

# Mass-size distribution and concentration of metals from personal exposure to arc welding fume in pipeline construction: a case report

Show-Yi YANG<sup>1, 2\*</sup>, Jia-Ming LIN<sup>1</sup>, Li-Hao YOUNG<sup>3</sup> and Ching-Wen CHANG<sup>1</sup>

<sup>1</sup>Institute of Environmental Health, College of Public Health, National Taiwan University, Taiwan

<sup>2</sup>Institute of Labor, Occupational Safety and Health, Ministry of Labor in Taiwan, Taiwan

<sup>3</sup>Department of Occupational Safety and Health, China Medical University, Taiwan

Received November 21, 2017 and accepted April 3, 2018

Published online in J-STAGE April 7, 2018

**Abstract.** We investigate exposure to welding fume metals in pipeline construction, which are responsible for severe respiratory problems. We analyzed air samples obtained using size-fractionating cascade impactors that were attached to the welders performing shielded metal and gas tungsten arc welding outdoors. Iron, aluminum, zinc, chromium, manganese, copper, nickel, and lead concentrations in the water-soluble (WS) and water-insoluble (WI) portions were determined separately, using inductively coupled plasma mass spectrometry. The mass-size distribution of welding fume matches a log-normal distribution with two modes. The metal concentrations in the welding fume were ranked as follows: Fe>Al>Zn>Cr>Mn>Ni>Cu>Pb. In the WS portion, the capacities of metals dissolving in water are correlated with the metal species but particle sizes. Particularly, Zn, Mn, and Pb exhibit relatively higher capacities than Cu, Cr, Al, Fe, and Ni. Exposure of the gas-exchange region of the lungs to WS metals were in the range of 4.9% to 34.6% of the corresponding metals in air by considering the particle-size selection in lungs, metal composition by particle size, and the capacities of each metal dissolving in water.

**Key words:** Pipeline construction, Water-soluble metals, Welding fume, Mass-size distribution, Respirable particulate matter

## Introduction

Welding fumes can cause adverse respiratory effects, which includes impaired pulmonary function, metal fume fever, cough, and an increased susceptibility to bronchitis and pneumonia in occupational cohorts<sup>1</sup>. Additionally, welding aerosols may induce lung injury depending on the inherent toxicity of the inhaled metal species<sup>1, 2</sup>.

Particulates are formed during welding through nucleation, condensation, and coagulation<sup>3</sup>. In nucleation mode, the primary particles having sizes ranging from a few nanometers to 0.1 μm are formed through vapor condensation. In the accumulation mode, the accumulated particles with sizes ranging from 0.1 to 1 μm include aggregates and agglomerates that are named based on their morphological appearance. Aggregates are fused clusters of primary particles, whereas agglomerates are groups of primary particles that are adhered together by electrostatic or van der Waals forces<sup>4</sup>. The particles with sizes of 1 to 20 μm (microspatters) are obtained from a liquid ejection<sup>5</sup>.

\*To whom correspondence should be addressed.

E-mail address: year@mail.ilosh.gov.tw

©2018 National Institute of Occupational Safety and Health

Particulate matter (PM) has been classified based on the regions of deposition in the respiratory tract. Inhaled particulate matter (IPM), thoracic particulate matter (TPM), and respiratory particulate matter (RPM) are defined by the European Committee for Standardization<sup>6)</sup>, the International Standards Organization<sup>7)</sup>, and the American Conference of Governmental Industrial Hygienists<sup>8)</sup>. IPM is deposited anywhere in the respiratory tract, whereas TPM may be deposited only in the tracheobronchial region. Both IPM and TPM are cleared by cilia within minutes and is translocated to the gastro-intestinal tract. RPM can reach the alveolar region, which does not expel RPM often. Therefore, the fate of RPM is determined by the particular respiratory maneuver and physicochemical characteristics of the PM.

When PM avoids respiratory clearance, the solubility of the PM determines the impact on the respiratory tract. The water solubility of the particles controls both the dissolution and residence time of the PM in the respiratory tract. Soluble particles dissolve into the surfactant layer of the alveoli or in the mucus layer<sup>9)</sup>. For instance, Mn and Ni from the WS portion that are deposited in the lungs are absorbed easily than those in the WI portion<sup>10, 11)</sup>. Moreover, the repairable and soluble Mn in particles is correlated with urinary Mn<sup>12)</sup>. However, Mn-exposure over a workshift has not been correlated with the level of Mn in blood or urine, but Mn in toenail would be a valid long term cumulative biomarker<sup>13)</sup>. Additionally, it has been suggested that WS metals are primary contributors that cause adverse cardiopulmonary health effects that is observed in rats<sup>14)</sup>. Respirable PM increases oxidative stress and can cause inflammatory adverse effects on the respiratory system<sup>15)</sup>. The chemical and physical properties of the particles, particularly their water solubility, may trigger biological effects<sup>9)</sup>.

Air sampling for PM and chemical analysis for metals are routinely performed as industrial hygiene activities to comply with the allowable exposure levels to welding fume. However, metal concentrations in the air do not correlate well with the levels of the corresponding metals in urine or blood in certain cases. In fact, the exposure levels occurring in lung would be relatively close to internal exposure, since the size selection of the particles in lungs and water solubility of the metals govern the dissolution in the surfactant of respiratory system. Therefore, we sample welding fume in pipeline construction in our study to characterize the mass-size distribution of PM, determine the metal species in the PM, analyze WS metals and WI metal particles based on their size, and estimate the exposure to WS metals in different lung compartments.

## Subjects and Methods

### *Study subject*

A total of 5 male welders were recruited for this field study, based on similar tasks assigned, welding techniques with shielded metal arc welding (SMAW) and gas tungsten arc welding (GTAW), and a welding duration of at least two hours. The study subjects were employed to construct a 1,900 km pipeline in an oil refinery factory in the Mailiao Industry Park, Taiwan. For avoiding personal technical variation, the study recruited welders that had at least five years of welding experience. Their tasks included butt-welding joints, as well as flange neck and fillet welding on carbon steel. The GTAW with a fillet of ER70S-G (Fe-98.04%, Mn-1.16%, Si-0.55%, Cu-0.16%, Cr-0.05% and Ni-0.04%), and the SMAW with a fillet of E7016 (Fe-98.04%, Mn-1.6%, Si-0.75%, Ni-0.3% and Cr-0.2%) were employed to weld of 20% carbon steel pipes and 80% stainless steel pipes with an anti-rust painting. Each run of welding was performed in triplicate, one layer at a time. The first layer was conducted using GTAW at 100 to 150 A, 13 to 24 V, and 5-15 cm/min; the second layer used SMAW at 100-140 A, 20-30 V, and 10-20 cm/min; and the third layer used SMAW at 130-180 A, 20-30 V, 10-20 cm/min. The combination of GTAW and SMAW is commonly used in vessel or work piece joining, especially for welding on thickness greater than 26 mm of work piece<sup>16)</sup>. The welders worked outdoors where temperatures were 24.1 °C to 35.5 °C, and wind speed was 6.9 m/sec on average with a maximum of 10.5 m/sec. To avoid wind turbulence, every welders was surrounded with a 2 to 3 meter of diameter temporary wind shield.

The study was approved by the Institutional Review Board of Tri-Service General Hospital in Taiwan, all participants were aware of the study aim and participated voluntarily and written informed consent was obtained from all study participants.

### *Air sampling*

Samples were collected by using a Marple Cascade Impactor Model 298 (Andersen Samplers, Inc., Atlanta, GA, USA). The Marple Cascade Impactor has eight stages with collection bands: <0.52 µm (assuming the lower limit: 0.1 µm)<sup>17)</sup>, 0.52-0.93 µm, 0.93-1.55 µm, 1.55-3.50 µm, 3.50-6.00 µm, 6.00-9.80 µm, 9.80-14.80 µm, 14.80-21.30 µm, and >21.30 µm (assuming the upper limit: 31.35 µm)<sup>18)</sup>. Each stage had a 34-mm perforated Polyvinyl chloride (PVC) membrane (0.8 µm pore size; SKC Corp) mounted. The Marple Cascade Impactor was mounted on

a participant's shoulder, and ran at flow rate of 2 l/min for two hours.

#### *Gravimetric analysis of welding fume*

Each of the PVC membrane was conditioned before and after air sampling at a temperature of  $21 \pm 2^\circ\text{C}$  and a relative humidity of  $50 \pm 3\%$  for 48 h. Then, the membrane was weighted twice using a balance with a 0.01 mg resolution (Mettler-Toledo MX5 scale, Mettler-Toledo, Columbus, OH, USA). The weight difference between pre- and post-sampling was calculated for each membrane to designate the mass of the PM.

#### *Analysis for metals in water-soluble and water-insoluble portions of welding fume*

To divide welding fume into WS and WI, the Ghio method<sup>19</sup> was followed. Each PVC membrane was placed into a 50-ml polypropylene tube with 40 ml of deionized water and agitated for 96 h. The membrane was removed, and the aqueous extract was centrifuged at  $2,500 \times g$  for 30 min, with the supernatant collected. This procedure was performed twice for each membrane. The two parts of the supernatant were pooled and then filtrated using a syringe filter holder (SWINNEX, Millipore, Billerica, MA, USA), cooperated with a mixed-cellulose ester (MCE) filter. Finally, the PVC membrane and the MCE filter were separately digested for analysis of WI metals.

And the pooled supernatant was for analysis of WS metals by following the NIOSH analytical method 7301 with two modifications<sup>20</sup>: (i) the hotplate was replaced with a microwave digester (CEM Corp., NC, USA), and (ii) the digesting acid was replaced with ultra-pure nitric acid (Sigma-Aldrich Inc., St. Louis, USA). Fe, Al, Zn, Cr, Mn, Cu, Ni, and Pb were determined with an Agilent 7500ce ICP-MS (WA, USA). Recovery was checked through two consecutive spikes of a quality control standard every ten samples analysis. The recovery for analysis ranged from 90% to 110%. The recovery of sample preparation was  $85.3 \pm 5\%$ , determined by spiking nickel chloride solution on three MCE filters, then extracting, filtrating, and analysis.

To express the capacity of a metal for dissolving in water, the proportion of a WS metal in total metal was calculated.

#### *Estimation of exposure to PMs and metals*

The concentration of inhalable, thoracic, and respirable PM ( $C_p$ ) from welding fume was estimated one at a time with the following equation (1):

$$C_p = \sum(C_i \times E_{ip}) \quad (1)$$

Where,  $C_p$ : concentration of the  $p$  form of PM (IPM, TPM, or RPM),  $C_i$ : concentration of the  $i$  size of PM in air,

$E_{ip}$ : collection efficiency of the  $i$  size of PM in the  $p$  form of PM, according to particle size-selective sampling criteria<sup>8</sup>.

The concentration of a metal in the IPM, TPM, or RPM ( $C_{jp}$ ) was also estimated one at a time with the equation (2):

$$C_{jp} = \sum(C_{ji} \times E_{ip}) \quad (2)$$

Where,  $C_{ji}$ : concentration of  $j$  metal in  $i$  size of PM in air, Furthermore, the concentration of a WS metal in the IPM, TPM, or RPM,  $WSC_{jp}$  was estimated one at a time with the following equation (3):

$$WSC_{jp} = \sum(C_{ji} \times E_{ip} \times F_{ji}) \quad (3)$$

Where,  $F_{ji}$ : WS proportion of the  $j$  metal in the  $i$  size of PM (%).

#### *Data analysis*

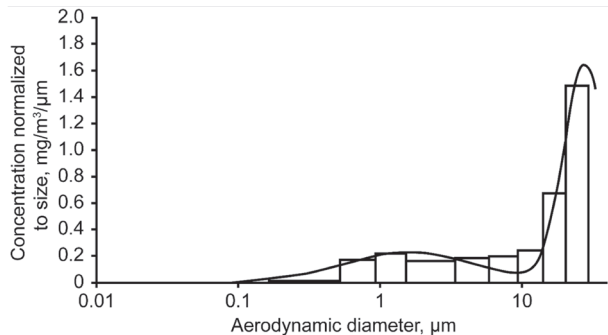
To test fitness of the mass-size distribution of welding fume to a log-normal distribution, a residual analysis was carried out with the Impactor package<sup>21</sup>. The histogram plot for the mass-size distribution of welding fume was drawn and characterized by the mass median aerodynamic diameter (MMAD) and the geometric standard deviation (GSD).

One-way ANOVA and two-way ANOVA were performed using SPSS version 14.0 (IBM SPSS Statistics, Chicago, IL, USA). ANOVA was applied to test capacity of metals in various size groups for dissolving in water. Scheffe's test was employed as a post-hoc analysis. The level for statistical significance was a  $p$ -value < 0.05.

## Results

#### *Mass-size distribution of welding fume*

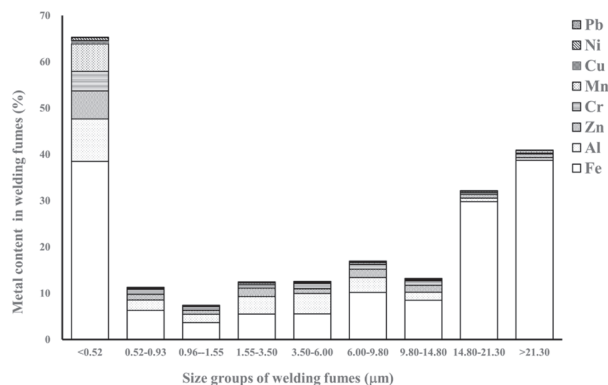
Figure 1 illustrates the mass-size distribution of welding fume. A bimodal log-normal distribution concludes that 39.7% of the PM have a size less than  $10 \mu\text{m}$  and 60.3% have a size greater than  $10 \mu\text{m}$ . The MMAD for PM with size less than  $10 \mu\text{m}$  is observed to be  $1.5 \mu\text{m}$  with a GSD of 3.16, whereas the MMAD for PM with size greater than  $10 \mu\text{m}$  is  $18.8 \mu\text{m}$  with GSD of 1.43. The concentration of the welding fume is  $4.45 \pm 0.31$  (mean  $\pm$  SD)  $\text{mg}/\text{m}^3$ . The length of welding employment of this current study subjects was  $22.2 \pm 13.9$  yr.



**Fig. 1. Bimodal mass-size distribution of welding fume during pipeline construction using SMAW (shielded manual metal arc welding) and GTAW (gas tungsten arc welding). PM>10 μm accounts for 60.3% of the total fume, whereas PM<10 μm accounts for 39.7% of the total fume.**

The MMADs for PM >10 μm is 18.8 μm with deviation (δg) of 1.43. The MMAD for PM <10 μm is 1.5 μm with δg of 3.16.

$$\% = \frac{\text{the mass of PM by size groups}}{\text{the mass of total fume}}$$



**Fig. 2. Metal composition (in percentage) categorized by the size group in welding fume, which is statistically different.**

**Table 1. Proportions of water-soluble metals over total mass of individual metals corresponding to the PM size groups<sup>(1)</sup>**

Size groups of PM, μm	Proportions of water-soluble metals <sup>(1)</sup> , mean ± SD, %							
	Fe	Al	Zn	Cu	Mn	Cr	Ni	Pb
<0.52	5.8 ± 2.7	18.4 ± 9.5	81.9 ± 14.0	36.5 ± 10.0	77.1 ± 8.9	31.2 ± 10.1	11.1 ± 3.6	36.9 ± 11.6
0.52–0.93	10.1 ± 5.4	18.3 ± 16.7	79.0 ± 17.9	27.5 ± 12.6	65.0 ± 18.3	22.1 ± 11.3	8.6 ± 2.7	46.3 ± 13.3
0.93–1.55	12.6 ± 7.0	16.3 ± 10.9	80.6 ± 16.8	28.1 ± 12.8	68.0 ± 19.6	24.6 ± 3.9	9.0 ± 1.7	52.1 ± 13.7
1.55–3.50	8.6 ± 4.3	20.1 ± 15.3	86.5 ± 12.7	35.8 ± 11.7	67.4 ± 16.6	22.5 ± 2.5	12.6 ± 9.8	40.8 ± 5.0
3.50–6.00	19.0 ± 12.6	31.2 ± 14.0	83.5 ± 13.5	33.6 ± 14.1	56.4 ± 12.1	28.8 ± 9.1	10.5 ± 5.4	62.4 ± 12.4
6.00–9.80	8.1 ± 3.0	22.9 ± 12.0	86.4 ± 15.1	34.0 ± 12.4	60.4 ± 18.7	19.7 ± 6.8	9.6 ± 3.6	57.3 ± 12.8
9.80–14.80	12.2 ± 7.6	18.7 ± 10.4	77.1 ± 13.5	31.0 ± 13.1	73.6 ± 11.3	20.5 ± 3.2	11.8 ± 5.9	55.0 ± 10.8
14.80–21.30	14.3 ± 11.0	19.1 ± 12.1	84.1 ± 15.6	25.2 ± 15.2	53.0 ± 15.1	19.4 ± 4.5	11.5 ± 5.5	44.8 ± 11.3
>21.30	15.3 ± 12.6	8.6 ± 6.0	71.9 ± 12.9	24.2 ± 9.9	37.9 ± 19.9	21.3 ± 8.9	7.2 ± 2.9	41.8 ± 8.7
Overall	11.8 ± 8.2	19.3 ± 12.3	81.2 ± 14.7	30.7 ± 16.5	62.1 ± 12.5	23.3 ± 7.4	10.2 ± 5.1	48.6 ± 11.4

PM: particulate matter.

$$(1)\% = \frac{\text{mass of water soluble metal}}{\text{total mass of individual metal by sizes of PM}}$$

*Metal composition along with size groups of welding fume*

Figure 2 illustrates the metal compositions in the nine size groups of the welding fume, which are statistically different (one-way ANOVA, n=9, p<0.05). Fe dominates every size group, especially the groups having sizes >21.3 μm, <0.52 μm, and in the range of 14.8 to 21.3 μm.

*WS proportion of metals by size groups of welding fume*

Table 1 depicts the WS proportion of metals in the welding fume. The WS proportion for Zn, Mn, Pb, Cu, Cr, Al, Fe, and Ni is 81.2%, 62.1%, 48.6%, 30.7%, 23.3%,

19.3%, 11.8%, and 10.2%, respectively. The WS proportion of metals significantly corresponds to the metal species, but the particle size (two-way ANOVA, p<0.001 for metal species, p=0.3 for particle size). The WS proportion of a metal denotes its capacity for dissolving in water.

*Estimation of exposure to PM and water-soluble metals in respiratory tract*

The concentrations of IPM, TPM, and RPM are 3.80 ± 0.26 mg/m<sup>3</sup> (85.4% of total PM), 2.02 ± 0.14 mg/m<sup>3</sup> (45.4% of total PM), and 1.37 ± 0.88 mg/m<sup>3</sup> (30.7% of total PM),

**Table 2. Estimation of exposure levels of inhalable, thoracic, and respirable PM and metals**

PM and metals	Air concentrations of PM and metals, $\times 10^{-3}$ mg/m <sup>3</sup>	Levels of exposure to PM and metals, mean $\pm$ SD, $\times 10^{-3}$ mg/m <sup>3</sup>					
		Inhalable PM		Thoracic PM		Respirable PM	
		Total	WS	Total	WS	Total	WS
PM	4,450 $\pm$ 307.1	3,800 $\pm$ 262.2	-	2,020 $\pm$ 139.4	-	1,370 $\pm$ 87.7	-
Fe	1,988.5 $\pm$ 103.4	1,388.0 $\pm$ 72.2	413.8 $\pm$ 15.7	419.6 $\pm$ 18.5	244.6 $\pm$ 8.3	198.8 $\pm$ 16.5	137.2 $\pm$ 4.9
Al	378.2 $\pm$ 13.6	329.4 $\pm$ 11.9	68.0 $\pm$ 3.3	280.6 $\pm$ 5.3	59.4 $\pm$ 2.9	214.1 $\pm$ 10.9	44.2 $\pm$ 2.2
Zn	219.6 $\pm$ 4.6	180.5 $\pm$ 3.8	147.1 $\pm$ 3.1	129.3 $\pm$ 2.7	105.8 $\pm$ 2.2	93.6 $\pm$ 5.1	76.0 $\pm$ 1.6
Cr	146.4 $\pm$ 2.9	125.9 $\pm$ 2.5	29.1 $\pm$ 0.6	101.7 $\pm$ 2.0	22.7 $\pm$ 0.5	76.0 $\pm$ 4.1	16.4 $\pm$ 0.3
Mn	134.2 $\pm$ 3.0	107.1 $\pm$ 2.4	63.6 $\pm$ 1.4	65.8 $\pm$ 1.4	32.9 $\pm$ 0.7	52.3 $\pm$ 3.0	24.2 $\pm$ 0.5
Ni	27.7 $\pm$ 0.4	23.5 $\pm$ 0.3	2.4 $\pm$ 0.0	18.3 $\pm$ 0.3	1.9 $\pm$ 0.0	13.6 $\pm$ 0.1	1.4 $\pm$ 0.0
Cu	24.3 $\pm$ 0.4	21.2 $\pm$ 0.3	6.4 $\pm$ 0.1	17.6 $\pm$ 0.3	5.2 $\pm$ 0.1	13.8 $\pm$ 0.7	4.0 $\pm$ 0.1
Pb	6.3 $\pm$ 0.1	5.3 $\pm$ 0.1	2.5 $\pm$ 0.0	4.0 $\pm$ 0.1	2.5 $\pm$ 0.0	3.0 $\pm$ 0.2	1.5 $\pm$ 0.0

PM: particulate matter; WS: water-soluble.

respectively.

The concentrations of Fe, Al, Zn, Cr, Mn, Ni, Cu, and Pb in air are observed to be 1.99, 0.38, 0.22, 0.15, 0.13, 0.03, 0.02, and 0.01 mg/m<sup>3</sup>, respectively, whereas the concentrations of Al, Fe, Zn, Cr, Mn, Cu, Ni, and Pb in RPM are 0.21, 0.20, 0.09, 0.08, 0.05, 0.01, 0.01, and 0.003 mg/m<sup>3</sup>, respectively.

Furthermore, the concentrations of WS Fe, Zn, Al, Mn, Cr, Cu, Pb, and Ni in RPM are 0.14, 0.08, 0.04, 0.02, 0.02, 0.004, 0.002, and 0.001 mg/m<sup>3</sup>, respectively (Table 2).

## Discussion

The mass-size distribution of welding fume corresponds to 39.7% of the PM having size  $<10$   $\mu\text{m}$  and 60.3% of the PM with size  $>10$   $\mu\text{m}$ . The proportion of coarse particles, which was obtained using the SMAW and GTAW techniques in the field study, is relatively high as compared to 30% of coarse particles that were obtained using flux-cored arc welding (FCAW), which was demonstrated in a laboratory experiment<sup>5</sup>. Coarse particles that were formed through liquid ejection (microspatter) are minute enough to remain airborne<sup>22, 23</sup>. Liquid ejection is caused due to boiling of the melted materials, surface evaporation, and vapor pressure gradients<sup>24</sup>. Improper arc length, high welding speed and current, and sudden temperature drop further facilitate liquid ejection<sup>25</sup>. Additionally, welding speed influences the formation of welding fume and accelerates agglomeration<sup>26</sup>. However, these factors vary depending on the welding techniques.

In this study, the welding process comprises one-third of GTAW and two-thirds of SMAW. SMAW is operated entirely manually using a stick filler having a metal core.

One of the most common quality issues of SMAW is spatter generation. Cleaning of the welded pipeline may result in resuspension of the microspatter particles. Besides, air sampling is performed outdoors in the breathing zone of a welder. Here, the sampling location is fixed, and the environmental conditions are controlled on the sampling experimental scale. The welder in pipeline construction site is surrounded by a wind shield that is 2–3 m in width and functions in a bent-forward posture in accordance with the welding operations. Thus, exposure to the liquid ejected from welding would be easy, and the dispersion of particles in air is likely to be restricted. Additionally, a portion of the coarse PM may be attributed to the suspended PM in the outdoor dust. Moreover, the Fe/Al ratio in the suspended PM that was sampled in Mailiao area<sup>27</sup> is sometimes observed to be close to the ratio observed in PM having size less than 10  $\mu\text{m}$ .

Particles having sizes less than 520 nm are not subjected to this study because the cascade impactor function is limited. In fact, the initial welding burst can generate nanoparticles having sizes ranging from 5 to 40 nm, which would be capable of forming chain-like agglomerates having a maximum diameter value of 500 nm<sup>28</sup>.

Measuring the particle size-selection in the lung compartments, the IPM and RPM concentrations are estimated using the mass-size distribution of the welding fume. The IPM and RPM concentrations are 3.80 and 1.37 mg/m<sup>3</sup>, respectively. These estimations are based on the data obtained from the 8-stage cascade impactor. Actually, there are specific samplers for sampling of IPM as well as RPM. For instance, Lehnert investigated the exposure of 215 welders to welding fume in 25 German companies using the PGP-EA sampler (IFA, German), which simultaneously



accumulated both inhalable as well as respirable PM. The result depicts that the median IPM and RPM levels (interquartile range, 25–75%) are 2.48 mg/m<sup>3</sup> (1.10–6.81 mg/m<sup>3</sup>) and 1.29 mg/m<sup>3</sup> (<0.45–4.01 mg/m<sup>3</sup>)<sup>29</sup>.

The metal composition in the welding fume varies with the PM size groups. Relatively large percentages of metals are observed in PM having sizes <0.52 µm, >21.3 µm, and in the range of 14.8 to 21.3 µm. The composition of metals in welding fume depends on the vapor pressure and boiling point of the metals, the welding temperature, the welding techniques, and the content of metals in welding materials and the electrode<sup>30</sup>. Fine particles contain abundant metal oxides that are formed through burst evaporation<sup>22</sup>, whereas coarse particles, which are formed through microspatter, contain metals that reflect the chemistry of the welding materials<sup>23</sup>.

Fe, Mn, Cr, and Ni would stem from the welding electrode and base metals, whereas Al and Zn are likely to originate from the antirust painting of the pipeline surface and additives to the welding fillers of the electrode or the materials.

The dissolving capacity of individual metals in water does not correspond significantly to the particle sizes in the welding fume, and the capacity decreases in the following order: Zn, Mn, Pb, Cu, Cr, Al, Fe, and Ni.

Furthermore, the solubility of individual metals varies with the welding technique, the composition of the leaching fluid, and the leaching time<sup>17</sup>. The SMAW electrode incorporates a filler, which enhances the formation of various metal compounds, such as metal fluorides, in the welding fume<sup>31, 32</sup>; KCrF<sub>3</sub>, KMnF<sub>3</sub>, K<sub>2</sub>NaCrF<sub>6</sub>, and KFeF<sub>3</sub>, which are water soluble, are the metal compounds that may be formed as a product of SMAW or FCAW<sup>33</sup>. Moreover, Mn-solubility significantly increases with time<sup>17</sup>. The dissolved amount of metals such as Mn increases with the extraction time during the initial 24 h. The solubility for a metal in particles commonly refers to its capacity to dissolve in aqueous media, but the lung simulation solutions such as Gamble's and Hatch's solution have been employed to mimic the interface of the alveoli. Hatch's solution containing ionized buffer and some proteins and enzymes was the reasonable choice in the study because the obtained metal content was higher in the extraction test<sup>17</sup>. In contrast, using deionized water as an extraction solution would sometimes underestimate the capacity of metals dissolving in body fluid. The water solubility of individual metals in the welding fume seems comparable only when the welding technique, welding material, and extraction method are well defined.

The solubility of metals dissolving in a body fluid is important to estimate the uptake and subsequent translocation of these metals. Further, metals absorbed in the lungs may cause extrapulmonary system effects, and the absorption of metals depends on their water/acid solubility<sup>34</sup>. Considering the particle size-selection in different lung compartments and composition of metals depending on their particle sizes and the metals dissolving in water, the percentages of WS metals from the welding fume that are available in the gas-exchange region of the lungs range from 4.9% to 34.6% depending on the metal species. Examples of such an estimation would be 18% (24.2 µg/m<sup>3</sup>) for Mn, 11.2% (16.4 µg/m<sup>3</sup>) for Cr, and 4.9% (1.4 µg/m<sup>3</sup>) for Ni.

WI particles deposited in the lungs are cleared by alveolar macrophages through phagocytosis. Some metals, such as Al, can show accumulation in the lungs with age. The deposited WI particles may be sequestered in the lungs at the alveolar space, where they can trigger inflammation<sup>35</sup> and transform into soluble ionic forms due to slower biochemical reaction and gradually clear from the lungs<sup>36</sup>. For instance, an insoluble nickel compound such as nickel subsulfide is more likely to be carcinogenic at the site of deposition<sup>37</sup>.

## Conclusion

In this study, we examined the exposure to welding fume when using SMAW and GTAW in outdoor pipeline construction. The log-normal mass-size distribution having two modes exhibited 39.7% of PM having sizes less than 10 µm, and 60.3% of PM having sizes greater than 10 µm. The mass of PM having size >10 µm was unusually high, which does not agree with the results observed in laboratory experiments. The resuspension of the particles due to cleaning processes of the welded pipeline are likely to have contributed to coarse PM along with the particles that were generated through liquid ejection.

The WS metals retained in welding fumes are critical in terms of hazards stemming from various welding techniques. In this study, PM having sizes <0.52 µm and >14.8 µm were rich in metals, whereas PM having sizes ranging from 0.52 to 14.8 µm contained a relatively low amount of metals. The WS metals varied with the metal species in a decreasing order as follows: Zn, Mn, Pb, Cu, Cr, Al, Fe, and Ni. If we consider the particle size selection in lung compartments, composition of metals based on PM sizes, and metals dissolving in water, WS metals available in the gas-exchange region of the lungs amounted to <35%

of the total airborne metals. Therefore, it becomes important to investigate the relation between the exposure levels of metals, instead of airborne concentrations, and concentration of metals in blood/urine, to accurately evaluate the health risk from exposure to welding fume.

## Acknowledgements

The authors acknowledge and express gratitude for the technical aid and advice provided by Tung-Sheng Shih, Rui-Shu Zhou, Chun-Wan Chen, and Shu-Jui Chang during the preparation of this manuscript. We thank the Institute of Labor, Occupational Safety and Health (ILOSH) in Taiwan for their instrumental support to this study.

## Conflict of Interest

No potential conflict of interest was reported by the authors.

## References

- Antonini JM, Lewis AB, Roberts JR, Whaley DA (2003) Pulmonary effects of welding fumes: review of worker and experimental animal studies. *Am J Ind Med* **43**, 350–60.
- Schoonover T, Conroy L, Lacey S, Plavka J (2011) Personal exposure to metal fume, NO<sub>2</sub>, and O<sub>3</sub> among production welders and non-welders. *Ind Health* **49**, 63–72.
- Zimmer AT (2002) The influence of metallurgy on the formation of welding aerosols. *J Environ Monit* **4**, 628–32.
- Sokolov SV, Tschulik K, Batchelor-McAuley C, Jurkschat K, Compton RG (2015) Reversible or not? Distinguishing agglomeration and aggregation at the nanoscale. *Anal Chem* **87**, 10033–9.
- Jenkins NT, Eagar TW (2005) Chemical analysis of welding fume particles. *Weld J* **84**, 87s–93s.
- European Committee for Standardization (CEN) (1993) Workplace atmospheres: size fraction definitions for measurement of airborne particles in the workplace (Report No. BS EN 481:1993). 1–7. CEN, British Standards Institute, London.
- International Standards Organization (ISO) Air quality-particle size fraction definitions for health-related sampling (ISO 7708) April 1995. <https://www.iso.org/standard/14534.html>. Accessed April 2008.
- Vincent JH (1999) Particle size-selective sampling for particulate air contaminants, Revised ed., 25, American Conference of Governmental Industrial Hygienists (ACGIH), Cincinnati.
- Lee BM, Kacew S (2002) Lu's basic toxicology: fundamentals, target organs and risk assessment, 6th ed. 430, CRC Press, New York.
- Aschner M, Erikson KM, Dorman DC (2005) Manganese dosimetry: species differences and implications for neurotoxicity. *Crit Rev Toxicol* **35**, 1–32.
- Roels H, Meiers G, Delos M, Ortega I, Lauwerys R, Buchet JP, Lison D (1997) Influence of the route of administration and the chemical form (MnCl<sub>2</sub>, MnO<sub>2</sub>) on the absorption and cerebral distribution of manganese in rats. *Arch Toxicol* **71**, 223–30.
- Ellingsen DG, Hetland SM, Thomassen Y (2003) Manganese air exposure assessment and biological monitoring in the manganese alloy production industry. *J Environ Monit* **5**, 84–90.
- Laohaudomchok W, Lin X, Herrick RF, Fang SC, Cavallari JM, Christiani DC, Weisskopf MG (2011) Toenail, blood, and urine as biomarkers of manganese exposure. *J Occup Environ Med* **53**, 506–10.
- Kodavanti UP, Hauser R, Christiani DC, Meng ZH, McGee J, Ledbetter A, Richards J, Costa DL (1998) Pulmonary responses to oil fly ash particles in the rat differ by virtue of their specific soluble metals. *Toxicol Sci* **43**, 204–12.
- Delfino RJ, Staimer N, Vaziri ND (2011) Air pollution and circulating biomarkers of oxidative stress. *Air Qual Atmos Health* **4**, 37–52.
- American Society of Mechanical Engineers (ASME) Multi process welding procedures QW200.4 and ASME QW451.1 December 2008. <http://www.gowelding.com/wp/asme-multi.htm>. Accessed February 2018.
- Berlinger B, Ellingsen DG, N  ray M, Z  ray G, Thomassen Y (2008) A study of the bio-accessibility of welding fumes. *J Environ Monit* **10**, 1448–53.
- Inc SKC Inhalable particulate mass: history, current technology, and data considerations December 2017. <https://www.skcinco.com/catalog/infopage.php?id=9020>. Accessed February 2018.
- Ghio AJ, Stonehuerner J, Dailey LA, Carter JD (1999) Metals associated with both the water-soluble and insoluble fractions of an ambient air pollution particle catalyze an oxidative stress. *Inhal Toxicol* **11**, 37–49.
- Cena LG, Chisholm WP, Keane MJ, Cumpston A, Chen BT (2014) Size distribution and estimated respiratory deposition of total chromium, hexavalent chromium, manganese, and nickel in gas metal arc welding fume aerosols. *Aerosol Sci Technol* **48**, 1254–63.
- Fernandez-Diaz JM, Rodriguez-Bra  na MA, Arganza BA (2006) Complete computer program to fit the data from an impactor to a sum of lognormal. Paper presented at the 7th International Aerosol Conference, Saint Paul, September 10–15.
- Jenkins NT, Mendez PF, Eagar TW (2006) Effect of arc welding electrode temperature on vapor and fume composition. Paper presented at the proceedings of the 7th international conference on trends in welding research, Pine Mountain, May 16–20.
- Kulkarni P, Baron PA, Klaus Willeke K (2011) Aerosol measurement: principles, techniques, and applications, 3rd

- ed., 31, John Wiley & Sons, New Jersey.
- 24) He X, Fuerschbach PW, DeRoy T (2003) Heat transfer and fluid flow during laser spot welding of 304 stainless steel. *J Phys D Appl Phys* **36**, 1388–98.
- 25) Farnsworth SR (2010) *Welding for dummies*, 291, Wiley, New Jersey.
- 26) Zimmer AT, Biswas P (2001) Characterization of the aerosols resulting from arc welding processes. *J Aerosol Sci* **32**, 993–1008.
- 27) Jhuang FC (2009) Chemical mass balance source apportionment of TSP in nearby regions of No.6 naphtha cracking project. Master thesis of Graduate School of Department of Safety Health and Environmental Engineering, National Yunlin University of Science and Technology, Taiwan.
- 28) Berlinger B, Benker N, Weinbruch S, L'Vov B, Ebert M, Koch W, Ellingsen DG, Thomassen Y (2011) Physicochemical characterisation of different welding aerosols. *Anal Bioanal Chem* **399**, 1773–80.
- 29) Lehnert M, Pesch B, Lotz A, Pelzer J, Kendzia B, Gawrych K, Heinze E, Van Gelder R, Punkenburg E, Weiss T, Mattenklott M, Hahn JU, Möhlmann C, Berges M, Hartwig A Brüning T, Weldox Study Group (2012) Exposure to inhalable, respirable, and ultrafine particles in welding fume. *Ann Occup Hyg* **56**, 557–67.
- 30) Burgess WA (1991) Potential exposures in the manufacturing industry—their recognition and control, 4th ed. 595, Wiley Interscience, New York.
- 31) Yoon CS, Paik NW, Kim JH, Chae HB (2009) Total and soluble metal contents in flux-cored arc welding fumes. *Aerosol Sci Technol* **43**, 511–21.
- 32) Jenkins NT (2003) Chemistry of airborne particles from metallurgical processing, PhD dissertation, Cambridge, Massachusetts, Massachusetts Institute of Technology.
- 33) Voitkevich VG (1995) Welding fumes—formation, properties and biological effects, 18–104, Abington Publishing, Cambridge, England.
- 34) Wallenborn JG, McGee JK, Schladweiler MC, Ledbetter AD, Kodavanti UP (2007) Systemic translocation of particulate matter-associated metals following a single intratracheal instillation in rats. *Toxicol Sci* **98**, 231–9.
- 35) Blum JL, Rosenblum LK, Grunig G, Beasley MB, Xiong JQ, Zelikoff JT (2014) Short-term inhalation of cadmium oxide nanoparticles alters pulmonary dynamics associated with lung injury, inflammation, and repair in a mouse model. *Inhal Toxicol* **26**, 48–58.
- 36) Goyer RA, Klaassen CD, Waalkes MP (2016) *Metal toxicology: approaches and methods*, 1st ed. 237, Elsevier, California.
- 37) Agency for Toxic Substances and Disease Registry (ATSDR) Toxicological profile for nickel August 2005. <https://www.atsdr.cdc.gov/toxprofiles/tp15.pdf>. Accessed February 2018.