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Effect of Particulate Matters from Shipping Activities around Si Racha Bay – Si Chang Island

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Abstract

Shipping activities around Si Chang harbor including tapioca flour pellet, cement, fertilizer, coal, and so on caused the deposition of particulate matters into marine environment and dispersion of fine particles to Si Racha and Si Chang coastal line. All particles were monitored to identify the emission sources by Scanning Electron Microscope (SEM). Chemical compositions were analyzed for the amounts of heavy metals adsorbed on the collected particulate matter by ICP-AES. Total Suspended Particulate (TSP) and Particulate Matter less than 10 micrometers (PM_{10}) were collected at the coast line of Queen Savangwadhana Hospital at Si Racha from April to July, 2008. The amounts of TSP varied from 0.0239-0.0950 $\mu g/m^3$ with average value at 0.0726±0.0191 $\mu g/m^3$. The concentrations of PM_{10} varied from 0.0142-0.0482 $\mu g/m^3$ with average value at 0.0195±0.01 $\mu g/m^3$. The microscopic structures of TSP provided the difference in shape and dimensions from 0.1 to greater than 100 micrometers while the microstructures of PM_{10} presented the spherical shape. The dimensions and shapes of all particulate matter samples were mostly similar to reference particulate of tapioca flour and fertilizer particles. The particulate with greater than 100 micrometers which provided similar shape to cement does not be seen in all samples. It could be summarized that the light particle of tapioca flour were dominated in the particulate matters dispersion from shipping activities at Si Chang harbor. The weighted mean concentrations (WMC ± WSD) of Zn, Ni, Cu, Hg, Pb and Se were 0.0476±0.0831, 0.0010±1.29 x 10^{-4}, 0.0071±4.96 x 10^{-5}, 0.0007±1.32 x 10^{-7}, 0.0006±1.78 x 10^{-7} and 0.0006±1.53 x 10^{-7} ng/g.m^3, respectively for TSP and 0.0119±1.46 x 10^{-4}, 0.0005±4.73 x 10^{-7}, 0.0024±1.31 x 10^{-5}, 0.0001±5.41 x 10^{-9}, 0.0004±1.55 x 10^{-7} and 0.0004±3.45 x 10^{-8} ng/g.m^3, respectively for PM_{10}.

Keywords: particulate matter; TSP; PM_{10}; SEM-EDS; heavy metals; ICP-AES

1. Introduction

Particulate air pollution causes serious problems around the world, especially in less-industrialized countries where the early stages of industrial growth are often pursued without much investment in environmental protection, leading to heavy air pollution in urban areas (Florig *et al*., 2002). Particulate matters from anthropogenic sources might concern various environmental impacts such as toxic effects of particle exposures on human health, reduction of visibility by light scattering from the particles, local, regional and global climatic effects related to back scattering of light to space and absorbing incoming solar radiation and outgoing thermal radiation and nuisance effects from deposited particles (Godish, 2003). Industries and transportations are the major sources of particulate matters. The emission of $PM_{2.5}$ and secondary particles were identified by positive matrix factorization, PMF from ship engine in the major ports around Seattle areas. Kim and Hopke (2008) also found other emission sources including diesel vehicle, airborne soil, residual oil combustion, sea salt, aged sea salt, metal powder and cement kiln. Contribution of particulate matter was one of main criteria pollutants that were greatest concerning to human health and leading to a social cost (Bickel and Friedrich, 2005; McCubbin and Delucci, 1996).

Elemental compositions of aerosols in Daihai Lake were determined for evaluation their change in physical and chemical properties which were varied from their origins, sources, transportation procedures and meteorological conditions. The ratio of elemental concentrations in TSP/$PM_{2.5}$ showed the abundant of crustal elements such as K, Ca, Ti, Fe as well as Pb, Ni and Sr dominated in TSP. The other elements including Cl, Ti, V, Cr, Mn, Cu, Zn, As, Se, Br and Zr were dominated in $PM_{2.5}$. In monsoon sensitive region as Daihai, the anthropogenic elements were lower while the crustal elements were in similar levels (Han *et al*., 2009).

For normal transportation, ambient air pollutions emitted from engine (Kim and Hopke, 2008), gaseous waste; carbon monoxide, nitrogen oxides, total organic gases and volatile organic compounds (Wang *et al*., 2009). For the transportation ports, ambient air pollutions were contaminated with emission from engine itself and shipping activities.

Si Chang Island is located in the Gulf of Thailand with 7 km far from Si Racha Bay; it belongs to Chonburi province (Fig. 1). Shipping activities occurred in the sea about 1 km far from the island to the east. Tapioca flour, coal, cement and fertilizer were the important import/export items that caused the emission of particulate matters during transportation. The distribution of fine

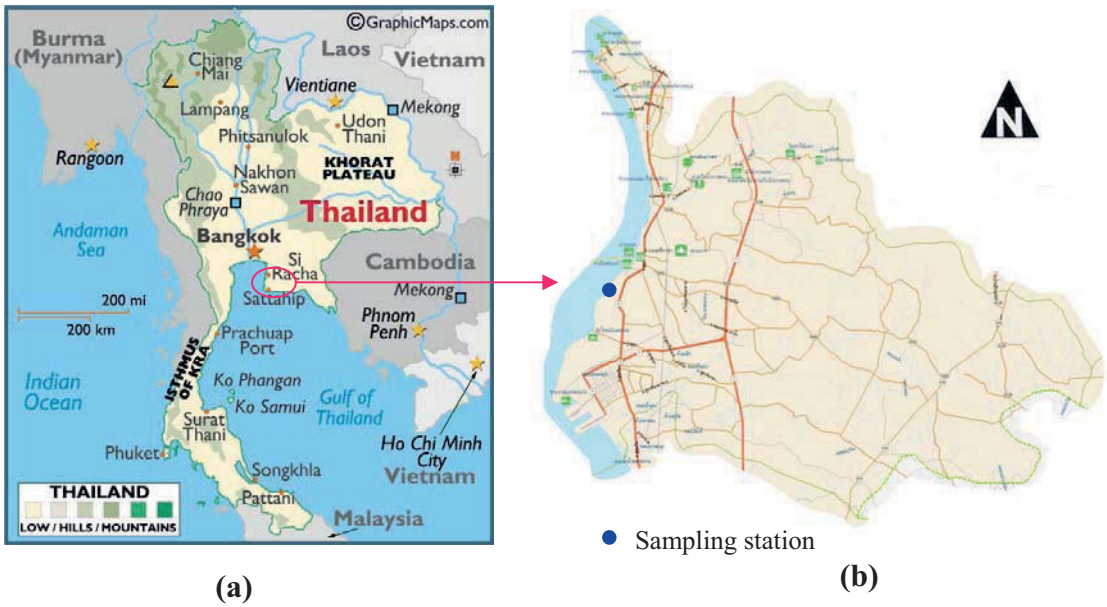

Figure 1. Map of Thailand presents the location of Si Racha and (b) Si Racha area and the sampling site.

particles can be seen in Fig. 2. The coarse particles settled down to the sea while the fine particles were carried by wind to reach the coast line of Si Racha.

The aims of this research are (1) to quantify the concentration of TSP and PM_{10} in ambient air around Si Racha area (2) to identify the type and compositions of particle by Scanning Electron Microscopes and X-ray Fluorescence Spectrometry and (3) to analyse the contamination of heavy metals adsorbed in particulate matters by ICP-AES. The obtained data may be applied for clarify the emission sources that were very important to reduce the emission from shipping activities.

2. Materials and Methods

2.1. Site selection and collection of particulate matters

Sample site was located beside the coastal area of Queen Savangwadhana Hospital at Si Racha (13° 9′ 43.20″ N and 100o 55′ 8.04″ E). Si Chang Island where shipping activities occurred was 7 km far from sampling site. The sampling site was approximately 250 m far from local road; it was semicircle to the sea and far away from other emission sources of particulate matter except only shipping activities (Fig. 2).

Gravimetric method was applied for collection of TSP and PM_{10}. For constant weight, the glass fiber filter (8 x 10 $inch^2$) was dried at 150–170°C not less than 24 h, kept in zip locked bag and cooled in desiccator for 2–3 h. The filter was accurately pre-weighted with analytical balance (Metler Toledo AG 204, ±0.0001 g) and installed in the filter holder. Air sample was pumped through 0.3 micron glass fiber filter of Andersen high volume air sampler for 24 h continuously at the flow rate of 40–60 ft^3/min.

For PM_{10}, the particle less than 10 micrometers in air sample was eliminated to settling chamber by impaction before passing through quartz fiber filter. Particulate matter which less than 10 micrometers were collected on the filter of Andersen air sampler. Quartz fiber filter was prepared in the same procedure as glass fiber filter.

Figure 2. Emission of particulate matters occurred during shipping processes

The filter was carefully fixed in the filter holder of high volume air sampler. Air sample was continuously collected for 24 h.

Before operation, both devices were pre-calibrated for leakage and air flow rate with Orifice flow rate transfer standard. The air flow rate, collection date and time, temperature and other climate parameters were recorded. Air samples were collected from April to June, 2008 particularly during shipping activities.

After 24 h, the filters were removed and wrapped together, the filter were then kept into zip lock bag. The filters were accurately weighted; the amounts of particle were calculated from the weight differences. Amounts of TSP and PM_{10} were presented in Tables 1-2 and Figs. 3–4.

2.2. Microscopic characterization of particulate matters by Scanning Electron Microscope

The shape and chemical structure of particles in the filters were observed using a Scanning Electron Microscope equipped with Energy Dispersive Spectrometer (Phillips: XL30 & EDAX). The filter samples and the reference (fertilizer, tapioca flour pellet and cement) fixed on a filter were cut onto the area of 0.5 x 0.5 cm^2. Gold and carbon were coated on the filter membrane with a Pirani 501 device for electrical conductance in SEM and EDS analysis, respectively. Analytical conditions were fit to 15 kV accelerating voltage and 100 s of effective counting time.

Besides EDS analysis, the electron beam was exposed over the selected area. Elements with atomic number less than 11 cannot be determined due to insufficient accuracy and presence of carbon in the substrate.

2.3. Chemical composition of heavy metals in particulate matters

Heavy metals adsorbed in particulate matters were determined by digestion a quarter of fitter paper with 20 ml deionized water (purified by Millipore Simplicity 185) in Sonorec ultrasonic bath (Super RK 514 BH) and the extract solutions were analyzed by Inductively Coupled Plasma Atomic Emission Spectrometry, ICP-AES (Jobin Yvon JY2000). The instrument was well calibrated with Standard Reference Materials (Spex Certiprep) of heavy metals prior analysis.

3. Results and Discussion

3.1. Concentrations of TSP and PM_{10} during April to July, 2008

Concentrations of TSP and PM_{10} were calculated from weight of particulate matter multiplied by air flow

Table 1. Concentrations of TSP during April to June 2008.

Collecting time		Air flow rate	Air volume	Particle weight	Concentration of TSP
Start	Stop	(m^3/min)	(m^3)	(µg)	(µg/m^3)
1/4/08	2/4/08	1.186	1707.84	0.0409	0.0239
3/4/08	4/4/08	1.186	1707.84	0.0649	0.0380
8/4/08	9/4/08	1.588	2286.72	0.0732	0.0320
11/4/08	12/4/08	1.186	1707.84	0.0732	0.0429
17/4/51	18/4/51	0.570	820.80	0.0378	0.0461
21/4/08	22/4/08	0.285	410.40	0.0133	0.0324
24/4/08	25/4/08	1.242	1788.48	0.0269	0.0150
28/4/08	29/4/08	1.242	1788.48	0.0513	0.0287
21/5/08	22/5/08	1.420	2044.80	0.0562	0.0275
22/5/08	23/5/08	1.130	1627.20	0.0556	0.0342
2/6/08	3/6/08	1.420	2044.80	0.0830	0.0406
5/6/08	6/6/08	1.420	2044.80	0.1115	0.0545
9/6/08	10/6/08	1.270	1828.80	0.0947	0.0518
11/6/08	12/6/08	1.420	2044.80	0.1942	0.0950

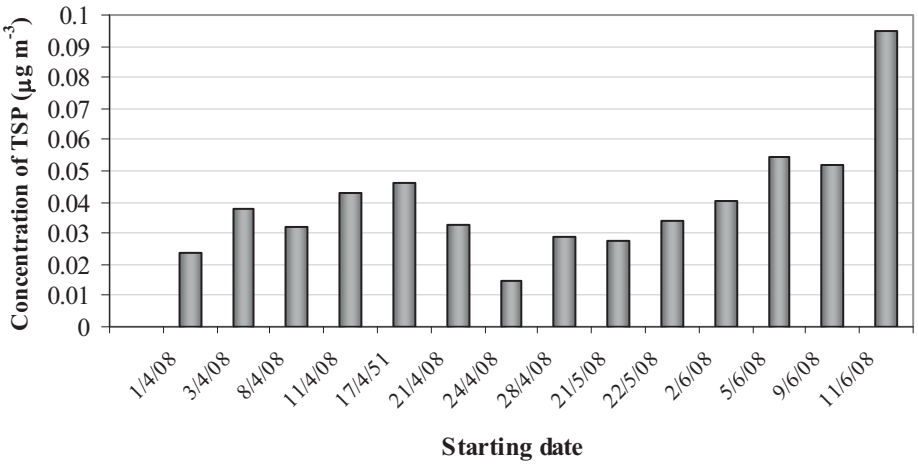

Figure 3. Variations of TSP during April to June 2008

rate and sampling time, the concentrations were given in µg/m^3. TSP varied from 0.0239–0.0950 µg/m^3 with average concentration of 0.0726±0.0191 µg/m^3 and PM_{10} ranged from 0.0142–0.0482 µg/m^3 with average concentration of 0.0195 ± 0.01 µg m^{-3}. The concentrations of TSP were approximately two times higher than PM_{10}; it might be because the strong wind could carry the coarse particles to reach the air sampler instead of settling down to the sea. The deviation amounts of both TSP and PM_{10} caused from the shipping activities, wind direction and the rainfall (Tables 1–2 and Figs. 3–4). There was no data from the end of April to the end of May because of heavy rain during this period. The amounts of both TSP and PM_{10} did not exceeded daily/annual standard values (less than 0.33 µg/m^3 for TSP and 0.12 µg/m^{-3} for PM_{10}).

However, the concentrations of both TSP and PM_{10} did not exceed Thailand standards for particulate

Table 2. Concentrations of PM_{10} during April to July 2008.

Collecting time		Air flow rate	Air volume	Particle weight	Concentration of PM10
Start	Stop	(m^3/min)	(m^3)	(µg)	(µg/m^3)
1/4/08	2/4/08	1.700	2539.80	0.0361	0.01421
8/4/08	9/4/08	1.700	2490.50	0.0441	0.01771
11/4/08	12/4/08	1.700	2448.00	0.0473	0.01932
17/4/08	18/4/08	1.700	2431.00	0.1114	0.04582
21/4/08	22/4/08	1.756	2528.64	0.0608	0.02404
24/4/08	25/4/08	1.756	2528.64	0.0331	0.01309
26/6/08	27/6/08	1.588	2286.72	0.0604	0.02641
27/6/08	28/6/08	1.588	2286.72	0.0553	0.02418
4/7/08	5/7/08	1.476	2140.20	0.0225	0.01051
5/7/08	6/7/08	1.476	2125.44	0.0233	0.01096
6/7/08	7/7/08	1.476	2154.96	0.0141	0.00654
7/7/07	8/7/08	1.476	2125.44	0.0353	0.01661
27/7/08	28/7/08	1.532	2206.08	0.0256	0.01160
28/7/08	29/7/08	1.476	2169.72	0.0398	0.01834
29/7/08	30/7/08	1.476	2125.44	0..692	0.03256

Note TSP and PM10 could not be collected in the same time from May to July because Air Sampler were broken down due to the strong wind and heavy rain.

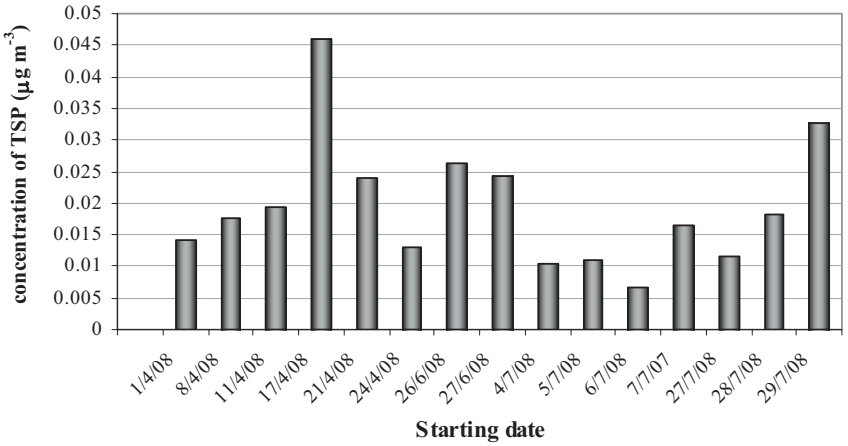

Figure 4. Variations of PM10 during April to July 2008

matters for 24 h and 1 year. Standard concentrations for TSP are 0.33 µg/m^3 (1 h) and 0.1 µg/m^3 (1 h). For PM_{10}, standard concentrations are 0.12 µg/m^3 (1 h) and 0.05 µg/m^3 (1 h) (Notification of National Environmental Board No. 10, 1995). Since the shipping activities occurred from ship to ship and generated high density of particulate matters (from visual inspection in Fig. 2) The concentrations of TSP and PM_{10} were very low compared with the standard values; since most of particles may settle down to the sea. The structure of particles from SEM indicated that most of particles were tapioca flour which was lighter than fertilizer and cement. These particles could be carried (by wind) far away from the emission sources and be finally collected by air sampler.

3.2. Microscopic structures and elemental compositions of particulate matters

The surface texture of fertilizer, tapioca flour pellet, cement from TSP and PM_{10} filter samples were illustrated in Figs. 5(a)–7(a), respectively. SEM micrograph of fertilizer standard was almost in spherical shape, the diameters were approximated around 10 micrometers. The amorphous shapes with almost 10 micrometers were found in tapioca flour pellet standard. The structure–like micrograph was obtained from cement standard. The spots were pointed the positions of incident X-ray with corresponding to EDS spectra of all samples given in the Figs. 5(b)-7(b), respectively.

The images and X-ray spectra of selected TSP and PM_{10} were given in Figs. 8–9. Both images provided spherical shape with 10 µm-diameter in TSP and less than 10 µm for PM_{10}. In agreement with the results from SEM observations and EDS analysis, it could be concluded that soil and rock dust represented a large proportion of particles adsorbed onto the glass and quartz filters. Particles from anthropogenic sources and marine environment were also found in filter samples. However, the EDS spectra of standard shows only C, N and O in Figs. 5(b)–7(b), whereas the elemental compositions of filter samples mostly contained oxygen and silicon and trace amount of sodium and aluminum. TSP also contained small amount of magnesium. Silicon might be originated from cruster or marine contributions. It could be summarized that of particulate matters from shipping activities may be the combination of fertilizer and tapioca flour.

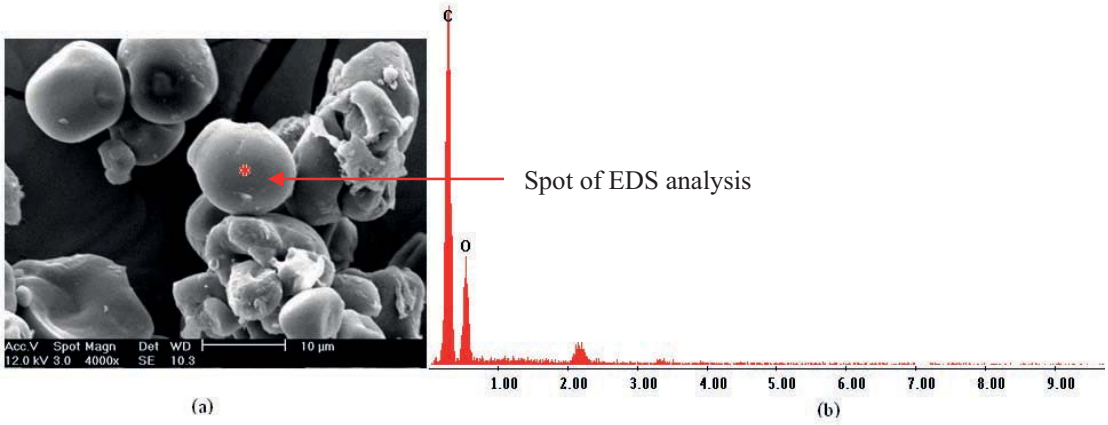

Figure 5. (a) SEM micrograph and (b) EDS profile of a fertilizer standard

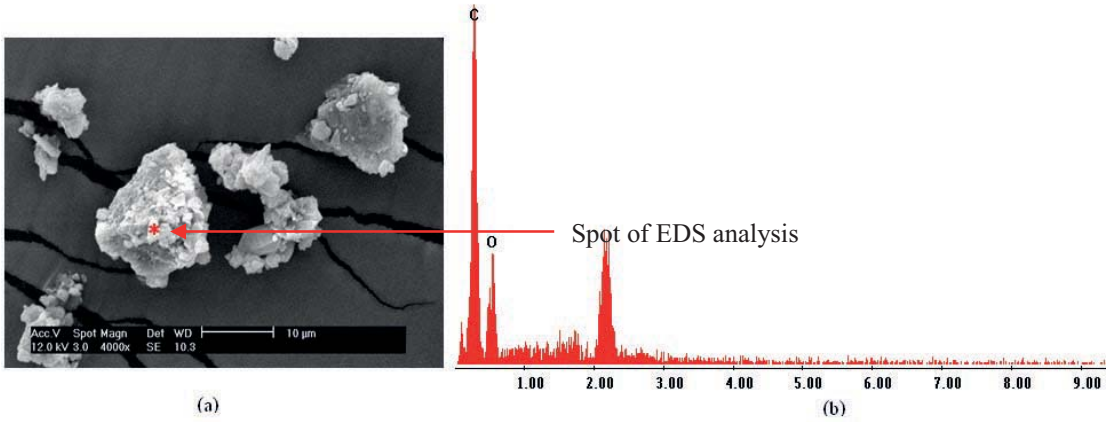

Figure 6. (a) SEM micrograph and (b) EDS profile of a tapioca flour pellet standard

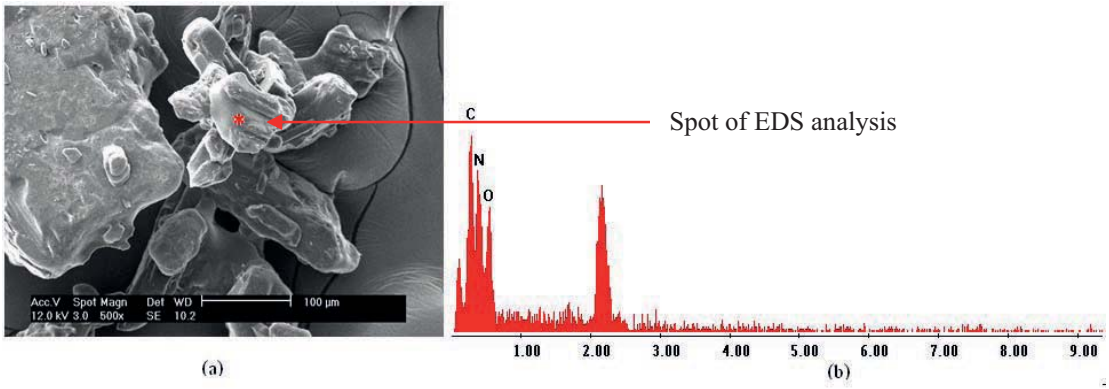

Figure 7. (a) SEM micrograph and (b) EDS profile of a cement standard

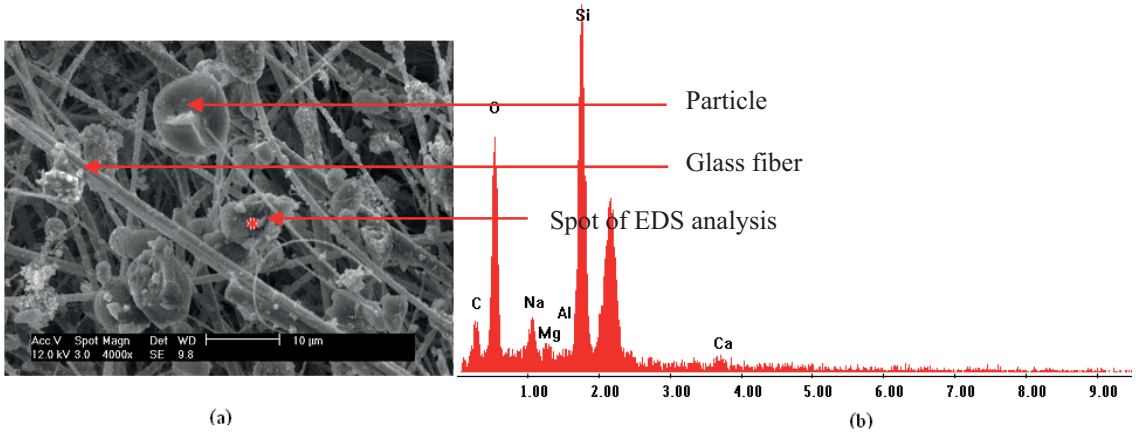

Figure 8. (a) SEM micrograph and (b) EDS profile of a TSP filter from a sampling site

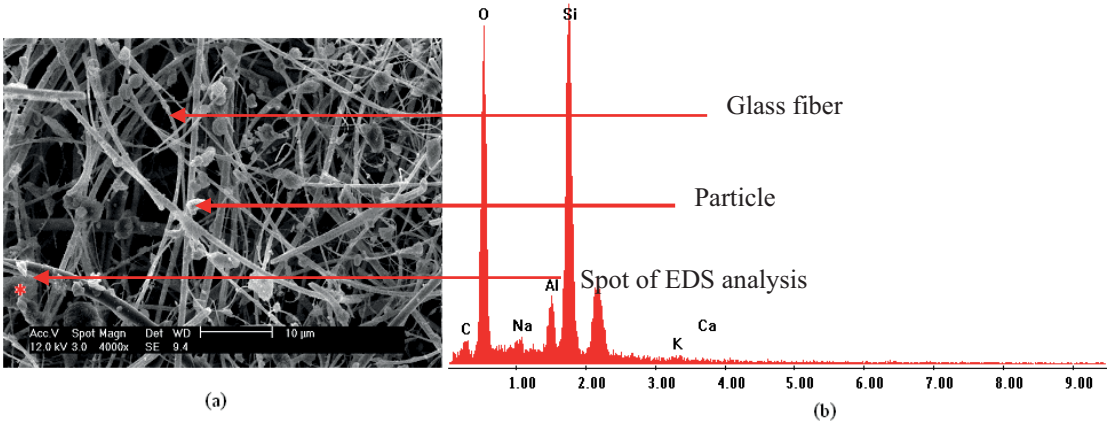

Figure 9. (a) SEM micrograph and (b) EDS profile of a PM_{10} filter from a sampling site

Table 3. Amounts of heavy metals in TSP

Collecting time		Amounts	Amounts of metals (ng/g.m^3)					
Start	Stop	of TSP (µg/m^3)	Zn	Ni	Cu	Hg	Pb	Se
1/4/08	2/4/08	23.90	0.2264	0.0008	0.0066	0.0017	0.0001	0.0005
3/4/08	4/4/08	38.00	0.0368	0.0010	0.0154	0.0013	0.0004	0.0005
8/4/08	9/4/08	32.00	0.1164	0.0005	0.0053	0.0012	0.0004	0.0004
11/4/08	12/4/08	42.90	0.0003	0.0005	0.0103	0.0008	ND	0.0005
17/4/51	18/4/51	46.10	0.0975	0.0007	0.0085	0.0010	0.0013	0.0011
21/4/08	22/4/08	32.40	0.1421	0.0002	0.0015	0.0016	0.0020	0.0022
24/4/08	25/4/08	15.00	0.0152	0.0002	0.0028	0.0004	0.0007	0.0005
28/4/08	29/4/08	28.70	0.0195	0.0013	0.0071	0.0004	0.0005	0.0005
21/5/08	22/5/08	27.50	0.0142	0.0007	0.0056	0.0004	0.0005	0.0004
22/5/08	23/5/08	34.20	0.0208	0.0008	0.0032	0.0003	0.0007	0.0005
2/6/08	3/6/08	40.60	0.0163	0.0006	0.0041	0.0005	0.0004	0.0004
5/6/08	6/6/08	54.50	0.0201	0.0002	0.0025	0.0003	0.0003	0.0004
9/6/08	10/6/08	51.80	0.0305	0.0056	0.0236	0.0003	0.0006	0.0004
11/6/08	12/6/08	95.00	0.0213	0.0003	0.0015	0.0004	0.0003	0.0004
Weighted mean concentrations (ng/g.m^3)			0.0476	0.0010	0.0071	0.0007	0.0006	0.0006
Weighted mean standard deviation (ng/g.m^3)			0.0831	1.29×10^{-4}	4.96×10^{-5}	1.32×10^{-7}	1.78×10^{-7}	1.53×10^{-7}

3.3. Chemical compositions of heavy metals in particulate matters

The amounts of selected heavy metals which were calculated in terms of mass of metals per mass of particle per unit air volume (ng/g.m^3) were seen in Table 3. Zinc was the most probable (0.9745 ng/g.m^3) among all metals in TSP. The weighted mean concentrations were 0.0476, 0.0010, 0.0071, 0.0007, 0.0006 and 0.0006 for Zn, Ni, Cu, Hg, Pb and Se, respectively.

Zinc strongly correlated with mercury and nickel strongly correlated with copper (seen in Table 4), it could be summarized that they originated from the same source. The strong correlation between lead and selenium could be the same source of the two elements. No correlation was found between any kind of heavy metals and amounts of TSP. It could be explained that only particulate matter emitted from shipping activities, heavy metals might originated from natural precursors or marine contributions.

Table 4. Correlation coefficients between amounts of TSP and various kinds of heavy metals

	TSP	Zn	Ni	Cu	Hg	Pb	Se
TSP	1						
Zn	0.0002	1					
Ni	0.0112	0.0110	1				
Cu	0.0005	0.0100	0.7169	1			
Hg	-0.0826	0.7140	-0.0565	0.00020	1		
Pb	-0.0213	0.0730	-0.0006	-0.01750	0.2141	1	
Se	-0.0164	0.1920	-0.0276	-0.03680	0.2679	0.8887	1

Table 3. Amounts of heavy metals in TSP

Collecting time		Amounts of	Amounts of metals (ng/g.m^3)					
Start	Stop	PM_{10} (µg/m^3)	Zn	Ni	Cu	Hg	Pb	Se
1/4/08	2/4/08	14.210	0.0049	0.0012	0.0011	0.0002	0.0004	0.0004
8/4/08	9/4/08	17.710	0.0166	0.0005	0.0045	0.0002	0.0003	0.0004
11/4/08	12/4/08	19.320	0.0076	0.0006	0.0086	0.0002	0.0004	0.0004
17/4/08	18/4/08	45.820	0.0136	0.0012	0.0062	0.0001	0.0008	0.0004
21/4/08	22/4/08	24.040	0.0055	0.0013	0.0019	0.0002	0.0003	0.0004
24/4/08	25/4/08	13.090	0.0017	0.0002	0.0004	0.0002	0.0006	0.0004
26/6/08	27/6/08	26.410	0.0028	0.0000	0.0005	0.0002	0.0003	0.0003
27/6/08	28/6/08	24.180	0.0149	0.0001	0.0009	0.0002	0.0004	0.0004
4/7/08	5/7/08	10.510	0.0135	0.0002	0.0006	0.0001	0.0002	0.0003
5/7/08	6/7/08	10.960	0.0060	0.0004	0.0003	0.0001	0.0002	0.0003
6/7/08	7/7/08	6.540	0.0067	ND	0.0003	0.0001	0.0003	0.0003
7/7/07	8/7/08	16.610	0.0163	0.0002	0.0007	0.0001	0.0003	0.0003
27/7/08	28/7/08	11.600	0.0858	0.0001	0.0006	0.0001	0.0002	0.0003
28/7/08	29/7/08	18.340	0.0065	0.0001	0.0007	0.0001	0.0002	0.0003
29/7/08	30/7/08	32.560	0.0039	0.0004	0.0005	0.0001	0.0004	0.0003
Weighted mean concentrations (ng/g.m^3)			0.0119	0.0005	0.0024	0.0001	0.0004	0.0004
Weighted mean standard deviation (ng/g.m^3)			1.46×10^{-4}	4.73×10^{-7}	1.31×10^{-5}	5.41×10^{-9}	1.55×10^{-7}	3.45×10^{-8}

Amounts of heavy metals in PM_{10}, Zn was dominated with 0.0119 ng/g.m^3, the others were almost in the same levels and much lower than in TSP. Heavy metals were easily adsorbed in coarse particles than ultra fine particles (Table 5). Similar to TSP, there were no correlations between PM_{10} and heavy metals. It is clarified that shipping activities were not the sources of heavy metals (Table 6).

4. Conclusion

Shipping activities pay an important role in marine and atmospheric impacts as well as human health. In Si Chang – Si Racha Bay shipping transportations, tapioca flour, fertilizer, cement and coal were the main particular items. The concentrations of TSP varied from 0.0239–0.0950 µg/m^3 with average concentration

Table 6. Correlation coefficients between amounts of PM_{10} and various kinds of heavy metals

	PM_{10}	Zn	Ni	Cu	Hg	Pb	Se
PM_{10}	1						
Zn	-0.0333	1					
Ni	0.1524	-0.0640	1				
Cu	0.1768	-0.0039	0.2002	1			
Hg	0.0037	-0.1050	0.0558	0.0774	1		
Pb	0.4287	0.1621	0.1965	0.2049	0.0772	1	
Se	0.0926	-0.0449	0.3689	0.3387	0.5469	0.3748	1

of 0.0726±0.0191 μg/m^3 and PM_{10} ranged from 0.0142–0.0482 μg/m^3 with average concentration of 0.0195±0.01 μg/m^3. The microscopic images indicated that particulate matters from tapioca flour were predominated in this port because of its fine particles and light weight. It could be carried by wind from the island to Si Racha Bay. The amounts of TSP and PM_{10} were lesser in dry season than in wet season. The amounts of both TSP and PM_{10} were not exceeded daily/annual standard values (less than 0.33 μg/m^3 for TSP and 0.12 μg/m^3 for PM_{10}) since the sampling station was located far from the emission sources.

The images from scanning Electron Microscope and the spectra from spots EDS were available for screening the shape, dimension and basic components of particulate matters. The particle images of samples compared with reference materials were able for roughly identification of the particles. The elemental compositions of particulate matter might be confirmed by XRF from the whole sample not only spot exposure.

Among the amounts of selected heavy metals (ng/g.m^3), zinc was the most probable among all metals (0.0975 ng/g.m^3) in TSP. The weighted mean concentrations (WMC±WSD) of Zn, Ni, Cu, Hg, Pb and Se were 0.0476±0.0831, 0.0010±1.29 x 10^{-4}, 0.0071±4.96 x 10^{-5}, 0.0007±1.32 x 10^{-7}, 0.0006±1.78 x 10^{-7} and 0.0006±1.53 x 10^{-7} ng/g.m^3, respectively. Zinc strongly correlated with mercury and nickel strongly correlated with copper, it might be originated from the same source. The strong correlation between lead and selenium could be the same origin of the two elements. No correlation was found between any kind of heavy metals and amounts of TSP. It could be explained that only particulate matter emitted from shipping activities, heavy metals might originated from natural precursors or marine contributions. Amounts of heavy metals in PM_{10} were 0.0119±1.46 x 10^{-4}, 0.0005±4.73 x 10^{-7}, 0.0024±1.31 x 10^{-5}, 0.0001±5.41 x 10^{-9}, 0.0004±1.55 x 10^{-7} and 0.0004±3.45 x 10^{-8} ng/g.m^3, respectively. Zn was dominated with 0.0119 ng/g.m^3, the others were almost in the same levels and much lower than in TSP. Heavy metals were easily adsorbed in coarse particles than the fine particles.

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