



# Article Characterization of Ambient Particulate Matters in an Industry-Intensive Area in Central Taiwan

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Abstract: Atmospheric particulate matters (PMs) were measured in an industry-intensive region in central Taiwan in order to investigate the characteristics and possible sources of PMs. The samplings were simultaneously conducted using a 10- and 3-stage Micro Orifice Uniform Deposit Impactor (MOUDI) from 2017 to 2018. In this study, the characteristics of PMs in this region were evaluated by measuring the mass concentration of PMs and analyzing water-soluble ions and metallic elements, as well as dioxins. Additionally, principal component analysis (PCA) was used to identify the potential sources of PMs. The results showed that the mean concentration of coarse (>1.8  $\mu$ m), fine (0.1–1.8  $\mu$ m), and ultrafine (<0.1  $\mu$ m) particles were 13.60, 14.38, and 3.44  $\mu$ g/m<sup>3</sup>, respectively. In the industry-intensive region, the size distribution of ambient particles showed a bi-modal distribution with a high concentration of coarse particles in the spring and summer, while fine particles were dominant in the autumn and winter. The most abundant water-soluble ions of PMs were NO<sub>3</sub><sup>-</sup>, Cl<sup>-</sup>, and SO<sub>4</sub><sup>2-</sup>, while the majority of metallic elements were Na, Fe, Ca, Al, and Mg in different particle sizes. The results of Pearson's correlation analysis for metals indicated that the particles in the collected air samples were related to the iron and steelmaking industries, coal burning, vehicle exhausts, and high-tech industries. The dioxin concentration ranged from 0.0006 to 0.0017 pg I-TEQ/Nm<sup>3</sup>. Principal component analysis (PCA) revealed that the contribution to PMs was associated with sea salt, secondary pollutants, and industrial process.

Keywords: PMs; size distribution; chemical composition; PCA

# 1. Introduction

Air pollution is a major concern in urban areas because of its threat to human health, including asthma and acute and chronic respiratory symptoms [1]. Ambient particulate matters (PMs) include small solid or liquid particles suspended in the air. In addition, PMs included particles with an aerodynamic diameter of less than 0.1  $\mu$ m, less than 2.5  $\mu$ m, and less than 10  $\mu$ m, also known as PM<sub>1</sub>, PM<sub>2.5</sub>, and PM<sub>10</sub>, respectively [2]. Epidemiological studies reported that the highest association noted was an 18% rise in asthma admissions correlated with a 10  $\mu$ g/m<sup>3</sup> increase in coarse particles on the same day of admissions [3]. Furthermore, ultrafine particles cause a stronger toxic effect than



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**Copyright:** © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). fine and coarse particles due to their high surface area [4,5]. The chemical compositions of PMs include water-soluble ions, metallic elements, and organic compounds, such as dioxin, which are correlated to toxicity and health [6,7]. Typically, the ionic constituents accounted for 35–60% of the PM<sub>2.5</sub> mass in Korea, and sulfates and nitrates were the dominant ionic species [8]. In southern Taiwan, Tsai et al. [9] reported that sulfate, nitrate, and ammonium were the major ionic species and contributed a large fraction of PM mass in different sizes. In Boston, over 80% of the elements mostly found in the coarse mode included Ca, Mn (road dust), and Cl (sea salt) [10]. In Mexico City, high concentrations of Al, Cr, Ni, Pb, and V were measured in late fall [11]. PCDD/Fs have been a source of much public concern over the last decade due to adverse health effects [6]. The dioxin concentration in the atmosphere could be affected by different seasons because of domestic heating, photolysis, and chemical reactions [12]. In rural Germany, PCDD/F concentrations were higher in the winter than in the summer due to domestic heating [13].

The main objectives of this study were to investigate the characteristics of ambient PMs in an industrial area. The different particle sizes of PMs, including coarse, fine, and ultrafine modes, were measured, as well as water-soluble ions, metallic elements, and dioxins in PMs. The principal component analysis (PCA) is a useful statistical method to assess possible sources of pollution and their contribution to ambient PMs. The results of this study are of great importance to understand the characteristic of PMs in an industrial area and will provide useful information for the future control strategy of ambient PMs.

## 2. Experiment and Methods

### 2.1. Sampling of PM

The atmospheric particle sampling site was designed according to the understanding of local emission sources. The atmospheric particle samplings were taken at the campus of Tunghai University in central Taiwan as a receptor for assessment. The sampling site was surrounded by Taichung Industrial Park (A: 500 m away), Central Taiwan Science Park (B: 4 km away), and Taichung Power Station (C: 10 km away) (Figure 1). The samples were collected continuously for 24 h on four consecutive days during all four seasons in 2017 and 2018. This study employed two 10-stage and 3-stage cascade impactors from Micro-Orifice Uniform Deposit Impactors (MOUDI) (Model 110-R, 100S4; MSP Corporation, in Shoreview, MN, USA). The corresponding cut-off sizes were 0.056, 0.10, 0.18, 0.32, 0.56, 1.0, 1.8, 3.2, 5.6, 10.0, 18  $\mu$ m, and 1.0, 2.5, and 18  $\mu$ m, respectively. A 47 mm Teflon filter was used to collect the samples with a flow rate of 30 L/min. The filters were conditioned at 25 ± 3 °C and 45 ± 5% relative humidity for 24 h before weighing. In addition, PCDD/Fs were collected by a PS-1 sampler that was equipped with quartz fiber filters.



**Figure 1.** Schematic diagram of sampling site (Tunghai University) and the surrounding pollution sources (A: Taichung Industrial Park; B: Central Taiwan Science Park; C: Taichung Power Plant). The figure is reproduced from Google map at 24°10′54.1″ N 120°36′24.7″ E.

#### 2.2. Analysis of Water-Soluble Ions and Metallic Elements

Water-soluble cations and anions were analyzed by ion chromatography according to the standard method of the Taiwan Environment Protection Agency (EPA): NIEA W415.54B. One-half of each Teflon filter was analyzed by ion chromatography (Metrohm, 883 Basic IC plus). Each filter was put into a polyethylene bottle with 10 mL deionized distilled water and was extracted using an ultrasonic bath for 60 min. The extract was filtered with an acetate filter with 0.22 µm pore size diameter. Five cations (Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>) and five anions (F<sup>-</sup>, Cl<sup>-</sup>, NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>) were analyzed by ion chromatography (Metrohm, 883 Basic IC plus). The anion species were detected by Metrosep C 4-150/4.0 (4 × 150 mm) analytical column with an effluent of 1.7 mM HNO<sub>3</sub>/0.7 mM C<sub>7</sub>H<sub>5</sub>NO<sub>4</sub> at a flow rate of 0.9 mL/min. The cation species were separated by Metrosep A Supp5-150/4.0 (4 × 150 mm) analytical column with an effluent of 1.0 mM NaHNO<sub>3</sub>/3.2 mM NaCO<sub>3</sub> at a flow rate of 0.7 mL/min. The detection limits were 0.20, 0.18, 0.11, 0.42, 0.36, 0.59, 0.46, 0.48, 0.15, and 0.5 µg/L for Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>, Ca<sup>2+</sup>, Ca<sup>2+</sup>, Ca<sup>2+</sup>, NO<sub>2</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup>, and F<sup>-</sup>, respectively.

The metals were analyzed by inductively coupled plasma mass spectrometry according to the standard method of NIEA A306.10C of the Taiwan Environment Protection Agency (EPA). The other half of the Teflon filter was analyzed to clarify the metal concentrations. The Teflon filters were placed in a microwave digestion furnace and were heated at 175 °C for 10 min, and then 24 metals (Zn, Cd, Co, Cr, Cu, Fe, Ga, In, Mn, Ni, Pb, Sr, Ag, B, Ba, Bi, Ca, K, Li, Mg, Na, Tl, As, and Al) were analyzed by inductively coupled plasma mass spectrometry (Thermo Scientific, iCAP RQ). The method detection limits for Zn, Cd, Co, Cr, Cu, Fe, Ga, In, Mn, Ni, Pb, Sr, Ag, B, Ba, Bi, Ca, K, Li, Mg, Na, Ni, Pb, Sr, Ag, B, Ba, Bi, Ca, K, Li, Mg, Na, Ti, As, and Al were 0.55, 0.89, 0.66, 0.88, 0.63, 0.92, 0.78, 0.98, 0.55, 0.66, 1.75, 0.56, 0.65, 1.63, 0.37, 0.78, 0.62, 0.57, 0.36, 1.45, 0.45, 1.34, 1.23, and 1.55 ug/L, respectively. For clarity, the samples are divided into coarse (>1.8  $\mu$ m), fine (0.1–1.8  $\mu$ m), and ultrafine (<0.1  $\mu$ m) particles.

#### 2.3. Analysis of PCDD/Fs

Analysis of ambient air samples for PCDD/Fs was performed according to the US EPA Reference Method TO9A [14]. Each sample was spiked with a known amount of the internal standard. All samples were extracted with toluene for 24 h, and this was then followed by a series of sample cleanup and fractionation procedures. The extract was transferred to a vial and further concentrated by a N2 gas stream. High-resolution gas chromatographs/high-resolution mass spectrometers (HRGC/HRMS) were used for PCDD/F analysis. The HRGC (Hewlett-Packard 6970 Series gas, Atalanta, CA, USA) was equipped with a DB-5 fused silica capillary column (L = 60 m, ID = 0.25 mm, film thickness = 0.25  $\mu$ m) (J&W Scientific, Atalanta, CA, USA) with a splitless injection, while the HRMS (Micromass Autospec Ultima, Manchester, UK) had a positive electron impact (EI<sup>+</sup>) source. The oven temperature program was set according to the following: start at 150  $^{\circ}$ C (held for 1 min), then increased by 30 °C/min to 220 °C (held for 12 min), and finally increased by 1.5 °C/min to 310 °C (held for 20 min). Helium was used as the carrier gas. Laboratory blank samples were also sampled and analyzed for quality assurance purposes. The PCDD/F mass of method blank samples for the tetra through hexa-chlorinated homologues ranged from 0.79 to 1.58 pg, while that for the hepta- and octa-chlorinated homologues ranged from 0.19 to 0.70 pg.

#### 2.4. Principal Component Analysis

Principal component analysis (PCA) is widely used as an assessment method to identify possible pollution sources. PCA is applied to characterize large sets of data by re-expressing to a rotated coordinate system in which the eigenvectors of the variance–covariance matrix are calculated to explain as much variance as possible. The principal component score, i.e., the weight of the eigenvector, can be obtained. These scores of the original variables, called the principal component loadings (PC loadings), can illustrate the relationship between the variable and the principal component. The first PC (PC1) will explain most of the variance of the original data variables, whereas the second PC (PC2) will explain fewer of the original data variables. By doing this, the raw data matrix can

be reduced to two or three principal component loadings that account for the majority of the variance. The PCA provides qualitative information about the nature of the source profile compositions and the relative importance of a given source to the observed concentration [15]. The main objective of PCA is to reduce a large number of variables to a smaller set of factors that retain most of the information in the original data set [16,17]. Each factor explains the maximum total variance of the data set, and this set is completely uncorrelated with the rest of the data. Thus, the elements with higher factor loading (>0.7) are interpreted as fingerprints of emission sources [18].

## 3. Results and Discussion

#### 3.1. Particulate Matter Mass Concentrations and Particle Size distribution

The mass concentration of coarse (>1.8  $\mu$ m), fine (0.1–1.8  $\mu$ m), and ultrafine (<0.1  $\mu$ m) particles are listed in Table 1. The order of the mass of particle sizes in spring and summer was coarse particles > fine particles > ultrafine particles, while those in autumn and winter were fine particles > coarse particles > ultrafine particles. The coarse particles were dominant in spring and summer, with an average mass concentration of 16.96 and 12.67  $\mu$ g/m<sup>3</sup>, accounting for 47.5 and 45.6%, respectively. The concentration of the fine particles was 15.39 and 18.17  $\mu$ g/m<sup>3</sup> in autumn and winter, accounting for 46.7 and 62.2%, respectively. In summer, the mass levels of coarse, fine, and ultrafine particles were 12.67, 9.14, and 5.96  $\mu$ g/m<sup>3</sup>, respectively, and were slightly higher than those in the Shanghai urban summer atmosphere (9.38, 8.82, and 2.02  $\mu$ g/m<sup>3</sup>) and the traffic site in Los Angeles (6.3, 5.8, and 1.7  $\mu$ g/m<sup>3</sup>) [19,20]. Therefore, the particle concentration in this industrial area is relatively polluted compared to the references [19,20], and the mass level was different in different seasons. In addition, the  $PM_{10}$ concentration was measured as being from 9.26 to 42.59  $\mu$ g/m<sup>3</sup>, which is close to values of the previous study in central Taiwan [21]. Our data showed slightly lower values than the  $PM_{10}$ concentration of 13.83–53.67  $\mu$ g/m<sup>3</sup> at Xitun station (one of the EPA air quality monitoring stations) due to the difference between the automatic and manual sampling methods. In addition, the  $PM_{10}$  data were much lower than the Taiwan EAP  $PM_{10}$  regulations (125  $\mu$ g/m<sup>3</sup>). The ambient PMs displayed a bimodal distribution, as shown in Figure 2. In spring and summer, the most prominent peak occurred in the range of Dp =  $3.2-5.6 \mu m$ , and the minor peak was in the range of  $Dp = 0.18-0.32 \mu m$ . In autumn and winter, the most prominent peak occurred in the range of Dp =  $0.32-0.56 \mu m$ , and the minor peak was in the range of Dp =  $3.2-5.6 \mu m$ . These results are similar to a previous study in that coarse particles were the predominant components in spring [22]. The cumulative mass fractions for fine particles were 41.8 and 33.4% in spring and summer, respectively, while they accounted for 51.8 and 62.5% in autumn and winter, respectively (Figure 3). This indicated that fine particle pollution is dominant in Taichung in autumn and winter. In addition, both coarse and fine particles were associated with local traffic [23].

Sampling	Coarse Particles (µg/m <sup>3</sup> )		Fine Particles (µg/m <sup>3</sup> )		Ultrafine Particles (µg/m³)	
lime	Mean	SD	Mean	SD	Mean	SD
Spring	16.96	3.9	14.82	5.7	3.94	0.9
Summer	12.67	3.3	9.14	1.7	5.96	3.7
Autumn	14.99	1.8	15.39	3.0	2.61	0.5
Winter	9.78	5.8	18.17	10.0	1.27	0.1

Table 1. The mass concentration of different particle sizes (coarse, fine, ultrafine particles).



Figure 2. Particle size distributions: (a) Spring; (b) Summer; (c) Autumn; (d) Winter.



Figure 3. Cumulative mass fractions of particulate matter: (a) Spring; (b) Summer; (c) Autumn; (d) Winter.

## 3.2. Water-Soluble Ions

Table 2 shows the water-soluble ion content in different particle sizes. The sequence of the major ionic species in the coarse particles was Cl<sup>-</sup> (1.63  $\pm$  0.41 µg/m<sup>3</sup>) > NO<sub>3</sub><sup>-</sup> (1.62  $\pm$  0.61 µg/m<sup>3</sup>) > SO<sub>4</sub><sup>2-</sup> (1.18  $\pm$  0.29 µg/m<sup>3</sup>) > Na<sup>+</sup> (0.90  $\pm$  0.52 µg/m<sup>3</sup>), while the sequence was SO<sub>4</sub><sup>2-</sup> (2.32  $\pm$  1.03 µg/m<sup>3</sup>) > Cl<sup>-</sup> (1.37  $\pm$  0.29 µg/m<sup>3</sup>) > NO<sub>3</sub><sup>-</sup> (1.30  $\pm$  1.02 µg/m<sup>3</sup>) > NH<sub>4</sub><sup>+</sup> (1.24  $\pm$  1.01 µg/m<sup>3</sup>) for the fine particles. Huang et al. (2013) reported that the dominant species were SO<sub>4</sub><sup>2-</sup>, Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup> at an industrial complex site in China. SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup>, and NO<sub>3</sub><sup>-</sup> are considered to be the major secondary aerosols

by anthropogenic sources [24]. Regarding ultrafine particles, the majority of components were  $SO_4^{2-}$  (0.44  $\pm$  0.20 µg/m<sup>3</sup>), Cl<sup>-</sup> (0.41  $\pm$  0.25 µg/m<sup>3</sup>), NO<sub>3</sub><sup>-</sup> (0.26  $\pm$  0.09 µg/m<sup>3</sup>), and Na<sup>+</sup> (0.23  $\pm$  0.28  $\mu$ g/m<sup>3</sup>), accounting for 62.05% of the total species. The Cl<sup>-</sup> in both the coarse and fine particles is generally derived from sea spray [25]. Motor vehicle emissions and fossil fuel combustion are the dominant contributors to  $NO_3^-$  and  $SO_4^{2-}$ , respectively [26,27]. In addition, the high concentration of Cl<sup>-</sup> and K<sup>+</sup> during the heating season may be due to increased coal combustion [28] and straw burning [29]. Notably, the K<sup>+</sup> concentration  $(0.16-0.35 \ \mu g/m^3)$  was not relatively high in this study because the sampling site is 10 km away from Taichung Power Station (see Figure 1). Compared with other cities, water-soluble constituents contributed an average of 11.57% in PM25 and 16.98% in PM1 in Drug city in India. Notably, similar to our study, the concentrations of  $SO_4^{2-}$  and  $NO_3^{-}$  were high in all size fractions and accounted for 32.76% and 13.38% of the total mass of the water-soluble ions in PM\_{2.5} [30]. In Palermo, Italy, a large fraction of PM\_{10} (31–47\% in weight) and PM\_{2.5} (29\% in weight) are made up of water-soluble ions.  $SO_4^{2-}$  and  $NH_4^+$  concentrations in the PM<sub>2.5</sub> fraction were higher than in  $PM_{10}$ .  $SO_4^{2-}$ ,  $NH_4^+$ , and  $NO_3^-$  ions also constituted 51–63% of the total measured water-soluble ions [31]. In the electronic processing industrial region of Kunshan, China, the most abundant ions are  $NO_3^-$  (30.96 µg/m<sup>3</sup>),  $SO_4^{2-}$  (24.67 µg/m<sup>3</sup>), and  $NH_4^+$  (19.98  $\mu g/m^3$ ), and are significantly higher than those found in this study [32].

 Table 2. The content of water-soluble ions in different particle sizes.

Water-Soluble	Coarse Particles (µg/m <sup>3</sup> )		Fine Particles (µg/m <sup>3</sup> )		Ultrafine Particles (µg/m <sup>3</sup> )	
TOTIO	Mean	SD	Mean	SD	Mean	SD
Na <sup>+</sup>	0.90	$\pm 0.52$	0.43	±0.39	0.23	$\pm 0.28$
$NH_4^+$	0.30	$\pm 0.10$	1.24	$\pm 1.01$	0.16	$\pm 0.05$
$K^+$	0.35	$\pm 0.16$	0.32	$\pm 0.11$	0.16	$\pm 0.21$
Ca <sup>2+</sup>	0.47	$\pm 0.18$	0.25	$\pm 0.17$	0.09	$\pm 0.08$
Mg <sup>2+</sup>	0.20	$\pm 0.17$	0.11	$\pm 0.10$	0.04	$\pm 0.05$
$\bar{F}$	0.35	$\pm 0.28$	0.35	$\pm 0.25$	0.13	$\pm 0.13$
Cl-	1.63	$\pm 0.41$	1.37	$\pm 0.29$	0.41	$\pm 0.25$
$NO_3^-$	1.62	$\pm 0.61$	1.30	$\pm 1.02$	0.26	$\pm 0.09$
$SO_4^{2-}$	1.18	$\pm 0.29$	2.32	$\pm 1.03$	0.44	$\pm 0.20$

## 3.3. Metallic Elements

The fraction of metals in coarse particles in different seasons is shown in Figure 4a. The data of the winter samples are not discussed due to contamination. In spring, the dominant species were found to be Na (18.1%), Fe (17.9%), and K (17.5%), contributing about 53.5% of the total metallic elements, while Na (25.1%), Ca (20.9%), and Al (13.9%) accounted for 59.9% of the total components in summer. In autumn, Fe contributed 22.6%, Al 22.2%, and Mg 21.0%, which accumulated to 65.79%. Notably, Na, Fe, Ca, and Al were mainly distributed in coarse particles that may have come from the earth crust elements and road/tires [33]. Figure 4b shows the fractions of metals in fine particles during different seasons. In spring, the most abundant elements were Na (31.6%), Ag (12.3%), and Fe (11.2%), while in summer, the proportions of Na, Ca, and Al were 29.1, 22.5, and 12.4%, respectively. In autumn, the dominant species were found to be Al (28.3%), K (16.7%), and Mg (15.4%), accounting for 60.4% of total elements. In addition, Ag was measured in fine particles that came from anthropogenic sources, such as the iron and steel industries, the cement industry, the combustion of coal and oil, and incineration [34–37]. According to the national emissions inventory of sources and emissions of silver [37], the principal sources (after emission control, if any) are the iron and steel industries (46%), the cement industry (25.7%), the combustion of coal and oil (13.6%), and incinerators (5.6%). Fe was also the most abundant element that was sourced from the Earth's crust [38]. Hence, the major elements in the fine particles were Na, Fe, Ca, and Al. In China, the crustal elements Al, Ca, K, Na, Mg, and Fe were most abundant at an industrial site, which may be due to more

vehicle emissions and dust resuspension [24]. Figure 4c shows the fraction of metals in ultrafine particles in different seasons. In spring, the dominant species were found to be Na (23.4%), K (18.8%), and Ca (18.1%). On the other hand, the fractions of Na, Ca, and Al were 30.8%, 23.4%, and 11.8%, respectively, in summer. In autumn, the dominant species were Al (24.5%), Fe (19.7%), and K (14.9%), accounting for 59.1% of the total metallic elements. As a result, the most abundant species in the ultrafine particles were Na, K, Ca, and Al.



Figure 4. The fraction of metallic elements in different seasons (a): Coarse particles; (b): Fine particles; (c): Ultrafine particles.

Pearson's correlation coefficients of heavy metals in coarse, fine, and ultrafine particles are summarized in Figure 5. By analyzing the value of the correlation coefficient between the metal elements, it is possible to understand the possible sources of air pollution around the sampling point. The significant correlations of Fe-Mn (r = 0.664-0.965), Fe-Zn (r = 0.638-0.826), and Mn-Zn (r = 0.127-0.779) were observed in coarse, fine, and ultrafine particles, with the exception of Zn-Mn (0.127) in ultrafine particles, suggesting possible iron and steelmaking industry activities near the sampling site. This result agrees with a previous study conducted by Mohiuddin et al. (2014), which analyzed the correlation of Fe-Mn-Zn from iron and steel industry sites [39]. In Agra, India, Fe shows a good correlation with Mn (0.605), Pb (0.506), Cr (0.957), and Al (0.580), indicating emissions sources such as the iron or steel industries and crustal materials contribute to ambient air pollution [40]. As shown in Figure 5, Pb displayed significant correlation (p < 0.05) with Zn (r = 0.607–0.684), Cr (r = 0.772–0.914), Fe (r = 0.555–0.720), and Al (r = 0.684-0.737) in both coarse and fine particles, illustrating the contribution of crustal emissions and coal burning. The results of the statistical analyses by PCA showed that the factors loaded with Co, Cd, Pb, Ni, Cu, and Zn are anthropogenic emissions from the combustion of fossil fuels and industry in the ambient air in the central Himalayan region, Nepal [41]. High correlations were observed between Al-Cu (r = 0.718-0.891), Al-Sr (r = 0.782-0.864), Al-K (r = 0.629–0.856), K-Sr (r = 0.380–0.662), and Sr-Cu (r = 0.531–0.726) in coarse, fine and ultrafine particles (see Figure 5), indicating the contribution from the vehicle exhausts. These results are similar to the findings from the Pittsburgh metropolitan area; that is, Al, K, Mo, Sb, Sr, and Cu are related to the characteristics of vehicle emissions. Notably, Mo, Sb, Sr, and Cu are related to brake and tire wear [42]. The sampling site (THU) is near the Central Taiwan Science Park (CTSP), which is a high-tech, industry-intensive area. Therefore, positive correlations were observed between Zn-Cu (r = 0.638–0.782), Cd-Cr (r = 0.679–0.943), Cd-Pb (r = 0.638–0.675), Cu-Ga r = (0.444–0.689), Pb-K (r = 0.493–0.652), and In-Bi (r = 0.409–0.600). Some correlations between two metals (Cd-V, Cu-Ga, Zn-Cu, Mo-As, Pb-As, Pb-Cd, K-V, K-Pb, and Cr-Cd) had a



greater confidence level and correlation coefficient in the CTSP [43] because the CTSP is a major contributor to Ga and As in ultrafine particles [44].

**Figure 5.** Correlations among metal elements for (**a**) coarse, (**b**) fine, and (**c**) ultrafine particles in sampling periods. \*:  $p \le 0.05$ .

#### 3.4. PCDD/F Concentration in Ambient Air

The measured species and concentrations of PCDD/F are listed in Table 3. In 2017, the total concentrations of PCDD/F were 0.2541 and 0.1387 pg/Nm<sup>3</sup> in the spring and summer, respectively. The corresponding I-TEQ concentrations were 0.0017 pg I-TEQ/Nm<sup>3</sup> in spring and 0.0006 pg I-TEQ/Nm<sup>3</sup> in summer 2017. This result is consistent with the findings of atmospheric PCDD/Fs ranging from 0.97 to 255 fg I-TEQ/m<sup>3</sup> in 11-year observations in Taiwan [45]. Those values were slightly lower than those in the previous study in southern Taiwan (0.0319–0.0847 pg I-TEQ/Nm<sup>3</sup>) [46,47]. A study in Southeast Asia showed that the ambient levels were 15 times greater in urban areas than in rural areas, varying from 23 to 565 fg TEQ m<sup>-3</sup> [48]. In addition, the present results were much lower than the regulation values of ambient air PCDD/F concentrations in Japan (0.6 pg I-TEQ/Nm<sup>3</sup>). As shown in Table 3, the main species of measured PCDD/F were 1,2,3,7,8,9-Hexachlorodibenzo-pdioxin (1,2,3,7,8,9-HxCDD), 1,2,3,4,6,7,8-Heptachlorodibenzo-p-dioxin (1,2,3,4,6,7,8-HpCDD), 1,2,3,7,8,9-Hexachlorodibenzofuran (1,2,3,7,8,9-HxCDF), 2,3,4,6,7,8-Hexachlorodibenzofuran (2,3,4,6,7,8-HxCDF), 1,2,3,4,6,7,8- Heptachlorodibenzofuran (1,2,3,4,6,7,8-HpCDF), and Octachlorodibenzofuran (OCDF), which were similar to the dominant congeners of OCDD, OCDF, 1,2,3,4,6,7,8-HpCDF, and 1,2,3,4,6,7,8-HpCDD in Taiwan's atmosphere [49].

**Table 3.** PCDD/F concentration  $(pg/m^3)$  in ambient air.

PCDD/F	Spring, 2017	Summer, 2017
2,3,7,8-TeCDD	ND	ND
1,2,3,7,8-PeCDD	ND	ND
1,2,3,4,7,8-HxCDD	ND	ND
1,2,3,6,7,8-HxCDD	ND	ND
1,2,3,7,8,9-HxCDD	ND	ND
1,2,3,4,6,7,8-HpCDD	0.0377	0.0177
OCDD	0.1042	0.0488
2,3,7,8-TeCDF	ND	ND
1,2,3,7,8-PeCDF	ND	ND
2,3,4,7,8-PeCDF	ND	ND
1,2,3,4,7,8-HxCDF	ND	ND
1,2,3,6,7,8-HxCDF	ND	ND
1,2,3,7,8,9-HxCDF	0.0068	ND
2,3,4,6,7,8-HxCDF	0.0013	ND
1,2,3,4,6,7,8-HpCDF	0.0341	0.0288
1,2,3,4,7,8,9-HpCDF	ND	ND
OCDF	0.0699	0.0434
PCDDs	0.1122	0.0722
PCDFs	0.1418	0.0665
PCDDs/PCDFs ratio	1.26	0.92
Total PCDD/DFs	0.2541	0.1387
PCDDs I-TEQ <sup>a</sup>	0.0005	0.0002
PCDFs I-TEQ <sup>a</sup>	0.0012	0.0003
PCDDs/PCDFs(TEQ) ratio	0.39	0.68
I-TEQ <sup>a</sup>	0.0017	0.0006

<sup>a</sup> The unit is pg I-TEQ/Nm<sup>3</sup>.

## 3.5. Source Identification by Principal Component Analysis

In order to estimate the possible emission sources for PMs, the PCA model was used in this study. The PCA for water-soluble ions is listed in Table 4. The total value was over 79.8% of the explained variance. The PC1 had high loadings of Cl<sup>-</sup>, Na<sup>+</sup>, F<sup>-</sup>, and Mg<sup>2+</sup> with a maximum percentage of variance of 46.94%. The Na<sup>+</sup>, Mg<sup>2+</sup>, and Cl<sup>-</sup> may come from sea salt, while F<sup>-</sup> may come from industrial processes [50]. The PC2 consisted of NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>, with 32.92% of the total variance comprised of secondary pollutants formed by photochemical reactions [51–53].

Water-Soluble Ions	Factor 1	Factors 2
Na <sup>+</sup>	0.831	0.324
$NH_4^+$	-0.711	0.657
K <sup>+</sup>	-0.449	0.138
Ca <sup>2+</sup>	0.184	-0.365
$Mg^{2+}$	0.781	0.064
$F^-$	0.821	-0.110
$Cl^{-}$	0.870	0.109
$NO_3^-$	0.163	0.950
$SO_4^{2-}$	0.487	0.719
% of Variance	46.942	32.921
Cumulative %	46.942	79.864
Possible sources	Sea salt Industrial process	Secondary pollutants

**Table 4.** Principal component analysis (PCA) for water-soluble ions. The bold means it emphasize the strong related ions to each Factors.

## 4. Conclusions

In this study, we successfully characterized the PMs with size distributions, ion and cation compositions, metals, and PCDD/F concentrations in an industry-intensive area in central Taiwan. Coarse particles were dominant in spring and summer, with an average mass concentration of 16.96 and 12.67  $\mu$ g/m<sup>3</sup>, accounting for 47.5 and 45.6%, respectively. Fine particle concentrations were 15.39 and 18.17  $\mu$ g/m<sup>3</sup> in autumn and winter, accounting for 46.7 and 62.2%, respectively. The sequence of the major ionic species in the coarse particles were Cl<sup>-</sup> (1.63  $\mu$ g/m<sup>3</sup>) > NO<sub>3</sub><sup>-</sup> (1.62  $\mu$ g/m<sup>3</sup>) > SO<sub>4</sub><sup>2-</sup> (1.18  $\mu$ g/m<sup>3</sup>), whereas the sequence was SO<sub>4</sub><sup>2-</sup> (2.32  $\mu$ g/m<sup>3</sup>) > Cl<sup>-</sup> (1.37  $\mu$ g/m<sup>3</sup>) > NO<sub>3</sub><sup>-</sup> (1.30  $\mu$ g/m<sup>3</sup>) in the fine particles. In addition, the largest metal components were Na, Fe, Ca, Al, and Mg in different particle sizes. The measured PCDD/F concentrations ranged from 0.0006 to 0.0017 pg I-TEQ/Nm<sup>3</sup>. According to the results of PCA, the contribution to PMs were associated with sea salt, secondary pollutants, and industrial process.

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