2. Rain Acidity in Hong Kong and Its Trend

‘Acid Rain’ is a term describing the acids and potential acid generators in atmospheric wet and dry deposition. Acidic wet deposition in form of rain, mist, fog and snow is always due to the presence of un-neutralized anions from natural gases. In nature, the presence of atmospheric CO_2 gives wet deposition a theoretical equilibrium pH of 5.6. The pH of 5.6, however, cannot be considered as a background level for unpolluted air due to some other acidity of natural origin such as volcanic eruption. Instead, the rain pH recorded in remote and pristine areas is referenced as the natural rain pH, which is of a value of 5.0 (Howells, 1995; Radojevic and Harrison, 1992). It is the anthropogenic emissions of acid-forming gases beyond the natural acidity that leads to acid rain. This definition of acid rain will be adopted throughout the present study.

2.1 Acid rain data

When rainwater chemistry is concerned, statistical analysis is primarily focused on the wet deposition data. This is because the composition of wet deposition is more representative than that of dry deposition. Wet deposition includes all the gaseous, particulates, oxidized and partially oxidized products in its matrix. Gases are usually absent from the dry deposition. Composition of dry deposition may also have greater variation and depends heavily on the meteorological conditions as well as the physical characteristics such as deposition velocity and surface friction.

Data of ionic composition and concentration of the rainwater from the two acid rain monitoring stations (Kwun Tong and Central/Western) was provided by the Hong Kong Environmental Protection Department (HKEPD). The data sets covered the period of 1993 to 1999. A total of eleven ionic species were included for the analysis (H^+, Ca^+, Mg^+, Na^+, NH_4^+, K^+, Cl^-, NO_3^-, SO_4^{2-}, formate and acetate).

Data description

The volume weighted mean (vw-mean) and mode pH of the rainwater collected in Kwun Tong and Central/Western was summarized in Table 2-1. The volume weighted mean rain pH was in the range of 4.48 to 5.39 and the mode pH from 4.21 to 6.50 from 1993 to 1999. Acid rain phenomenon is thus observed in Hong Kong.

Table 2-1. Summary of the rain pH data collected by the EPD's acid rain monitoring stations

Year	Station	Frequency	vw-Mean pH	Mode pH
1993	Kwun Tong	28	4.78	4.41
	Central/Western	24	4.72	4.21
1994	Kwun Tong	21	4.88	4.90
	Central/Western	17	4.86	5.10
1995	Kwun Tong	19	5.39	6.50
	Central/Western	18	4.74	4.30
1996	Kwun Tong	22	4.85	4.90
	Central/Western	21	4.84	4.40
1997	Kwun Tong	27	5.13	4.80
	Central/Western	19	5.07	4.70
1998	Kwun Tong	29	4.60	4.70
	Central/Western	28	4.48	4.40
1999	Kwun Tong	32	4.87	4.60
	Central/Western	28	4.75	4.40

Remarks:

Frequency: assume 52 weeks per annum; vw-mean pH = $\frac{\sum(rainvolume * pH)}{\sum rainvolume}$

Mode pH: the pH recorded for most rainfall in the year

Data screening

Prior to statistical analysis, data was screened to eliminate extremes and abnormalities. Initial screening was through charge balance between the cations (H^+, Ca^+, Mg^+, Na^+, NH_4^+, K^+) and the anions (NO_3^-, SO_4^{2-}, formate and acetate) assuming the neutrality of charge of all cations and anions as in ideal condition. To allow slight discrepancy from reality of the neutrality, a cut-off interval of $\pm$ 20% was adopted in the present study in accordance with other reported studies (Fujita *et al.*, 2000; Ayers and Yeung, 1996; USEPA, 1994). Results of the screening on the data-points were summarized in Table 2-2. A 73.0% and 75.5% of the data is retained for Kwun Tong and Central/Western, respectively. Data quality is good as a whole according to the charge neutrality.

Table 2-2. Summary of the results of data screening, numbers represent discrete data points and percentage of the total raw data points is given in brackets.

	Data Points (1993-1999)	
	Kwun Tong (KT)	Central/Western (CW)
Raw	178 (100%)	155 (100%)
Charge balance ($\pm$ 20%)	130 (73.0%)	117 (75.5%)

In facilitating the statistical analysis to be followed, the ion concentration was all converted to μeq/l with respect to its charge. Results of the mean (arithmetic) and its standard error (S.E.) were summarized in Table 2-3. The volume-weighted means were also listed for reference. In general, the mean values of the raw and screened data in both stations showed no statistical differences as the 95% confidence limit (1.96 S.E.) overlapped with each other. This, in accordance with the ion balance, suggested the quality of the data is very good.

2.2 Statistical analysis

2.2.1 Factor analysis

The factor analysis serves for two purposes. Firstly, the dimensionality of the problem could be reduced from the original 11 variables into key factors (groups) by substituting a limited number of linear combinations. Secondly, the factor analysis could interpret differences between sites in terms of the factors (groups). A commercial statistics package, STATISTICA was employed in carrying out the analysis.

Table 2-3. The mean and volume-weighted mean of the ion concentration (μeq/l) of wet deposition in the Year 1993 to 1999 collected in (A) Kwun Tong and (B) Central/Western.

(A)	Kwun Tong raw (n=178)		Kwun Tong screened (n=130)	
	Mean ± SE	vw-mean	Mean ± SE	vw-mean
Anion				
H^+	34.275 ± 3.616	24.625	26.465 ± 2.441	17.930
Ca^{2+}	38.358 ± 8.437	15.692	39.200 ± 9.515	14.760
Mg^{2+}	12.898 ± 2.098	7.085	14.761 ± 2.422	7.907
Na^+	62.565 ± 8.420	39.550	67.750 ± 9.271	41.255
K^+	7.454 ± 0.444	6.742	7.453 ± 0.482	6.719
NH_4^+	34.939 ± 3.333	18.168	36.281 ± 3.526	18.408
Cation				
Cl^-	70.989 ± 9.508	44.923	77.229 ± 10.47	47.746
SO_4^{2-}	74.065 ± 8.564	39.578	77.552 ± 9.603	39.330
NO_3^-	24.283 ± 3.387	10.967	25.218 ± 3.720	10.832
Formate	4.570 ± 0.112	4.377	4.713 ± 0.120	4.488
Acetate	3.452 ± 0.055	3.389	3.528 ± 0.050	3.472

B	Central/Western raw (n=155)		Central/Western screened (n=117)	
	Mean ± SE	vw-mean	Mean ± SE	vw-mean
Anion				
H^+	46.147 ± 5.065	29.847	37.821 ± 4.422	24.110
Ca^{2+}	25.087 ± 4.081	11.354	28.184 ± 4.614	12.464
Mg^{2+}	13.782 ± 2.470	7.957	15.951 ± 2.804	8.993
Na^+	71.824 ± 11.44	43.142	80.076 ± 12.62	48.950
K^+	7.282 ± 0.464	6.577	7.592 ± 0.528	6.756
NH_4^+	30.401 ± 3.341	15.176	32.614 ± 3.709	16.019
Cation				
Cl^-	83.046 ± 12.85	50.621	91.819 ± 14.19	56.871
SO_4^{2-}	69.749 ± 6.592	39.314	75.013 ± 7.397	41.045
NO_3^-	23.585 ± 3.336	10.097	25.973 ± 3.776	10.554
Formate	4.543 ± 0.126	4.368	4.644 ± 0.136	4.478
Acetate	3.493 ± 0.102	3.378	3.561 ± 0.111	3.460

The mean was the arithmetic mean of all data points with SE, the standard error of mean. The vw-mean concentrations of ions were calculated by $[X]_{vw} = \sum_i (V[X]) / \sum_i V$, where the index *i* run over the entire set of sample concentration [X] and the respective rain volume.

The principal component analysis (PCA) combined with the varimax rotation was employed for the factor analysis. In this study, a factor is signified in the analysis if its corresponding eigenvalue is larger than 1. Factors with an eigenvalue less than 1 were also examined but could not be interpreted. The eigenvectors obtained in this way are determined uniquely only up to rotation. Varimax rotation chooses eigenvectors such that they minimize the unique variation in the sense of least squares. Theoretically, varimax rotation is justified over oblique (non-orthogonal) rotations because of the model assumption that the factors are independent. The rotation amplifies the difference in factor loadings, enhancing high loadings to become higher and low ones to lower in magnitude.

Tables 2-4 and 2-5 summarized the results of the factor analysis for the rainwater ions collected at Kwun Tong (KT) and Central/Western (CW), respectively. Three loading factors are identified in both stations regardless of data screened or not. The cumulative variance accounted for by the three factors is 85.99% and 90.68% of the total in the raw data set for KT and CW, respectively. This increases to 87.29 and 92.93%, correspondingly, after screening, improving the raw data set by 1.30 and 2.25 %, respectively.

Kwun Tong (1993 – 1999, Table 2-4)

Three Factors are identified from the original data set in KT accounting for 86% of the total variance. Factor 1 by itself accounts for 59.8% of the total variance in the original data set. However, it is poorly resolved and comprised a total of 8 ionic species - NH_4, SO_4, NO_3, Ca, Mg, Na, Cl and K (charge omitted hereafter). Factor 2 consists of formate and acetate, which are members of volatile organic carbons and accounts for 15.5% of the total variance. Factor 3 has H, the free acid as the sole variable in the factor group accounting for 10.7% of the total variance. Results after screening give identical factor groups with similar variance as well, viz. 62.2%, 15.2% and 9.9%, respectively. This contrasts with the findings of Sequeira and Lai (1998) who identified three distinct Factors - partially neutralized acids, sea salt and based ions, and K, for the year 1990 to 1992 data in KT. In the present study, all the three Factor groups of Sequeira and Lai are mixed as a single Factor (Factor 1). Formate and acetate, which is lacked in Sequeira and Lai's data set, is identified as another unique Factor group (Factor 2) and free acids (H) as the third Factor (Factor 3).

Central/Western (1993-1999, Table 2-5)

Three factor groups are also identified from the original data set accounting for 90.7% of the total variance. Factor 1 comprises of K, formate and acetate and accounts for 57.9% of the total variance. Factor 2 is of partially neutralized acids (PNA) in nature consisting of NH_4, NO_3 and H with a strong SO_4 association (0.6332). This partially neutralized acids accounts for 17.8% of the total variance. Factor 3 is sea salt and base ions (Mg, Na, Cl, Ca) and accounts for 14.9% of the variance. Factor 3 is also strongly associated with NH_4, SO_4 and K as revealed by their eigenvalues of 0.6186, 0.6966 and 0.6247, respectively.

Table 2-4. Results of Factor Analysis on rainwater composition at Kwun Tong, 1993-1999. Figure in bold indicates a significant loading (>0.70) of the variable for the factor.

Raw data (n=178)

	Varimax rotated factor loading pattern		
Variables	Factor 1	Factor 2	Factor 3
NH_4^+	***.800688***	.126925	.343545
SO_4^{2-}	***.950994***	.046656	.220113
NO_3^-	***.889832***	.020289	.347629
Ca^{2+}	***.904184***	-.013484	.071413
H^+	.023606	.039073	***.908888***
Mg^{2+}	***.949608***	.088150	-.081164
Na^+	***.883604***	.088249	-.206052
Cl^-	***.891849***	.079039	-.203938
K^+	***.911573***	.063154	-.040469
Formate	.130280	***.928223***	.028636
Acetate	.002137	***.926430***	.019933
Eigenvalue	6.58005	1.70362	1.17562
% of total Variance	59.81870	15.48749	10.68748
Cumul. Eigenvalue	6.58005	8.28368	9.45930
Cumulative %	59.81870	75.30618	85.99366

Screened data (n=155)

	Varimax rotated factor loading pattern		
Variables	Factor 1	Factor 2	Factor 3
NH_4^+	***.811190***	.141268	.286566
SO_4^{2-}	***.959125***	.049005	.163475
NO_3^-	***.907182***	.013767	.263932
Ca^{2+}	***.926959***	.005728	-.036308
H^+	.046303	.042006	***.971433***
Mg^{2+}	***.964228***	.085281	-.012033
Na^+	***.899456***	.077882	-.058113
Cl^-	***.907617***	.077330	-.060092
K^+	***.951987***	-.015115	-.030283
Formate	.128459	***.907732***	.124588
Acetate	-.015723	***.914641***	-.056019
Eigenvalue	6.84309	1.67269	1.086740
% of total Variance	62.20993	15.20635	9.87946
Cumul. Eigenvalue	6.84309	8.51579	9.60253
Cumulative %	62.20993	77.41628	87.29574

Table 2-5. Results of Factor Analysis on rainwater composition at Central/Western, 1993-1999. Figure in bold indicates a significant loading (>0.70) of the variable for the factor.

Raw data (n=155)				Screened data (n=117)			
	Varimax rotated factor loading pattern				Varimax rotated factor loading pattern		
Variables	Factor 1	Factor 2	Factor 3	Variables	Factor 1	Factor 2	Factor 3
NH_4^+	.045235	***.705681***	.618552	NH_4^+	.653284	.680027	.029535
SO_4^{2-}	.287090	.633165	.696563	SO_4^{2-}	***.707058***	.625207	.290801
NO_3^-	.087635	***.895643***	.286352	NO_3^-	.277662	***.934089***	.088849
Ca^{2+}	.506827	.293257	***.726690***	Ca^{2+}	***.741045***	.260535	.528567
H^+	.006103	***.861133***	-.245212	H^+	-.180236	***.932051***	-.012705
Mg^{2+}	.198388	.138456	***.944634***	Mg^{2+}	***.958962***	.109966	.195121
Na^+	.112506	.053027	***.963240***	Na^+	***.971543***	.051704	.111365
Cl^-	.125656	.052264	***.959027***	Cl^-	***.965902***	.051380	.129311
K^+	***.707270***	.161517	.624672	K^+	.641269	.145507	***.700739***
Formate	***.928251***	.048762	.081776	Formate	.081169	.044357	***.924036***
Acetate	***.955973***	.031596	.159907	Acetate	.169777	.015223	***.952143***
Eigenvalue	6.36916	1.96450	1.64213	Eigenvalue	6.47481	2.12003	1.62844
% of total Variance	57.90147	17.85911	14.92848	% of total Variance	58.86194	19.27301	14.80408
Cumul. Eigenvalue	6.36916	8.33366	9.97579	Cumul. Eigenvalue	6.47481	8.59484	10.22329
Cumulative %	57.90147	75.76058	90.68905	Cumulative %	58.86194	78.13495	92.93903

Interestingly, the factor group shows swapping in that Factor 1 and Factor 3 in the original data set interchanged with each other in the screened data set. In the screened data set, K, formate and acetate made up the Factor 3 with a 14.8% of the total variance instead of Factor 1 in the original data set with a variance of 57.9%. Factor 2, the partially neutralized acids, remains the same in both original and screened data sets with similar variance of 17.8 and 19.2%, respectively. Sea salt, base ions and SO_4 associated with NH_4 accounts for 58.8% of the variance as Factor 1 in the screened set. Again, contrasting to the same report of Sequeira and Lai (1998), the mixed group in 1990-1992 is resolved better to 3 factors in this study. However, as indicated above, the strong association with other PNA species suggests that mixed group with poor resolution could not be totally excluded.

Potential sources of chemical species in the rainwater

The PCA is intended to resolve the variables into identifiable groups. This in turn, assists in assessing the potential sources of the variables. Despite different factor loadings is found in KT and CW, consistent groups of variables contributing to Hong Kong rain acidity and hence, their source, are revealed.

The SO_4, NO_3, NH_4 and H are found closely associated with each other and described as PNA. The PNA exists as a contributing Factor group as in CW or mixed with the sea salt and base ions component as in KT. The SO_4 and NO_3 are inarguably of anthropogenic origins. Both are closely associated with H and major acidifying agents of Hong Kong rainwater. NO_3 is the oxidation product of NOx, which is ubiquitously generated during the combustion by oxidation of nitrogen present in the air. Traffic would be one of the major sources of NOx. The oxidation of NOx to NO_3 in atmospheric condition is 10 times faster than the oxidation of SO_2 to SO_4 (Howells, 1995). Since NO_3 is highly water soluble, wet deposition in forms of rain or moisture becomes the major pathway for its deposition from the atmosphere. These chemical properties make NO_3 or its parental gaseous precursors, NOx, commonly known to be of local emission origin. SO_4 is the oxidation product of SO_2 from fossil combustion. However, the dry oxidation of SO_2 to SO_4 or wet oxidation of SO_2 to SO_4 via the bisulfite (HSO_3) intermediate in the ambient atmospheric condition is much slower than that of NOx. As a result, The SO_2 and its intermediate oxidative products have much longer residence time in the atmosphere. This makes them more susceptible to be transported by the movement of the air mass comparing with NOx. Thus, the parental gaseous source of SO_4 in the rainwater could be of local emission as well as from neighboring regions and transported by air mass to Hong Kong.

In rural or agricultural regions, NH_4 is sourced from fertilizers. This is, however, very unlikely the case in Hong Kong as agricultural activity is not a major economic activity in the past ten years as a whole. Land use in KT and CW are mainly residential mixed with industrial and commercial nature, respectively. Such land usage could definitely not be a source of fertilizers. One very possible source would be the secondary aerosols as $(NH_4)_2SO_4$ from a distant source. Another possibility will be from the sewage which is discharged daily directly to Victoria Harbor without secondary treatment. Primary sewage contains high NH_3, which could easily volatized in alkaline pH (Gray, 1989; Idelovitch and Michail, 1981).

Sea salt (Na, Cl, Mg, Ca and K) is also a major component of the rainwater collected in KT and CW, probably due to their proximity to the Victoria Harbor. The sea salt contributes by itself as a single factor as in CW or mixed with the PNA as in KT. While the Na, Mg and Cl are presumed from the seawater (discussed further in section 2.3 later), Ca and K could be from some other terrestrial sources as evidenced from the egeinvalues of Ca and K found in CW (Table 2-5). While eigenvalue of a variable greater than 0.7 suggests a significant loading of the variable into the factor group, eigenvalue above 0.5 and close to 0.7 suggests a strong correlation of the variable in the loading factor group as well. For Ca, the values are 0.5068 and 0.7267 loaded in Factor 1 and 3, respectively, in the original data set. The corresponding values for Ca in the screened set are 0.7410 and 0.5286. In case of K, the values in the original data set are 0.7073 and 0.6247 in Factor 1 and 3, respectively. Those in the screened set are 0.6417 and 0.7007, correspondingly. These imply some other sources of Ca and K in the rainwater apart from seawater.

Calcium is basically crustal in nature originated locally or as a result of some long or short range transport. The heavy construction works, road repair and maintenance, as well as the paved road dust in the urban areas contribute as major local sources of Ca.

Potential terrestrial source of K is associated with biomass or waste burning. The hill fires and the then still operating incinerators in early and mid 90s could be possible local sources. K loading could also be sourced from local urban and roadside soil, which has generous K supply (Jim, 1996).

One particular factor group so identified in both KT and CW is the formate and acetate, which accounts for 15 to 18 % of the total variance. According to USEPA (2000), acetaldehyde and formaldehyde are compounds commonly emitted from mobiles and vehicles. The acetaldehyde and formaldehyde is easily oxidized to acetate and formate. Thus this factor group should reflect the contribution by the local traffics as well. The significant association of K and the strong association of Ca with this factor group in CW (Table 2-5) provide further evidence on the potential local source of K and Ca in relation to traffic such as the uplifting of the roadside soils and the paved road dust as a result of the turbulence created by the exhaust of traffic.

The PCA results suggest that factors contributing to rain acidity is spatially dependent as revealed from the different factor groups and their loading variables as well as the relative contribution to the total variability in KT and CW stations. Factor groups can also shift with time as demonstrated by comparing the results of the present study (1993-1999) with the previous one (1990-1992). This implies that rain acidity recorded in a station will be significantly affected by its ambient conditions such as nature of human activities related to the landuse of the district as well as the district's development in temporal sense.

The present Factor Analysis suggests that:

(1) PNA and sea salt are still the major factor group accounting for about 60% of the total variance of the rainwater composition. This implies that SO_4 and NO_3 are key acidifying chemical species.

(2) The organic species, formate and acetate, comprises a significant contributing factor to the local rain (15 to 19% of the total variance). In this regard, vehicular and automotive discharge should be noted as they are the potential sources of the formate and acetate.

2.2.2 Trend Analysis by using Seasonal Kendall

Trend analysis is commonly employed to examine the trend of change over time. The Seasonal Kendall's tau is a common statistics for testing randomness against trend. The total (wet + dry) deposition was thus employed for trend analysis in accounting the significant contribution from the wet deposition and reducing the error from missing data points in the wet deposition data set between raining and non-raining seasons.

The total deposition of NO_3 and SO_4 were shown in Table 2-6. For the year 1993 to 1999, the NO_3 and SO_4 in the wet deposition accounts for 54.1 to 79.2% and 64.7 to 83.1%, respectively, of the total annual deposition in KT. The corresponding percentage in CW ranges from 59.6 to 80.6% and 68.4 to 85.9%, respectively. In general, the wet and dry deposition accounts for about 70% and 30%, respectively, of the total annual deposition (Table 2-6). Wet deposition is thus dominant over dry deposition in terms of the concentration levels. The wet deposition or raining in Hong Kong is of random and seasonal nature. There are, however, long periods of the non-raining seasons (Dec to May), in which no data will be available as result of insufficient rainwater collected for trend analysis. The total deposition (sum of wet and dry deposition) suffixes the dry deposition data in the non-raining season between consecutive raining seasons to reduce the estimation error due to the missing points. With this rationale, the corresponding weekly data of wet and dry deposition were summed and the monthly average calculated before the Seasonal Kendall test. Table 2-7 summarizes the S-tau statistics and the p-level of the various ions in the deposition.

Despite the fact that acid rain is evidenced from the two monitoring stations, there is no significant trend of change in the acidity over the year 1993 to 1999 as revealed from the H in the rainwater. In both stations, no significant trend of change is also observed for cations such as Ca, K, Mg and NH_4 and for anions formate and acetate. Integrating with the results of Factor Analysis (Section 2.2.1), it implies that input from sources of Ca, K, Mg and NH_4 should be more or less constant throughout these years. A significant decreasing trend is found for Na and Cl in CW but not in KT. The magnitude of decrease for Na and Cl as reflected by the S-tau (0.172 and 0.153, respectively) are close to each other. This suggests that they are likely related to some sodium chloride source, which consumption has been reduced over the years in CW.

Table 2-6. The relative distribution of (A) NO_3^- and (B) SO_4^{2-} in the wet and dry deposition in Hong Kong

(A) – NO_3^- (kg/ha)	Kwun Tong, 1993 – 1999			Central/Western, 1993 – 1999		
	Wet (% of total)	Dry (% of total)	Total	Wet (% of total)	Dry (% of total)	Total
1993	10.63	5.63	16.26	10.89	5.16	16.05
	(65.4)	(34.6)		(67.8)	(32.2)	
1994	9.79	3.59	13.38	10.10	2.43	12.53
	(73.2)	(26.8)		(80.6)	(19.4)	
1995	7.04	5.98	13.02	7.88	4.16	12.04
	(54.1)	(45.9)		(65.4)	(34.6)	
1996	12.19	5.87	18.06	9.52	4.72	14.25
	(67.5)	(32.5)		(66.9)	(33.12)	
1997	20.91	5.50	26.41	9.98	3.96	13.94
	(79.2)	(20.8)		(71.6)	(28.4)	
1998	18.20	5.80	24.01	13.00	5.52	18.53
	(75.8)	(24.2)		(70.2)	(29.8)	
1999	9.90	7.30	17.20	10.34	7.01	17.35
	(57.6)	(42.4)		(59.6)	(40.4)	

(B) – SO_4^{2-} (kg/ha)	Kwun Tong, 1993 – 1999			Central/Western, 1993 – 1999		
	Wet (% of total)	Dry (% of total)	Total	Wet (% of total)	Dry (% of total)	Total
1993	39.14	21.36	60.5	46.62	12.22	58.84
	(64.69)	(35.31)		(79.23)	(20.77)	
1994	36.32	14.05	50.37	30.63	5.01	35.64
	(72.11)	(27.89)		(85.94)	(14.06)	
1995	32.05	16.00	48.05	27.53	9.93	37.46
	(66.70)	(33.30)		(73.49)	(26.51)	
1996	33.53	13.95	47.48	29.42	10.63	40.05
	(70.62)	(29.38)		(73.46)	(26.54)	
1997	44.34	9.03	53.37	35.00	7.62	42.62
	(83.08)	(16.92)		(82.12)	(17.88)	
1998	36.45	9.00	45.45	29.43	9.83	39.26
	(80.20)	(19.80)		(74.96)	(25.04)	
1999	26.05	10.82	36.87	22.66	10.46	33.12
	(70.65)	(29.35)		(68.42)	(31.58)	

Table 2-7. Summary of the S-tau statistics resulted from Seasonal Kendall analysis for the ions in the total (wet + dry) deposition in Hong Kong. Significant values printed in bold ($p<0.05$).

	Kwun Tong, 1993 – 1999		Central/Western, 1993 – 1999	
	S	p-level	S	p-level
H*	0.032	0.671	-0.135	0.069
Ca	0.038	0.610	-0.122	0.101
Mg	-0.104	0.162	-0.107	0.151
Na	0.053	0.472	***-0.172***	***0.020***
K	0.010	0.889	-0.045	0.547
NH_4	-0.009	-0.902	-0.022	0.769
Cl	0.053	0.472	***-0.153***	***0.039***
SO_4	-0.119	0.108	-0.013	0.865
NO_3	***0.187***	***0.012***	0.129	0.084
Formate	0.016	0.829	-0.057	0.441
Acetate	0.018	0.814	-0.048	0.518

H: only wet deposition data used for H

Concerning the two key acidic species, the total deposition of SO_4 is more than 2-fold of the NO_3 loading (Table 2-6). The SO_4 is still the most important acidifying agents in Hong Kong rainwater. No significant trend of change in SO_4 level is observed in both stations. As discussed before, sources for SO_4 could be more extensive. Apart from sea salt SO_4, most SO_4 are oxidative products of SO_2. The SO_2 could be from local emission and oxidized. Local sources include power stations where coal is burned and industries with installations such as boilers that are powered by fossil fuel. The SO_2 could also be regionally originated across the border and transported to Hong Kong in fully oxidized SO_4 or partially oxidized intermediates. Both the local and regional factors could contribute SO_4 to the local rainwater but unfortunately, could not be differentiated from the sole rainwater composition data. The result of the trend analysis demonstrated the complexity of the situation regarding to SO_4. KT has been designated as a mixed residential and industrial zone since the 80s. In the 80s and early 90s, boilers powered by fossil fuel are very common basic installations in the factories. This implies a prominent local source of SO_2 and SO_4 in KT itself. KT has shifted from an industrial nature to more commercial nature since late 80s. As a result, more and more factory buildings are renovated or even re-built to commercial (office) and storage buildings. The development of e-business further speeds up the shift of the industrial nature of KT to service (server and data center) nature. Number of active operating factories decreases and in turn, the consumption of the fossil fuel in the district. Emission of local SO_2 from the factories in KT should therefore decrease correspondingly. This speculation is supported by the total annual SO_4 deposition data. KT has a range of 36 to 60 kg/ha and in a chronological decreasing trend (Table 2-6) when comparing with the mixed residential and commercial nature of CW, which has the SO_4 loading lies in a more constant range of 33 to 40 kg/ha from 1994 to 1999. The SO_2 emission inventory also shows a significant reduction in the

SO_2 emission from an annual 197,564 tonnes in 1993 to 59,792 tonnes in 1999 (http://www.info.gov.hk/epd/index.htm). This is equivalent to about 70% SO_2 reduction over the years in the territory. The SO_2 emission inventory also shows that the emission from motor vehicle reduced from 11124 tonnes in 1993 to 1629 tonnes in 1999 accounting for about 85% reduction from the motor vehicles. In other words, both local in terms of individual district such as KT or territory wise, a decreasing trend in the oxidative product, SO_4, is expected. However, as revealed from the S-tau statistics (Table 2-7), there is no significant trend of change in the total deposition from 1993 to 1999 in both stations. In this regard, the SO_4 from parental SO_2 originated from neighboring regions and transported to Hong Kong by air mass should not be neglected.

Another key acidic species, NO_3 shows a significant increasing trend in KT but no trend of change observed in CW. Such discrepancy is speculated to be a result of the spatial variation between the two sites. The NO_3 is the major oxidative product of NOx which are ubiquitously generated in the combustion process. According to the NOx emission inventory reported by HKEPD (http://www.info.gov.hk/epd/index.htm), the total NOx emission has been reduced from 176,241 tonnes in 1993 to 99,556 tonnes in 1999, accounting for 76,685 tonnes or about 44% reduction. Majority of the NOx reduction was contributed by the Power Generation, which has a reduction in the NOx emission from 113,300 tonnes in 1993 to 41,744 tonnes in 1999 accounting for a 71,556 tonnes reduction. This 71,556 tonnes is equivalent to 93.3% of the total 76,685 tonnes reduction. Motor vehicle is the second most important NOx source as revealed from the HKEPD emission inventory. However, unlike that of SO_2 which had a 85% reduction from the motor vehicle, NOx emission from the motor vehicle had just a very slight reduction from 39,555 tonnes in 1993 to 37020 tonnes in 1999 (about 6.5% reduction) in the territory. No significant reduction in the total NO_3 deposition is observed as well in Table 2-6. In KT, the total NO_3 deposition fluctuates in the range of 13 to 26 kg/ha in 1993 to 1999. In case of CW, the range is 12 to 18 kg/ha. These data suggest that the traffic effect around the monitoring site could possibly more significant than the significant reduction of NOx from power stations in districts nearby.

The significance of the traffic loading in terms of the daily vehicle kilometrage in the territory is illustrated in Table 2-8. There is actually an overall 18 % increase in 1999 in the vehicle kilometrage compared with 1993 in the whole territory according to the Transport Department (1994, 2000).

Table 2-8. The summary of the daily vehicle kilometrage in Hong Kong

	1993	1999	% change
Hong Kong Island	4,953,422	5,616,789	+13
Kowloon	8,037,592	7,873,646	-2
New Territory	12,605,308	16,755,877	+33
Total	25,596,322	30,246,312	+18

The motor vehicle contribution is also reflected from the data reported as the Annual Average Daily Traffic (AADT) by the Transport Department (1994, 2000). Five counting stations for vehicle flow in the proximity of the two acid rain monitoring station were selected and their AADT in 1993 and 1999 was summarized in Table 2-9.

Table 2-9. The Annual Average Daily Traffic (AADT) of the roads nearby the acid rain monitoring station at Kwun Tong and Central/Western

Road (Road Code)	1993	1999	% change
Kwun Tong			
Kwun Tong Rd underpass western end to Kwun Tong Rd RA (3236)	28510	31470	+10
Yue Man Square to Kwun Tong Rd (3276)	6010	5200	-13.5
Tung Yan St to Kwun Tong Rd RA (3474)	27540	26310	-4.5
Tung Yan St to Yue Man Square (3481)	19920	18540	-7
Kwun Tong Rd RA to Kwun Tong Rd underpass eastern end (3643)	35610	34920	-2
Central/Western			
Water St to Pok Fu Lam Rd (1227)	12190	11520	-5.5
Des Voeux Rd W to Queen’s Rd W (1621)	9150	8550	-6.6
Des Voeux Rd W to Queen’s Rd W (1622)	19650	16490	-16
Western St to Pok Fu Lam Rd (2010)	19960	19360	-3
Water St to Western St. (2027)	13620	14480	+6

In general, the traffic loading is more or less the same in terms of AADT between the year 1993 and 1999 in both KT and CW. The traffic loading around the acid rain monitoring station in KT is also heavier than that in CW as reflected by the AADT. The discrepancy in the NO_3 trend could, therefore, be related to the different traffic loadings in the two districts with the facts of (i) no effect in the deposition data observed regardless of a prominent reduction in the NOx emission from local power station, (ii) insignificant reduction in terms of NOx emission and traffic flow in KT and CW and (iii) KT has generally a heavier traffic load than CW. The increasing trend of NO_3 also signifies the importance of its relative contribution to local rain acidity. This will be discussed further in Section 2.3.

2.2.3 Multiple Regression Analysis

The stepwise multiple regression is conducted to estimate or predict the free acidity (H) in relation to the acidic and base ions. Results of the stepwise multiple regression is summarized in Table 2-10. In explaining the variance of the free acidity (H), screening has significantly improved the variance calculated, from about 52% to 77% and 58% to 91% in Kwun Tong and Central/Western, respectively. In line with the findings from the PCA (Section 2.2.1) and Trend Analysis (Section 2.2.2), NO_3 and SO_4 are still the major acidifying species as reflected by their positive coefficients in the regression analysis. The regression indicates that Cl could also be an important species for the acidity. Similarly, Na is a potential base next to Ca and NH_4.

One particular phenomenon noted worthy is that the coefficient of NO_3 is greater than SO_4 in magnitude, viz. 0.9500 vs. 0.9049 in KT and 0.9885 vs. 0.7806 in CW, in the screened data sets. This implies that in estimating or predicting the free acidity in rainwater, the relative contribution by NO_3 will be higher than that by SO_4. In other words, NO_3 contribution to the rain acidity becomes more important. Such deduction, once again, is in line with results of the Trend Analysis.

In predicting the future possible trend of rain acidity in Hong Kong, it relies, therefore, on the effective control of the key acidifying species. The main acidifying species are SO_4^{2-} and NO_3^-, oxidative products of SO_2 and NOx, respectively. The government had already mandated the use of ultra-low sulfur diesel to further reduce the local emission of SO_2. Traffic emission is a major source of NOx as well as acetaldehyde and formaldehyde which are potential organic acid generators in the atmosphere. The relative contribution of NO_3^- in the rainwater is also increasing over the years indicating the traffic loading is playing a more important role. Provided with maintaining the existing governmental policy on traffic control, there should have no deterioration trend in future in the rain acidity. In this regard, a more stringent control on the exhaust by reducing the number of vehicles and/or providing better maintenance should further improve the situation.

Table 2-10. Summary for the coefficients of the stepwise multiple regression for the prediction equations.

H=	bo	+b_1NO_3	+b_2SO_4	+b_3Ca	+b_4NH_4	+b_5Na	+b_6Cl	+ error	R^2
Kwun Tong (1993 - 1999)									
Raw data	17.51	1.3028	0.5530	-0.7410	-0.4432		-0.1679		0.5158
Screened data	**11.42**	0.9500	0.9049	-0.8343	-0.9042	-1.130	0.8143		0.7651
Central/Western (1993 - 1999)									
Raw data	16.57	0.6308	0.7612	-0.7267	-0.2740	-1.2322	0.9231		0.5777
Screened data	22.50	0.9885	0.7806	-0.5167	-0.9495	-0.8031	0.5862		0.9086

Charge omitted for all ionic species

Intercept in bold denotes values not significantly different from zero ($p<0.05$)

2.3 Sea-salt and Non Sea-salt components in rainwater

Both the Factor Analysis and Multiple Regression suggests that NO_3 and SO_4 are playing a very dominant role in the local rain acidity. While NO_3 is universally considered to be of anthropogenic, SO_4 could be from anthropogenic source as well as natural sources. One of the potential natural sources of SO_4 in Hong Kong will be from marine salt as both stations are close to the Victoria Harbour. The determination of the sea salt (ss) and non-sea salt (nss) fraction will further help identifying the potential SO_4 source. A classical protocol in determining the sea salt fraction in rainwater by Keene *et al*. (1986) was followed. Table 2-11 summarized the results of regressions and test of significance between pure sea-salt constituents and the reference species (Na or Mg) in the wet deposition collected in Kwun Tong and Central Western.

Four possible cases can be deduced from Table 2-11. (1) Both the slopes and intercepts of the ion ratios in Table 2-11 are not significantly different from the reference seawater ratio, both species are originated from sea-salt. (2) Significant non-zero intercept but the slope is same as that of the seawater ratio, the magnitude of the non-sea salt component would be constant and independent of sea-salt effects. (3) Intercept insignificant difference from zero but a significant difference of the slope from seawater ratio indicates a variable source for the ion species elsewhere from seawater. (4) Both the slope and intercept are significantly different from the seawater ratio and zero respectively, fractionation and/or a variable non-sea-salt effect were present (Keene *et al.*, 1986).

Reviewing Table 2-11, screened data provide a better estimate than raw data. Pattern is similar in the screened data in both stations. When Mg is referenced, Cl, K and SO_4 are in Case (4) and Ca in Case (3) for the screened data in both stations. Similar results are obtained using Na as reference for sea salt except that Cl falls into Case (3) rather than (4). Cl depletion is evidenced from the smaller slope value than that of seawater. Ca falls in Case (3) in both stations with a slope significantly higher than that of seawater. This suggests a dominant variable non-sea salt input, which will likely be related to the heavy construction activities in Hong Kong over the years. The K and SO_4 are in Case (4) in both stations indicating potential variable sources of the two species apart from seawater. The relative input of the two species in rainwater from seawater and these other possible sources varies over time and conditions. Anthropogenic source of SO_4 from combustion of fossil fuel is the general consensus. Land based sources for K could be from the roadside soil, paved road dust or biomass burning such as hill fire in Hong Kong as discussed in the Factor Analysis. The overall results indicate clearly the influence of non-sea salt component in the local rainwater.

Table 2-11. Summary of regressions and test of significance between sea-salt constituents and reference species (Mg and Na) in the wet deposition collected in Kwun Tong and Central Western

Ion Ratio		Sea Water Ratio*	Kwun Tong		Central /Western	
			Raw	Screened	Raw	Screened
$Cl^-: Mg^{2+}$	Corr. Coeff		0.909	0.960	0.931	0.957
	Slope ± S.E.	5.14	**4.118 ± 0.142**	**4.151±0.106**	4.841±0.154	**4.842±0.137**
	Inter.		**17.856±4.379**	**15.955±3.766**	**16.324±5.166**	**14.584±5.226**
$Ca^{2+}:Mg^{2+}$	Corr. Coeff		0.810	0.825	0.829	0.836
	Slope ± S.E.	0.194	**3.257±0.178**	**3.239±0.196**	**1.370±0.0747**	**1.376±0.084**
	Inter.		-3.645±5.469	-8.617±6.956	**6.212±2.511**	6.229 ± 3.216
$K^+:Mg^{2+}$	Corr. Coeff		0.846	0.906	0.759	0.767
	Slope ± S.E.	0.0961	**0.179±0.009**	**0.180±0.007**	**0.143±0.010**	**0.144±0.011**
	Inter.		**5.145±0.261**	**4.789±0.264**	**5.315±0.332**	**5.288±0.431**
$SO_4^{2-}:Mg^{2+}$	Corr. Coeff		0.860	0.883	0.792	0.798
	Slope ± S.E.	0.532	**3.512±0.157**	**3.500±0.165**	**2.113±0.132**	**2.104±0.148**
	Inter.		**28.774±4.824**	**25.885±5.828**	**40.628±4.429**	**41.453±5.676**
$Mg^{2+}:Na^+$	Corr. Coeff		0.906	0.958	0.933	0.960
	Slope ± S.E.	0.227	0.226±0.008	**0.250±0.007**	**0.201±0.006**	**0.213±0.0058**
	Inter.		-1.223±1.021	-2.195±0.931	-0.690±1.000	-1.129±1.0248
$Cl^-:Na^+$	Corr. Coeff		0.997	0.999	0.999	0.999
	Slope ± S.E.	1.160	**1.126±.007**	**1.128±.0047**	**1.122±0.005**	**1.123±0.005**
	Inter.		0.543±0.851	0.815±0.657	**2.474±0.761**	1.877±3.875
$Ca^{2+}:Na^+$	Corr. Coeff		0.641	0.698	0.712	0.744
	Slope ± S.E.	0.0439	**0.643±0.058**	**0.716±0.065**	**0.254±.020**	**0.272±0.023**
	Inter.		-1.842±7.438	-9.334±9.134	**6.838±3.222**	6.400±4.005
$K^+:Na^+$	Corr. Coeff		0.767	0.808	0.690	0.714
	Slope ± S.E.	0.0218	**0.040±0.003**	**0.042±0.003**	**0.028±0.002**	**0.030±0.003**
	Inter.		**4.923±0.327**	**4.605±0.381**	**5.272±0.378**	**5.199±0.480**
$SO_4^{2-}: Na^+$	Corr. Coeff		0.750	0.782	0.723	0.737
	Slope ± S.E.	0.121	**0.763±.0507**	**0.810±0.057**	**0.417±0.032**	**0.432±0.037**
	Inter.		**26.304±6.501**	**22.674±8.023**	**39.826±5.122**	**40.426±6.498**

*Keene *et al*., 1986**;** Unit of concentration are μeq/l; Correlation coeff: correlation coefficient;

Slope ± S.E. : slope of regression plus or minus the standard error; inter ± S.E. : intercept of the regression plus or minus the standard error. Figures in bold are significantly different from the seawater ratio or intercept of zero ($p<0.05$).

In differentiating the nss and ss components from the rainwater, the equations of Keene *et al*. (1986) were also adopted.

$$[\text{sample}]_{ss} = ([\text{sample}]_{bk} / \text{Ref}_{bk}) \times \text{Ref}_{samp}$$

$$[\text{sample}]_{nss} = [\text{sample}]_{T} - [\text{sample}]_{ss}$$

where	$[\text{sample}]_{ss}$	is the calculated sea-salt components of a particular chemical constituent concerned in a precipitation sample
	$[\text{sample}]_{bk}$	is the ion concentration in bulk sea water
	Ref_{bk}	is the concentration of the reference species (Na^+ or Mg^{2+}) in bulk seawater
	Ref_{samp}	is the concentration of the reference species in the wet deposition sample
	$[\text{sample}]_{nss}$	is the calculated ion concentration of non-sea salt components
	$[\text{sample}]_{T}$	is the total measured concentration of ions

According to Keene *et al*. (1986), Mg or Na should be used as the reference estimator for the other ions because they are conservative tracers of sea salts at all sites. The choice rests on the one with lower concentration relative to the seawater ratio (Mg:Na = 0.227). Thus if the slope of Mg:Na in Table 2-11 is below 0.227, Mg is preferred and vice versa for Na. For the screened data in Table 2-11, Na and Mg should be used as estimator for Kwun Tong and Central/Western, respectively. In this accord, both estimators will be used for the calculations. Table 2-12 summarizes the calculated ss and nss components for the various sea salt ions in the local rainwater. Generally, the nss fraction calculated for Ca, SO_4 and K is in very close agreement (within 3%) in the screened data set regardless of using Na or Mg as estimator. The only difference is the calculated nss-Cl fraction. Using Mg as estimator, 7.104 and 10.647 μeq/l are calculated as nss fraction in KT and CW, respectively. The corresponding value is –0.11 and 0.089 μeq/l using Na as estimator. The majority of the Cl is, however, seawater originated. The significance of Table 2-12 is that nss component is prominent for Ca, SO_4 and K, which accounts for about 86 to 90% of the total wet deposition. This is also in line with the findings so far. nss-Ca could be traced to the heavy construction activities in the territory. The nss-K could be originated locally or regionally. Local sources include the urban and roadside soil, and biomass burning such as hill fires in winter as discussed before in the Factor Analysis section. The common practice of agricultural burning as means of fertilizing the topsoils in the neighbouring fields could also be potential regional source of nssK, which is then transported to Hong Kong by the air mass. About 90% SO_4 in the rainwater was contributed by the nss component (Table 2-12) and the ratio of ss-SO_4/nss-SO_4 ratio is well below 0.2 (Table 2-13). Anthropogenic source of SO_4 is very prominent.

Table 2-12. Summary of the concentration of sea-salt and non-sea-salt components of anions and cation (μeq/l) in the raw and screened data set at two sites (1993-1999) with respect to Mg and Na as estimator for the (A) raw and (B) screened data.

(A) Raw			Mg^{2+} estimator		Na^{+} estimator	
	$[sample]_T$	$[sample]_{bk}$	$[sample]_{ss}$	$[sample]_{nss}$	$[sample]_{ss}$	$[sample]_{nss}$
Kwun Tong						
Na	39.550	581.238	31.245	**8.305**	39.550	0.000
Cl	44.923	677.452	36.417	**8.506**	45.878	-0.955
Ca	15.692	25.569	1.374	**14.317**	1.736	13.955
SO_4	39.578	70.118	3.769	**35.809**	4.786	34.792
K	6.742	12.666	0.681	**6.061**	0.862	5.880
Mg	7.085	131.800	7.085	**0.000**	8.978	-1.893
Central/Western						
Na	43.142	581.238	35.090	**8.052**	43.142	0.000
Cl	50.621	677.452	40.898	**9.723**	50.044	0.576
Ca	11.354	25.569	1.544	**9.810**	1.894	9.460
SO_4	39.314	70.118	4.233	**35.081**	5.220	34.094
K	6.577	12.666	0.765	**5.813**	0.940	5.637
Mg	7.957	131.800	7.957	**0.000**	9.793	-1.836

(B) Screened			Mg^{2+} estimator		Na^{+} estimator	
	$[sample]_T$	$[sample]_{bk}$	$[sample]_{ss}$	$[sample]_{nss}$	$[sample]_{ss}$	$[sample]_{nss}$
Kwun Tong						
Na	41.255	581.238	34.870	**6.385**	41.255	0.000
Cl	47.746	677.452	40.642	**7.104**	47.856	-0.110
Ca	14.760	25.569	1.534	**13.226**	1.811	12.949
SO_4	39.330	70.118	4.207	**35.124**	4.992	34.339
K	6.719	12.666	0.760	**5.959**	0.899	5.820
Mg	7.907	131.800	7.907	**0.000**	9.365	-1.457
Central/Western						
Na	48.950	581.238	39.659	**9.291**	48.950	0.000
Cl	56.871	677.452	46.224	**10.647**	56.782	0.089
Ca	12.464	25.569	1.745	**10.720**	2.149	10.316
SO_4	41.045	70.118	4.784	**36.261**	5.923	35.122
K	6.756	12.666	0.864	**5.891**	1.067	5.688
Mg	8.993	131.800	8.993	**0.000**	11.112	-2.119

$[sample]_T$ is the total measured concentration of ions;
$[sample]_{bk}$ is the ion concentration in bulk sea water;
$[sample]_{ss}$ is the calculated sea-salt components;
$[sample]_{nss}$ is the ion concentration of non-sea salt components

Table 2-13. Summary of the acidifying potential and neutralizing potential by (A) Mg^{2+} and (B) Na^{+} estimator for the screened data

Site	Year	[nss-Ca^{2+}]	[NH_4^+]	[nss-SO_4^{2-}]	[NO_3^-]	[ss-SO_4^{2-}] / [nss-SO_4^{2-}]	[NO_3^-] / [nss-SO_4^{2-}]	NP/AP
Kwun Tong								
(A)	93/99	13.226	18.408	35.371	10.832	**0.119**	**0.306**	**0.678**
	93	17.518	16.151	36.220	8.507	**0.171**	**0.235**	**0.691**
	94	11.894	16.561	37.959	8.970	**0.106**	**0.236**	**0.652**
	95	9.313	15.134	26.754	5.960	**0.187**	**0.223**	**0.706**
	96	10.363	19.043	36.754	11.733	**0.119**	**0.319**	**0.685**
	97	16.811	25.037	42.001	16.631	**0.084**	**0.396**	**0.818**
	98	18.104	17.845	29.620	11.428	**0.118**	**0.386**	**0.736**
	99	11.753	17.164	35.032	12.239	**0.088**	**0.349**	**0.557**
(B)	93/99	12.949	18.408	34.586	10.832	**0.138**	**0.313**	**0.690**
	93	16.414	16.151	33.145	8.507	**0.280**	**0.257**	**0.782**
	94	11.895	16.561	37.941	8.970	**0.106**	**0.236**	**0.607**
	95	9.523	15.134	27.307	5.960	**0.163**	**0.218**	**0.741**
	96	10.127	19.043	36.079	11.733	**0.140**	**0.325**	**0.610**
	97	16.540	25.037	41.236	16.631	**0.104**	**0.403**	**0.718**
	98	17.912	17.845	29.073	11.428	**0.139**	**0.393**	**0.883**
	99	10.724	17.164	32.180	12.239	**0.185**	**0.380**	**0.628**

Site	Year	[nss-Ca^{2+}]	[NH_4^+]	[nss-SO_4^{2-}]	[NO_3^-]	[ss-SO_4^{2-}] / [nss-SO_4^{2-}]	[NO_3^-] / [nss-SO_4^{2-}]	NP/AP
Central/Western								
(A)	93/99	10.719	16.019	36.261	10.554	**0.132**	**0.291**	**0.571**
	93	12.036	18.089	44.959	10.875	**0.136**	**0.242**	**0.540**
	94	12.390	16.479	36.983	12.218	**0.124**	**0.330**	**0.587**
	95	6.552	13.052	33.353	7.990	**0.109**	**0.240**	**0.474**
	96	10.041	14.376	31.107	8.957	**0.130**	**0.288**	**0.609**
	97	10.407	15.236	32.567	7.747	**0.191**	**0.238**	**0.636**
	98	18.562	20.956	41.525	17.307	**0.116**	**0.417**	**0.672**
	99	9.179	17.715	37.530	14.618	**0.102**	**0.389**	**0.516**
(B)	93/99	10.315	16.019	35.122	10.554	**0.169**	**0.300**	**0.577**
	93	11.139	18.089	42.456	10.875	**0.203**	**0.256**	**0.548**
	94	12.219	16.479	36.490	12.218	**0.139**	**0.335**	**0.589**
	95	6.585	13.052	33.426	7.990	**0.106**	**0.239**	**0.474**
	96	9.685	14.376	30.105	8.957	**0.167**	**0.298**	**0.616**
	97	9.815	15.236	30.904	7.747	**0.255**	**0.251**	**0.648**
	98	18.233	20.956	40.595	17.307	**0.142**	**0.426**	**0.677**
	99	8.652	17.715	36.057	14.618	**0.147**	**0.405**	**0.520**

Concentration are all in μeq/l

AP: [NO_3^-+nss-SO_4^{2-}], NP: [nss-Ca^{2+}+NH_4^+]

In evaluating the extent of acidification, several indexes have been reported for assessment. These include the ratios of NO_3/nss-SO_4 index for evaluating the condition of acidification, [nss-Ca+NH_4] which was defined as neutralizing potential (NP) by Tsuruta (1989) and is an index demonstrating the effect of air masses of continental origin (Fujita *et al*., 2000) and [nss-SO_4+NO_3] as acidifying potential (AP) which is an index demonstrating the effect of human activities. These values are calculated in Table 2-13. The condition of acidification as revealed from the ratio of NO_3/nss-SO_4 increases from about 0.24 in 1993 to 0.40 in 1999 in both stations (Table 2-13). The ratio of NO_3/nss-SO_4 is much less than one. This indicates that the main acid contributor in the Hong Kong rainwater is still SO_4, which has an annual vw-mean concentration about three to four-fold that of NO_3 in terms of H equivalent in both stations (Table 2-13). From Table 2-13 and base on the Na estimator, the annual vw-mean concentration of nss-SO_4 varies around 30 ueq/l over the years. The annual vw-mean of NO_3, however, increases from about 8 ueq/l in early 90s to about 12 ueq/l in late 90s at KT. For CW, the annual vw-mean of NO_3 also increases from about 10 ueq/l in early 90s to about 14 ueq/l in 1999. The increase in the ratio of NO_3/nss-SO_4 is therefore contributed by the increase in NO_3 over the years. This agrees with the findings so far that NO_3 is playing a more and more contributing role to rainwater acidity. No specific trend is observed in the NP/AP ratio, which signifies the influence of base ions (nss-Ca and NH_4) of continental origin over the anthropogenic acidifying species of NO_3 and nss-SO_4. Rain acidification results when $NP<AP$ or $NP/AP < 1$. From 1993 to 1999, the value of NP/AP fluctuates around 0.70 in Kwun Tong and 0.60 in Central/Western. This indicates that the condition of $NP<AP$ leading to acidification. In other words, anthropogenic emission of NO_3 and nss-SO_4 are not totally neutralized by the continental originated base cations in the air mass. Comparing with other East Asian countries, Hong Kong is slightly better than Tokyo, which has an average ratio of 0.51 and positioned in the moderate group among the 18 East Asian cities with respect to the NP/AP ratio and with reference to the 1993 data (Fujita *et al*., 2000).

2.4 Loadings of SO_4 and NO_3 in relation to wind direction

A simple estimation for the source origin of the acidic species is the association of the SO_4 and NO_3 level in relation to wind direction. Although wet deposition accounts for about 70% of the total deposition (Table 2-6), the rainout or washout event associated with wet deposition is more spatially restricted to the space or air column directly above the receptor and less susceptible to the movement of air mass in the atmosphere. The gases, aerosols and particulates associated with dry deposition, on the contrary, are more susceptible to movement of air mass. Dry deposition data from the year 1993 to 1999 was, therefore, extracted into two groups of wind direction, North (N) and South (S) recorded in Wagland Island and with wind speed greater than 2 m/s. North covered the direction from 315^{o} to 45^{o} while South covers 135^{o} to 225^{o}. The t-test was employed to test whether significant difference found in the SO_4 and NO_3 level with respect to wind direction in the two stations. Results are summarized in Table 2-14. Spatial variation is observed. In KT, there are no significant difference in the SO_4 and NO_3 level regardless of the wind direction (Table 2-14A). Only the NO_3 in 1999 shows a significant higher level in northerly wind conditions. In CW, SO_4 is only found significantly higher in northerly wind than the southerly in 1998 and 1999 while NO_3 was found higher in northerly condition in the year 1995, 1996 and 1999 (Table 2-14B). The consistency found among the data points showing significant difference is that higher SO_4 and NO_3 loading was associated with northerly wind. This could not be conclusive yet since only 6 out of the 28 data points shows such significance.

It would be rather difficult and inaccurate to infer sources of origin from solely the deposition data. As discussed before, the two stations show spatial and temporal variations as a result of various interacting factors such as the meteorological conditions particularly the microclimates, the nature of the land use and human activities, and the topological conditions. Acid deposition can be viewed as the end-point of a series of complex interactions among the atmospheric chemical species of natural and anthropogenic origins. Some of these species could be oxidized sequentially to more acidic forms as a result of the chemical, meteorological and physical interactions while others could be neutralized or remain unchanged. In tracking and differentiating the local (within the territory) and regional (across the border) sources and assess their apportionment, it is recommended to employ a more vigorous modeling approach for atmospheric transport to tackle the problem. The model should be integrated in nature in accounting the relevant physical, meteorological and chemical phenomena of the species covering the source emissions, dry and wet deposition removal and the potential chemical transformation in the atmospheric conditions.

2.5 Hong Kong’s position

To conclude, the acid rain phenomenon is observed in Hong Kong over the years of 1993 to 1999. The acid rain condition in Hong Kong is just moderate when compared with other major cities (Table 2-15). Rain acidity in Hong Kong in terms of pH is moderate (KT-4.87 and CW-4.75) in 1999 and comparable with Taiwan (4.8-5.1) and Compton of London (4.9). It is less acidic than West Point of New York (4.47) and Woburn of London (4.6).

In terms of the extent of acidification as indicated by the ratio of NO_3/nss-SO_4, the relative contribution of NO_3, likely from traffics, has been increasing in temporal sense from about 0.23 to 0.40 from 1993 to 1999 (Table 2-13). Hong Kong is still amongst the lowest when compared with other metropolis such as Tokyo (0.71), New York (0.50) and London (0.85-0.99).

Rain acidification is also reflected from the ratio of neutralization potential (NP)/acidifying potential (AP) as discussed before. Rain acidification results when NP<AP or NP/AP<1. The high value of NP/AP (1.18 or NP>AP) in Beijing is explained by the common sand storm phenomenon, which is rich in crustal Ca (Fujita *et al*., 2000). London had NP/AP value greater than 1 (1.04 and 1.36) as well indicating a strong influence of continental base cations. Hong Kong has a NP/AP value in the range of 0.52 to 0.88 in the year 1998 and 1999. The average NP/AP value of the two stations is 0.78 in 1998 and 0.58 in 1999. This is similar to Tai Chung in Taiwan (0.70) and slightly better than Tokyo (0.51) but much better than New York (0.24).

Table 2-14. Results of the t-test on the dry deposition SO_4 and NO_3 level with respect to wind direction in (A) Kwun Tong and (B) Central/Western, significant difference indicated in bold.

(A) Kwun Tong			SO_4 (μeq/l)			NO_3 (μeq/l)		
	Direction Range	n	Mean ± SD	t	p	Mean ± SD	t	p
1993	N	6	234.0±236.4	-1.285	0.217	38.1±22.5	-0.790	0.441
	S	12	348.2±143.3			50.6±35.1		
1994	N	4	174.5±115.0	-2.043	0.068	47.1±48.1	-0.200	0.846
	S	8	340.4±139.5			51.1±22.0		
1995	N	13	234.7±311.9	-0.194	0.848	71.2±92.8	0.925	0.367
	S	8	256.9±96.4			40.0±23.2		
1996	N	11	146.0±82.5	-0.526	0.605	44.9±31.4	-0.092	0.927
	S	9	164.7±74.2			46.0±18.3		
1997	N	10	148.7±75.9	1.232	0.231	57.1±27.5	0.176	0.862
	S	14	114.2±61.4			54.9±33.0		
1998	N	11	132.9±55.4	-1.271	0.222	71.9±37.2	0.128	0.899
	S	7	181.1±106.3			69.0±61.5		
1999	N	20	113.0±83.2	1.992	0.054	54.5±29.2	**2.560**	**0.015**
	S	16	68.1±38.7			31.8±22.5		
(B) Central Western								
1993	N	5	138.1+67.5	1.616	0.126	30.3+9.2	0.692	0.499
	S	13	90.1+52.2			24.3+18.0		
1994	N	2	124.9+23.6	-0.185	0.860	67.1+10.1	1.476	0.190
	S	6	130.5+38.9			34.3+29.5		
1995	N	11	169.2+94.7	1.805	0.089	53.8+22.4	**3.580**	**0.002**
	S	8	104.3+41.7			21.8+13.5		
1996	N	9	137.0+45.2	1.123	0.278	46.3+14.8	**2.921**	**0.010**
	S	9	110.3+55.3			28.3+11.2		
1997	N	5	144.9+41.5	-0.222	0.827	43.4+11.0	-0.570	0.576
	S	16	161.5+163.2			58.7+58.8		
1998	N	18	170.6+77.6	**2.950**	**0.007**	72.1+31.9	1.697	0.103
	S	7	77.3+47.4			42.9+53.2		
1999	N	17	163.2+138.5	**2.642**	**0.014**	84.1+77.6	**2.210**	**0.036**
	S	12	54.3+38.1			32.1+28.4		

n: valid sample number in the year

N: defined as wind direction in the range of 315° to 45°

S: defined as wind direction in the range of 135° - 225°

Table 2-15. The rain acidity in major cities

Major City[*]	Year	Rain pH (vw-mean)	$NO_3^-/nssSO_4^{2-}$	NP/AP[#]
Hong Kong	**1998**	**KT: 4.60**	**0.39**	**0.88**
		CW: 4.48	**0.43**	**0.68**
	1999	**KT: 4.87**	**0.38**	**0.63**
		CW: 4.75	**0.41**	**0.52**
Beijing	1993	6.0	0.32	1.18
Taiwan	1999	4.8-5.1[+]	No data	0.70 (Tai Chung, Fujita *et al.*, 2000)
Tokyo	1993	4.51	0.71	0.51
West Point (New York)	1999	4.47	0.50[++]	0.24[+++]
Woburn (North of London)	1999	4.6	0.99	1.04[++++]
Compton (South of London)	1999	4.9	0.85	1.36[++++]

*Beijing, Taiwan and Tokyo: data from Fujita *et al.*, (2000) Precipitation chemistry in East Asia.

Taiwan: data (except NP/AP) from Taiwan EPA, 1999 (http://www.epa.gov.tw/news/en880728-1.htm)

West Point (New York): data from USEPA, 1999 (http://nadp.sws.uiuc.edu/nadpdata/)

Woburn and Compton; data from The United Kingdom Precipitation Composition Monitoring Network, 1999 (www.aeat.co.uk/netcen/airqual)

[#] NP/AP: neutralizing potential/acidifying potential, same definition as Table 2-13, nss values for Hong Kong were from Na estimator (Table 2-12)

[+] annual average as reported

[++] total SO_4 used for the calculation

[+++] total Ca and SO_4 used for the calculation

[++++] total Ca used for the calculation

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