PERGAMON

Chemosphere 45 (2001) 791–799

CHEMOSPHERE

www.elsevier.com/locate/chemosphere

Particle size distribution and anion content at a traffic site in Sha-Lu, Taiwan

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Received 8 November 2000; received in revised form 23 April 2001; accepted 8 May 2001

Abstract

Ambient air particle concentrations were sampled by two total suspended particle (TSP) samplers, $PM_{10}/PM_{2.5}$ specific sampler and micro-orifice uniform deposit impactor (MOUDI) during July–October 2000 at a traffic sampling site in central Taiwan. The average TSP concentration (194 μg/m^3) was about a factor of two higher than that of the fraction <2.5 μm (93.2 μg/m^3). The mean level of the fraction <10 μm collected by MOUDI (93.2 μg/m^3) was about $1\frac{1}{2}$ times higher than that of the size class <2.5 μm (43.8 μg/m^3). Furthermore, this fraction showed a certain correlation with the TSP concentration. The particle size distribution was bimodal in the ambient air at the traffic site. The major peaks appear at particle diameters between 0.56–1.0 and 3.2–5.6 μm. The percentages of anions contained in TSP were 0.24% F^-, 13.7% Cl^-, 0.52% Br^-, 12.0% NO_3^-, 18.9% NO_2^-, and 54.6% SO_4^{2-}. The Cl^-, NO_2^-, and NO_3^- size distributions were all unimodal and the major peaks appeared at 3.2–5.6 μm. The SO_4^{2-} size distribution was bimodal, with major peaks at 0.32–0.56 and 3.2–5.6.

Keywords: Anion; Fine particles; Coarse particles; Size distribution

1. Introduction

Particulate air pollution has long been considered to be harmful to human health (Pedersen et al., 1999). Recent epidemiological studies have raised concerns over the potential health effects of airborne particulate matter (Dockery and Pope, 1994). In environmental epidemiology, effects of ambient particles on daily mortality, hospital admissions or lung function have been described for a large variety of atmospheric conditions (Brunekreef et al., 1995). Particulate matter is a mixture of components that are formed by a large variety of mechanisms associated with both natural and anthropogenic origins (Alves et al., 2000). Paved road dust on the surface of streets consists of a complex mixture of soil dust, deposited motor vehicle exhaust particles, tire dust, brake-lining wear dust, plant fragments, and other biological materials (Miguel et al., 1999). In all these studies the mass concentration of particles has been measured, either the total mass concentration of suspended particles (TSP) or the mass concentration of particles smaller than 10 or 2.5 μm in aerodynamic diameter ($PM_{2.5}$, PM_{10}). Studies in which measurements in different particle size ranges have been performed suggest stronger associations with health effects for PM_{10} and $PM_{2.5}$ than for TSP.

Legislation has introduced a separate criterion for ambient concentrations of fine particles, those below $PM_{2.5}$, to extend the existing PM_{10} standard. This allows also to distinguish between sources (Maricq et al., 1999). The size of ambient particles ranges from a few

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0045-6535/01/$ - see front matter
PII: S0045-6535(01)00129-1

nanometers to several micrometers. Particles up to 0.1 μm in diameter originate from either atmospheric gas-to-particle conversion or combustion processes. These particles coagulate rapidly to form larger particles. Particles in the size range above 1 μm originate from mechanical suspension processes in the lower troposphere (Tuch et al., 1997). Kleeman et al. (1999) described that four major classes of particles are observed: (1) large mineral dust and road dust particles that accumulate only small amounts of secondary aerosol products; (2) primary combustion particles (released initially from diesel vehicles, non-catalytic gasoline-powered vehicles and food processing; (3) sea-salt products; (4) sulfate-containing non-sea-salt background particles. It also indicated that paved road dust has a mass distribution, which peaks at particle diameters larger than 2.5 μm. Particles emitted from diesel engines and non-catalyst-equipped gasoline-powered engines are predicted to contribute significantly to the ambient particle size and composition distribution between 0.1 and 0.3 μm particle diameter.

The traffic sampling is influenced by many particle sources such as vehicle engines, road dust and resuspended dust. In this study, we used a micro-orifice uniform deposit impactor (MOUDI), a $PM_{10}/PM_{2.5}$ specific sampler and a PS-1 sampler to determine the relationship between particle concentrations and size distributions at a traffic sampling site in central Taiwan.

2. Experimental methods

2.1. Nomenclature

- $PM_{2.5}$ (PM10/2.5) – particle concentration collected by $PM_{10}/PM_{2.5}$ specific sampler with a size less than 2.5 μm.
- $PM_{2.5}$ (M) – particle concentration collected by MOUDI with a size less than 2.5 μm.
- PM_{10} (PM10/2.5) – particle concentration collected by $PM_{10}/PM_{2.5}$ specific sampler with a size less than 10 μm.
- PM_{10} (M) – particle concentration collected by MOUDI with a size less than 10 μm.
- Fine particle – particle diameter less than 2.5 μm.
- Coarse particle – particle diameter larger than 2.5 μm (TSP concentration minus $PM_{2.5}$).

2.2. Sampling program

In this study, the ambient particle concentrations were collected by a MOUDI, $PM_{10}/PM_{2.5}$ specific sampler ($PM_{2.5}$ and PM_{10}) and two PS-1 samplers (total suspended particle sampler). Further details are given below. The MOUDI sampler can distinguish different suspended particles with a diameter from 0.056 to 10 μm. The $PM_{10}/PM_{2.5}$ specific sampler can collect and differentiate between particle diameters lesser than 2.5 μm ($PM_{2.5}$) and lesser than 10 μm (PM_{10}). The PS-1 sampler collects suspended particles less than 50 μm. Particle concentrations were determined in the period July–October 2000 (Table 1). All the sampling groups were operated under sunny and cloudy days, thus there was no precipitation in any of the sampling groups. The MOUDI was set on the sidewalk near Chung-Chi Road; the $PM_{10}/PM_{2.5}$ specific sampler was placed on the pedestrian overpass (5 m high) over Chung-Chi Road and two PS-1 samplers (TSP1 and TSP2) were positioned on the safety island of Chung-Chi Road. TSP1 was operated at 24-h intervals and TSP2 at 12-h intervals to characterize differences between the day and night periods during the sampling time. The sampling site was located at the Chung-Chi Road in front of the Hungkuang Institute of Technology (HKIT), Sha-Lu, Taichung, Taiwan, about 20 km from the sea (Taiwan Strait). HKIT has 10,000 students and about 3000 vehicles per hour pass the sampling site.

2.3. MOUDI sampler

MOUDI (Model 100, MSP) is a cascade impactor intended for general purpose aerosol sampling. Each stage on the MOUDI consists of an impaction plate above and a nozzle plate below. The use of micro-orifice nozzles extends the sizes of the lowest stage to 0.056 μm without creating an excessive pressure drop across the impactor stages. By rotating alternate stages of the MOUDI, deposits are distributed uniformly on the circular impaction areas of about 1 inch in diameter. The flow rate is 30 l/min and cut-size diameter ranges from 18 μm at the first stage to 0.056 μm at the last. The available cut-size diameters were 18, 5.6, 3.2, 1.8, 1.0, 0.56, 0.32, 0.18 and 0.1 and 0.056 μm) (choice of eight). A Teflon filter (Omega Specialty Instrument CAT#M-0454700, 0.45 μm filter) coated with silicone grease (ITW Fluid Products Group) was used as the surrogate surface and absorbent grease in this study. The MOUDI was set on the sidewalk near Chung-chi Road.

2.4. $PM_{10}/PM_{2.5}$ specific sampler

The Model 310 $PM_{10}/PM_{2.5}$ specific Air SamplerTM (USATM) is a general-purpose air sampler for atmospheric aerosol sampling and for mass concentration. Air is sampled at 300 l/min from the ambient atmosphere through an omni-directional, cylindrical inlet. Particles greater than 10 μm aerodynamic equivalent diameter are removed from the sampled air stream by the PM10 classifier and discarded. Particles less than 10 μm flow to the $PM_{2.5}$ classifier downstream. Particles in the 2.5–10 μm range are collected on a 62 mm × 165 mm filter and those smaller than 2.5 μm are collected on a

Table 1
Sampling dates, equipment and meteorological conditions during particle collection

Date	Sampling devices	Wind speed (m/s)	Weather	Precipitation
7/20–7/21	TSP1, TSP2, $PM_{10}/PM_{2.5}$ specific sampler	3.31	Sunny	None
7/21–7/22	TSP1, TSP2, $PM_{10}/PM_{2.5}$ specific sampler	2.57	Sunny	None
7/23–7/24	TSP1, TSP2, $PM_{10}/PM_{2.5}$ specific sampler	1.87	Cloudy	None
7/26–7/27	TSP1, TSP2, $PM_{10}/PM_{2.5}$ specific sampler	3.31	Sunny	None
7/27–7/28	TSP1, TSP2, $PM_{10}/PM_{2.5}$ specific sampler	2.45	Cloudy	None
7/28–7/29	TSP1, TSP2, $PM_{10}/PM_{2.5}$ specific sampler	2.75	Sunny	None
8/2–8/3	TSP1, TSP2, MOUDI, $PM_{10}/PM_{2.5}$ specific sampler	4.03	Sunny	None
8/3–8/4	TSP1, TSP2, MOUDI, $PM_{10}/PM_{2.5}$ specific sampler	1.14	Sunny	None
8/15–8/16	TSP1, TSP2, MOUDI, $PM_{10}/PM_{2.5}$ specific sampler	2.78	Sunny	None
8/16–8/17	TSP1, TSP2, MOUDI, $PM_{10}/PM_{2.5}$ specific sampler	2.54	Cloudy	None
8/17–8/18	TSP1, TSP2, MOUDI, $PM_{10}/PM_{2.5}$ specific sampler	3.01	Cloudy	None
8/18–8/19	TSP1, TSP2, MOUDI, $PM_{10}/PM_{2.5}$ specific sampler	3.14	Sunny	None
9/30–10/1	TSP1, TSP2, MOUDI, $PM_{10}/PM_{2.5}$ specific sampler	5.61	Sunny	None
10/02–10/03	TSP1, TSP2, MOUDI, $PM_{10}/PM_{2.5}$ specific sampler	3.99	Sunny	None
10/23–10/24	TSP1, TSP2, MOUDI, $PM_{10}/PM_{2.5}$ specific sampler	4.22	Sunny	None

200 mm × 250 mm final filter. The filters used in this study were Teflon filters (Whatman International, Cat. No. 1882886). Teflon filters showed the least artifact errors in a previous study (Fang et al., 1997a,b). The $PM_{10}/PM_{2.5}$ specific sampler was set on the pedestrian crosswalk about 5 m above Chung-Chi Road.

2.5. PS-1 sampler

The PS-1 (GPS1 PUF Sampler, General Metal Work) is a complete air-sampling system designed to simultaneously collect suspended airborne particles at a flow rate up to 280 l/min. The flow rate was adjusted to 200 l/min in this study. Teflon filter (Toyo Roshi Kaisha, Lot No. 1008075) with a cut-off of 10.3 mm was used to filter the suspended particles in this study. The two PS-1 samplers were set on the safety island near Chung-Chi Road. The distance between the two samplers was about 10 m.

2.6. Wind sensor

Wind speed and direction was measured with a Model 034 Wind Sensor which combines wind speed and wind direction measurements in a single unit. Model 034 is designed to continuously operate in adverse environments at wind speeds as high as 110 mph. The wind sensor was set on the over-pass across the Chung-Chi Road near the $PM_{10}/PM_{2.5}$ specific sampler.

2.7. Anion analysis

After sampling, the samples from PS-1, $PM_{10}/PM_{2.5}$ specific sampler and MOUDI were put into a 100 ml glass bottle. 10 ml of de-ionized water (specific conductivity >18 μs/cm) was added to the bottom before ultrasonic processing for 30 min. About 0.3 ml solution was then injected into an ion chromatography (DIONEX DX-100 Ion Chromatography) with the DIONEX 4500I anion column for anion analysis. The elute was 1.8 mM Na_2CO_3 + 1.7 mM $NaHCO_3$ and the flow rate was adjusted to 1.2 ml/min. The anions analyzed in this study were F^-, Cl^-, Br^-, NO_3^-, NO_2^- and SO_4^{2-}. At least 10% of the samples were analyzed in spiking with a standard anion solution (Merck INC) for recovery test. The recovery varied between 90% and 110% in this study.

3. Results and discussion

3.1. Particle concentrations in ambient air

The particle concentrations determined at the traffic site is given in Table 2. Values for TSP1 (24 h) ranged from 144–293 μg/m^3 (average 194 μg/m^3). TSP2 varied between 167–324 μg/m^3 (average 215 μg/m^3) for day time, from 146–290 μg/m^3 (average 192 μg/m^3) for night time resulting in calculated 24-h levels of 157–307 μg/m^3 (average 204 μg/m^3). Night-time levels were about 10% lower than for day time. This is due to less traffic at night. The calculated 24-h levels from TSP2 were in average ca. 5% higher than the data from TSP1. The most likely explanation is that traffic during the night sampling time was less than during the day period. The mean TSP2 concentration was about 1.05 times higher than the mean TSP1 concentration. The reasonable explanation is that the long sampling time for TSP1 (24 h) at the traffic site caused filter clogging and reduced the flow rate, thus underestimating the total suspended particle concentrations in the ambient air.

Table 2
Particle concentrations determined at the traffic site in Sha-Lu in μg/m^3

Date	TSP 1	TSP 2			$PM_{2.5}$ (PM10/2.5)	$PM_{2.5–10}$ (PM10/2.5)	$PM_{2.5}$ (M)	$PM_{2.5–10}$ (M)
		Day	Night	Average				
7/20–7/21	147	169	147	158	33.6	12.3	–	–
7/21–7/22	187	200	190	195	47.5	15.6	–	–
7/23–7/24	217	237	213	225	57.7	25.1	–	–
7/26–7/27	179	197	177	187	30.2	18.3	–	–
7/27–7/28	205	241	201	221	54.4	23.1	–	–
7/28–7/29	144	167	146	157	28.3	15.5	–	–
8/2–8/3	278	301	268	284	59.4	22.6	83.0	57.8
8/3–8/4	161	182	173	178	32.1	18.7	107	52.6
8/15–8/16	165	200	176	188	29.4	13.3	69.9	27.3
8/16–8/17	194	210	180	195	35.5	15.7	75.2	40.5
8/17–8/18	200	216	191	204	48.9	20.4	58.6	36.3
8/18–8/19	166	191	169	180	33.4	16.6	33.0	38.7
9/25–9/26	293	324	290	307	72.6	23.4	220	58.2
10/2–10/3	203	210	188	199	46.3	18.3	82.1	24.3
10/16–10/17	169	183	169	176	12.2	33.1	110	58.7
Average	194	215	192	204	41.4	19.5	93.2	43.8

TSP: total suspended particles.
$PM_{2.5}$ (PM10/2.5): particle concentration collected by $PM_{10}/PM_{2.5}$ specific sampler with a size less than 2.5 μm.
$PM_{2.5}$ (M): particle concentration collected by MOUDI with a size less than 2.5 μm.
PM_{10} (PM10/2.5): particle concentration collected by $PM_{10}/PM_{2.5}$ specific sampler with a size less than 10 μm.
PM_{10} (M): particle concentration collected by MOUDI with a size less than 10 μm.

The $PM_{2.5}$ (PM2.5/10) concentrations ranged from 28.3–72.6 μg/m^3 (average 41.4 μg/m^3) and those of the MOUDI were from 33.0–220 μg/m^3 (average 93.2 μg/m^3). $PM_{2.5–10}$ levels of PM2.5/10 were within 12.3–25.1 μg/m^3 (average 19.5 μg/m^3) and of the MOUDI between 27.3–58.7 μg/m^3 (average 43.8 μg/m^3). The significantly higher level ($P < 0.05$, two-sided t-test) of about a factor of 2 is caused by the height difference of 5 m between the sampling sites. Nevertheless, the fine particle concentration $PM_{2.5}$ has to be considered as high. The proposed reasons are that there were much more particles less than 10 μm resuspended and exhausted by vehicles on the ground level and these particles were collected immediately by the MOUDI sampler just near the Chung-Chi Road. However, these high-concentration particles might coagulate and condense to larger particles (Hinds, 1999). Thus, these larger particles cannot be collected by the $PM_{10}/PM_{2.5}$ specific sampler on the 5-m height pedestrian overpass.

In the following only the MOUDI data are discussed as a worse-case scenario. At the traffic sampling site, the coarse particles (diameter larger than 2.5 μm) were 62.3–198 μg/m^3 (average 115 μg/m^3). The fine particles (diameter less than 2.5 μm) were 33.0–220 μg/m^3 (average 93.2 μg/m^3) (Table 3). The particles >10 μm were 0.7–136 μg/m^3 (average 61.2 μg/m^3). The

Table 3
Coarse and fine particle concentrations in ambient air at Sha-Lu, Taiwan in μg/m^3

Date	TSP	Coarse particle $\{TSP - PM_{2.5}(M)\}$	$PM_{2.5}$ (M)	$PM_{10}\{PM_{2.5}(M) + PM_{2.5–10}(M)\}$	Particle > 10 μm
8/2–8/3	281	198	83.0	146	136
8/3–8/4	160	62.3	107	160	0.7
8/15–8/16	177	107	69.9	97.2	79.3
8/16–8/17	195	120	75.2	116	79.2
8/17–8/18	202	143	58.6	94.9	107
8/18–8/19	173	140	33.0	71.8	101
9/25–9/26	300	80.3	220	278	22
10/2–10/3	201	119	82.1	107	94.6
10/16–10/17	173	62.3	110	169	3.6
Average	199	115	93.2	138	61.2

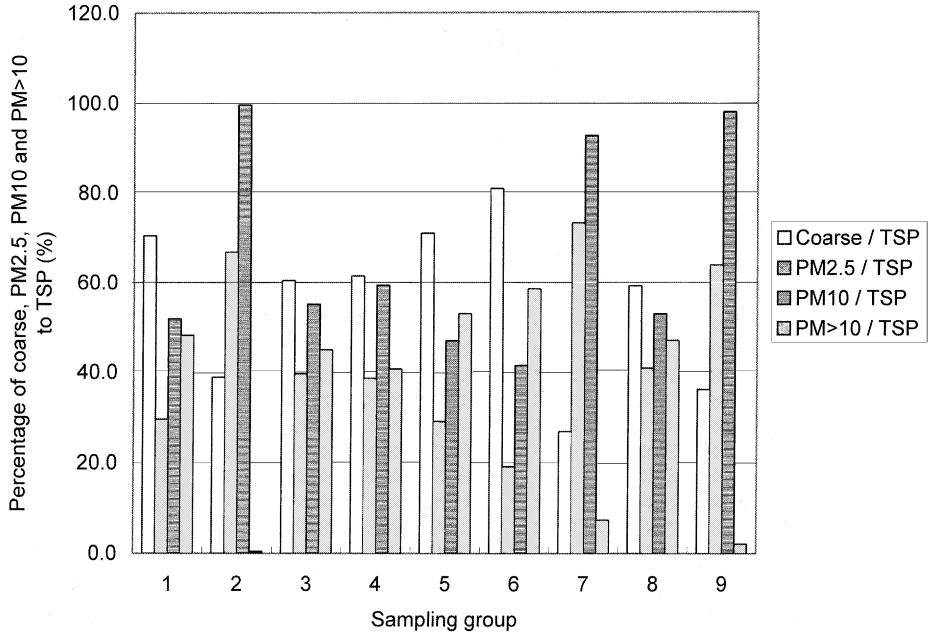

Fig. 1. Relative concentrations of coarse, $PM_{2.5}$, PM_{10} and >10 μm particles at a traffic site in Sha-Lu, Taiwan.

average coarse particle concentration was about 20% higher than that of the mean fine particle concentration and caused a high coarse particle burden in ambient air.

The percentages of coarse and fine particles to TSP are shown in Fig. 1. The percentages of coarse (M)/TSP ranged from 26.8% to 80.9% and averaged 56.1%. The percentages of PM_{10} (M)/TSP ranged from 41.5% to 99.5% and averaged 66.4%. The percentages of $PM_{2.5}$ (M)/TSP ranged from 29.0% to 73.2% and averaged 44.5% in the ambient air. This indicated that $PM_{2.5}$, $PM_{2.5-10}$ and particle diameter larger than 10 μm occupied 44.5%, 21.9% and 34.2%, respectively of the total suspended particles (TSP) at the traffic sampling site in central Taiwan.

The correlation between the $PM_{2.5}$ fraction compared to TSP and PM_{10} fraction is displayed in Fig. 2. $PM_{2.5}$ correlated clearly with PM_{10} ($PM_{2.5} = 0.84\ PM_{10} - 22.5$,

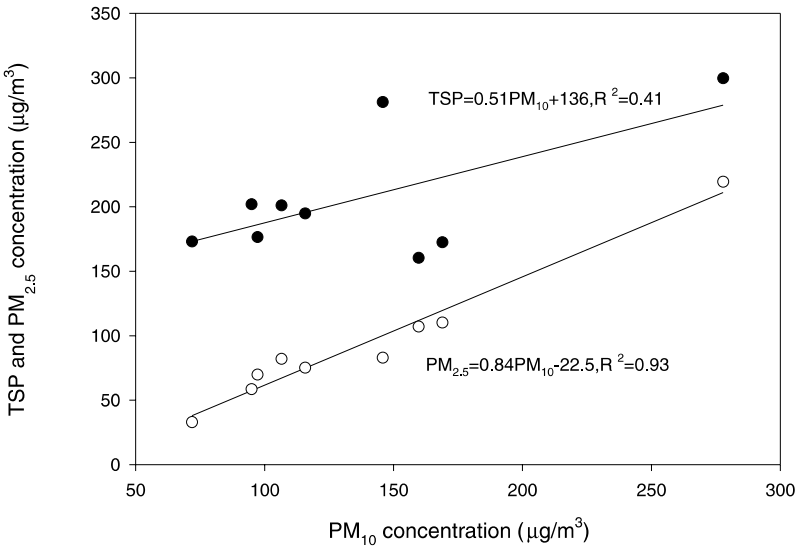

Fig. 2. Correlation between quantity of PM_{10} and TSP or $PM_{2.5}$.

Table 4
Mean concentration of particles in ambient air at Sha-Lu compared to different sites in Canada (Brook et al., 1997)

Sampling site	This study	Vancouver	Watertown	Victoria
TSP	199	–	–	–
$PM_{2.5}$	93.2	15.5	17.4	11.5
$PM_{2.5–10}$	44.4	9.2	17.4	6.3

Results are given in μg/m^3.

$r^2 = 0.93$) but not TSP ($r^2 = 0.41$). A Canadian study (Brook et al., 1997) reported an r^2 of 0.56 for the relation PM_{10} and TSP and of 0.63 for PM_{10} and $PM_{2.5}$, which is fairly consistent with this study. However, overall levels were in general higher in Sha-Lu, Taiwan than in Canada (see Table 4 for a comparison).

3.2. Size distribution of suspended particles

The average size distribution of suspended particles in the traffic site is shown in Fig. 3. The particle size distribution was bimodal in the ambient air at the traffic site. The major peaks in the particle diameter occurred between 0.56–1.0 and 1.0–10 μm in the ambient air.

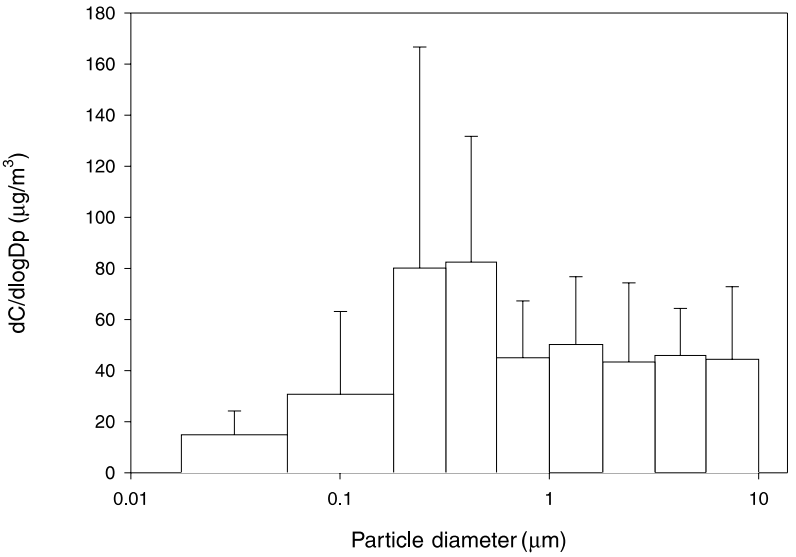

Fig. 3. Mean particle size distribution and S.D. found at a traffic site in Sha-Lu, Taiwan.

Table 5
Average anion concentrations in different particle size fraction and S.D.'s for a traffic site at Sha-Lu, Taiwan

Particle diameter (μm)	F^- (±S.D.)	Cl^- (±S.D.)	Br^- (±S.D.)	NO_2^- (±S.D.)	NO_3^- (±S.D.)	SO_4^{2-} (±S.D.)
	MOUDI ($N = 9$)					
10	<5.0	259 (±12.4)	<5.0	87 (±17.4)	170 (±18.3)	250 (±32.5)
5.6	<5.0	462 (±22.4)	<5.0	154 (±21.2)	243 (±22.4)	295 (±18.9)
3.2	<5.0	643 (±20.2)	<5.0	451 (±32.0)	721 (±154)	702 (±54.5)
1.8	<5.0	133 (±17.7)	<5.0	201 (±38.1)	388 (±20.8)	278 (±20.8)
1.0	<5.0	59 (±6.4)	<5.0	96 (±10.1)	198 (±16.3)	1230 (±239)
0.56	<5.0	38 (±6.0)	<5.0	88 (±7.1)	192 (±30.6)	2110 (±144)
0.32	<5.0	<5.0	<5.0	87 (±15.5)	173 (±17.5)	1750 (±120)
0.18	<5.0	<5.0	<5.0	59 (±10.7)	92 (±8.9)	862 (±36.5)
0.056	<5.0	<5.0	<5.0	45 (±6.7)	48 (±6.4)	253 (±29.7)
	$PM_{10}/PM_{2.5}$ specific sampler ($N = 15$)					
2.5–10	35 (±6.1)	1270 (±145)	56 (±5.5)	897 (±58.1)	1970 (±161)	2050 (±158)
<2.5	42 (±4.2)	256 (±24.0)	57 (±6.1)	465 (±28.7)	870 (±35.2)	5420 (±388)
	TSP ($N = 15$)					
<50	45 (±6.5)	2580 (±341)	97 (±7.1)	2250 (±241)	3550 (±441)	10300 (±548)

Results are given in ng/m^3.

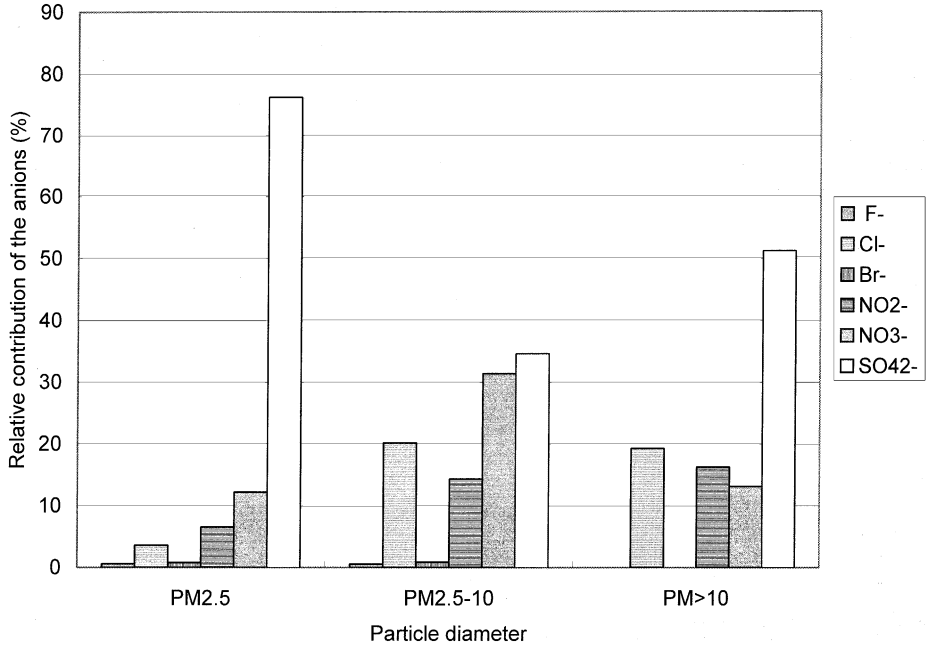

Fig. 4. Relative contribution of the measured anions.

3.3. Anion concentration versus size contribution

Anion concentrations in the different particle size distributions are given in Table 5 together. Fluoride and chloride levels were below the detection limit in the fractions from the MOUDI sampler (detection limit is 5 ng/m^3). In general, the concentration distribution order was $SO_4^{2-} > NO_3^- > Cl^- > NO_2^- > Br^- > F^-$ for $PM_{2.5}$ and distribution order as $PM_{2.5-10}$. This is consistent with the data for anion size distribution obtained below.

The average relative contributions of the measured anions in the $PM_{2.5}$, $PM_{2.5-10}$, $PM_{>10}$ fractions as well as TSP are given in Fig. 4. As can be seen, the relative contribution of chloride increased with increasing particle diameter and was abundant in particles of size 2.5 μm. F^- and Br^- were below the detection limits in particles >10 μm. The contribution of Br^- was about the same over the whole size range. Furthermore, levels of NO_3^- were higher for coarser particles (up to ca. 3 μm) while the opposite was observed for SO_4^{2-} where increased levels down to a size of 0.3 μm were observed. The present study (Hidy et al., 2000) sampled on nine healthy, non-smoking individuals living and working in the metropolitan area of Birmingham: the average Cl^-, NO_3^- and SO_4^{2-} in $PM_{2.5}$ concentrations were 45, 71 and 3870 ng/m^3, respectively. The results were not consistent with this study. This is because the sampling site in this study is located at the heavy traffic highway and about 30 km away from the sea. The exhaust NO_x from vehicles could be transferred to NO_3^- and NO_2^-. In addition, the nitrate-containing sea salt will also increase the Cl^-, NO_2^- and NO_3^- concentrations in this study.

The anion distribution in relation to particle size is shown in Fig. 5. The Cl^-, NO_2^- and NO_3^- ions dominated in the size range 3.2–5.6 μm, while a distribution with a maximum in the range 0.32–0.56 μm and a second smaller increase for the range 3.2 μm were found. The present study (Liu et al., 2000) indicated that the total nitrate size distribution dominated in the size range 0.2–2.6 μm, which was consistent with this study.

4. Conclusion

Ambient air particle concentrations were sampled by two TSP samplers, $PM_{10}/PM_{2.5}$ specific sampler and micro-orifice uniform deposit impactor (MOUDI), during July–October 2000 at a traffic sampling site in central Taiwan. The results indicate that the average 24-h TSP concentration is 194 μg/m^3. In addition, the result also indicates that the coarse particle concentration ($PM_{2.5-10}$) is about 20% higher than the fine particle concentration ($PM_{2.5}$) in the traffic site. The anion contents in the traffic-site particles are much higher than other sampling site. The maximum size distribution of anions is 0.32–0.56 μm and a second smaller increase for the range 3.2 μm. The combining of exhaust vehicles' NO_x with the nitrate-containing sea salt is the major reason leading to the high concentration of these anion species.

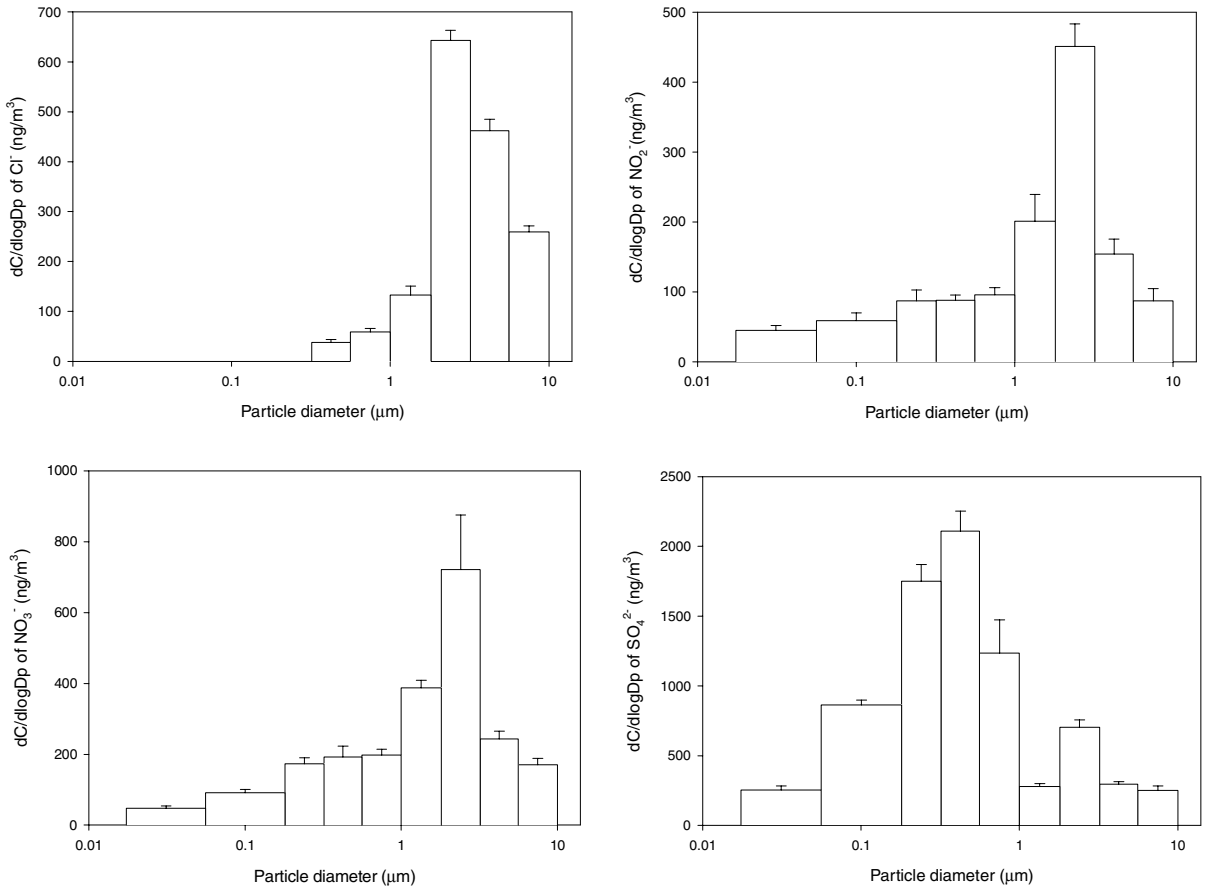

Fig. 5. Anion concentration in relation to particle size in samples collected by the MOUDI sampler.

Acknowledgements

The authors gratefully acknowledge the National Science Council of the R.O.C. (Taiwan) for the financial support under project no. NSC 89-2211-E-241-007.

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