



Characteristics of Heavy Metals Emitted from a Heavy Oil-Fueled Power Plant in Northern Taiwan

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ABSTRACT

The characteristics and distribution of metal contents emitted from a power plant fueled by heavy oil and its impact to the ambient atmosphere near the power plant was investigated. The current investigation measured toxic (As, Cd, Cr, Hg, Ni and Pb), anthropogenic (Ba, Cu, Mn, Sb, Se, Sr, Ti, V and Zn) and crust (Al, Ca, Fe, K and Mg) elements from a 2,000 MW heavy oil-fired power plant. Results showed the emission concentration from the power plant contributed to 17,976 kg/yr annual emission of anthropogenic elements, which was significantly higher than those from some electrical arc furnaces and coke ovens in Taiwan. For toxic metals, As, Cd or Ni concentration do not exceed target values established by the European Council (2004/107/EC) for As (6 ng/m³), Cd (5 ng/m³) and Ni (20 ng/m³). This study also applies nonparametric statistical analyses for evaluating the relationship between metal concentrations and operational parameters (including emitted CO₂, O₂, flue gas emission temperature, flue gas velocity, moisture, heavy oil consumption rate, boiler steam temperature, boiler operational pressure, and electricity). Findings show negative correlations between most toxic metals (As, Cd, Cr and Hg) and operational parameters, though some pairs were not statistically significant. The current study provides only preliminary statistical results between metal concentrations and operational parameters due to small sample sizes. Further investigation requires larger sample sizes.

Keywords: Heavy oil power plant; metal; Emission factor; Ambient atmosphere; Operational parameters.

INTRODUCTION

Scientists recognize certain trace metals, such as iron (Fe), zinc (Zn), copper (Cu), chromium (Cr), iodine (I), cobalt (Co), molybdenum (Mo), and selenium (Se), as essential elements for human health and plant nutrition. However, the toxic properties of many metals are a point of concern. (Sarkar, 2002; Saxena *et al.*, 2008). Findings show that exposure to transition metals associated with ambient particles originating from industrial sources elicit airway inflammation in healthy subjects (Ghio and Devlin, 2001; Schaumann *et al.*, 2004; Wang *et al.*, 2008). Highly concentrated airborne metals in industrial areas could play a role in regional prevalence of allergic conditions

(Heinrich *et al.*, 2002; Kuo *et al.*, 2007). This hypothesis is supported by evidence that sensitized mice exposed to PM_{2.5} extracts rich in Zn, Mn, Pb, Cu and Cd, exhibited increased airway responsiveness to metacholine and lung inflammatory cells (Gavett *et al.*, 2003). Metals from industrial emissions or traffic have been associated with heart rate variability (Magari *et al.*, 2002), increase in hospital emergency visits for respiratory conditions, and increased risk of lung cancer after long term exposures (Pope Iii *et al.*, 2002). In urban and industrial environments, however, particles not only contain metals, but also include toxic organic compounds, such as PAHs, whose concentrations are greater in fine and ultrafine particles. This fact has caused people concern about various pollution sources around their habitations.

Coal and oil combustion facilities produce fine particles in the submicron size range (Jang *et al.*, 2007), enriched by heavy metals. Jang *et al.* (2007) indicated that heavy oil combustion resulted in relatively pronounced ultra-fine particles smaller than 1.0 μm, difficult to remove by

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existing air pollutant control devices (APCD). The diverse PM composition responsible for these health effects is still unknown and attracts researchers' attention, heightening awareness of pollutants emitted from power plants. Some literatures address heavy metal emissions from coal/oil power plants worldwide, such as Finland (Hatanpaa *et al.*, 1997), Greece (Petaloti *et al.*, 2006), Spain (Moreno *et al.*, 2007), Turkey (Yatkin and Bayram, 2008), India (Reddy *et al.*, 2005) and Korea (Jang *et al.*, 2007), however, few literatures refer to Taiwan. This study investigates the characteristics and emission factors of heavy metals from a power plant fueled by heavy oil. Besides the influence of power plant emission on the nearby atmosphere, the current work also evaluates the relationship between heavy metal and operational parameters to provide more information for further research on the effect of flue gas from the power plant.

EXPERIMENTAL SECTION

The current investigation selects the largest power plant (2,000 MW) near the Pacific Ocean and located in northern Taiwan, for this study (Fig. 1). Sixteen samples were collected from the power plant stack fueled by low-sulfur heavy oil from March 2006 through January 2007. The power plant is equipped with four sets of boilers configured with lower-NO_x burners and with electrostatic precipitators as air pollution control devices. The oil consumption ranged from 41.1 kL/hr to 94.5 kL/hr. All samples were collected from the flue gas stack according to Taiwan EPA NIEA A302.72C. To ensure contamination-free sampling and transportation, this research also took one blank trip and one blank field when conducting the field sampling. After completing the flue gas sampling, this work brought the samples back to the laboratory and placed them in a refrigerator at a temperature below 10°C.

Six ambient air samples were collected using a PS-1 sampler (Graseby Andersen, GA) according to the Taiwan EPA Reference Method NIEA A102.11A. Fig. 1 also shows the sampling site. The samples were collected separately in August and November. Three sampling sites A, B, and C were chosen as considering the upwind and downwind effect and available sampling location. The wind blew from the southwest 8% of the time on August and from the northeast around 30% of the time on November according to the wind rose. The sampling flow rate was specified at ~0.225 m³/min, and each sample was collected continuously on three consecutive days. The PS-1 sampler was equipped with a quartz fiber filter for sampling heavy metals. The filters were digested following the procedure of NIEA A301.11C, which is the EPA reference method in Taiwan, and metal elements were quantified by ICP-AES (Jobin-Yvon, JY38 Plus). Additionally, nonparametric correlations and the median test analysis for all samples were analyzed using Statistical Package for Social Sciences, version 15.0 (SPSS, Chicago, IL).

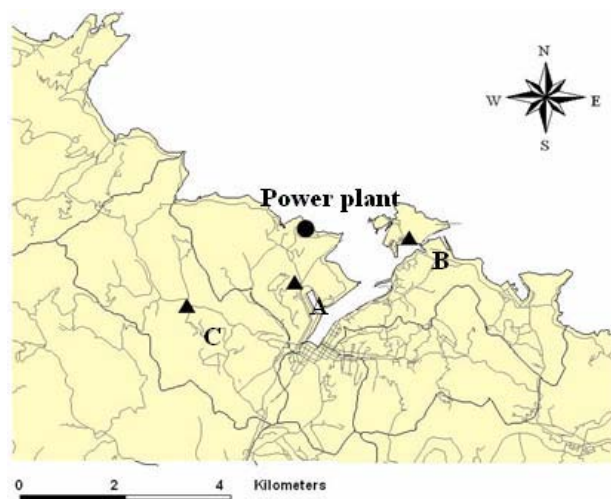


Fig. 1. The sampling sites were located in northern Taiwan.

RESULTS AND DISCUSSION

Metal Concentrations in Flue Gas Stack of the Heavy Oil-Fueled Power Plant

The current work classified metal elements into toxic (As, Cd, Cr, Hg, Ni and Pb), anthropogenic (Ba, Cu, Mn, Sb, Se, Sr, Ti, V and Zn) and crust (Al, Ca, Fe, K and Mg) groups (Wang *et al.*, 2003). Findings reveal higher mean concentrations in two crust elements of Al and K (1637 and 1153 $\mu\text{g}/\text{m}^3$), respectively, and two anthropogenic elements, Zn and Ba (1445 and 1128 $\mu\text{g}/\text{m}^3$). The four metals account for 82.2% of total metal element mass concentrations. Findings show Mercury (Hg) as having the lowest metal element mass concentrations of the twenty investigated, with a mean value of 0.15 $\mu\text{g}/\text{m}^3$. The concentrations of toxic elements Cd, Cr, Ni and Pb, were higher than those in Reddy's study (Reddy *et al.*, 2005), however, As and Hg were lower than those in Reddy's study (Reddy *et al.*, 2005). The capacity in this study was 2,000MW, much higher than in Reddy's study (200 MW for a coal-fired power plant and 6 MW for a fuel-oil based power plant). Given a compatible power plant (1170 MW), the concentrations of As Cd, Cr, Hg, Ni and Pb were much lower than those in Aunela-Tapola's study (Aunela-Tapola *et al.*, 1998).

Table 1 lists metal concentrations in the flue gas stack of a power plant among different seasons, the first report among English literatures for a power plant fueled by heavy oil in Taiwan. Findings show most mean metal concentrations in fall as having the lowest concentration compared to those in other seasons except Fe, Cd, Cr, Mn, Ni, Sb, Se and Ti. Furthermore, findings show Cd and Hg as statistically significant with seasons. The result also indicates seasonal statistical significance for some operational parameters, flue gas temperature, flue gas velocity, fuel consumption, boiler steam temperature and electricity. Additionally, no significant differences exhibited in all variables among the four stacks via the nonparametric Kruskal-Wallis *H* tests (data not shown). These findings also qualified the analysis results.

Table 1 Concentrations of metal in the flue gases of the power plant.

Elements ($\mu\text{g}/\text{Nm}^3$, dry)	Spring (n = 4)			Summer (n = 4)			Fall (n = 4)			Winter (n = 4)			<i>p</i> ^a			
	Mean	SD	Range	Mean	SD	Range	Mean	SD	Range	Mean	SD	Range				
<i>Toxic metals</i>																
As	0.18	0.000	0.10-0.30	0.93	0.001	0.10-1.80	0.18	0.000	0.15	0.10-0.30	0.45	0.000	0.40	0.40-0.60	0.098	
Cd	0.75	0.001	ND ^b -2.00	ND	ND	ND	5.68	0.005	3.85	1.40-13.6	1.58	0.002	1.35	ND-3.60	0.028*	
Cr	10.1	0.012	3.00-27.9	7.43	0.005	1.50-13.3	3.93	0.001	3.65	2.60-5.80	1.90	0.001	2.00	0.50-3.10	0.113	
Hg	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.60	0.001	0.25	ND-1.90	0.017*	
Ni	32.2	0.027	13.0-72.3	10.4	0.009	8.5	2.0-22.4	12.3	0.008	12.4	4.80-19.6	15.8	0.012	11.4	7.10-33.2	0.195
Pb	7.76	0.004	3.00-13.0	7.20	0.005	6.85	2.20-12.9	4.93	0.001	4.65	3.80-6.60	5.23	0.003	5.35	2.30-7.90	0.599
Sum	51.1			25.9			27.0				25.5					
<i>Crust metals</i>																
Al	4776	8.351	465-17300	495	0.186	362-770	82.2	0.030	78.4	50.0-122	1193	1.355	996	40.4-2740	0.109	
Ca	772	0.226	570-973	680	0.311	351-1061	262	0.130	256	142-395	1015	0.993	890	59.7-2220	0.207	
Fe	420	0.205	245-710	241	0.219	122-569	243	0.169	169	141-494	193	0.133	190	53.4-337	0.300	
K	1669	0.818	496-2400	1201	0.754	569-2290	106	0.015	104	92.5-124	1636	1.907	1343	17.0-3840	0.229	
Mg	120	0.023	94.9-150	150	0.107	90.8-311	93.3	0.074	71.0	35.3-196	117	0.103	115	10.5-229	0.760	
Sum	7757			2768			787				4154					
<i>Anthropogenic metals</i>																
Ba	1459	0.620	726-2220	810	0.806	5.20-1910	4.90	0.003	4.15	2.60-8.70	2236	2.687	1776	1.50-5390	0.210	
Cu	5.38	0.002	2.80-6.90	5.63	0.006	0.80-15.1	1.25	0.002	0.65	0.10-3.60	2.28	0.002	1.95	0.40-4.80	0.086	
Mn	35.8	0.010	28.1-50.7	26.1	0.010	17.0-39.2	32.3	0.009	34.8	20.0-39.8	18.5	0.017	15.8	1.70-40.7	0.446	
Sb	0.71	0.000	0.30-1.20	0.28	0.000	0.10-0.50	0.38	0.000	0.15	0.10-1.10	0.35	0.000	0.35	0.30-0.40	0.207	
Se	0.41	0.000	0.15-0.70	0.25	0.000	ND-0.80	0.45	0.000	0.40	0.20-0.80	0.06	0.000	0.07	ND-0.10	0.092	
Sr	7.25	0.003	4.39-10.6	5.93	0.002	5.55	4.10-8.50	1.28	0.001	1.25	0.60-2.0	6.60	0.008	4.50	0.30-17.1	0.135
Ti	1.38	0.000	0.80-1.70	1.53	0.000	0.80-1.90	2.90	0.002	1.85	1.50-6.40	3.33	0.003	3.20	0.60-6.30	0.390	
V	12.7	0.009	5.97-25.4	6.08	0.004	2.30-9.70	5.60	0.006	3.65	1.30-13.8	8.6	0.010	4.85	1.90-22.7	0.388	
Zn	1969	0.799	847-2630	1479	0.875	666-2660	87.0	0.047	82.6	35.0-148	2243	2.645	1812	10.4-5340	0.163	
Sum	3492			2335			136				4519					
CO ₂ (%)	12.4	0.33	12.0-12.8	12.5	0.31	12.1-12.8	12.5	0.10	12.4	12.4-12.6	12.6	0.14	12.7	12.4-12.7	0.503	
O ₂ (%)	5.48	0.38	5.00-5.90	5.35	0.34	5.00-5.80	5.10	0.34	4.95	4.90-5.60	5.28	0.38	5.20	4.90-5.80	0.377	
FGT(°C)	152	5.29	147-158	137	13.5	127-157	130	4.53	128	127-137	126	2.84	125	124-130	0.021*	
FGV(m/s)	21.0	3.62	16.6-25.3	12.1	6.01	8.53-21.1	13.5	1.35	13.4	12.1-15.3	14.9	1.28	15.2	13.0-15.9	0.038*	
MT(%)	12.1	0.77	11.1-12.7	11.2	1.42	9.75-12.7	11.7	0.44	11.7	11.1-12.1	11.4	0.08	11.4	11.3-11.4	0.525	
FC (kL/h)	90.5	3.18	87.5-94.5	52.6	23.6	37.5-87.8	60.5	1.72	59.8	59.3-63.0	67.5	5.27	68.2	61.6-72.1	0.021*	
BST (°C)	538	2.31	535-541	535	8.71	522-540	508	5.02	508	502-513	511	4.43	510	507-517	0.009*	
BP(kg/cm ²)	178	12.2	171-197	121	39.8	93.4-180	152	6.70	151	145-161	154	5.06	154	148-159	0.069	
E (MWH)	399	3.33	396-404	226	115	164-398	266	7.89	267	255-275	299	12.5	301	284-312	0.026*	

a: Kruskal-Wallis *H* tests, b: ND: not detectable, *p* < 0.05

FGT: flue gas temperature, FGV: flue gas velocity, MT: moisture, FC: fuel consumption, BST: boiler steam temperature, BP: boiler pressure, E: electricity.

For toxic metals, Cr, Ni and Pb concentrations reached the highest in spring. However, Hg concentrations are only detected in winter and not detectable in other seasons. The sums of toxic metals and crust metals in spring were obviously much higher than those in other seasons. The sums of crust metals and anthropogenic metals were much lower in fall than those in other seasons. Dust storms always occur in the dry springtime (Fang *et al.*, 2009; Shen *et al.*, 2009), which will probably lead to more toxic metals from the power plant contributing to nearby ambient air. It warrants more concern in the future.

Metal Concentrations in Ambient Air around the Heavy Oil-Fueled Power Plant

As concentration ranged from 0.50–2.00 ng/m³, with an average of 1.30 ng/m³. Cd concentration ranged from 0.10–0.70 ng/m³, with an average of 0.45 ng/m³. Ni concentration ranged from 2.00–13.0 ng/m³, with an average of 5.35 ng/m³. Pb concentration ranged from 8.9–46.7 ng/m³, with an average of 26.2 ng/m³. Concentration was between Moreno's result (Moreno *et al.*, 2007) of PM₁₀ and PM_{2.5} (a mean of 1.9 ng/m³ and 1.0 ng/m³, respectively) in Puertollano, an important coal mining town including fossil fuel burning power plants as well as petrochemical and fertilizer complexes. Cd, Ni or Pb concentration was higher than that in Moreno's result (Moreno *et al.*, 2007) (Cd in PM₁₀ and PM_{2.5} both were 0.1 ng/m³; Ni in PM₁₀ and PM_{2.5} were 4.1 ng/m³ and 3.0 ng/m³, respectively; Pb in PM₁₀ and PM_{2.5} were 12 ng/m³ and 9.3 ng/m³, respectively). However, Cd, Ni or Pb was lower than that in Yatkin's study (Yatkin and Bayram, 2007), whether in an urban or suburban sampling site (except for PM_{2.5} sample in suburban winter). Lippmann *et al.* (2006) indicated Ni was a particularly influential component of ambient FPM (fine ambient particulate matter) in terms of cardiac responses to inhalation ambient air FPM. The Cd or Ni concentration did not exceed target values established by the European Council (2004/107/EC) for As (6 ng/m³), Cd (5 ng/m³) and Ni (20 ng/m³) (Petaloti *et al.*, 2006). The Pb concentration was also much lower than that (1.0 µg/m³) in the ambient air quality standard of the Taiwan EPA (2004/0060) (EPA). The Hg concentration was 0.10 ng/m³ in all samples, the lowest concentration compared with other studies (Kim and Kim, 2001; Liu *et al.*, 2002; Sakata and Marumoto, 2002; Wang *et al.*, 2005).

Fig. 2(a) and 2(b) show the toxic metal profiles measured from ambient atmosphere of the heavy oil-fueled power plant in August and November, respectively. The Pb was the dominant metal among the six toxic metals for August and November at three sampling sites, except for November samples at the B site. Fig. 2(c) illustrates the averaged profiles of toxic metals in ambient atmosphere. The pattern was totally different from that in the power plant flue gas. The highest toxic metal concentration was Cd in flue gas samples; however, Pb was highest in ambient samples. The influence of metal concentrations from power plant flue gas to ambient atmosphere is seemingly not direct; other possible influential factors warrant further investigation.

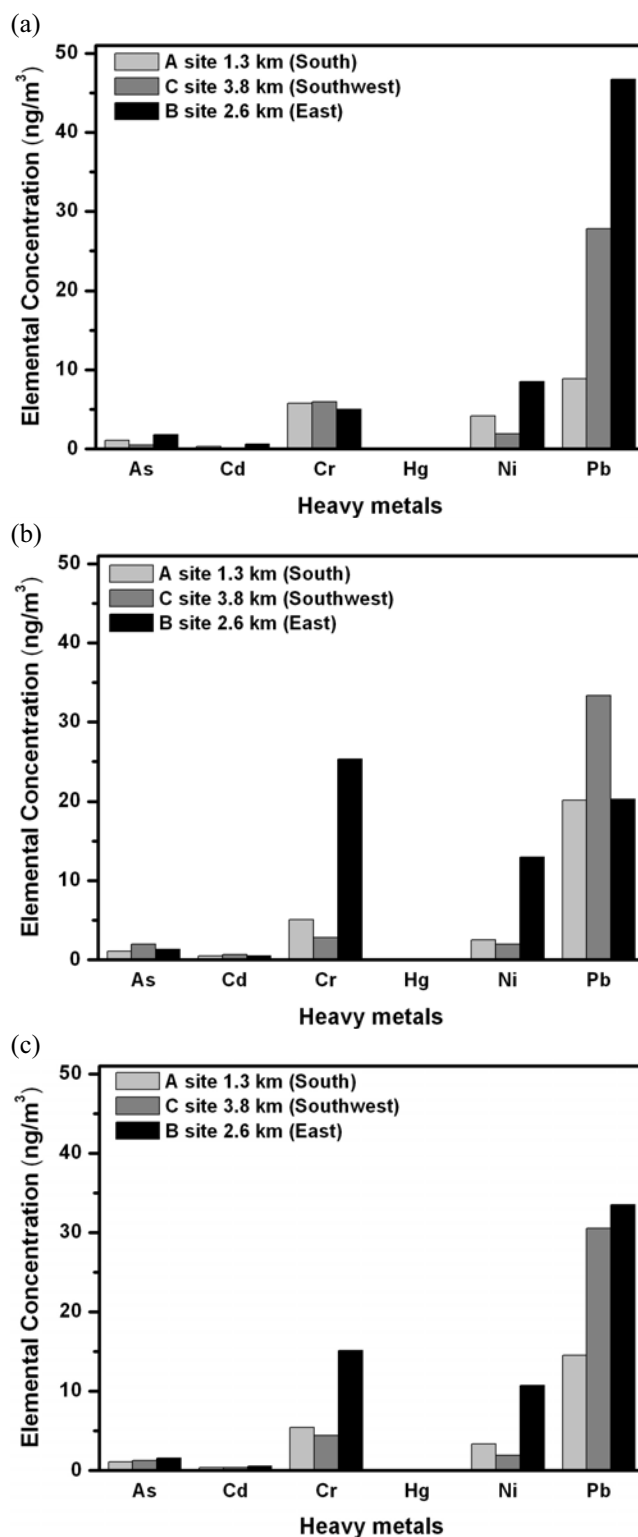


Fig. 2. (a) Toxic metals profiles in the ambient atmosphere in August (b) Toxic metals profiles in the ambient atmosphere in November (c) Averaged toxic metals profiles in the ambient atmosphere

Al, Fe and Ca are the most common metal elements formed in the earth's crust (Taylor, 1964). The enrichment factor (EF) pattern is a reference value of contamination

status (Kim *et al.*, 2002). The EF provides information to judge enrichment (or depletion) of a given element relative to the reference element (Al, in this study). The Taylor's study defines EF as follows: $EF = \{X/Al\}_{\text{sample}} / \{X/Al\}_{\text{crust}}$ (X denotes a metal of interest). The EF values were grouped into three categories in this study: (1) <10, Al, Ca, Fe, Mg, Mn, Sr and Ti, (2) 10–1000, As, Ba, Cr, Cu, K, Ni, Pb, Sb and V, (3) >1000, Cd, Se and Zn. Fig. 3 also shows the EF for the twenty analyzed metal elements for various studies.

For toxic metals, the values of EFs for As, Cd and Ni were highest in Reddy's study on the coal fired power plant (Reddy *et al.*, 2005). The 220 MWs power plant was located in western India with an independent bituminous coal generation unit and equipped with tangential burners. The EF for Cr was highest in a 6 MW-installed capacity oil-based power plant also located in western India (Reddy *et al.*, 2005). Lower EFs for the power plant in this study were probably due to higher combustion efficiency or higher APCD efficiency. Findings show the EF for Pb in Yatkin's study (Yatkin and Bayram, 2008) to be highest. The sampling site was in the city center near a motorway in Izmir, the third most populous city in Turkey. Heavy loading transportation obviously contributed more Pb to the atmosphere than other industries. Contrasted to toxic metals, the highest EFs found in this study were Al, Ba, K, Mg, Mn, Sr and Zn, perhaps because of the power plant's special geographic effect.

Correlation of Metal Concentrations and Operational Parameters

This study applied nonparametric statistical analyses to evaluate the relationship between metal concentrations and operational parameters. The operational parameters included emitted CO₂, O₂, flue gas emission temperature,

flue gas velocity, moisture, heavy oil consumption rate, boiler steam temperature, boiler operational pressure, and electricity. The sampling size was not large enough to reach statistical significance; therefore, we chose the nonparametric Spearman correlation coefficient test. The results show no significant difference for all metal concentrations among four stack flue gases (data not shown). This finding indicates no obvious emission concentration variations for the mean annual values. Findings show significant Spearman's correlation coefficients among parts of the metals (Table 2), but As, Cd, Cr, Hg and Ti correlated less with other metals. The highest coefficient in nineteen metal pairs occurred between Ba and K, with $r = 0.968$ and $p < 0.001$. All statistically related metals showed significant positive correlations, however, findings show only a negative correlation between As and Se ($r = -0.564$, $p = 0.023$). Seventeen pairs among sixty significant pairs have high coefficients with $r > 0.800$ and $p < 0.001$, especially, the crust metals, Al, Ca and K related pairs, account for eleven pairs.

Statistically significant associations did not exhibit between metal concentrations and flue gas velocity. Significant positive correlations were found between Ni and fuel consumption ($r = 0.753$, $p = 0.001$), boiler pressure ($r = 0.679$, $p = 0.004$) and electricity ($r = 0.662$, $p = 0.005$). However, significant negative correlations were found between As and moisture ($r = -0.606$, $p = 0.013$), and boiler pressure ($r = -0.625$, $p = 0.01$). Ca, Cr, Cu, Mg, Mn, Ti, V and Zn showed no significant correlation with all operational parameters. Furthermore, the values of Cr and Pb were significantly related to decreasing CO₂ emission ($r = -0.528$, $p = 0.036$ and $r = -0.553$, $p = 0.026$, respectively). Table 2 indicates negative correlations between most toxic metals (As, Cd, Cr and Hg) and

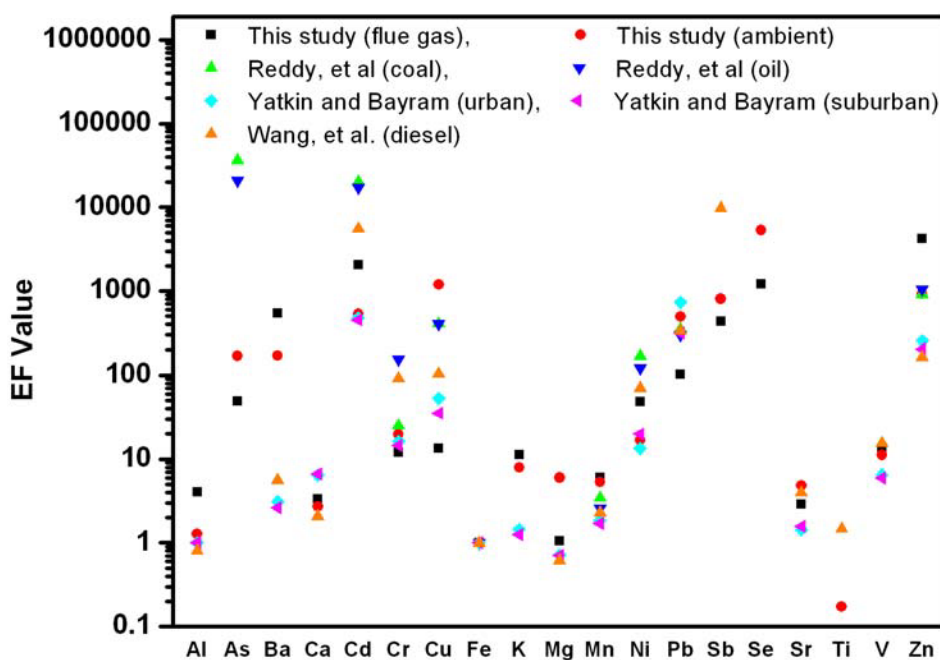


Fig. 3. The enrichment factor (EF) ratio for each metal element.

operational parameters, though some of the pairs were not statistically significant. Especially, for all operational parameters, Hg only negatively significantly correlated with flue gas temperature ($r = -0.526$, $p = 0.036$). Comparing the pairs of operational parameters, findings show significant positive correlations between electricity and flue gas velocity ($r = 0.906$, $p < 0.0001$), fuel consumption ($r = 0.950$, $p < 0.0001$) and boiler pressure ($r = 0.909$, $p < 0.0001$). This study provides only preliminary statistical results between metal concentrations and operational parameters because of small sample size. Further investigation requires larger sample sizes. Nevertheless, to our knowledge this is the first report to interpret correlations between metal concentrations and operational parameters.

CONCLUSIONS

Comparing with other studies on power plants, metal emission concentration from the power plant was not the highest. However, comparing with local data in Taiwan, the value for anthropogenic element emission from the power plant (17,976 kg/yr) was much more significant (10.8, 104, and 4.81 fold higher than those emitted from the coal power plant, electrical arc furnace, and coke oven) compared with three reference emission sources (Wang et al., 2003). The value for crust element emission from the power plant (14,744 kg/yr) was not so significant (0.16, 7.16, and 0.24 fold higher than those emitted from the coal power plant, electrical arc furnace, and coke oven) compared with three reference emission sources (Wang et al., 2003). Metal concentration from the power plant was not so high; however, the total amount of metal emissions from power plants cannot be neglected owing to the high volume of flue gas. For proper environmental management of metals, especially toxic metals, establishing a complete source inventory of metal emission is necessary. The government should particularly pay more attention to power plants to address the information shortage.

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