In the figures the gray area is for lead aerosols and the hatched area is for particles. As is readily seen, the size distribution of lead aerosols was quite different from that of particles. For particles, both fine and coarse modes are present, while only the coarse mode is seen for lead aerosols. For particles, as shown in Figure 2(a), the mass median aerodynamic diameter (MMAD) and geometric standard deviation (GSD) are 8.86 $\mu$m and 1.71 for the fine mode, and 13.24 $\mu$m and 1.82 for the coarse mode, respectively. MMAD and GSD for lead aerosols are 14.24 $\mu$m and 2.04. MMAD is smaller for particles compared with lead aerosols. The lognormal parameters reported above and hereafter were obtained from the DISTFIT computer program (TSI Inc., St. Paul, Minn.).

Lead content varied from 9.7–41% for particles with $d_{50}$ larger than 1.0 $\mu$m, and 0.1–1.9% for submicron particles. Overall, lead content in particles was about 14%. Qualitative X-ray diffraction and X-ray energy dispersive analyses for samples collected on PTFE filters show that particles contain PbSO$_4$, Fe$_2$O$_3$, CaO, and SiO$_2$ in addition to PbO. The coexistence of CaO, FeO, and SiO$_2$, which mainly came from road dust, and PbSO$_4$, which mainly came
from the acid filling area, serves to explain partially why lead content of the particles in this area was low.

In the sputtering area of the capacitor plant, shown in Figure 2(b), characteristics of lead aerosol and particle size distributions in the coarse mode was similar to that in Figure 2(a). MMAD of particles was 24.6 μm, which was smaller than that of lead aerosols, 30.9 μm. Coarse particles were mainly generated from the paper tape removing process in the same area. Because of the high temperature operation, a large number of submicron particles were generated, which show up as a peak in the fine mode. Lead content ranged from 4–26% for particles larger than 1.0 μm in d<sub>ae</sub> and 4–21% for submicron particles. Considering all size ranges, lead content in the particles was 17%. Qualitative X-ray energy dispersive analysis shows particles mainly contained tin and zinc, which mixed internally with a small amount of Pb. Ambient aerosol constituents were small with respect to total dust content.

As shown in Figure 2(c), lead content was high and particles as well as lead aerosols were mainly in the coarse mode in the furnace area of the lead powder plant. Lead content ranged from 40–69% for particles larger than 1.0 μm in d<sub>ae</sub> and 12–13% for submicron particles. Lead content in the particles was 58%. Qualitative X-ray diffraction and X-ray energy dispersive analyses showed particles mainly contained PbO. Ambient aerosol constituents were minimal. MMAD of particles was 17.9 μm, which is smaller than that of lead, 22.0 μm.

Three impactor samples each were analyzed for both particle and lead aerosol size distributions at each plant. There were nine total samples. Considering all samples, lead content in particles was 7.7–18.4% in the battery plant, 13.7–17.3% in the capacitor plant, and 58.3–73.6% in the lead powder plant. Comparing the MMAD of the coarse mode of lead aerosols with that of particles, Figure 3 shows that the MMAD of lead aerosols was always less than (about 77%) that of particulates.

Variation of the MMAD of lead aerosols in the coarse mode with total PbA concentration is shown in Figure 4. Except one data point at the battery plant (marked with an asterisk in Figure 4), both battery and lead powder plants are seen in Figure 4 to have much smaller MMAD of lead aerosols than the capacitor plant. Excluding the exceptional data point, the MMAD of lead aerosols for the battery plant ranged from 12.6–17 μm and was nearly independent of total PbA concentration. The exceptional data, which had total PbA of 395 μg/m<sup>3</sup> and MMAD of 30.3 μm, was taken about 40 cm away from a cutting machine to increase
the particle concentration in the sample intentionally. Indeed, the mechanical cutting process generated large particles. The MMAD of lead aerosols for the lead powder plant ranged from 13–22 μm and was also nearly independent of total PbA concentration. MMAD of lead aerosols for the capacitor plant ranged from 25.2–33.1 μm. With few exceptions, most of GSDs of lead aerosols for all plants ranged from 1.8–2.6 and can be considered as nearly independent of total PbA concentration. In view of this conclusion, the assumption made in Froines et al. (2) that lead aerosol distribution is constant over the range of 50–200 μg/m³ is not unreasonable.

![IMM Agead MMD with total PbA concentration, coarse mode (*) = exceptional data point, the impacter is close to the cutting machine)](image)

**Respirable Lead and Lead Smaller than 1.0 μm**

Figures 5 and 6 show the concentration of lead aerosols smaller than 1.0 μm in dm and respirable lead aerosol versus total PbA concentration, respectively. Respirable lead aerosol concentration was integrated from the new American Conference of Governmental Industrial Hygienists’ (ACGIH) curve, (16) and the lead mass distribution was determined by the Marple cascade impactor as

\[
\text{respirable lead concentration} = \int \eta(d_m) M(d_m) d_m
\]

where \(M(d_m)\) is the lead mass distribution, \(\eta(d_m)\) is the collection efficiency defined in the ACGIH criteria for respirable dust sampling. It is seen in Figure 5 that in the PbA concentration range of 0–800 μg/m³ considered, small lead aerosol concentration \((d_m \leq 1.0 \mu m)\) increases with increasing total PbA concentration. It varied from 1.7–10.8 μg/m³ for PbA ranging from 28–395 μg/m³ in the battery plant; 9.7–13.4 μg/m³ for PbA ranging from 110–249 μg/m³ in the sputtering area, or 2.1–5.6 μg/m³ for PbA ranging from 340–419 μg/m³ in the charging area of the capacitor plant; and 5.3–16.0 μg/m³ for PbA ranging from 214–783 μg/m³ in the lead powder plant. Exceptionally high lead concentration of small particles at the sputtering area was mainly caused by submicron particles generated during the process. The finding that small-particle lead concentration increases with increasing total PbA concentration is different from Hodgkins et al. (5) in which small-particle lead aerosol concentration was found to be relatively constant at 1.9 μg/m³ when PbA ranged from 12.1–91.2 μg/m³.

In Figure 6 respirable lead aerosol concentration except the charging area of the capacitor plant is shown to be well correlated with total PbA concentration in all work areas of the three plants.

![IMM Agead lead versus total PbA concentration](image)

Fraction of small-particle lead aerosols \((d_m \leq 1.0 \mu m)\) and respirable lead aerosols in total PbA is further examined in Figure 7. Typical fraction of small-particle lead \((d_m \leq 1.0 \mu m)\) aerosol in total PbA was less than 4% except in the charging and sputtering areas, where submicron particles were generated in the high temperature process. The fraction for these areas is seen to decrease with increasing total PbA concentration, when the contribution of fine mode to total mass concentration becomes less important. In other work areas there was no clear trend regarding the variation of the fraction with total PbA concentration.

![IMM Agead lead versus total PbA concentration](image)
will be in the 1–3% range as shown in Figure 7. The fine mode in the sputtering area adds to the respirable lead fraction to let it fall in the 5–10% range. Higher than 15% of respirable lead in the casting area of the battery plant further indicates that there is fine particle mode present.

Respirable lead concentration was shown to be well correlated with total PbA concentration in all work areas of the three plants except the charging area of the capacitor plant. The fraction of respirable lead in total PbA was shown to be independent of total PbA and within 5–10%, except in the charging area of the capacitor plant where the MMD of Pb was very large, and the charging area of the battery plant where fine particle contribution to total PbA concentration was very high. Therefore, it is speculated that whether total PbA or respirable PbA concentration is used to correlate, PbB concentration generally will make no significant difference.

Future research will focus on establishing the relationship between PbA and PbB using the personal sampling method. Also for workplaces where PbA concentrations are high, control measures are now being taken to reduce PbA concentration levels.

CONCLUSIONS

This article discusses the physical and chemical characteristics of lead aerosols in different work areas of three plants using the area sampling method. A range of total PbA concentration from 28–783 µg/m³ was examined. The following is concluded from the study.

It is seen that particle generation by different processes influences particle morphology, lead content, and lead size distribution. Concentration of lead aerosols is further influenced by the materials used, room ventilation conditions, and the distance between a sampler and pollutant sources. High temperature processes such as sputtering and casting tend to generate submicron spherical particles that may contribute significantly to fine particle mass. Processes involving mechanical actions generate large irregular particles, where small aerosol mass concentration is low, such as in the charging area of the capacitor plant.

Size distribution of lead aerosols is different from that of particles. In the coarse mode, the MMD of lead aerosols is about 77% of that of particles. In estimating lead aerosol deposition within the human respiratory tract, such differences must be taken into account if lead aerosols are internally mixed with other particles, such as in the sputtering area of the capacitor plant.

Variation of MMD and GSD of lead aerosols in the coarse mode with total PbA concentration was found to be small. This confirms earlier work by Froines et al. who found that lead aerosol distribution is constant over the range of 50–200 µg/m³.

Small-particle lead (dₚ ≤ 1.0 µm) aerosol concentration is typically larger than 1.0 µg/m³ and can be larger than 10 µg/m³ in high temperature processes and high PbA concentration situations. The concentration increases with increasing total PbA concentration. The conclusion differs from that of Hodgkins et al. who found that small-particle lead aerosol concentration was relatively constant at 1.0 µg/m³ when PbA ranged from 12.1–91.2 µg/m³.

REFERENCES