Significant Marine Source for SO_2 levels in Hong Kong

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Abstract

Understanding the source of pollutants is essential for formulating appropriate policy measures to reduce ambient levels and lower the associated health impacts. A study of concentrations of SO_2 has been performed in Hong Kong. The work reported here develops an improved pollution rose, called circular pollution wind map (CPWM), and combines it with both principal component analysis (PCA) and singular value decomposition (SVD) techniques to analyze the pollutant data recorded in 14 air quality monitoring stations and wind data from two automatic weather stations in Hong Kong.

Variations of SO_2 over the Hong Kong Special Administrative Region of China (HKSAR) are discussed from the standpoint of monthly variations, inter-annual variabilities and spatial distributions, as well as wind effects. We identified two dominant modes of SO_2 variation. The first is associated with weak southerlies and southwesterlies, and is localized around the Kowloon peninsula. Analysis of speciated PM_{10} data shows that this mode is associated with the nearby combustion of residual fuel oil from marine sources in the vicinity of the large container terminal at Kwai Chung (a kilometer or so to the west of Kowloon).

What makes this mode especially important is that the pollution is released close to ground level and within a few kilometers of some of the highest population densities in the world (with a total nearby population over three million). In contrast, emissions from the other major <u>local</u> source, coal burning power plants, are released quite high into the air from which point they are substantially diluted before reaching sensitive receivers. Since the health risks associated with SO_2 and other pollutants such as PM_{10} are directly related to the concentration in which they reaches sensitive receivers, the significance of the local marine sources is of considerable importance for polices to reduce the health impacts of local air pollution. Yet, most attention has been focused on the power plant (with the push for installation of flue gas desulphurization by the government), while no controls are being imposed on the quality of fuel ocean going cargo ships may burn while in port, nor the fuel used by coastal vessels while in Hong Kong waters.

A second mode of SO_2 variation is associated with moderate to strong northwesterlies, and affects most of the territory of the HKSAR. Analysis of speciated PM_{10} data indicates that this mode is associated with a mixture of sources, but mainly related to the regional transport from the northwest (from the Pearl River Delta across the border in mainland China). This second mode indicates that the local source of SO_2 is not the only source which needs to be addressed. Nonetheless, our focus here is on the local emission of SO_2 by marine vessels (along with other hazardous pollutants like nickel, vanadium and elemental carbon) from near ground level close to a large proportion of the population of the HKSAR, where its healthy impact is likely to be greater than the relative quantity of SO_2 released might at first seem to suggest.

The proportion of the total SO_2 concentrations contributed by each of the source regions was quantitatively estimated by SVD. It is found that residual oil combustion from marine vessels around Kwai Chung terminal container port and local power plants are responsible, respectively, for 36% and 7% of the total SO_2 concentrations measured at general air quality monitoring stations in Hong Kong. However, with regard to specific locations in the heart of the urban area, local SO_2 sources, in particular marine vessels burning high sulphur residual fuel oil, appear to be the major source. And it is this source that is the primary focus of attention in this paper.

Keywords: residual oil combustion; PM_{10} ; power plant; PCA; terminal container.

Introduction

Hong Kong is a booming international port and urban metropolis, with a densely packed population of about 7 million people. It has the typical local and regional air pollution problems associated with most large cities. Specifically, air quality is seriously affected by nitrogen dioxide (NO_2), ozone (O_3), and particulate matter with aerodynamic diameter smaller than 10 μm (PM_{10}), [also referred to as respirable suspended particulates (RSP) in Hong Kong].

As is the case in many other urban cities, levels of sulphur dioxide (SO_2) in the HKSAR have generally been decreasing in response to mandates for the use of higher quality diesel fuel (especially in transport) and structural changes in the economy from manufacturing to services. For example, SO_2 has not been widely produced in Hong Kong except from power plants since the banning in 1990 of high sulphur fuel oil by industry (EPD, 1991).

Yet, SO_2 levels remain a problem, with average annual levels at the monitoring stations for 2000-2002 ranged between about 10 and 24 μ/m^3**.** Further, study of SO_2 levels offers important opportunities for tracing *local* and *regional* polluted air masses. Through this work we will show that ground-level SO_2 measured in Hong Kong is now primarily produced regionally in the heavily industrialized areas of the Pearl River Delta and, from marine vessels locally while they are resident in the harbour around Hong Kong.

Levels of SO_2 in Hong Kong have been studied in the past. For example, Cheng and Lam (1998) followed the concentration of SO_2 at three stations in Hong Kong over a nine-year period. They found that high concentrations of SO_2 were related to SSW-WNW winds, and attributed the source of the SO_2 pollution to power plants, industry, and diesel motor vehicles in Hong Kong. A similar study of five sites in 1996 (Wang et al., 2001) associated the transport of SO_2 with strong northwest winds from the Pearl River Delta as well as contributions from local power plants. More recently, Liu and Chan (2002) analyzed a pollution episode on December 29 & 30, 1999 and concluded that the high concentrations of SO_2, PM_{10}, and NO_2 came mainly from vehicles and power plants in Hong Kong. Nevertheless, these were mainly emission based estimates, and there has not really been any direct chemical source apportionment studies relating power plant emissions to air quality measurements.

Yet, Hong Kong is the world's largest container port, with annual throughput of 20.45 TEU in 2003 (Notteboom, 2004; TEU = Twenty-foot container Equivalent Unit). While emissions from fuel oil combustion on board container ships are known to be a significant source of SO_2 (Corbett et al. 1999, Streets et al. 2000), its importance in Hong Kong remained unclear. Table 1 summarizes the SO_2 local emission inventory for Hong Kong (HKEPD, 2003) from 1990 to 2000. The data show a decrease in emissions from power plant and motor vehicles but an increase in emissions from marine sources. Further, we note that pollutants from power plants are released and at a high level over 200m (Hong Kong Electric 2003, CLP Power 2003) with increased buoyancy while pollutants from marine sources are released only a few tens of meters above sea level. In term of ground-level SO_2 impact, it is unclear whether the power plants or marine sources would be more significant.

The objective of this study is to use chemical speciated data to identify significant sources of SO_2 in Hong Kong. In so doing, we will also better understand the role of marine SO_2 sources, and the spatial and temporal extent of these marine emissions.

2. Data

As shown in Figure 1 the Hong Kong Environmental Protection Department (HKEPD) operates 14 air quality monitoring stations (AQMSs) throughout the HKSAR. Each station is either 'general' or 'roadside'. Roadside stations are located at street level to capture the air quality characteristics as experienced by pedestrians, while 'general stations' are located on rooftops, and often in elevated parts of the urban area and are intended to indicate ambient air quality. There are 3 roadside stations in Hong Kong located at Mongkok (MK) Kowloon, and Central (CL) and Causeway Bay (CB) on the north shore of Hong Kong Island; they are in area of both very high levels of road traffic and dense population. Hourly measurements of standard criteria pollutants, including SO_2, NO_2 and PM_{10}, etc. are taken at each the AQMS. Here we examine the hourly SO_2 data from 2000 to 2002.

In addition to real time hourly measurements, EPD does 24-hour high-volume filter-based sampling of particulate matter at the AQMSs once every six days. Analysis of the resulting PM_{10} samples provides details of the particulate composition, which we have examined (for 1999-2001) for the 25 stable species shown in Table 2. The 9 general AQMSs for which speciated PM_{10} data were obtained are: Tung Chung (TC), Yuen Long (YL), Tsuen Wan (TW), Kwai Chung (KC), Tai Po (TP), Central Western (CW), Shatin (ST), Shum Shu Po (SP) and Kwun Tong (KT). The high-volume filter-based sampling were not taken for the stations at Tap Mun (TM) and Eastern (EN).

Wind measurements were taken at both Waglan Island (WGL) operated by the Hong Kong Observatory and Tap Shek Kok (TSK) operated by the Hong Kong University of Science and Technology near opposite ends of the HKSAR (Figure 1). Both sites provide hourly wind speed and direction measurements, and have data available for the period of 2000 to 2002.

3. Winds and SO_2 over Hong Kong

3.1 Winds

Waglan Island (WGL) is a rural and remote island situated in the southeastern corner of the HKSAR. Winds measured there are considered representative of the large-scale winds prevailing over Hong Kong. The percentage frequency of hourly winds measured in different directions for Waglan Island averaged over the period 2000 to 2002 is shown in Figure 2a. The majority of winds (over 70%) come from the north, northeast, or east. The less frequent southerly and southwesterly winds (about 15% of the total) occur mainly in the summer in association with the Asian monsoon. Compare with urban wind stations, the winds at Waglan are typically stronger, with a much higher frequency of occurrence for wind over 4 m/s (the white bars in Figure 2a).

In contrast, the Tap Shek Kok wind station located in an open area at the western side of Hong Kong has much weaker winds with the majority less than 4 m/s as shown in Figure 2b. Noting the directions of background winds measured at Waglan Island (north, north-east, and east) the same winds arrive at Tap Shek Kok after encountering the urban high rise setting of Hong Kong, with accompanying modifications. At this location, weak southeasterly winds are most frequent, followed by easterly, northeasterly, and northerly winds. While winds from other directions, namely the west, southwest and northwest are less common (about 20% of the total), these directions are important, as they are often associated with the transport of pollutants from the Pearl River Delta region into Hong Kong and a worsening of air quality (see sections 3 and 4).

The diurnal variations in speed and direction of hourly wind data averaged over the period of 2000 to 2002 are shown in Figure 3 for both Waglan Island and Tap Shek Kok. For the background measurements of Waglan Island, stronger winds (6 to 7 m/s) moving steadily from the east are observed, with minimal diurnal variation. In contrast, winds at Tap Shek Kok are weaker (2-3 m/s) but show significant diurnal variations. The early easterly winds in the morning rotate in a clockwise direction from about 10 am local time, turning north and northwest in the afternoon, southwest and south in the late afternoon, and finally back to easterly winds around 6 pm in the evening.

The presence of such a clear diurnal cycle at Tap Shek Kok makes it an ideal site for monitoring the heat island effect associated with atmospheric conditions in Hong Kong. Despite the low frequency of west winds noted in Figure 2b, Figure 3 shows a clear occurrence of daily west winds, which are stronger than winds from other directions at about 3 m/s. The strength of the westerly winds suggests that the diurnal land-sea breeze circulation could be responsible for transporting pollutants from the Pearl River Delta into Hong Kong.

3.2 SO_2 variations

The concentrations of SO_2 in Hong Kong as shown in Figure 4, are averaged over a 3-year period (2000 to 2002). Some monthly variations of SO_2 concentration are evident, with a peak occurring in the summer months of July and August, a secondary peak in January, and an annual minimum in October. The geographical spread of SO_2 levels from 2000 to 2002 is shown in Figure 5a and 5b for all 14 AQMSs across the HKSAR. Levels are highest at Kwai Chung (KC) as well as in urban areas such as Tsuen Wan (TW), Shum Shu Po (SP) and Central Western (CW), and at the roadside stations, Mongkok (MK), Central (CL) and Causeway Bay (CB). Concentrations of SO_2 are lowest at Tap Mun (TM) the most remote site, located in a rural area in the northeast of Hong Kong as indicated in Figure 5b.

Diesel fuel above 0.5% sulphur was banned in industrial use in Hong Kong since 1990, eliminating what was, until then, the dominate local source of SO_2 (EDP, 1991). If the most significant source of SO_2 emissions remaining is diesel powered motor vehicles, we would expect the highest measurements from roadside stations, which is not the case. Similarly, if SO_2 from the Pearl River Delta accounts for most of SO_2, we would expect the north-western site at Yuen Long (YL) to record the highest SO_2 levels, which it has not. However, the highest level of SO_2 is found at Kwai Chung (KC), the station adjacent to the container terminal (Figure 1). Perhaps most tellingly, if regional transport is most important, we would expect the annual peak to occur in winter, not in the summer when the prevailing background winds are southeasterlies.

The yearly mean SO_2 concentrations at each AQMS are shown in Figure 6. Some interesting inter-annual variations evident over the three years examined. SO_2 levels decreased substantially at the three roadside stations (Central CL, Mongkok MK, Causeway Bay CB). This corresponds well to the introduction of ultra low sulphur diesel (ULSD) in 2001, which reduces the statutory tolerance limit of sulphur from 0.035% to 0.005% in Hong Kong (HKEPD, 2004). At Yuen Long (YL) which is most sensitive to pollution transported from the Pearl River Delta region, the steady decrease of SO_2 levels may indicate changes in industrial energy use and or stricter emission controls in mainland China. In contrast, there is an increase in SO_2 concentration in a number of sites, especially Kwai Chung (KC) that shows not only the highest concentration, but also an increasing concentration of SO_2 over the entire study period up to 2002.

3.3 Relationship between Winds and SO_2 (circular pollution wind map)

The relationship between winds and SO_2 concentrations from 2000 to 2002 in Yuen Long is summarized in the circular pollution wind map in Figure 7. The wind speeds and directions are measured at Tap Shek Kok (TSK), with wind speeds indicated by the dotted concentric circles at 5, 10, and 15 m/s and wind direction given by angle. The colors in the figure indicate the average SO_2 concentration measured at Yuen Long for different wind speed-direction pairs at TSK.

Frequency of occurrences of winds at particular speeds and directions are shown by the contours. The area bounded by the innermost thick black contour corresponds to the top 25% wind speed-direction pairs in terms of frequency of occurrence, and moving outward, the other contours bound the most frequent 50%, 75% and 95% wind speed-direction pair at TSK. In other words, there is a less than 5% chance to observe any wind speed-direction pair at TSK outside the outermost contour in Figure 7. Hence, the wind frequency contours maybe used as a proxy measure of the uncertainty of each wind speed-direction pair under consideration.

As indicated by the red areas in Figure 7, the highest concentrations of SO_2 (over 40 $\mu g/m^3$) measured at Yuen Long are associated with *moderate* northwesterly wind. Generally the SO_2 levels are higher when the wind has northerly or westerly components, while more frequent winds from the south, southeast, and east carry concentrations of SO_2 lower than 20 $\mu g/m^3$. This shows elevated SO_2 levels at Yuen Long are associated, not with weak winds carrying pollutants from local sources, but moderate northwesterly winds of 5 to 10 m/s. This suggests that transport (and not local sources) is the most important factor contributing to the high SO_2 levels at Yuen Long. In particular, high SO_2 levels are associated with northwesterly transport, pointing to the significance of SO_2

sources in the Pearl River Delta to the northwest of Yuen Long. (Measurements such as the high value of SO_2 found at 11 m/s and 240 degrees, are not considered indicative since they lie outside the top 95% frequency contour, and thus are an quite infrequent occurrence.)

To further show the importance of northwesterlies in the transport of SO_2 over Hong Kong, the circular pollution wind maps with respect to the winds recorded at Tap Shek Kok for SO_2 observations at all the AQMSs is plotted in Figure 8. The panels are placed in rough accordance with the locations of the sites over Hong Kong. The wind frequency contours are shown on each plot as well as in the lowest empty panel for clarity. As indicated by the red and yellow regions on the circular plots, higher levels of SO_2 are associated with moderate to strong northwesterly winds at almost all stations, not just in Yuen Long (YL). An exception is the remote northeastern station at Tap Mun (TM), which has low levels of SO_2 regardless of winds. The prevalence of high SO_2 levels for moderate northwest winds for nearly all stations shows that transport of SO_2 by northwesterlies from the Pearl River Delta affects virtually all of the HKSAR, including even urban areas such as Central (CL) and Causeway Bay (CB). The strongest northwest transport signature is seen for Tung Chung (TC). Beside the Pearl River Delta sources of SO_2, Hong Kong's Chek Lap Kok airport also lies to the immediate northwest of Tung Chung. The airport has been identified as an important source of SO_2 by Yu et al. (2004). The high concentration of SO_2 at Tung Chung for moderate northerlies shown in Figure 8 may be related to a combination of transport of SO_2 from the Pearl River Delta and that of the airport.

However, besides moderate northwesterlies, Figure 8 shows that high levels of SO_2 are also associated with weak southwesterlies for a number of AQMSs including Kwai Chung (KC), Shum Shu Po (SP), Tsuen Wan (TW), Kwun Tong (KT), and Central Western (CW). These stations are mostly in the city center area around the harbor. In particular, the highest levels of SO_2 are found at Kwai Chung are associated with *weak* winds (below 5 m/s) from the south and west, suggesting the significance of *local* source(s) to the south and west of Kwai Chung. It was this insight which led us to realize that, while regional transport sources of SO_2 are important, local sources can also be very important. Noting that Hong Kong's large container terminal is situated just to the south-southwest of the Kwai Chung AQMS (see Figure 1), we suspected that the increases in SO_2 during weak southwesterly winds was associated emissions from marine sources (including both ocean going vessels while in port, and coastal vessels which service the larger ships and the port area). The presence of elevated SO_2 levels at AQMSs around the harbor further suggests the significance of emissions from marine vessels. In subsequent sections, we shall attempt to verify this hypothesis using statistical and chemical speciation techniques.

4. Contributing factors for SO_2 variations

Principal component analysis (PCA) is routinely used to identify the dominant modes of variations in atmospheric and environmental datasets. Generally speaking, PCA is a linear method to analyze the variability of the internal structure of the dataset. Sometimes, it is performed as linear data reduction method, which extracts the few dominant modes from the measured variable and these modes then used to explain the whole dataset.

We carried out detailed examinations of the variability of SO_2 levels in the hourly data from 2000 to 2002 for all 14 AQMSs. First, the time series data from each station were normalized, and PCA was performed on the correlation matrix (as opposed to the covariance matrix) to better indicate overall pollutant transport patterns. Further investigation was undertaken using rotated principal component analysis (RPCA) on the same data set. Singular value decomposition (SVD) was also used in conjunction with the speciated particulate data, in order to identify the exact source of the pollutants. Further references of the methodology and applications on PCA, RPCA and SVD can be found in Richman (1986), Bretherton et al. (1992) and Hartmann (2002).

4.1 Principal Component Analysis (PCA) of SO_2

A plot of the percentage variance of the 14 PCA modes from the hourly SO_2 data is shown in Figure 9. The first four PCA modes explain 46.2%, 10.0%, 8.0% and 6.3% of the total variance respectively, with the first three modes well above the one-fourteenth level of 7.1%. The error bars in the figure are associated with the North criteria, which is associated with 95% confidence of the eigenvalues; details can be referred to Hartmann (2002). The analysis indicates that the first mode is distinct from the following modes, explaining a large proportion of the variance.

The first PCA mode explaining 46% of the total variance from the 2000 to 2002 hourly data for SO_2 at 14 AQMSs in Hong Kong is shown in Figures 10a through 10d. The eigenvectors, or ‘station loading vectors’ are plotted in Figure 10a. The spatial distribution of SO_2 over Hong Kong shown in Figure 10c.

It is clear that the station loading values (Figure 10c) for this mode are similar at all sites except at Tung Chung (TC) and Tap Man (TP), indicating that the first PCA mode is associated with coherent territory-wide variations of SO_2. We then averaged and plotted the hourly time-series from the PCA analysis for each month and show it in Figure 10b. Although the data are too short to allow for definitive conclusions about the long term trend, the 2000-2002 period does show an *apparent decrease.*

A circular pollution wind plot of the hourly PCA time-series and hourly wind conditions at Tap Shek Kok are shown in Figure 10d, indicating the wind conditions associated with variations of SO_2 for the first PCA mode. It shows that this mode is associated with moderate to strong northwesterlies *and* weak to moderate southwesterlies when SO_2 levels are enhanced. Thus, this mode is related to both of the wind speeds and directions previously linked to high SO_2 levels.

The second PCA mode (which accounts for 10% of the total variance) is shown in Figure 11. The station loading vectors in Figures 11a and 11c suggest that the SO_2 levels associated with the second mode coherently around the Kowloon peninsula near Kwai Chung (KC), Tsuen Wan (TW), Shum Shu Po (SP), Mongkok (MK) and Shatin (ST). The circular pollution map in Figure 11d shows that increases in SO_2 in this second PCA mode are related to the occurrence of weak south and southwest winds. As indicated in the monthly averaged time-series shown in Figure 11b, the second PCA mode is strongest in the summer. The prevalence of *weak* winds from the south and southwest, as well as the clustering of the affected sites around the Kowloon peninsula to the north and east of Kwai Chung (KC) suggest that this mode is associated with *local* SO_2 sources in or around the harbor area.

4.2 Rotated Principal Component Analysis (RPCA) of SO_2

The analysis in the previous section reveals a fundamental limitation of the PCA method because of the orthogonal requirement of identified modes, which often results in mixing of the underlying modes (Richman, 1986). The presence of both northwesterlies and southwesterlies in the first mode and the emergence of weak southwesterlies in the second mode suggest mixing of modes has occurred. Following Richman (1986), varimax rotation was applied to the first two PCA modes and the resulted RPCA modes are shown in Figures 12 and 13. As expected, the rotated modes are better-separated (i.e. more localized spatially) in the physical sense.

The first rotated mode accounts for about 30% of the total variance, and Figures 12a and 12c show that the mode is centered around AQMSs in the city center around the harbor, specifically Kwai Chung (KC), Shum Shu Po (SP), Tsuen Wan (TW), Shatin (ST), Central Western (CW) and Mongkok (MK). The monthly average time-series in Figure 12b shows seasonal peaks in the summer months, and the circular pollution wind map in Figure 12d indicates that weak south and southwest winds are associated with enhanced levels of SO_2 for this RPCA mode. Structurally, this mode is similar to the second PCA mode of the unrotated analysis, and is likely related to local SO_2 marine emissions at or near the container terminal adjacent to the Kwai Chung site as was discussed earlier (see also chemical speciation analyses in the next section).

About 26% of the total variance is explained by the second RPCA mode, depicted in Figure 13, which is only slightly less than the first RPCA mode. In contrast to the first mode, the second mode is localized at all sites away from the Kowloon peninsula, namely Yuen Long (YL), Tung Chung (TC), Tai Po (TP), Kwun Tong (KT), Tap Mun (TM), Eastern (EN), Central (CL) and Causeway Bay (CB). The monthly averaged RPCA time-series shows a seasonal cycle which is stronger in the winter and weaker in the summer. The circular pollution map indicates that this mode is associated with the transport of SO_2 by moderate to strong northwesterlies (Figures 13 b and d), suggesting that this mode is associated with regional transport of SO_2 from Guangdong. A general decreasing trend can also be seen in Figure 13b, indicating that the relative importance of this northwesterly transport mode has been decreasing over the period 2000 to 2002.

4.3 Speciated SO_2 and PM_{10} data at Kwai Chung

To help identify the pollution sources contributing to SO_2 levels measured at the AQMSs, simple receptor modeling was carried out using chemical speciation data from 24-hour high volume PM_{10} samples collected by the HKEPD every six days at the urban sites. Kwai Chung (KC) was chosen as a specific site of interest due to the highest measured SO_2 levels, as well as the probable highly localized sources of pollutants. Daily average SO_2 concentrations from PM_{10} sampling days at Kwai Chung were combined with the speciated PM_{10} data for the 25 chemical species indicated in Table 2, and PCA methodology was applied.

The eigenvectors from the first PCA mode, which is associated with 41% of the total variance, are shown in Figure 14a. This mode is related to large loads of most of the analyzed species, with the exception of SO_2. Hence, the first PCA mode is not of primary interest in the present study. The relatively equal proportion of various species indicates that this mode may be a mixture of a number of different pollution sources**.**

The second PCA mode is related to 21% of the total variance, with the eigenvectors plotted in Figure 14b. There is notable coherent variation present in only a few chemical species, namely nickel (Ni), vanadium (V), elemental carbon (EC), barium (Ba), and sulphur dioxide (SO_2). It is well known (Watson et al, 1998) that vanadium, nickel, and elemental carbon together with sulphur dioxide are ‘signature compounds’ for residual oil combustion. Since residual oils are banned for land-based used in the HKSAR but commonly used on marine vessels (Streets et al. 2000), this supports the view that the main source of SO_2 at Kwai Chung is from marine vessels at or near the container terminal.

4.4 Singular Value Decomposition of speciated PM_{10} and SO_2

Finally, to confirm that residual oil combustion associated with marine sources is actually an important factor contributing to the SO_2 pollution in the HKSAR, the singular value decomposition (SVD) technique is applied to study the correlation between the speciated PM_{10} and the SO_2 datasets. Unlike PCA, which is focused on the identification of dominant variations within a dataset, SVD analysis is useful for identifying coupled (correlated) modes in two datasets (Bretherton et al., 1992; Newman et al., 1995). The coupled mode will appear as singular vectors in the analysis. Thus, SVD was undertaken using daily averaged SO_2 readings and the available speciated PM_{10} data for 9 AQMS sites, Tung Chung (TC), Yuen Long (YL), Tsuen Wan (TW), Kwai Chung (KC), Tai Po (TP), Shatin (ST), Central Western (CW), Shum Shu Po (SP) and Kwun Tong (KT)). Once again, only 25 chemical species were considered, chosen for their chemical stability (Table 2). Our interest is in finding the modes in the SO_2 and speciated PM_{10} time series that have the highest correlation with each other.

Figure 15 shows the percentage covariance value in SVD modes. The results from the first SVD mode, which explains 36% of the correlation between the SO_2 and speciated PM_{10} data sets, are shown in Figure 16. The loading scores for first singular vector of PM_{10} for each chemical species at each measurement site are plotted in Figure 16a. The 9 lines shown for each species correspond to the 9 AQMS sites ordered from left to right: Tung Chung (TC), Yuen Long (YL), Tsuen Wan (TW), Kwai Chung (KC), Tai Po (TP), Shatin (ST), Central Western (CW), Shum Shu Po (SP) and Kwun Tong (KT).

Figure 16b shows on the ordinate the loading scores of the first singular vector of SO_2 for each AQMS site. The pair of singular vectors in Figure 16 shows that SO_2 variations at Kwai Chung (KC), Tsuen Wan (TW), Shatin (ST), Shum Shu Po (SP), Mongkok (MK) and Kwun Tong (KT) are associated with residual oil combustion signatures (i.e. prominence of V, Ni, EC in Figure 16a). In addition, the residual oil combustion signatures are also associated with marine sea-salt signatures (Na^+ and Cl^-). The prominence of the loading vectors (Figure 16b) near Shum Shu Po (SP) and Kwai Chung (KC) is consistent with the hypothesis that the residual oil combustion is related to marine vessels at or near the Kwai Chung container terminal**.** It is also important to note that the overall structure of the first PM_{10} singular vector in Figures 16a and 16b (i.e. the relative amplitude of the dominant species) is similar to the second PCA mode shown in Figure 14b, and the SO_2 singular vector is similar to the first RPCA mode shown in Figure 12. This shows that the coupled variations identified by the SVD technique are each important modes of variation in SO_2 and PM_{10}.

Plots of the spatial distribution of the loading scores for the first SO_2 singular vector, and V, Ni and EC in the first PM_{10} singular vector are shown in Figures 17 a to d respectively. It is clear that the area of dominance for this mode for all these four species is the urban city area around the Kwai Chung (KC). This spatial distribution confirms the presence of a significant residual oil combustion source in or around the container terminal vicinity. More importantly, because of the large number of people residing in this area (over 3 million), Figure 17 highlights the significance of marine emissions as an pollution source impacting on a large portion of the local population.

To understand the seasonal variation of the first SVD modes, monthly averages were computed from the expansion coefficients of the two singular vectors. Figures 16c and 16d depict the monthly average expansion coefficients of the first singular vector for PM_{10} and SO_2 respectively. The first SVD mode is clearly strongest in the summer and weakest during the winter, suggesting that SO_2 levels associated with maritime residual oil combustion are more significant in summer. This seasonal difference may be related to the more common occurrence of southwesterlies during the summer. The southwesterlies are more effective in bring the marine pollutions towards the city area where most of the affected AQMSs are located.

Looking at the subsequent SVD modes, Figure 15 shows the percentage covariance explained by each SVD mode, with the first mode that we have attributed to residual oil combustion by far the largest covariance at 36%. The second, third and fourth singular modes accounted for 17%, 15% and 7% of the covariance between PM_{10} species and SO_2, respectively. From chemical speciated results, we found that the second SVD mode also have substantial contributions of Na^+, Mg^+ and Cl^- (details not shown, see Wu 2003), suggesting it may be partially mixed with the first mode (similar to the mixing of the first two PCA modes in section 4.1). In contrast, the third mode is more prominent at Yuen Long (YL), Tung Chung (TC) and Tai Po (TP), and is identified as being related to northwesterly transport of PM_{10} and SO_2 into Hong Kong from the Pearl River Delta. Finally, the fourth PM_{10} and SO_2 SVD mode accounted for only 7% of the total covariance (not shown, see Wu 2003). However, interestingly, this mode is characterized with high levels of Selenium and Arsenic which are signature elements of coal fire power plant emissions. The results suggests that in terms of primary SO_2 pollution measured at the ground level, the impact of marine sources is much more significant than that of the coal fire power plants.

5. Summary

A variety of statistical methods have been used here to study SO_2 variations in the HKSAR. Two dominant modes have been identified. One is associated with weak south and southwest winds, and is localized near Kwai Chung (KC), Shum Shu Po (SP), Tsuen Wan (TW), Shatin (ST), Central Western (CW) and Mongkok (MK). Analysis of speciated PM_{10} data show that this mode is associated with local SO_2 emissions from the combustion of residual oil combustion by marine sources in and around the container terminals in Kwai Chung.

According to EPD data, maritime SO_2 emissions have risen moderately but steadily in recent years from about 3,100 tons in 1996 to about 3,300 tons in 2000, making marine pollution sources an ongoing concern. Residual fuel oil is of very high sulphur content and generally of poor quality (e.g., with respect to viscosity and expected completeness of combustion; Streets et al. 200). The fact that the marine combustion of low quality fuel and emission takes place in relatively close proximity with a large percentage of the local population means that the environmental health implications of this combustion from nearby local pollution sources are likely to be far more serious than the absolute output level might otherwise suggest. For example, using Hong Kong Census data, Lau et al. (2004) estimated that over 3.8 million people live in the urban airshed directly affected by emissions from the container port.

Marine pollution may be controlled through the use on-shore electric power connections or by mandates to use low sulphur diesel fuel while a ship operates (or generates its own power) while in port. However, the solution for Hong Kong probably lies in mandates for the use of higher quality fuel since much of the container off-loading takes place at anchorages rather than at docks, and also because of the extensive use of coastal marine vessels for moving containers from anchorage to shore. One option would be for Hong Kong to work with the Mainland Chinese government to declare the entire Pearl River Delta as a Sulphur Emission Control Area (SECA) (Arthur Bowing, personal communication, 28/02/05); this is particularly important considering the rapid growth in population and also container throughput in the Pearl River Delta.

Besides marine emissions, the second mode of SO_2 variation is associated with moderate to strong northwest winds, and affects most of the AQMS sites in Hong Kong. Analysis of speciated PM_{10} data suggest that this mode is associated with a mixture of various sources, and mainly related to its transport from areas to the northwest of Hong Kong. Finally, our analysis did not identify road vehicle or power plants emissions as important sources of contribution to the local SO_2 levels in Hong Kong. This is in contrast to earlier work and reflects the effectiveness of environmental policies measures introduced over recent years. Emissions of SO_2 from vehicular sources has decreased from about 6,199 tons in 1996 to 1,121 tons in 2000 (Table 1). This result matches well with decreasing levels of SO_2 at the roadside AQMS sites as well as our overall findings.

For power plants, there has been a steady decrease in SO_2 emissions from 72,799 tons in 1996 to 56,803 tons in 2000 (Table 1). Although this is still much larger than the marine and vehicular sources in overall amount, due to the height and the temperature of the stack gases release, such emissions are mostly carried away from Hong Kong by the upper level winds, and they may not reach ground level receptors as easily or effectively as those from marine sources. In short, our study shows that power plant emissions are not a major factor affecting the local SO_2 concentrations in Hong Kong. In particular, signature species for power plants (e.g. selenium and arsenic) were only found in the 4th SVD mode, accounting for only 7% of the total covariance (compared with 36% due to marine vessels).

Present levels of SO_2 in Hong Kong have strong association with hospital admissions and death (Wong et al. 2002). We also know that even moderate reductions in ambient SO_2 levels lead to reductions in the health impacts (Hedley et al. 2003). Hence, it is important to better understand marine emission sources. With regard to environmental health, we need to come to better understand the overall contribution of marine sources of SO_2 to other health significant species (e.g., elemental carbon). With regard to pollution control policies, we need to better understand the cost implications of different options for replacing the use of residual fuel oil by marine ships while they are resident in Hong Kong waters as well as the use of low sulphur fuel by coastal vessels while working in Hong Kong waters. .

6. Acknowledgment

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Figure Captions

Figure 1: Locations of air quality stations and meteorological stations used in this study. The contours shown are in increments of 200 m. (source from Center for Coastal & Atmospheric Research in Hong Kong)

Figure 2: (a) Percentage frequency of hourly winds at Waglan Island (WGL) from 2000 to 2002 for different wind directions and speeds. The black bars correspond to winds less than 4 m/s, and the white bars correspond to winds more than 4 m/s. (b) Same as Figure 2a, but for winds at Tap Shek Kok (TSK).

Figure 3: Diurnal variation of hourly mean wind averaged from 2000 to 2002, at Waglan Island (white bars) and Tap Shek Kok (dark bars); the upper panel shows the wind speed, and the lower panel shows the wind direction.

Figure 4: Monthly mean SO_2 concentration ($\mu g/m^3$) in Hong Kong averaged over 2000 to 2002.

Figure 5: 2000 to 2002 mean SO_2 concentration ($\mu g/m^3$) for the 14 Air Quality Monitoring Stations shown with (a) a bar chart, (b) a spatial plot with the radius shown representing the SO_2 concentration level.

Figure 6: Annual mean SO_2 concentrations ($\mu g/m^3$) at the 14 AQMS from 2000 to 2002; the black, gray and white bars correspond to concentrations from 2000, 2001 and 2002, respectively.

Figure 7: Circular Pollution Wind Map - SO_2 concentration at Yuen Long (YL) in 2000-2002 as a function of wind measured at Tap Shek Kok (TSK). The color-coded SO_2 concentration (in $\mu g/m^3$) is measured at YL, averaged for different wind speeds (given by the radius with 1m/s resolution) and wind directions (given by the angle with 10 degree resolution) measured at TSK. The black concentric circles correspond to winds of 5, 10 and 15 m/s. The contours correspond to 95% (outermost contour), 75%, 50% (thick red) and 25% (thick black) of the measured wind speed and direction pairs.

Figure 8: Circular pollution wind maps for the SO_2 concentrations (in $\mu g/m^3$) for all the 14 AQMSs in Hong Kong, using winds measured at TSK. The panels are placed roughly in accordance with the location of the AQMS site in Hong Kong (e.g., YL in the northwest, TC in the southwest, TM at the northeast). The lowest right panel shows the circular frequency wind map for wind observations at TSK; counting inward from the outermost contour 95%, 75%, 50% and 25% frequency contours are indicated. These wind frequency contours are repeated on the other panels for clarity.

Figure 9: Plot of the percentage variance explained of the 14 PCA modes of the hourly SO_2 data.

Figure 10: The first PCA mode (46% variance) of the 2000-2002 hourly SO_2 data from the 14 AQMS sites in Hong Kong: (a) the eigenvector (or the station loading vector) (b) the monthly averaged PC time-series, (c) a spatial plot of the eigenvector with positive values shown in deep gray and negative values shown in light gray, and (d) a circular pollution wind map combining the winds at TSK and the PC time-series of the first PCA mode.

Figure 11: Similar to Fig. 10, but for the second PCA mode (10% variance) of the 2000-2002 hourly SO_2 data.

Figure 12: Similar to Fig. 10, but for the first RPCA mode (30% variance) of the 2000-2002 hourly SO_2 data.

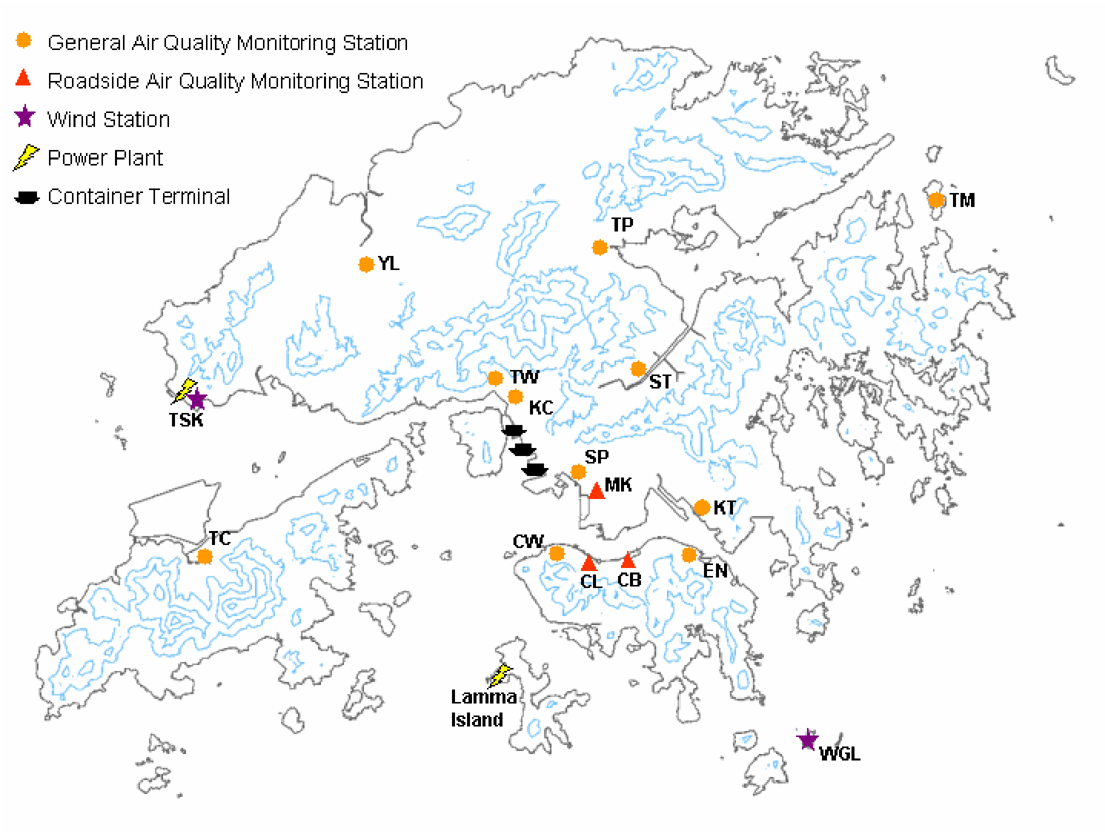
Figure 13: Similar to Fig. 10, but for the second RPCA mode (26% variance) of the 2000-2002 hourly SO_2 data.

Figure 14: The first two eigenvectors of the speciated PM_{10} and SO_2 data at KC: (a) first mode (41% variance) (b) second mode (21% variance).

Figure 15: The SVD covariance spectrum on PM_{10} and SO_2 is plotted.

Figure 16: The singular vectors of the first SVD mode (36% variance) for the speciated PM_{10} and SO_2 data over Hong Kong: (a) the PM_{10} (left) singular vector, and (b) the SO_2 (right) singular vector. Please refer to the text for details.

Figure 17: (a) Spatial distribution of the loading scores for the first SO_2 singular vector (positive values in deep gray and negative values in light gray); (b) Spatial distribution of Vanadium in the first PM_{10} singular vector; (c) Spatial distribution of Nickel in the first PM_{10} singular vector; (d) Spatial distribution of elemental Carbon in the first PM_{10} singular vector.



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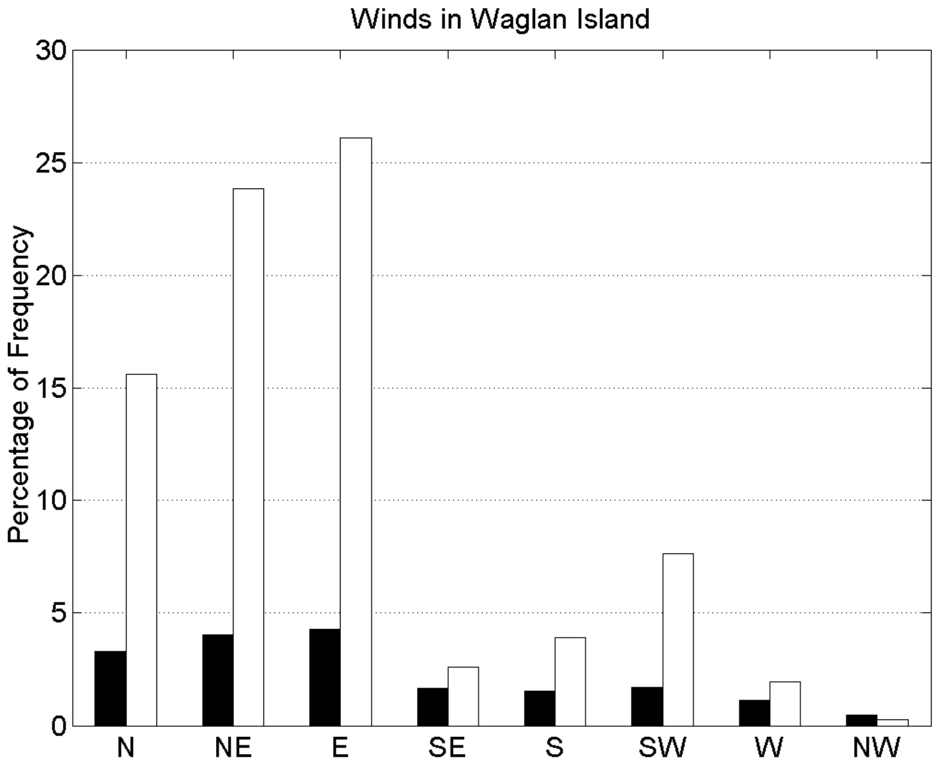

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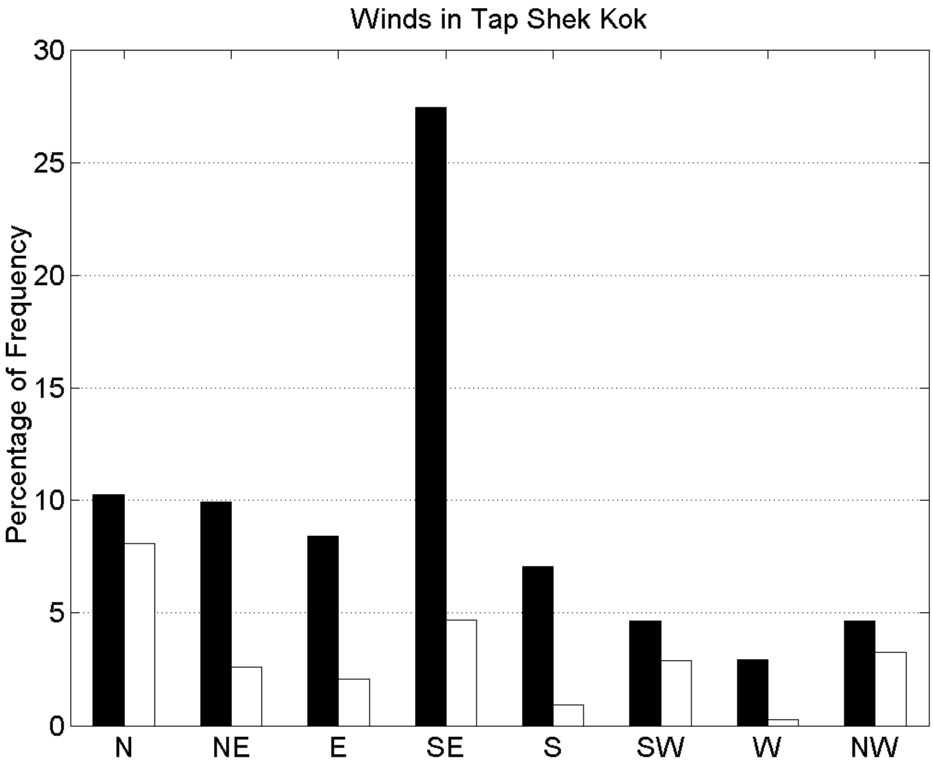

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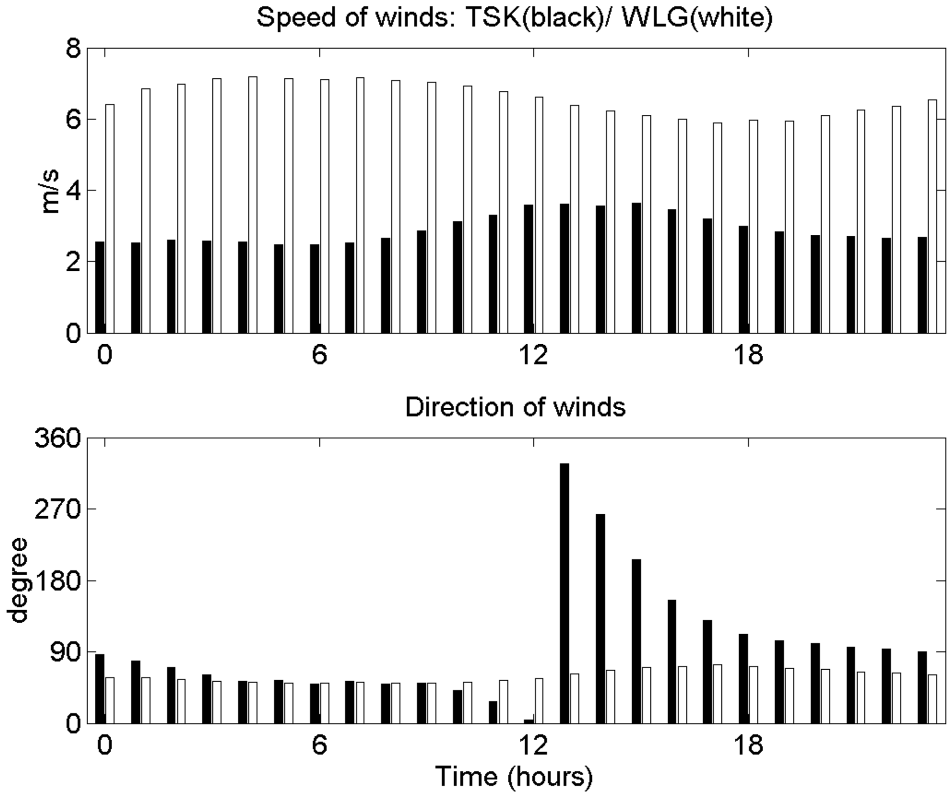

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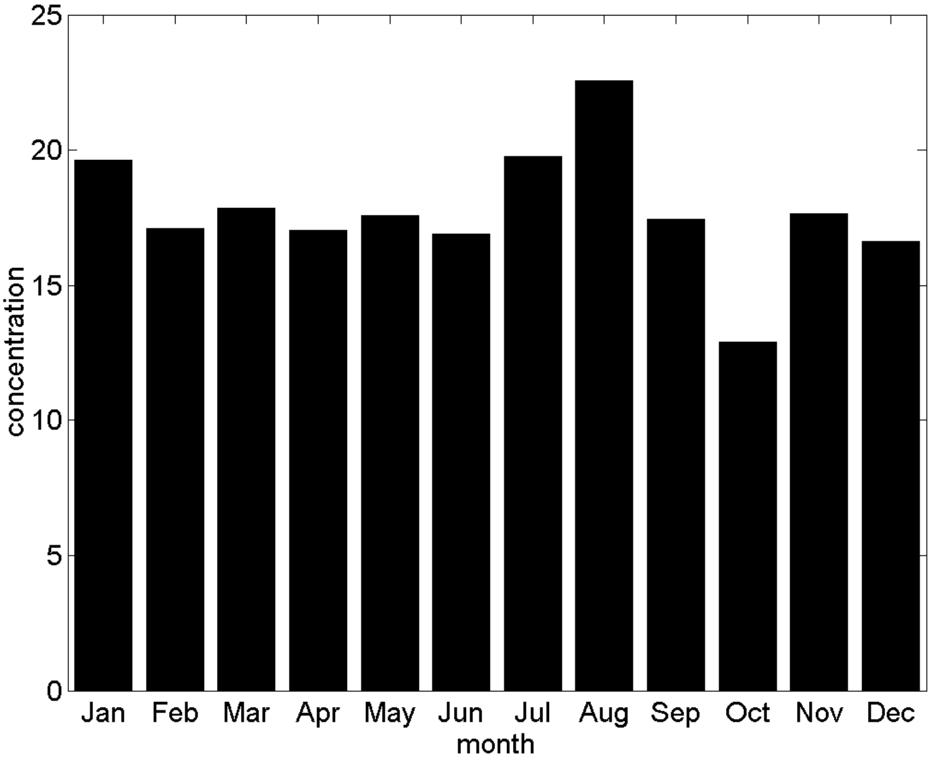

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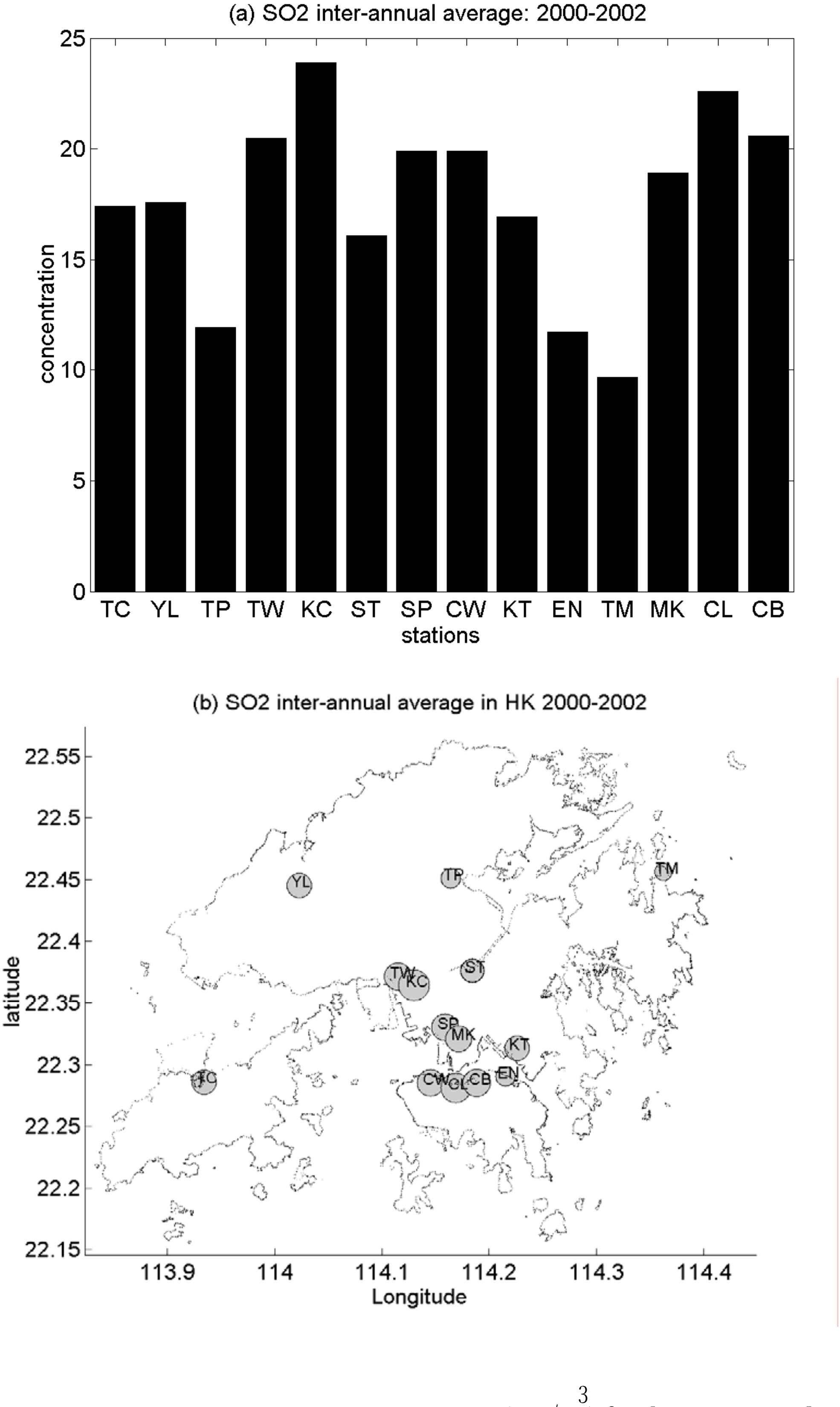

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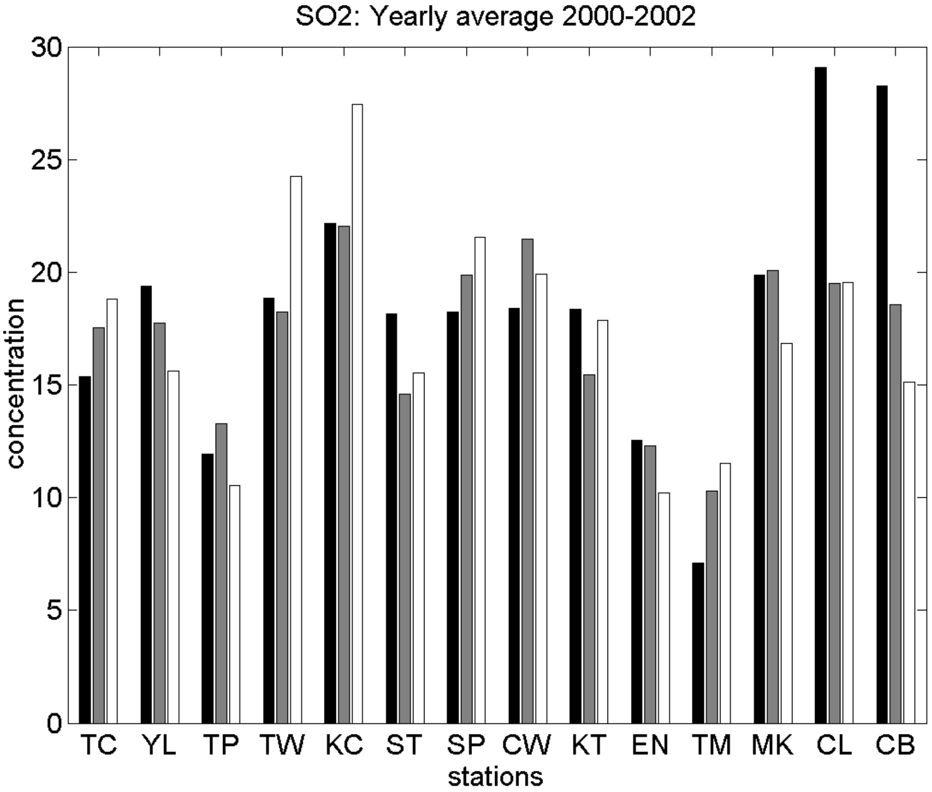

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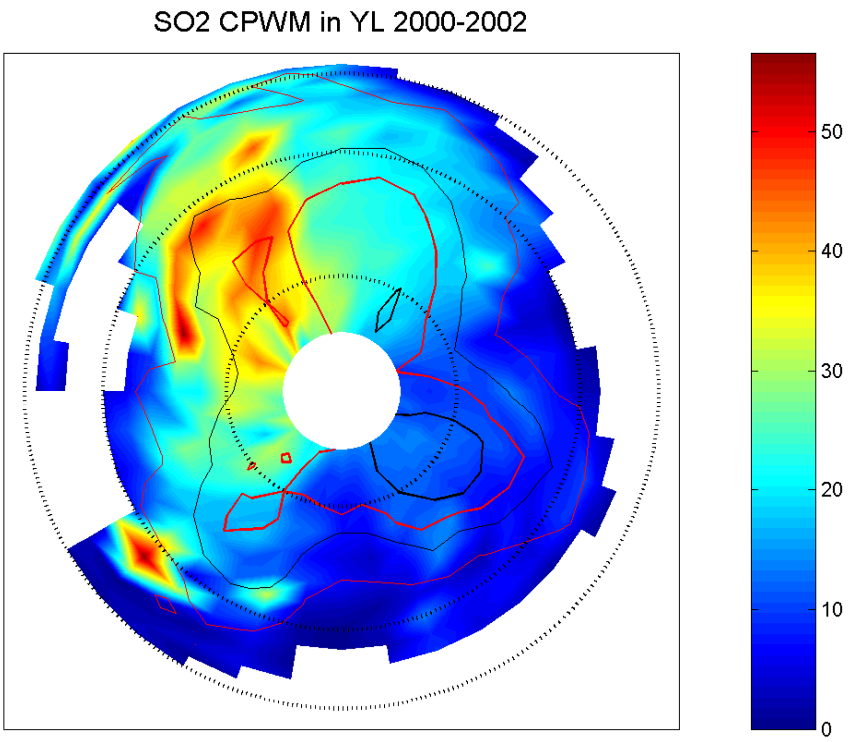

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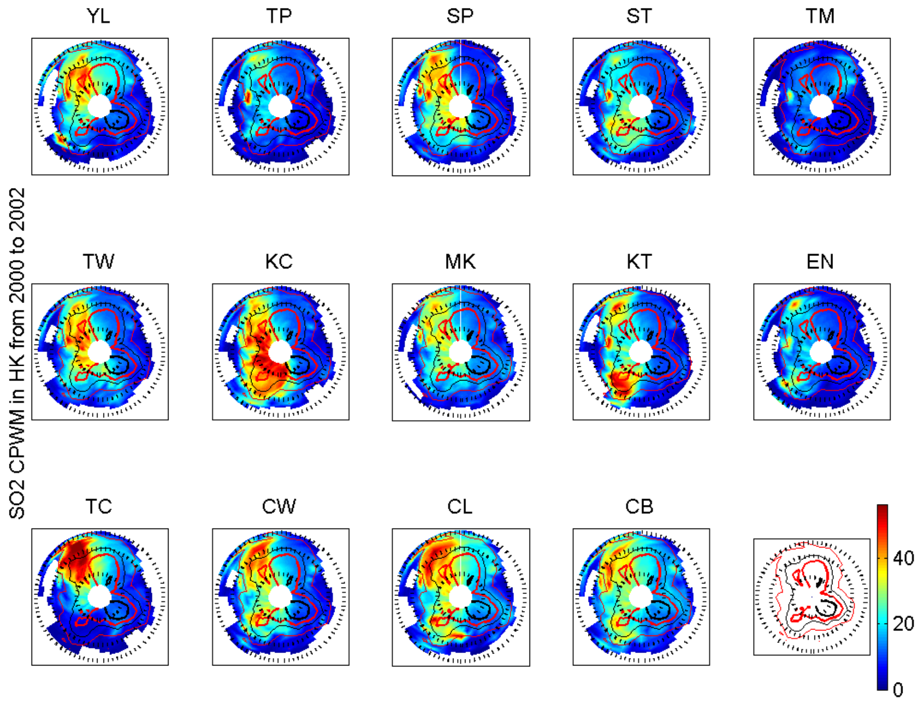

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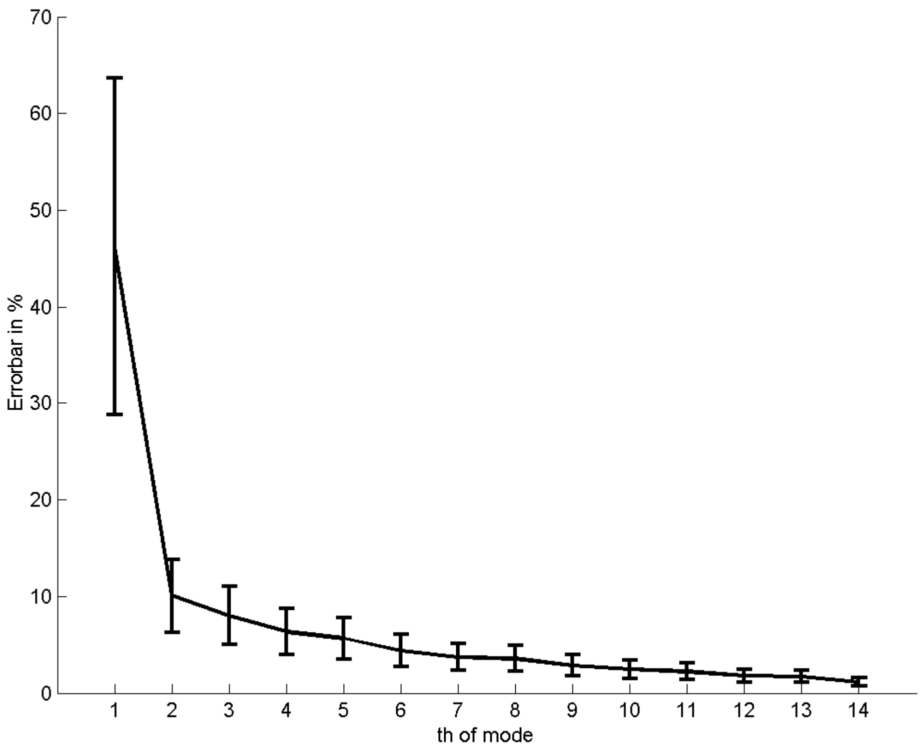

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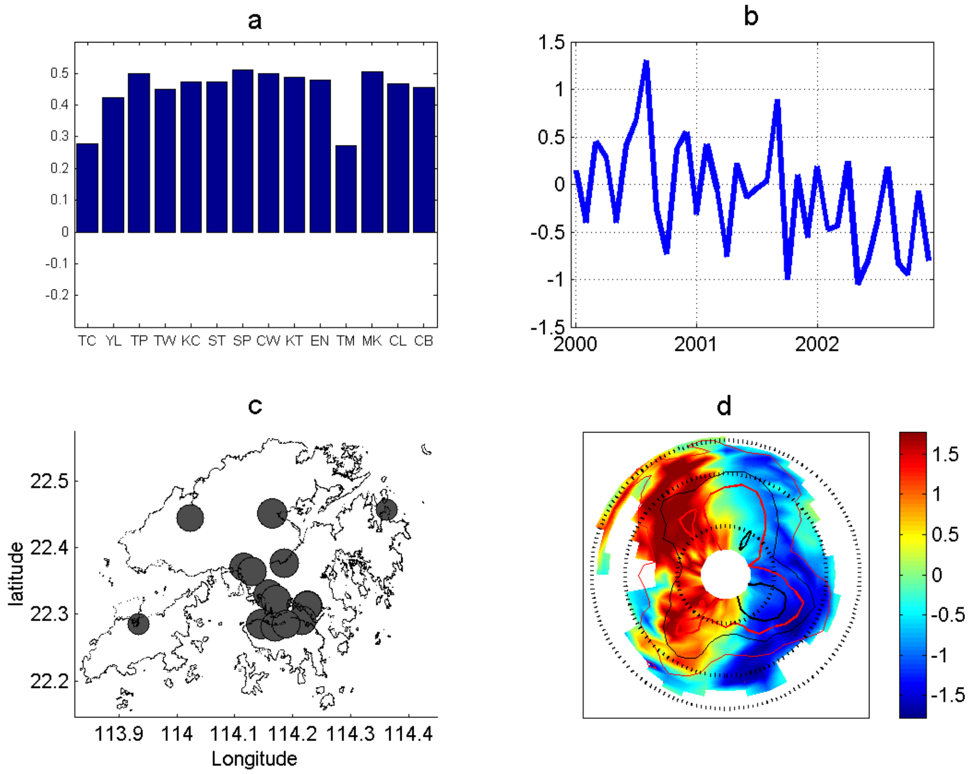

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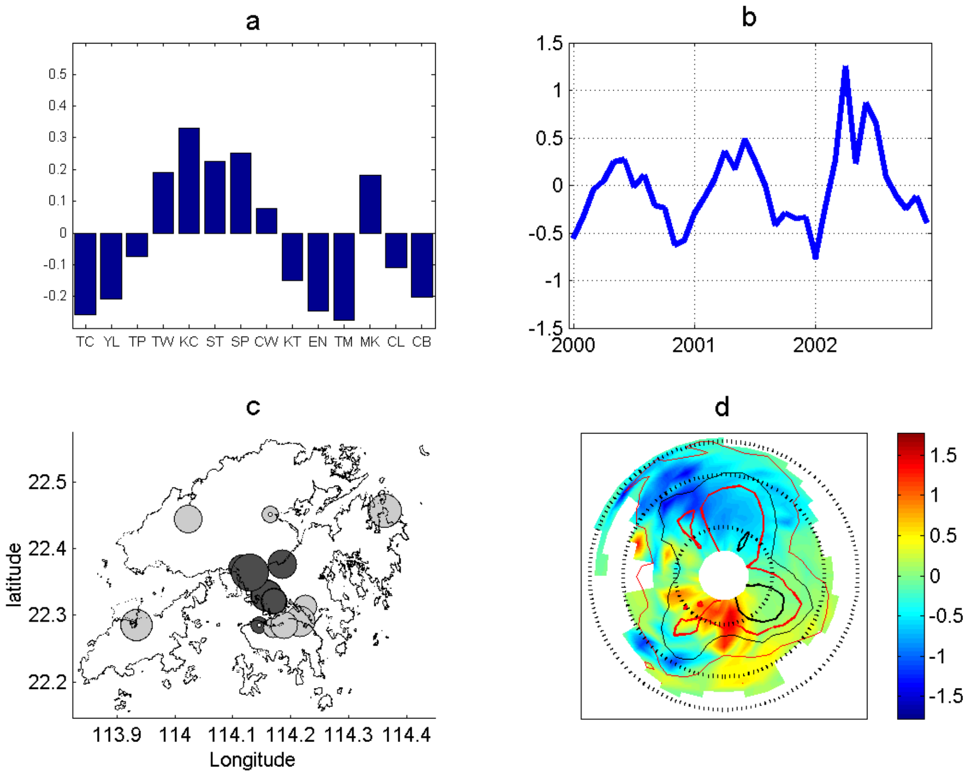

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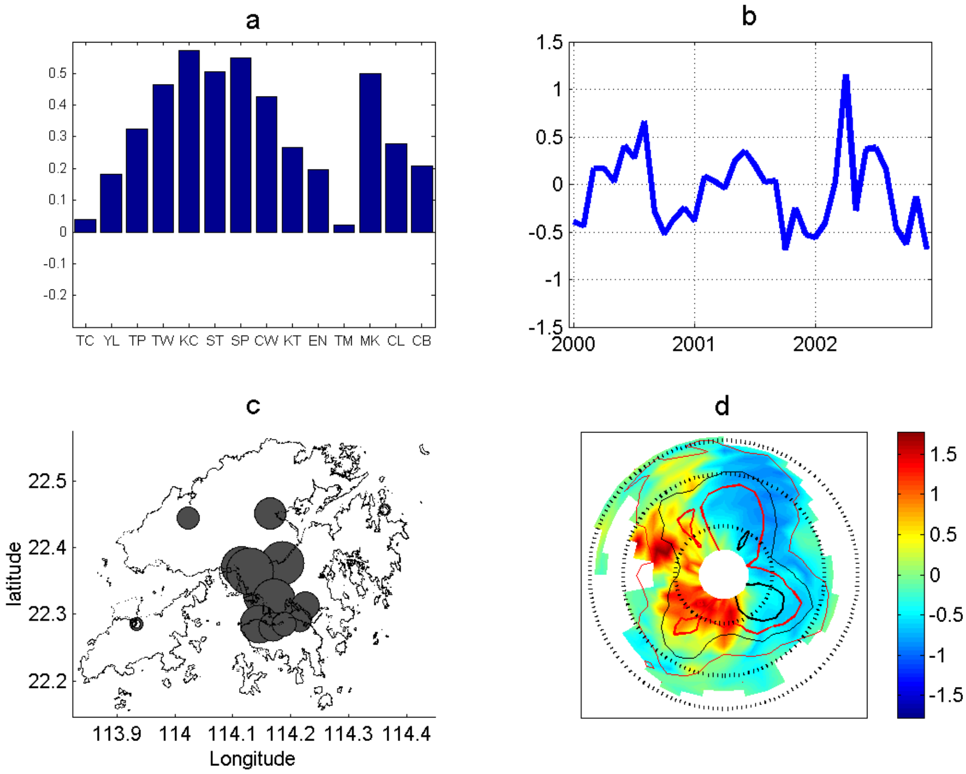

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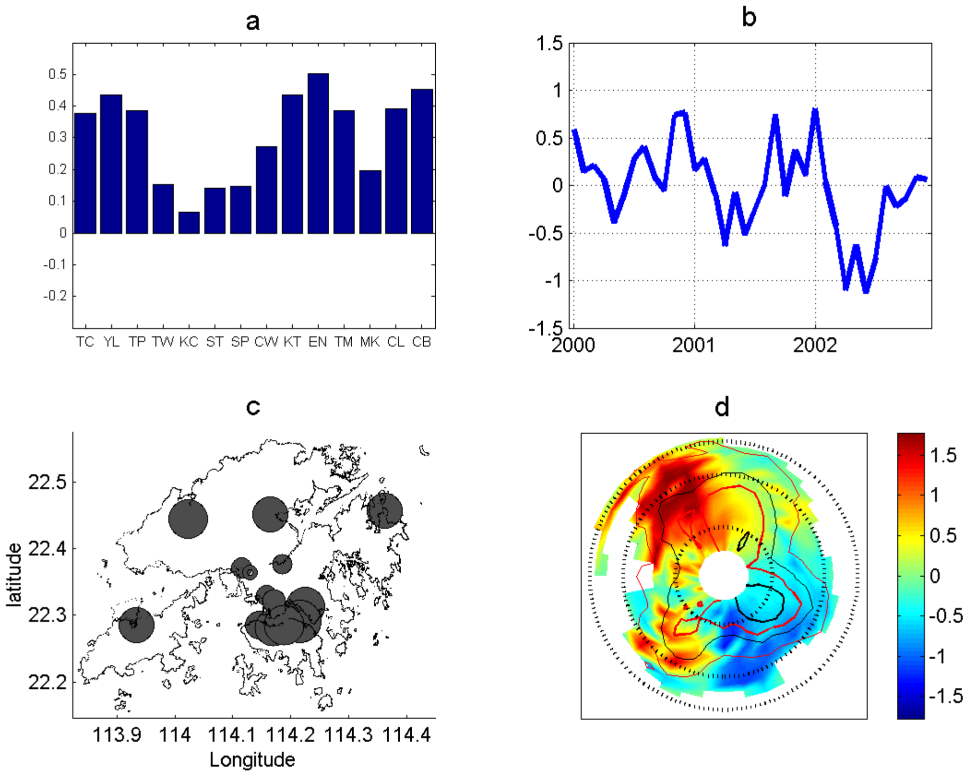

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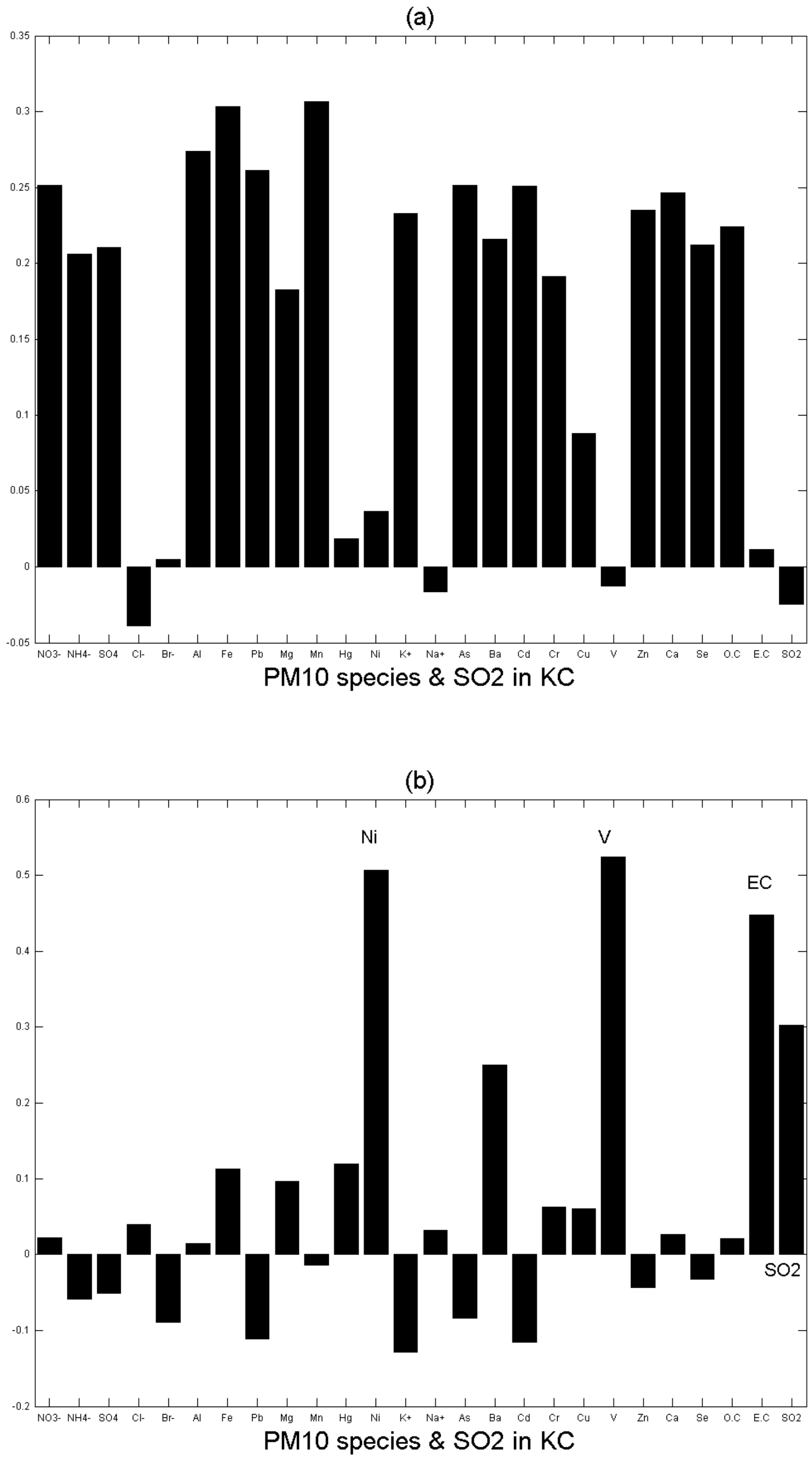

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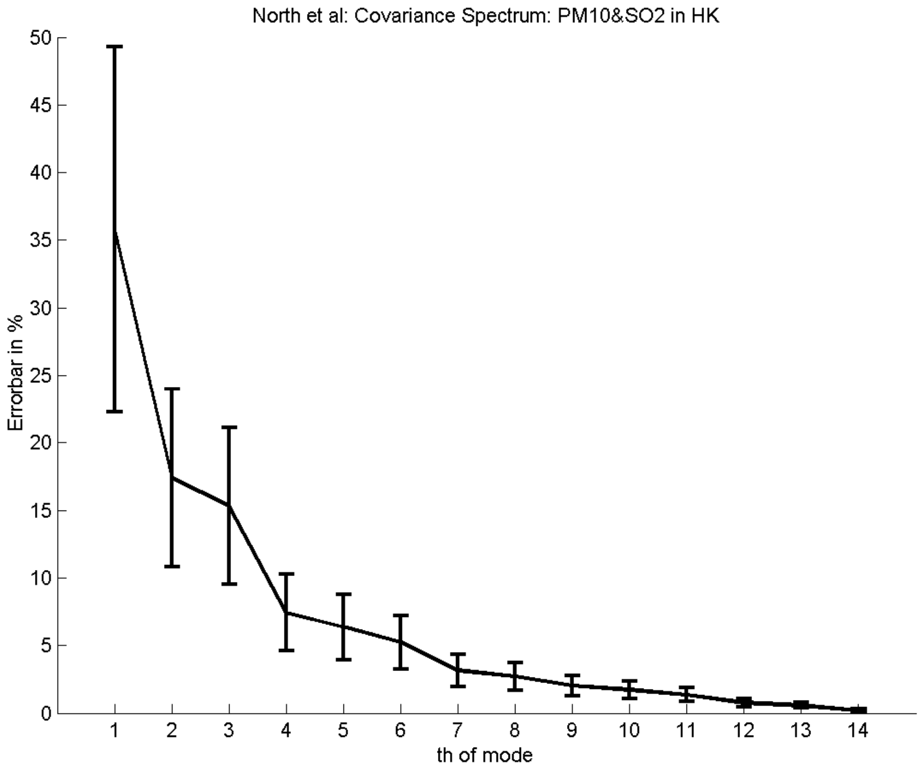

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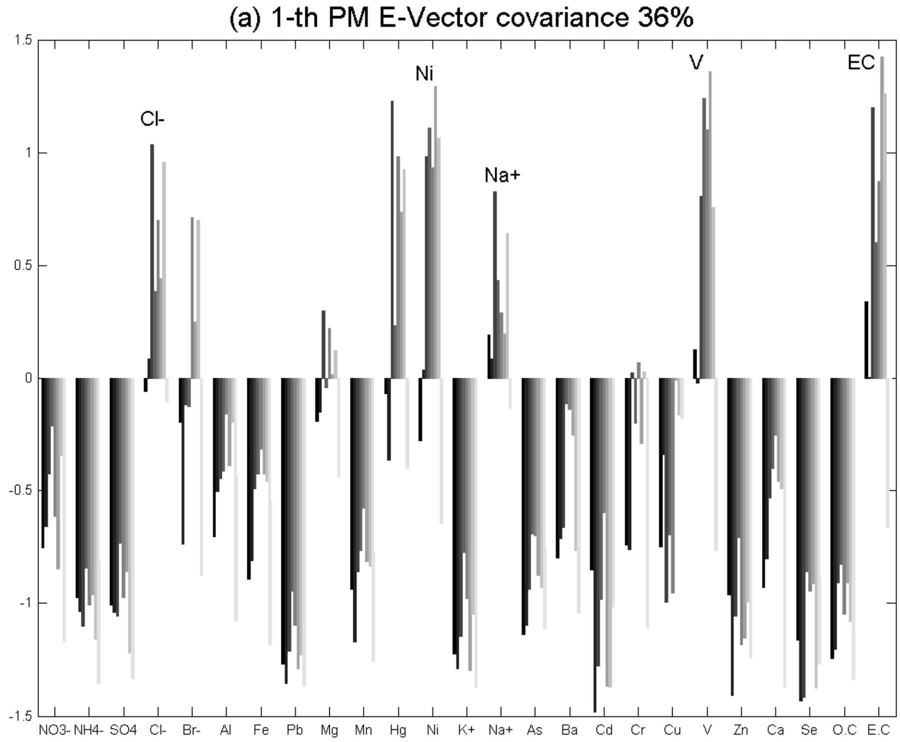
(a) 1-th PM E-Vector covariance 36%
Cl-
Ni
Na+
V
EC
1.5
1
0.5
0
-0.5
-1
-1.5
NO3-
NH4-
SO4
Cl-
Br-
Al
Fe
Pb
Mg
Mn
Hg
Ni
K+
Na+
As
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Cd
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Cu
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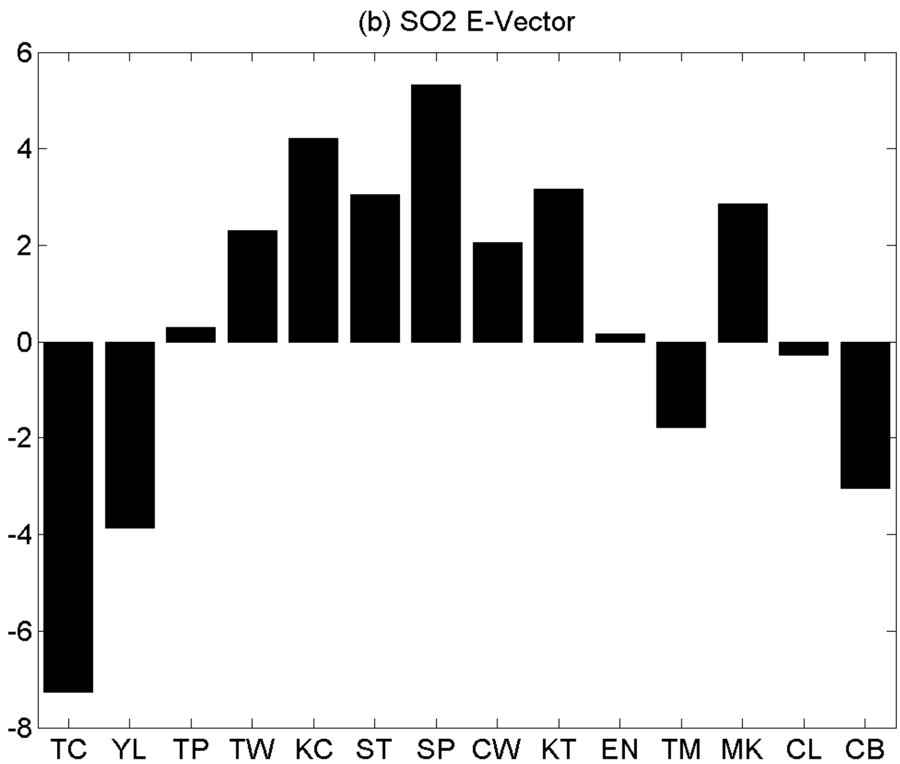
Zn
Ca
Se
O.C
E.C


(b) SO2 E-Vector
6
4
2
0
-2
-4
-6
-8
TC
YL
TP
TW
KC
ST
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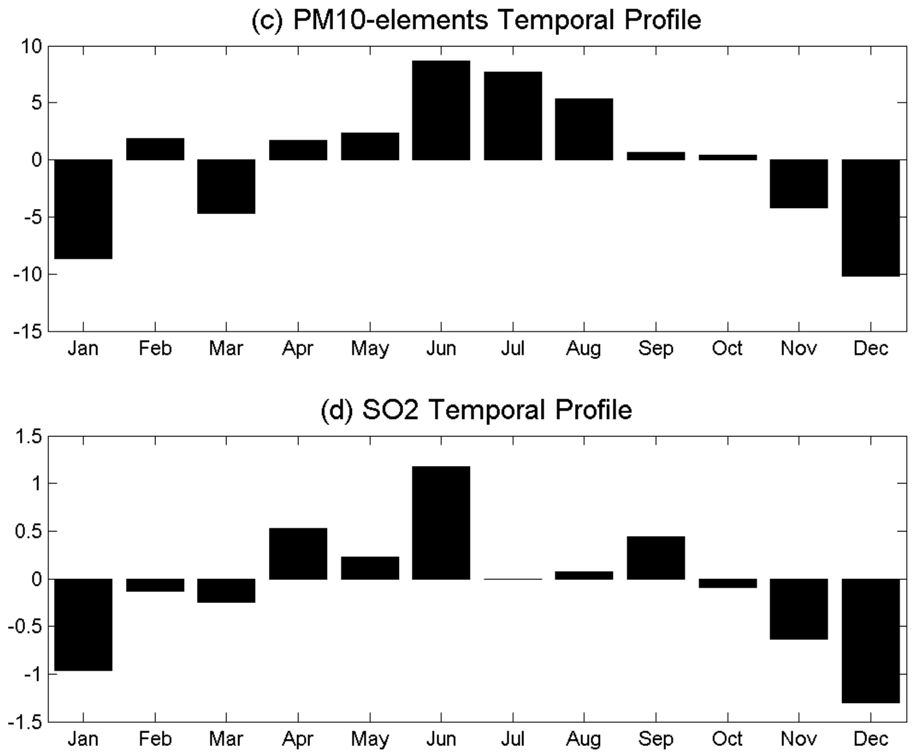
EN
TM
MK
CL
CB



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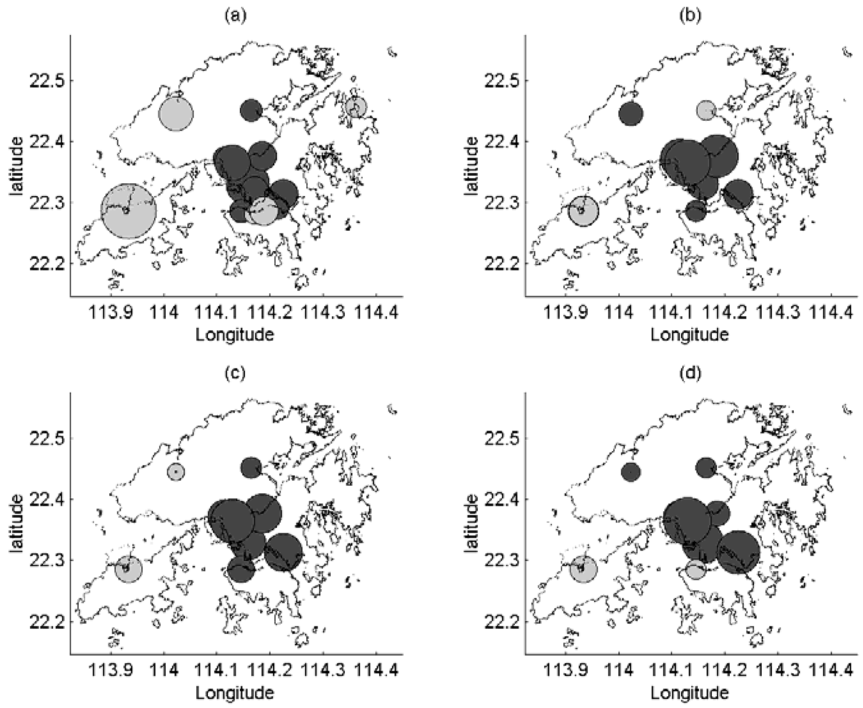

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(c) Spatial distribution of Nickel in the first PM_{10} singular vector;

(d) Spatial distribution of Elemental Carbon in the first PM_{10} singular vector.

Tables

	Power Plant	**Vehicle**	**Marine**
1990	118300	10251	1722
1991	131600	11432	2062
1992	145200	11602	2346
1993	162600	11124	2596
1994	116600	11332	2819
1995	103976	6070	3078
1996	72799	6199	3136
1997	54434	1620	3286
1998	60961	1538	3170
1999	47750	1629	3071
2000	56803	1121	3264

Table 1: SO_2 Emission Inventory in Hong Kong (tonnes)

Chemical	***Abbreviation***	***Chemical***	***Abbreviation***	***Chemical***	***Abbreviation***
Aluminum	Al	Copper	Cu	Organic Carbon	OC
Ammonium ion	NH_4^-	Elemental Carbon	EC	Potassium ion	K^+
Arsenic	As	Iron	Fe	Selenium	Se
Barium	Ba	Ammonium ion	$NH4^-N$	Sulfate	SO4
Bromide ion	Br^-	Lead	Pb	Sodium ion	Na^+
Cadmium	Cd	Magnesium	Mg	Vanadium	V
Calcium	Ca	Manganese	Mn	Zinc	Zn
Chloride ion	Cl^-	Nickel	Ni		
Chromium	Cr	Nitrate ion	NO_3^-N		

Table 2: Chemical species identified in the composition analysis of PM_{10} samples from AQMSs around Hong Kong