

Assessment of Toxic Air Pollutant Measurements in Hong Kong

Final Report

**Submitted
March 2003**

**Revised
November 2003**

Submitted to the
Environmental Protection Department, HKSAR
for Provision of Service
under
Tender Reference AS 01-285

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Acronyms

AQMS	Air Quality Monitoring Station
ACE	Acenaphthene
ACY	Acenaphthylene
Al	Aluminum
ALAPCO	The Association of Local Air Pollution Control Officials
AQO	Air Quality Objective
As	Arsenic
BaP	Benzo(a)pyrene
BbF	Benzo[b]fluoranthene
Be	Beryllium
BeP	Benzo[e]pyrene
BghiP	Benzo[ghi]perylene
BTEX	Benzene, Toluene, Ethylbenzene, and Xylenes
BTX	Benzene, Toluene, and Xylenes
CADAMP	California Dioxin Ambient Monitoring Plan
CARB	California Air Resources Board
CCMS	Committee on Challenges of Modern Society
Cd	Cadmium
CEPA	California Environmental Protection Agency
CO	Carbon monoxide
Cr	Chromium
Cr(VI)	Hexavalent Chromium
CW	Central/Western
CWTC	Chemical Waste Treatment Centre
DDT	Dichlorodiphenyl Trichloroethane
DEFRA	Department for the Environment, Food and Rural Affairs
DNPH	2,4-Dinitrophenylhydrazine
DRI	Desert Research Institute
EC	Elemental Carbon
ENSR	ENSR Environmental International Inc
EPA	Environmental Protection Agency
FIA-AA	Flow Injection Analysis - Atomic Absorption
FSP	Fine Suspended Particulate (term used in Hong Kong), same as PM _{2.5}
GC	Gas Chromatography
GL	Government Laboratory
HAAS	Hazardous Ambient Air Standards
HAP	Hazardous Air Pollutant
Hg	Mercury
HI	Hazard Index

HK	Hong Kong
HKEPD	Hong Kong Environmental Protection Department
HKPU	The Hong Kong Polytechnic University
HKUST	Hong Kong University of Science and Technology
HpCDD	Heptachlorodibenzo-p-dioxin
HPLC	High Pressure Liquid Chromatography
HQ	Hazard Quotient
HRGC	High Resolution Gas Chromatography
HT	Hok Tsui
HT2.5	Chemical dataset speciated by the DRI using PM _{2.5} samples collected at Hok Tsui between October 2000 and November 2001
HxCDD	Hexachlorodibenzo-p-dioxin
HxCDF	Hexachlorodibenzofuran
IARC	International Agency for Research on Cancer
ICP-AES	Induced Coupled Plasma-Atomic Emission Spectroscopy
IRIS	Integrated Risk Information System
I-TEF	International Toxicity Equivalency Factors
KC	Kwai Chung
KT	Kwun Tong
LPG	Liquid Petroleum Gas
MATES	The Multiple Air Toxics Exposure Study
MDL	Method Detection Limit
MK	Mongkok
MK10	Chemical dataset speciated by the GL using PM ₁₀ samples collected at Mongkok as part of HKEPD's filter-based PM monitoring program
MK2.5	Chemical dataset speciated by the DRI using PM _{2.5} samples collected at Mongkok between October 2000 and November 2001
Mn	Manganese
MOE	The Ministry of the Environment
MPCA	Minnesota Pollution Control Agency
MS	Mass Spectrometry
MSW	Municipal Solid Waste
MTBE	Methyl Tert-Butyl Ether
Nap	Naphthalene
NAPS	The Canadian National Air Pollution Surveillance
NATO	North Atlantic Treaty Organization
NCDENR	North Carolina Department of Environment and Natural Resources
NDAMN	National Dioxin Air Monitoring Network
NENT	North East New Territories
Ni	Nickel
NO ₂	Nitrogen Dioxide
NRC	National Research Council

NSW	New South Wales
NW	Nim Wan
O ₃	Ozone
OC	Organic Carbon
OCDD	Octa Chlorinated Dibenzo-P-Dioxin
OCDF	Octa Chlorinated Dibenzo Furan
OEHHA	Office of Environmental Health Hazard Assessment
p,p'-DDD	p,p'-Dichlorodiphenyl Dichloroethane
p,p'-DDE	p,p'-Dichlorodiphenyl Dichloroethene
PAH	Polycyclic Aromatic Hydrocarbon
PAMS	Photochemical Assessment Monitoring Station
Pb	Lead
PCB	Polychlorinated Biphenyl
PCDD	Polychlorinated Dibenzo-P-Dioxin
PCDF	Polychlorinated Dibenzofuran
PCE	Perchloroethylene
Phe	Phenanthrene
PIXE	Photon Induced X-Ray Emission
PM	Particulate Matter
PM ₁₀	Particulate Matter with aerodynamic diameter ≤ 10 μm, same as RSP
PM _{2.5}	Particulate Matter with aerodynamic diameter ≤ 2.5 μm, same as FSP
POM	Polycyclic Organic Matter
POP	Persistent Organic Pollutant
PRD	Pearl River Delta
PUF	Polyurethane Foam
REL	Reference Exposure Level
RSP	Respirable Suspended Particulate (term used in Hong Kong), Same As PM ₁₀
Se	Selenium
SENT	South East New Territories
SL	Sha Ling
SO ₂	Sulphur Dioxide
SSP	Sham Shui Po
ST	Shatin
STAPPA	State and Territorial Air Pollution Program Administrators
SWG	Special Waste Group
TAP	Toxic Air Pollutant
TC	Tung Chung
TCDD	Tetrachlorodibenzo-P-Dioxin
TCDF	Tetrachlorodibenzofuran
TDI	Tolerable Daily Intake
TEF	Toxic Equivalency Factor

TEQ	Toxic Equivalent
TKO	Tseung Kwan O
TM	Tap Mum
TNRCC	The Texas Natural Resource Conservation Commission
TO	Tai Po
TO-4	USEPA Air Toxic Analysis Method 4: Determination of Organochlorine Pesticides and Polychlorinated Biphenyls in Ambient Air
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TO-13	USEPA Air Toxic Analysis Method 13: Determination of Benzo(a)Pyrene [B(a)P] and Other PAHs in Ambient Air using GC and HPLC Analysis
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TO-14A	USEPA Air Toxic Analysis Method 14A: Determination of VOCs in Ambient Air using specially prepared Canisters with subsequent analysis by GC
TOMPS	Toxic Organic Micro Pollutant
TPU	Tertiary Planning Unit
TSP	Total Suspended Particulates
TW	Tsuen Wan
TW10	Chemical dataset speciated by the GL using PM ₁₀ samples collected at Tsuen Wan as part of HKEPD's filter-based PM monitoring program
TW2.5	Chemical dataset speciated by the DRI using PM _{2.5} samples collected at Tsuen Wan between October 2000 and November 2001
UATMP	Urban Air Toxics Monitoring Program
ug	Microgram, also shown as µg in text.
UK	United Kingdom
ULSD	Ultra-Low Sulphur Diesel
US	United States
USEPA	U.S. Environmental Protection Agency
UV	Ultraviolet
V	Vanadium
VOC	Volatile Organic Compound
WENT	West New Territories
WHO	World Health Organization
WHO-TEF	Toxic Equivalency Factor (assigned by WHO)
XRF	X-Ray Fluorescence
YL	Yuen Long
Zn	Zinc

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1 Executive Summary

Toxic air pollutants (TAPs), also known as hazardous air pollutants, are air pollutants that are known or suspected to cause cancer or other serious health effects such as reproductive effects or birth defects, or adverse environmental effects¹. In 1993, the Hong Kong Environmental Protection Department (HKEPD) commissioned Eureka Laboratories to perform a screening level emission inventory of TAPs for Hong Kong. In the report, Eureka (1996) concluded that Hong Kong has a TAP problem typical of modern urban cities, and recommended the development of a TAP management program in Hong Kong.

Subsequently in mid-1997, the HKEPD started a TAP monitoring program and six groups of TAPs were routinely monitored at the Tsuen Wan and Central/Western Air Quality Monitoring Stations (AQMSs). The six TAP groups include polychlorinated biphenyls (PCBs), dioxins/furans, polycyclic aromatic hydrocarbons (PAHs), volatile organic compounds (VOCs), carbonyls, and hexavalent chromium (Cr(VI)).

Besides routine TAP monitoring at Tsuen Wan and Central/Western, there have also been a number of ad hoc studies commissioned by the HKEPD in which concentrations of many TAPs were measured. These include a 12-month study with additional sampling at Mongkok and Hok Tsui from November 2000 to October 2001, and a study from April 2001 to March 2002 by the HKEPD Special Waste Group (SWG) in Nim Wan, Sha Ling and Tseung Kwan O, among others.

At the time of the Eureka report, the lack of actual TAP measurements dictated that the TAP conditions in Hong Kong could only be evaluated using estimated emission inventories. Now, following five years of monitoring, a measurement-based evaluation of TAP conditions in Hong Kong has been performed. The objective of this study is to review the TAP measurements, taking into consideration recent and established scientific findings, to better understand the TAP situation in Hong Kong and to provide recommendations for future monitoring efforts. The study was divided into five tasks:

- (1) Review current strategy and protocol of the TAP monitoring program in Hong Kong and those overseas.
- (2) Review the available TAP data, including data in the open literature, for the current situation of TAPs in Hong Kong and for episodic and trends analyses, location and seasonal variations.
- (3) Conduct a health impact assessment based on the measured ambient / roadside TAP concentrations in Hong Kong with reference to overseas experience.
- (4) Review TAP concentration trends against control measures introduced by the HKEPD.
- (5) Review TAP measurement data against the priority TAP list generated in the 1995 Eureka study, identifying measurement gaps and making recommendations for future monitoring efforts.

We also note that the health risks associated with criteria pollutants (e.g., O₃, NO₂ and SO₂) will be examined although these pollutants are typically not considered as TAPs. Our analyses show that the current levels of these criteria pollutants in Hong Kong are associated with substantial health risks for the public, and hence we feel that our report would be incomplete if these pollutants were excluded solely because they are not officially designated as TAPs. Furthermore, there is a movement internationally in officially designating these criteria pollutants as TAPs. For example, the Canadian government has just included O₃, NO, NO₂ and

¹ <http://www.epa.gov/air/toxicair/newtoxics.html>.

SO₂ in its list of toxic substances (Environment Canada, 2003).

In this executive summary, we shall outline the key findings for each of the tasks listed above, along with our recommendations for future TAP monitoring efforts.

1.1 Review of TAP Programs in Hong Kong and Overseas

The current strategy and protocol of a number of overseas TAP monitoring programs and networks have been reviewed (Chapter 3). These include the California Air Toxics Program, the New York Toxics Air Monitoring System, the U.S. Urban Air Toxics Monitoring Program, the U.S. National Air Toxics Monitoring Network, the U.S. National Dioxin Air Monitoring Program, the California Ambient Dioxin Air Monitoring Program, the Japanese Persistent Organic Pollutant Monitoring Program, the United Kingdom Toxic Organic Micro Pollutants Network, and the Canadian National Air Pollution Surveillance Network. We found that while the overall objectives of these programs are quite similar, there can still be substantial differences in the scope, focus and specific objectives in these TAP programs. In particular, we found the U.S., and in particular California, have the most extensive and intensive ambient monitoring programs for toxic air pollutants in the world thus far.

The TAP monitoring program in Hong Kong is quite similar to the California Air Toxics Program. The sampling frequency, data quality control and analysis procedures of the two programs are alike. It is of interest to note that the number of TAPs analyzed and reported is substantially higher in Hong Kong than in California. This is likely related to the fact the Hong Kong program has much shorter history, and the HKEPD is still gathering information through studies like this to better understand the overall TAP situation.

In terms of network design, the main objective of the current network design was to measure the ambient TAP levels for health-related population exposure assessment, which is similar to the objectives of most other monitoring programs. Specifically, it includes (HKEPD 2001a):

- (1) To determine representative concentrations in areas of high population densities, and hence the exposure of the broad mass of population to toxic air pollutants of concern and to assess the consequential risks to human health, as well as other environmental effects, resulting from this exposure.
- (2) To determine the general background as well as the highest concentrations expected to occur in the area covered by the monitoring network, and hence to determine the ranking of Hong Kong when compared to other metropolitan cities.
- (3) To provide data for policy formulation of environmental control strategies and legislation, air pollution control plans, etc.
- (4) To observe long term trends in ambient levels of toxic air pollutants.
- (5) To validate and provide input for atmospheric dispersion models.
- (6) To study and evaluate pollution interactions and patterns, including long-range atmospheric transport.

Under these objectives, Central/Western and Tsuen Wan were selected to represent typical residential/commercial environments in Hong Kong. The two TAP monitoring stations started operating in July 1997. Samples were taken regularly for measurement of ambient TAP concentrations. The target TAPs fall into six categories as noted earlier in section 1. The sampling schedules varied among the TAPs. For example, VOC samples were collected once every six days; carbonyl samples were collected once every twelve days; dioxins, PCBs, and PAHs samples were collected roughly once every month before July 1999 and roughly once every twelve days after July 1999.

Besides the two regular TAP monitoring stations (Tsuen Wan and Central/Western), the TAP monitoring program in Hong Kong included a number of ad hoc monitoring exercises. These include the dioxin monitoring exercise commissioned by the HKEPD and carried out by ENSR (HKEPD, 2002), the Tai Mo Shan Persistent Organic Pollutant (POP) study (Louie and Sin, 2003), and the 12-month PM_{2.5} study. The latter involves the setting up of two additional TAP stations, one at a roadside location in Mongkok and the other in a rural area in Hok Tsui from November 2000 to October 2001. The roadside (Mongkok) site was designed to obtain the exposure levels of TAPs by pedestrians close to traffic; monitoring at this site allows quantification of TAPs from vehicular sources in the close-to-worst-case scenarios. The rural (Hok Tsui) site was to provide background TAP levels in Hong Kong. Situated on the coastline of Southern China, Hok Tsui has distinct seasonality, receiving continental outflow in the winter and maritime inflow in the summer.

Furthermore, in addition to TAP-oriented monitoring programs, the HKEPD also collects high volume TSP and RSP samples every six days for chemical analyses in its general Air Quality Monitoring Network. Metal species and one PAH, benzo(a)pyrene (BaP), are among the aerosol constituents monitored. Some metal species such as cadmium, lead, and nickel, are known air toxics. In 2001, high volume TSP samples were collected at one roadside and nine general stations while high volume RSP samples were collected at one roadside and six general stations (HKEPD, 2001b). A list of the general air quality monitoring stations and characteristics of each station can be found in the annual air quality report prepared by the HKEPD (e.g., HKEPD, 2001b).

1.2 Review of Available TAP Data

A summary combining open literature and HKEPD data has been used to provide an overview of current TAP levels in Hong Kong (Chapter 4). In particular, the TAP measurements in Hong Kong are compared with similar measurements in other places, when data is available. Moreover, the spatial variability as well as the temporal trends of the various TAPs were studied.

Through our analyses, we confirmed the conclusion from Eureka (1996) that Hong Kong has a TAP problem typical of modern urban cities. Following the chemical grouping used in the sampling, the analyses are separated in seven sub-groups: diesel particulate matters, elements, dioxins, PCBs, PAHs, carbonyls and VOCs.

1.2.1 Diesel Particulate Matter

Diesel Particulate Matter (PM) has long been recognized as harmful to human health. In terms of health risks to the local population, we find that diesel particulate matter is by far the most important air pollutant in Hong Kong. Local vehicular emission is one of the major sources of diesel PM, and our findings suggest that efforts to reduce vehicular emissions should be continued and strengthened.

The WHO has suggested an annual air quality guideline of 2.3 $\mu\text{g}/\text{m}^3$ on the basis of its non-carcinogenic health endpoints (WHO, 2000). The WHO also estimates that diesel PM has a cancer risk of 16-71 per million for every 1 $\mu\text{g}/\text{m}^3$ of diesel PM. The California EPA recommends a chronic reference exposure level of 5 $\mu\text{g}/\text{m}^3$ (OEHHA, 2002).

Despite considerable research efforts devoted to chemical characterization of diesel PM, a unique tracer has not yet been found (Birch and Cary, 1996). As elemental carbon (EC) constitutes a large portion of diesel PM mass and it can be quantified at low levels, use of EC as a surrogate for diesel PM has been proposed and used in practice (USEPA, 2002). However,

we note that diesel PM is not the only source of EC in the ambient environment since combustion of other carbonaceous fuels also contributes to EC loading in the air. As a result, use of EC as a surrogate could overestimate the contribution of diesel PM, but it is an accepted and widely used approach up to this point.

We have found that the EC levels in Hong Kong are generally high. Many locations exceed the $5 \mu\text{g}/\text{m}^3$ chronic reference exposure level set by the Californian EPA. The roadside monitoring station has higher EC levels than other monitoring sites (the annual mean EC level at Mongkok is about $11 \mu\text{g}/\text{m}^3$, more than twice the level of $5 \mu\text{g}/\text{m}^3$ at Tsuen Wan), consistent with the understanding that vehicular emissions are the major source of EC.

The level of organic carbon has clear seasonality with higher concentrations in the winter and lower concentrations in the summer. This result suggests that long-range transport is a major carbonaceous aerosol source in Hong Kong. In contrast, seasonality of EC is much weaker or not discernable, and is variable among the monitoring sites. Many of the AQMSs with higher EC concentrations (e.g. Kwai Chung and Tsuen Wan) are to the north of the container terminal and hence they are downwind of ocean-going vessel emissions in the summer, but upwind in the winter. The variable EC seasonality may be a result of two counteracting factors, i.e., influence of marine emissions versus dirtier air masses transported from the north.

For the long-term, although a weak but steadily declining trend in EC concentrations has been observed in TSP and RSP at the roadside² (HKEPD 2003), we found no discernable annual trend during the four annual cycles (1998-2001) at nine of the ten general AQMSs. This suggests that factors other than vehicular emissions have an important effect on the EC concentrations at our general stations. Also worth noting is a rising trend seen for three annual cycles from 1998 to 2000 at Kwai Chung (chemical speciated data were not available for Kwai Chung after 2000), which has the highest EC concentration amongst all the general stations. This is likely related to EC emissions from residual fuel oil combustion by marine vessels at or close to the nearby container terminals.

Our study identified marine sources as one of the major sources contributing to the EC budget of Hong Kong. As EC is one of the most important parameters used in health risk assessment, further clarification of the role of marine contributions to the EC budget in Hong Kong is recommended in future studies. Along the same lines, we also recommend the resumption of elemental speciation of RSP samples collected at the Kwai Chung AQMS.

1.2.2 Toxic Elemental Species

Twelve elemental species are listed as priority toxic air pollutants in Hong Kong in the Eureka study (aluminum, arsenic, beryllium, cadmium, chromium and Cr(VI), lead, manganese, mercury, nickel, phosphorus, vanadium, and zinc). Eight of these are on the 33 species priority HAP list compiled by the U.S. Environmental Protection Agency (USEPA). Three elemental species (arsenic, hexavalent chromium, and nickel) have been identified to be carcinogens by the WHO (WHO, 2000). Hexavalent chromium has also been identified as a toxic air contaminant by the California Air Resources Board (CARB, 1987). Phosphorus was not available in the TAP database as it was found to be below its method detection limit (MDL) in a 2001 study by the HKEPD. Hence, our analyses are focused on the other elemental species listed above.

² No elemental analyses were performed for RSP at the Central and Causeway Bay stations, and the relocation of the Mongkok station make it difficult to compare the trend of EC before and after 1999.

In this group, the most important TAP is hexavalent chromium, Cr (VI). Hexavalent chromium concentration levels are similar at Tsuen Wan and Central/Western and remain relatively unchanged throughout the available data. Their annual means range from 0.20 to 0.28 ng/m³, close to its MDL of 0.2 ng/m³. For this reason, measurement uncertainty of Cr(VI) is expected to be high. The level of Cr(VI) in Hong Kong is approximately two times that in California. No seasonality could be discerned, possibly reflecting the large measurement uncertainty and/or its short lifetime in air.

All other toxic elemental species were determined from their total amounts in RSP samples at ten monitoring stations in Hong Kong. The two carcinogenic substances, arsenic and nickel, have concentrations at least six times lower than their California chronic exposure limits. The maximum 3-month averaged lead levels in Hong Kong are seven times lower than the current Hong Kong air quality objective for lead, and the maximum annual level is two times lower than the annual UK standard. Beryllium and mercury have their majority measurements below their respective MDLs, 0.10 ng/m³ and 0.49 ng/m³, indicating that their levels in RSPs were low across all the monitoring stations and over the time of more than four years.

Six elemental species, aluminum, arsenic, cadmium, lead, manganese, and zinc, have a clear seasonality with higher levels in winter and lower levels in the summer at all ten sites. In addition, Yuen Long sees the highest concentrations among all the sites for these elements. Both observations point to more polluted air masses of continental origin transported to Hong Kong impacting the ambient concentration levels for these elements, and may be the controlling factor for their seasonal variability.

Using the roadside station at Mongkok to gauge the importance of local vehicular emissions, we found that arsenic, cadmium, lead, manganese, and zinc displayed similar levels at Mongkok to those at other stations. This indicates that the local vehicular traffic is not an important source for the above elements. Aluminum and chromium have higher concentrations at the Mongkok roadside station but these two elements are found more in the coarse mode. Their higher levels at the roadside is probably related to re-suspension by passing vehicles rather than direct vehicular emissions.

Nickel and vanadium showed elevated levels at Kwai Chung in comparison with other stations, pointing to a source local to Kwai Chung. This source is most likely the cargo terminals in the vicinity, and the large number of ocean-going vessels not present at other sites. Combustion of heavy oil, which is more prominent in these transportation trades than others, is known to be a major source of vanadium and nickel. While the level of these elements is low in comparison to established guidelines, weak rising trends are identified for both species.

Finally, we note that pollution roses show that north / northwesterly regional transport is often associated with many of these toxic elements. With the fast growth of the industrial and economic activity over the Pearl River Delta Region, this in turn suggests that continual monitoring of TAPs continues to be important.

1.2.3 Dioxins

Polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are collectively called “dioxins”. PCDDs and PCDFs are two of the twelve Persistent Organic Pollutants (POPs) targeted by the Stockholm Convention. Dioxins are chemically stable semi-volatile solids. There are 210 known congeners, of which 75 are PCDDs and 135 are PCDFs. Congeners with chlorine atoms in the 2, 3, 7, and 8 positions are of particular environmental concern due to their toxicity. There are 17 of such congeners, all of which are tracked by the dioxin monitoring program in Hong Kong.

A number of air quality guidelines and source emission standards for dioxins have been set by various government agencies around the world and by the WHO. The guidelines applicable to ambient environment vary from 0.02 to 40 pg I-TEQ/m³. Hong Kong has no ambient air standard for dioxins.

The urban dioxin levels in Hong Kong generally fall within the lower end of the range of dioxin concentrations measured in other urban centres around the world. There appears to be a weak decreasing trend in dioxin levels at both Central/Western and Tsuen Wan from 1998 to 2001, but noting the large uncertainty relating to the method detection limits, this result has to be taken as preliminary.

A clear seasonal cycle is found in dioxin concentrations, with much higher levels in the winter when Hong Kong is mainly affected by continental air, and lower levels in summer when Hong Kong is under the influence of oceanic air. Pollution wind maps also show that higher levels of dioxins are associated with weak to moderate north / northwesterlies. Spatially, Tsuen Wan and Hok Tsui recorded the highest and lowest dioxin levels in 2000/01, respectively, suggesting a north to south dioxin gradient, but the difference amongst the four TAP monitoring stations was quite small.

On the other hand, results from ad hoc studies showed more clearly a northwest to southeast dioxin gradient, with two to four times higher levels of dioxins found at Yuen Long, Nim Wan and Sha Ling (as compared to urban areas in Tsuen Wan and Central/Western). Correlation analysis shows that the congener profiles of dioxins (excluding OCDD) are the same for samples taken on the same day at different sites.

Our analyses combine to suggest that the dioxin levels in Hong Kong are mostly associated with regional transport by weak to moderate winds from emission sources outside Hong Kong. Contributions from the Chemical Waste Treatment Center, local vehicular sources or from landfill / composting sites are small.

Noting that dioxin levels are generally higher in the northwestern part of Hong Kong, monitoring resources for dioxins on a site identified to be a receptor location for regional transportation but upwind of local sources (e.g. Yuen Long) is recommended. Since vehicular emission is not an important contributor to dioxins, future dioxins monitoring in a roadside environment is probably not necessary.

Finally, we also note that our examination of the dioxins monitoring effort showed that a large number of measurements were below the analytical method detection limits. This affects the accuracy of the mean dioxin level estimated, and this issue should be addressed when planning future monitoring programs.

1.2.4 Polychlorinated Biphenyls

Polychlorinated biphenyls (PCBs) are organic compounds with two chlorinated benzene rings linked by a C-C bond. There are 209 PCB isomers, in which one to ten chlorine atoms can be attached to a biphenyl. About 130 isomers are likely to occur in commercial products. The WHO has identified twelve co-planar PCB congeners as “dioxin-like PCBs” on the basis of their similar toxicity to dioxins (UNEP, 1999; Dyke and Stratford, 2002). The WHO has assigned each of these a toxic equivalency factor (WHO-TEF).

Japan has set an air quality standard of 0.6 pg-TEQ/m³ for the sum of dioxins (PCDDs and PCDFs) and dioxin-like PCBs (Japan MOE, 1999). In the US, California suggests a chronic inhalation exposure limit for PCBs to be 1.2 µg/m³ (OEHHA, 2002), while North Carolina

recommends an annual average of $0.083 \mu\text{g}/\text{m}^3$ to be the guideline for acceptable ambient levels (NCDENR, 2002). Hong Kong has no ambient air standard for PCBs.

The PCB levels in Hong Kong are a few times higher than the world background level, but they are lower in comparison with available data from other major cities worldwide where measurements can only be found for the earlier 1990s. The annual mean PCB levels at Tsuen Wan and Central/Western appear to be lowering. Spatially, concentrations at Central/Western, Tsuen Wan, and Mongkok varied rather coherently, with Tsuen Wan and Central/Western having similarly low PCB levels and Mongkok slightly higher. Amongst the four TAP sites, the highest levels of PCBs are found at the rural Hok Tsui site. In addition, higher levels of PCBs at Hok Tsui are found in the summer, suggesting local volatilization may be significant near Hok Tsui. This in turn suggests the presence of a local PCB source near the Hok Tsui site in Cape D'Aguiar. Further ad hoc study to verify this hypothesis is recommended.

The current regular TAP monitoring program only monitors the concentration of total PCBs. However, measurements of individual PCB congeners, and more specifically, the dioxin-like PCB congeners, are necessary for assessment of the health risks contributed by PCBs, and they cannot be estimated from total PCB concentrations. Limited measurements from the Special Waste Group (SWG) of the HKEPD study suggest that the toxicity equivalent attributable to dioxin-like PCBs could be significant (up to about 20%) in comparison with that attributable to dioxins. For this reason, analyses of dioxin-like PCB congeners are recommended for future PCB monitoring efforts.

1.2.5 Polycyclic Aromatic Hydrocarbons

Polycyclic aromatic hydrocarbons (PAHs) are organic compounds of two or more fused benzene rings, in linear, angular or cluster conformations. Because of limited emissions data, for assessment purpose PAH data has been limited to either a group of 7 or 16 individual PAH species referred to as 7-PAH and 16-PAH, respectively (USEPA, 2002b). The 7-PAH compounds are classified as either probable or possible human carcinogens by the International Agency for Research on Cancer (IARC). The 16-PAH group also includes the 7-PAH group. Five of 16-PAH compounds were listed as Category II air toxics in the Eureka study. They are anthracene, benzo[a]anthracene, chrysene, benzo[a]pyrene (BaP), and dibenzo[a,h]anthracene.

For this group, the most important TAP is benzo[a]pyrene (BaP) because of its high cancer risk, hence it is also the PAH compound most studied. It is often selected as a marker for the PAH group due to its stability and relatively constant contribution to the carcinogenic activity of particle-bound PAH (European Communities, 2001b). The WHO suggested a cancer risk value of 87 for every ng/m^3 of BaP (WHO, 2000). Air quality guidelines set by different countries vary a lot from the most stringent values of $0.1 \text{ ng}/\text{m}^3$ recommended by Sweden, France, and Croatia, to the a few tens ng/m^3 in the US (MPCA, 1999; NCDENR, 2002). Hong Kong has no ambient air standard for BaP or PAHs in general.

Levels of the PAH marker compound BaP were analyzed for both RSP and TSP samples collected at ten general air quality monitoring stations in Hong Kong. Comparison of the measurements indicates that BaP mostly reside in small particulates, making the BaP in RSP samples statistically similar to the BaP in TSP samples. This implies that simultaneous monitoring of BaP in both TSP and RSP are redundant, and that the monitoring of BaP in TSP samples can be stopped.

The BaP annual average concentrations in Hong Kong range from 0.10 to $0.54 \text{ ng}/\text{m}^3$. Comparing with the 0.5 to $1.0 \text{ ng}/\text{m}^3$ guideline that is being considered by European Communities, the Hong Kong levels are lower than or at the lower limit of this benchmark, and

most other guidelines set by countries in Europe. On the other hand, the BaP levels at all monitoring sites exceed the most stringent BaP guideline (0.1 ng/m^3) proposed by Sweden.

In the four complete annual cycles from November 1997 to October 2001, no rising or declining trend is seen for BaP. However, a rising annual trend in the total PAH is clearly present. The increase in the two most abundant PAHs, acenaphthene and acenaphthylene, largely account for the annual rising trend in the total PAH amount. The reason for this increase is still unclear.

BaP and other heavy PAHs of 5-rings or larger have a clear seasonal pattern with higher concentrations observed in the winter than in the summer. Such seasonality is anticipated as a result of lower mixing heights in the winter and higher wet removal rates in the summer. However, such seasonality could not be unambiguously identified for the lighter PAHs with 3- or 4-rings.

Spatially, Yuen Long consistently has the highest BaP concentrations among the ten monitoring stations in the three annual cycles with available measurements. Pollution roses show that higher BaP levels are associated with northerly wind, suggesting regional transport of BaP from the North. During the summer, Mongkok has the highest BaP concentrations, indicating the influence of local traffic emissions.

1.2.6 Carbonyls

Carbonyls are a class of organic compounds containing a ketone or an aldehyde group. They mainly exist in the gas phase because of their small molecular sizes and consequently high volatility. Many members of the carbonyl chemical class are known to be toxic. For example, formaldehyde is a probable human carcinogen (IACR, 1995), causing eye irritation at levels as low as tens of ppbv and respiratory tract irritations at levels as low as 100 ppbv (NRC 1981). In terms of chronic health impact for the population, we find that formaldehyde is the third most important TAP in Hong Kong (Table 1-1). Acetaldehyde, furfural, and crotonaldehyde are animal carcinogens (WHO 2000), while acrolein causes eye irritation and odor annoyance and exacerbates asthma.

Carbonyls in ambient air have direct emission sources such as combustion processes and can also be formed through oxidation of hydrocarbon precursors. The relative importance of primary and secondary sources depends both on location and the time of day. The photooxidation processes that lead to formation of carbonyls are also responsible for the accumulation of ozone in the air. Carbonyls are monitored either for assessing their exposure risk to the public health or for better understanding of the photochemical pollution problem. Formaldehyde and acetaldehyde are the two most abundant carbonyl compounds found in both urban and rural environments. Of all the carbonyl compounds, formaldehyde has received the most attention because of its established toxicity and abundance in the air.

We note that some carbonyls had blocks of recurrent low concentration values, which were close to $\frac{1}{2}$ MDLs. This suggests the levels are low and the reported values are not necessarily that of their ambient concentrations. Out of the 17 carbonyl compounds reported, eight were detected above their MDLs in more than two-thirds of the samples using the current sampling and analysis methodology. They are formaldehyde, acetaldehyde, acetone, propanal, methyl ethyl ketone, valeraldehyde, butyraldehyde / isobutyraldehyde and benzaldehyde. Amongst these, the acetone measurements are likely problematic due to low collection efficiency of the standard method (Moore, personal communications).

A clear rising trend is found in the formaldehyde annual average concentrations, but such a

trend is not found for other carbonyls. The rise of formaldehyde levels in the ambient air is possibly related to the suspected increasing presence of MTBE in gasoline sold in Hong Kong (Kirchstetter et al. 1999a, b, and Koshland et. al. 1998).

Of the carbonyls examined, seven show a clear seasonality with higher concentrations in the winter and lower concentrations in the summer. They include acetaldehyde, propanal, acetone, benzaldehyde, methyl ethyl ketone, valeraldehyde, and n+i-butanol. On the other hand, formaldehyde does not have a clear seasonal cycle. The lack of clear seasonality for formaldehyde is likely attributed to its short lifetime in the ambient air as it is the most reactive and shortest lived carbonyl compound. It also suggests that the dominating sources and sinks for formaldehyde may be different from those for carbonyls with a clear seasonality. Moreover, the increasing trend in background levels also makes it more difficult to identify the seasonal cycle in formaldehyde.

As no carbonyl measurements were made for Mongkok or Hok Tsui, detailed analysis of the spatial distribution of carbonyls over Hong Kong cannot be provided. We only note that carbonyl variations at Tsuen Wan and Central/Western were similar in magnitude, with most carbonyls found at slightly higher concentrations at Tsuen Wan (than at Central/Western). This may be related to the fact that vehicular emissions, a major source of carbonyl compounds, have an increased contribution at the Tsuen Wan station.

In comparison to other cities, the levels of the two most abundant carbonyls (formaldehyde and acetaldehyde) in Hong Kong are similar to those found in a number of urban areas in the US. When compared with the California chronic health exposure limits, formaldehyde levels exceed the exposure limit whereas acetaldehyde is far below the suggested exposure limit. Nevertheless, with increased use of ethanol to replace MTBE as a fuel additive, increased levels of acetaldehyde are expected in the US. As Hong Kong imports its fuel from the global market, it is of interest to see whether changes in long-term averaged acetaldehyde and formaldehyde levels will be observed in the next few years.

Finally, we note that although acrolein is one of the 33 priority TAPs listed by the USEPA, there is still no good method for measuring its ambient concentrations. Measurement problems with acrolein have been reported elsewhere (Vermont Air Pollution Control Division, 1998) and the USEPA has recently issued an addendum stating that the TO-11 and TO-11A methods are not useful for the determination of acrolein (USEPA 2002c). Due to the lack of ambient measurements, it is not possible to make a health assessment for this compound at present.

1.2.7 Volatile Organic Compounds

Volatile organic compounds (VOCs) are organic chemicals with vapor pressures greater than 0.01 kPa. Some VOCs are toxic and a few of them (e.g., 1,3-butadiene and benzene) are carcinogenic. Carbonyls are also VOCs by definition, but they are typically not included in most discussions when VOCs are referred to, mainly because different sampling and analysis techniques are used for determination of their concentrations in the ambient environment. VOCs is an important group of TAPs as we find that 18 of the 34 health-based priority TAPs are VOCs (Table 1-1), with 1,3-butadiene, benzene and tetrachloroethylene as the most significant ones for Hong Kong. (Ethylene oxide, methyl alcohol and butanol, three pollutants identified in the Eureka report, were not available in the TAP database as they were found to be below MDL in 2001 study by the HKEPD.)

Benzene levels at the two urban background sites (Tsuen Wan and Central/Western) are similar to those observed in other urban areas in industrialized countries around the world. These levels are lower than those reported for a few cities in South America and in Mediterranean

areas (e.g., Athens) and are about one-half of the $4.8 \mu\text{g}/\text{m}^3$ benzene air quality guideline suggested by European Communities. Seasonally, benzene shows a clear seasonality, but this trend is not found for the other aromatic hydrocarbons.

As expected, the roadside environment has higher levels of benzene and other aromatic hydrocarbons, demonstrating that vehicular emissions are major sources for these compounds. When compared with the levels reported in roadside environments in other countries and other cities in the Pearl River Delta region, the roadside BTEX (i.e., Benzene, Toluene, Ethylbenzene, and Xylenes) levels in Mongkok are generally lower.

The 1,3-butadiene levels at all sites, including the roadside, are below the $2.25 \mu\text{g}/\text{m}^3$ UK guideline value. There is a clear spatial gradient with Mongkok > Tsuen Wan > Central/Western > Hok Tsui, consistent with the spatial variation of traffic density. There is also a large difference (sixteen fold) in the annual average concentration between the roadside environment and the rural environment. No clear seasonality is observed for 1,3-butadiene.

Tetrachloroethylene displayed a clear spatial variation, highest at Central/Western, medium at Mongkok and Tsuen Wan, and lowest at Hok Tsui. This result suggests that there are more dry-cleaning activities near Central/Western than other TAP sites.

With respect to the long term, a weak decreasing trend can be noted for benzene, with levels decreasing respectively by 15 and 22% at the Tsuen Wan and Central/Western stations, from 1997/98 to 2001/02. This may be related to control measures started in 2000 to reduce benzene content in unleaded gasoline. Nevertheless, similar decreases are also noted for other aromatics (toluene and xylenes) not directly targeted by the gasoline control measures. The latter is also consistent with possible emissions reductions associated with the overall economic slow down in the same period.

The annual average levels of 1,3-butadiene in 2001 were higher than those in 2000 for both Tsuen Wan and Central/Western. Similar findings were found for other LPG-related VOCs, including butane, iso-butane, 1-butene / iso-butylene, 1-butyne, pentane, 1-pentene and propane. This may be partly related to the gradual fleet change of taxis from diesel to LPG. However, we caution that the two years of available data is not enough to establish any trend, and further monitoring is needed to confirm the levels of these species in the long term.

A large decrease of tetrachloroethylene was found in 2002 at Central/Western, and a small decrease observed at Tsuen Wan. These decreases may relate to control measures implemented to reduce tetrachloroethylene emissions from dry cleaning facilities since November 2001, and is consistent with our understanding that emissions from dry cleaning facilities are more prominent near this site (see next section). Again, we must caution that the data is not extensive enough to establish any definite trend, and further monitoring is needed in the long term.

Three halogenated VOCs, carbon tetrachloride, chloroform, and methylene chloride, show an unexpected surge in their concentrations since December 2000. Such a sudden increase is noteworthy because of the potential health impact of these species, and it is important to further investigate whether such an increase is an artifact of laboratory contamination or is real. This question aside, the levels of halogenated VOCs are within chronic reference exposure levels established by the CARB.

1.3 Health Impact Assessment

A health impact assessment is a combination of procedures, models and tools by which a policy, program or project may be judged as to its potential effects on the health of the population, and

the distribution of these effects within the population. It is useful as a planning tool for integrated decision-making.

1.3.1 Cancer Risks

Cancer risk estimates were estimated for the Hong Kong population, assuming 70 years of exposure. The cancer risks attributed to various TAPs are similar to results of the MATES II Report (2000). In particular, diesel fumes accounted for 8,158 cases of cancer, or about 80% and by far the most dominant contribution. Other important contributors are metals including hexavalent chromium, arsenic, cadmium, and organic compounds including benzene, 1,3-butadiene, carbon tetrachloride, formaldehyde, 1,1,2,2-tetrachloromethane and tetrachloroethene, which is within our expectations. The proportional contribution of TAPs towards health risks in Hong Kong is not expected to be too different from that in other major cities, because the sources of air pollutants are generally similar with traffic and industry contributing the majority of pollution.

We have also tried using both an overall and a district based approach to calculate the cancer risk from diesel, with both yielding similar results. The district based approach yields a cancer estimate of 8,100 cancer cases, similar to that based on the mean of two urban stations (Central/Western & Tsuen Wan, 8,158 cancer cases). Preliminary estimates using fine particulate data increased the estimate to 8560, only 6% higher than the 8158 cases calculated using respirable particulate data.

The cancer risk of dioxins from ambient concentrations of 2,3,7,8-TCDD was comparatively small, at 1.8 (USEPA) and 2 (CEPA) cases per million respectively. Based on data obtained from collocated samples measured from January to December 2002 by the HKEPD, the ratio of dioxins to dioxin-like PCBs was estimated to be 1:0.24. This was done by comparing the sum of the mean monthly values of dioxin-like PCBs ($0.015 \text{ pg I-TEQ m}^{-3}$) to the mean monthly values of total dioxins ($0.062 \text{ pg I-TEQ m}^{-3}$).

The ratio of dioxins to dioxin-like PCB is based on the best data available and hence a best estimate. However, the correlations between the 11 measurements (January to December 2002, except April 2002) were low ($r=0.073$) and statically insignificant ($P=0.83$). Therefore, one has to exercise caution in estimating the total health effects of dioxins and dioxin-like PCBs based on measurements of dioxins only.

Another dataset collected by the Special Waste Group (SWG) of the HKEPD was used to calculate the cancer risk of the thirteen POP species. We find that p,p'-DDD and p,p'-DDE contributed to over 95% of all cancers attributed to the 11 POPs. p,p'-DDE is the predominant contributor of cancer, accounting for 60%-83% of all cancer cases, while p,p'-DDD accounts for 14%-31% of cancer cases.

1.3.2 Acute Non-Cancer Risks of FSP

Many urban health studies have found that FSP is the most important contributor in terms of both acute and chronic health risks for the population (Dockery et al 1993 and MATES II Study 2000). Similar results are found in this study.

The annual mean concentration of FSP at Tsuen Wan is $34 \text{ } \mu\text{g}/\text{m}^3$. For acute risks, we used a relative risk of 1.14 (total mortality) per $10 \text{ } \mu\text{g}/\text{m}^3$ of FSP following the WHO guidelines (WHO, Air Quality Guidelines for Europe Second Edition). Assuming an annual number of 32,827 deaths (average of 1995 – 1998), a total of 4,596 deaths per year could be attributed to a $10 \text{ } \mu\text{g}/\text{m}^3$ increase of FSP concentrations. The calculation of 15,626 deaths attributable to FSP was based on reductions of FSP from 34 to zero $\mu\text{g}/\text{m}^3$. The mean annual number of deaths (for

the period 1995-1998) was 32,827, suggesting that 47.5% of deaths could be attributed to FSP. However, this is a theoretical number and not a realistic one because it would be impossible to achieve a zero concentration of FSP, and it should also be noted that the proportions of deaths attributable to several risk factors can theoretically add up to larger than 100%.

1.3.3 Other Non-Cancer Risks

Several TAPs and criteria pollutants (diesel PM, NO₂, O₃, SO₂ and formaldehyde) were found to have exceeded the reference values in some measurements. Superficially, the effects appear to be relatively minor (respiratory tract and eye irritation). In reality, the consequences on respiratory health are substantial, as damage to the respiratory system is documented in most of the TAPs and the effect on health is at least additive. (The effects of different TAPs may be synergistic, which is not unlikely.)

The level of NO₂ is of particular concern. The short- and long-term exposures to criteria pollutants (including PM₁₀) have been shown to be associated with increased mortalities and respiratory diseases in local studies (Wong et al 2002a, 2002b). The relative risks of NO₂ and O₃ are especially high. Although overseas studies have focused on RSP, and recently, FSP, NO₂ is highly correlated with the former, and might have accounted for much of the short-term health effects.

Besides, these chemicals have other long-term effects – diesel and formaldehyde are carcinogenic. Stringent control of these TAPs should bring about considerable benefits to the health of the population. We recommend that a high priority be accorded to control these TAPs (in particular, NO₂, diesel fumes and formaldehyde) exceeding the acute and chronic reference exposure levels (RELs).

For the acute inhalation non-cancer risks, three criteria pollutants (NO₂, O₃, SO₂) contributed to Hazard Quotients (HQs) and Hazard Indices (HIs) of greater than one in many of the AQMSs. The level of NO₂ was again of particular concern. The effects may appear to be minor (respiratory tract and eye irritation), but could contribute to increased mortalities and respiratory diseases found in local time-series studies (Wong et al, 1999, 2002a, 2002b). It should also be noted that the typical concentration of SO₂ over Hong Kong is quite low, and the high HQs of SO₂ were related to a few episodic events.

For chronic inhalation non-cancer risks, NO₂ was again the major contributor. The other pollutants of concern included diesel fumes, formaldehyde and naphthalene. The respiratory system was the organ system most affected and the findings also corresponded well with the results from an earlier local study on respiratory health of school children (Yu et al, 2001). High HI for eyes was noted in Central/Western and Tsuen Wan and could be attributed mainly to the level of formaldehyde found at those two stations. The only other high HI was for the effects on the nervous system at Mongkok where three metals and nine VOCs contributed to an HI of 1.15. Manganese was the main metal pollutant (HQ = 0.52) and methylcyclohexane was the major VOC involved (HQ = 0.29). We recommend that a high priority be accorded to controlling those TAPs which contributed to HQs and HIs that exceeded one (NO₂, diesel fumes and formaldehyde, which are the same TAPs exceeding the acute and chronic RELs).

1.3.4 Combined Effects of Different Chemicals

It should be noted that when a person is exposed to more than one carcinogenic substance, the cancer risk increases. It is very unlikely that the overall effect is less than the individual effects combined, and it is possible that the combined effect of simultaneous exposure to different carcinogenic substances is greater than the sum of the individual effects. Hence, the combined risk resulting from simultaneous exposure to several carcinogenic TAPs may be unacceptably high even though the risk of exposure to a single chemical might stay within the “acceptable limit”. The same holds true for non-cancer risks.

1.4 Concentration Trends against Control Measures Introduced

We have reviewed the TAP concentration changes against air toxics control measures introduced by the HKEPD since 1998. Before going into the details, we must caution that the TAP monitoring record is really too short for any conclusive statement on long-term impact of the control measures. Many factors affect the TAP levels measured at the monitoring sites, including local meteorology and emission changes. Hence, review and findings provided in this study must be considered preliminary and provisional; longer term monitoring and evaluation must be carried out to verify or challenge these findings.

1.4.1 Diesel Particulate Matters

A weak but discernible decreasing trend is noted in the level of NO_x and particulate matter including TSP, RSP and FSP concentrations recorded at the roadside (HKEPD 2003). This is consistent with the continual efforts by the HKEPD to reduce particulate emissions from the vehicular fleet. Noting the dominant role of diesel PM and NO₂ in our chronic and acute health risk estimates, these control measures move in the right direction and should be further enhanced.

However, no clear and consistent trend can be noted for the variation of EC and PM levels at the general stations, suggesting that factors / sources other than local vehicular traffic are important for controlling EC and PM levels at these AQMSs. We note that sources like marine shipping activities at or near the container terminal at Kwai Chung may be related to the apparent rise in EC (and PM) concentrations at the Kwai Chung AQMS during the past few years. It is recommended that the HKEPD study the emission profiles of various EC and PM sources, and if their contributions are found to be significant, consider additional control measures to reduce their emissions.

1.4.2 Benzene and Vapor Recovery

A weak decreasing trend is found for benzene, with the 2001/02 concentrations lowered by 15 and 22% at Central/Western and Tsuen Wan stations respectively, from their 1997/98 mean. Two control measures were introduced to reduce benzene concentrations in air. The first is a vapor recovery rule at petrol filling stations implemented in April 1999, and the second is a change in benzene levels in unleaded gasoline from 5% to 1% effective April 2000. Lack of measurements before 1997 makes it difficult to assess the effectiveness of the first control measure. Nevertheless, the second control measure is consistent with the reduction of benzene observed in the last few years.

However, this may partly also be related to reduced emissions in association of the overall economic slow down during the same period. The latter is consistent with the observation that similar decreases are seen in the levels of other aromatics (toluene and xylenes). Further monitoring is needed to see if these changes are part of longer-term decrease.

1.4.3 Tetrachloroethene

A vapor recovery regulation has been in place from November 2001 to reduce the level of tetrachloroethene. Using recently updated data for 2002, a substantial decrease of about 30% is found at Central/Western for 2002 when compared with the average concentration for the previous three years. Even at Tsuen Wan, a decrease of 13% is noted. These decreases are consistent with the tetrachloroethene control measures introduced, and suggest that these control measures may have helped in the reduction of ambient tetrachloroethene concentration. The larger decrease in Central/Western is also in line with our finding that emissions from dry cleaning facilities are more prominent near this site. Nevertheless, the record is still too short and this finding must be corroborated with subsequent measurements.

1.4.4 Phasing Out of Leaded Petrol

The phasing out of leaded petrol began in 1991 with the eventual ban of leaded petrol implemented in April 1999. By the beginning of the TAP monitoring program in 1997, the lead level in Hong Kong was already very low. This is a clear cut example showing that the control measures are having an impact, and lead levels in vehicular emissions have become minimal.

The lead concentrations recorded at the Mongkok roadside station are similar to those recorded in the other urban AQMSs, indicating that local vehicular traffic is no longer an important source of ambient lead levels in Hong Kong. In contrast, the consistently higher lead concentrations at Yuen Long in comparison with all other stations and the associated circular pollution wind map further suggest that regional transport has significant impact on lead levels in Hong Kong.

1.4.5 Reduction of Sulphur Content in Diesel

Reductions of sulphur content in diesel and the introduction of ultra-low sulphur diesel (ULSD) were implemented mainly as part of the overall effort to reduce secondary PM formation in Hong Kong. In addition, these measures also have a direct impact on ambient SO₂ levels, particularly at the roadside. For example, the average roadside SO₂ concentration in 2002 (18 µg/m³) dropped by 36% compared with the 2000 value (28 µg/m³), likely as a result of the introduction of ULSD for the vehicle fleet in late 2000 (HKEPD 2003).

1.4.6 Fleet Change of Taxis from Diesel to LPG

The LPG taxi pilot scheme began with 30 taxis in November 1997. By the same time in 2001, about 75% of the fleet had been converted to LPG. A review of VOCs commonly found in LPG showed that their concentrations in the 2000/01 cycle were higher than that of the 1999/00 cycle³. With recently updated VOC data, we found that the annual average levels in 2000/01 and 2001/02 are higher than those identified for 1999/00 at both measurement sites. These increases are consistent with the larger number of LPG taxis on the road. However, the increases are gradual and two years of data is insufficient to establish any long-term impact. Further monitoring is needed to assess the longer term results of the LPG conversion. Of particular concern in this group of LPG-related TAPs is 1,3-butadiene, which is one of the most important TAP species identified in terms of the overall health risk assessment. It is of interest to find out how much 1,3-butadiene is in LPG fuel, and whether this amount can be reduced to minimize its ambient level in the atmosphere.

³ Most of these LPG-related VOCs (e.g., propane, pentane, butane, iso-butane, 1-butene / iso-butylene, 1-butyne and 1-pentene) were not analyzed or below MDLs before 1999.

1.4.7 Others

Last but not least, the clear rise in formaldehyde (one of the top air toxics in our revised priority list) is believed to be related to the increased presence of the MTBE fuel additive as a result of the gradual phasing-out of leaded gasoline. In Hong Kong, with the phase-in of unleaded petrol and the reduction of benzene levels from 5% to 1%, MTBE is likely added into the gasoline as an octane enhancer substitute for the leaded compound and benzene removed. Unfortunately, no MTBE data is available for gasoline sold before 2000. As a result, the suggestion of increased MTBE in gasoline in recent years has yet to be verified. If this is actually the case, this is an unintended impact of control measures for other substances. To better understand this issue and similar ones which may occur in the future, it is recommended that the HKEPD obtain more information from fuel manufacturers about fuel additives / octane enhancers in their products.

Because of concern regarding the contamination of surface and ground water, use of MTBE as fuel oxygenate is being phased out in California and its use is declining in other states. As a replacement, increased use of ethanol as fuel oxygenate is expected, and acetaldehyde levels are expected to increase due to the change in fuel additives. Hence, information about ethanol or other fuel additive(s) are important for a better understanding of the impact of fuel changes, and it is also important for the HKEPD to continue close monitoring and see whether there are changes in acetaldehyde and formaldehyde levels in the next few years.

1.4.8 Control Experiences from California

Finally, we note that California has traditionally been very aggressive in its control of air pollutants. Hence, we have summarized some of its control measures in section 6.9. Also, noting the dominance of diesel particulate matter in the substances contributing to both chronic and acute health risks for the Hong Kong population, we recommend that the HKEPD look into these control measures to try to reduce the particulate content in the atmosphere. Following the example in California, the HKEPD should examine on-road as well as off-road control measures. In particular, marine vessels appear to be a significant source in contributing to elemental carbon variations over much of the Kowloon Peninsula. We recommend that the HKEPD confirm this result through source identification studies, and consider follow-up measures to limit emissions.

1.5 Review of TAP Priority List

Based on a screening level emissions inventory, the Eureka Report (1996) had categorized three groups of TAPs for priority considerations. With the TAP measurements and the subsequent risk analyses (Chapter 5), a list of 34 priority TAPs has been revised as shown in Table 1-1. This table is calculated based on a lifetime of 70 years' exposure to the mean concentration of corresponding TAPs measured at Tsuen Wan and Central / Western from Nov 2000 to October 2001, and using the higher risk estimate from Cal EPA, USEPA and WHO. The exceptions are p,p'-DDD and p,p'-DDE, which were not monitored regularly at Tsuen Wan or Central / Western. The additional cancer cases for p,p'-DDD and p,p'-DDE are based on data obtained in an ad hoc POP study conducted at Tseung Kwan O, Sha Ling, and Nim Wan.

The increase in cancer risks associated with the first 14 TAPs in this list is more than ten in a million, while the increase in cancer risks associated with the others in the list is between one and ten in a million people. In particular, ten out of the eleven TAPs (excluding lead) listed as Category I TAPs in the Eureka Report are also identified in this new list, and ten of them are originally identified as Category II TAPs in the Eureka Report. In addition, fifteen new TAPs not listed in the Eureka Report are found to have ambient concentrations associated with a risk

greater than one in a million for increased cancer cases. Within this group, the TAP with the highest risk is carbon tetrachloride.

On the other hand, there are a number of TAPs listed in the Eureka report for which unit risk factors were unavailable from any databases, although some were classified as possible human carcinogens or worse (e.g. acrolein). Hence, they are not included in our new risk-based priority list. These include many of the Category III compounds listed in the Eureka report.

1.6 Recommendation for Future Monitoring Efforts

The HKEPD has been monitoring many more TAPs than the ones listed in the Eureka Report (1996). This is prudent considering the report was compiled with emission estimates and not actual monitoring data, and it is better to obtain as much information as possible to define the baseline TAP situation in Hong Kong. The fact that fifteen new TAPs not originally listed in the Eureka Report was identified as priority TAPs in this study shows that the open-minded approach to monitoring taken earlier was important in establishing baseline conditions clearly. Nevertheless, with the baseline conditions better understood through this review, it is an opportune time to consider redeployment of our resources in more extensive and focused monitoring efforts for the TAPs we now know to be important in Hong Kong.

1.6.1 Routine Monitoring and Ad Hoc Studies

Currently, the TAP monitoring program in Hong Kong consists of regular monitoring at two urban sites (Tsuen Wan and Central/Western), as well as ad hoc studies such as the dioxins monitoring exercise carried out by ENSR, the Tai Mo Shan POP study, and the 12-month PM study. While ad hoc studies can be targeted for any particular focus of interest, it is useful to define clearly the monitoring objective of the regular monitoring program.

With respect to the six objectives of the TAP program listed in section 1.1, we recommend that the routine monitoring network focus mainly on population exposure and health-risk assessments studies. In addition, we recommend that other monitoring objectives be supplemented through ad hoc studies as necessary.

In particular, parameters that require substantial resources for analyses but are found to be at low concentrations can be moved out of the routine monitoring programs. On the other hand, ad hoc studies can be conducted once every few years to help gauge the ambient concentrations of these and other emerging TAPs such as mutagens, endocrine disruptive agents, organo-mercury, POPs, etc. that can affect the health of the population. If and when the concentration levels of any of these pollutants are found to be high enough to warrant more frequent monitoring, they can then be moved into the routine monitoring program.

Moreover, ad hoc studies can include items such as hot-spot identification, tracking of TAP movements, source profile identification, and monitoring of new TAP species of concern, etc. In particular, the current monitoring frequency is not suited for the monitoring of episodic events, for detailed tracking of regional and long-range transport, or for identification of pollutant interactions which often can take place in time-scales much shorter than the 24-hour sampling period. Finally, we also recommend ad hoc studies targeted on obtaining local and regional chemical signature profiles for various emission sources. Such findings will be useful for the formulation of effective TAP control measures.

1.6.2 Addition of a TAP Monitoring Station at Yuen Long

Our analyses show that there are substantial variations for many TAPs over Hong Kong. In

particular, concentrations of many TAPs are found to be significantly higher in the northwest and lower in the southeast (see Chapter 4). As a substantial portion of Hong Kong's population lives and/or works in that region, it is prudent that the ambient TAP levels in the northwest part of Hong Kong be better defined. Hence, we recommend the establishment of another routine TAP monitoring station and FSP measurements at Yuen Long.

1.6.3 TAP Monitoring in Other Airsheds

To better gauge the health risk associated with population exposure, a better definition of the spatial distribution of TAP in these areas is very much needed. Instead of the establishment of routine monitoring stations, ad hoc and less sophisticated but still systematically and densely placed (i.e. level-1 saturation monitoring network) studies are recommended to help define the baseline TAP levels in these areas. This would help map out the TAP gradients across different airsheds over Hong Kong. This type of level-1 survey can be repeated once every few years to help track the gradual change of TAP levels over the different airsheds in Hong Kong as a result of changing emission characteristics of the Pearl River Delta region.

1.6.4 Sampling Parameters

The list of parameters monitored by the HKEPD is much longer than the ones recommended in the Eureka Report (1996). This is necessary as the report was compiled with emission estimates and not actual monitoring data. With the baseline condition better understood through this review, we recommend the HKEPD to modify the list of TAPs monitored in its routine monitoring effort – putting more focus on priority parameters and less effort on TAPs that are of low concentrations or of questionable accuracies.

Diesel Particulate Matter, FSP and Priority TAPs Identified in This Study

Our review identified diesel particulate matter (PM) as the most important parameter contributing to the overall health risk of the population. As most of the diesel PM is in the fine mode, expansion of the current FSP network (e.g. extending to Yuen Long) can help in providing more detailed information for subsequent health risk-assessment in Hong Kong. Along the same lines, we recommend that for the list of priority TAPs identified in this review, monitoring efforts should be continued and even strengthened (if resources are available). The establishment of a routine monitoring station at Yuen Long will be of great benefit in this respect.

Elemental Analyses of RSP

Also, although strictly speaking not part of the TAP program, the elemental analyses performed on RSP samples have proven very useful for the identification of marine-related pollution sources. We recommend the continuation of these analyses for RSP samples, particularly for AQMSs such as Kwai Chung which has characteristics distinct from the other sites. On the other hand, with the limited resources available, it is probably not necessary to continue these elemental analyses for all the AQMSs since not all of them have unique characteristics.

Acrolein, Acetone and Styrene

We have found that the analysis results of some of the TAPs appeared to be questionable, including measurements of acrolein, acetone and styrene. The USEPA recently issued an addendum stating that the TO-11 and TO-11A methods are not useful for determination of acrolein (USEPA 2002c). Problems in measuring acetone and styrene appear known to analytical chemists but are not well-documented in the literature (Moore, personal communications). In particular, the acetone measurements are likely problematic due to low

collection efficiency of the standard method while the problem with styrene is related to its high reactivity. Both of these complications can lead to analytical results not representative of ambient levels. Analyses of these pollutants should be stopped or at least flagged in the data base until better analytical methods can be developed here or elsewhere.

Other TAPs with Concentrations That Are Near or Below Detection Limits

Our review finds that the concentration levels of a number of TAPs are at or below their method detection limits. For example, nine of the 17 carbonyl species reported were routinely below their MDLs due to low concentrations in the ambient environment. Whether or not to continue the analyses for these TAPs of low ambient concentrations should depend on whether additional resources are needed for the analyses and reporting of these TAP species. If no additional cost / resources are required, then monitoring should be continued to allow for the detection of any unexpected increase in concentration levels.

However, if additional cost is associated with the measurement of TAPs that are often at or below method detection limits, we recommend that they be eliminated from routine monitoring efforts. The resources saved can then be used within the TAP program to help the establishment of a new routine monitoring station at Yuen Long, or for ad hoc studies as outlined earlier. After the elimination of these parameters in the routine monitoring effort, it is prudent that their ambient levels still be monitored once every few years (e.g., every 5 years and preferably using methods with lower detection limits) as part of an ad hoc baseline review study to make sure that any unexpected and systematic increase in their levels would not remain unnoticed.

1.6.5 Sampling Issues

Currently, the samples are collected over 24 hour period once every six or twelve days. The concentration levels of many of the TAPs, even some on the priority list (e.g. dioxins), are below or near detection limits. The HKEPD should find ways to improve the detection limits to allow for better quantification of TAP levels in the ambient atmosphere.

1.6.6 Management of Health Data and Establishment of a Health-Related Database

In a separate study on air pollution and health conducted by the Chinese University of Hong Kong (CUHK), problems with the quality of the health outcome data were identified. Mortality data, obtained from Census and Statistics Department via the HKEPD, was incomplete in terms of the dates of deaths, although this is an important estimate of health outcome. Similarly, missing data in Tertiary Planning Units (TPUs) was commonly found in the mortality data. As a result, daily time series cannot be performed and data from the Hospital Authority could not be easily divided into districts according to the residential address of the patients. This hinders district-based analysis.

To allow better analyses of district based health analyses, we recommend a better monitoring network for FSP to improve definition of local population exposures. Moreover, we also recommend the establishment of an online health and environmental database which would enable an accurate risk estimate using a time series and spatial approach. In particular, data (with personal information removed) must be routinely updated in this online health and environmental database, and be readily accessible by researchers in Hong Kong. A stable and open access health information system similar to the current air pollution database would go a long way in aiding the understanding of environmental and public health issues in Hong Kong.

1.6.7 Control Policy

Several TAPs and criteria pollutants (diesel particulate matter, NO₂, O₃, SO₂ and formaldehyde) were found to have exceeded their reference values in some measurements. Superficially, the effects appear to be relatively minor (respiratory tract and eye irritation). In reality, the health consequences on respiratory health are substantial, as damage to the respiratory system is documented in most of the TAPs – the criteria pollutants, carbonyls, and some VOCs and metals, and the effects on health is at least additive.

The level of NO₂ is of particular concern. The short- and long-term exposures to criteria pollutants (including RSP) have been associated with increased mortalities and respiratory diseases in local studies. The relative risks of NO₂ and O₃ are especially high. Although overseas studies have focused on RSP, and recently FSP, NO₂ is highly correlated with the former, and might have accounted for much of the short-term health effects. Besides, these chemicals have other long-term effects – diesel and formaldehyde are carcinogenic. Stringent control of these TAPs should bring about considerable benefits on the health of the population.

We recommend that a high priority should be accorded to control those TAPs exceeding the acute and chronic reference exposure levels and/or with HIs or HQs exceeding one (in particular, NO₂, diesel fumes and formaldehyde).

1.6.8 Impact Related to Future Development

This is the first systematic study in Hong Kong on TAP monitoring and health risk assessment. Notwithstanding the above limitations, the impacts of TAPs on community health in terms of cancer risks and non-cancer risks have been estimated. Results of this study set a baseline health risk estimate for the general population, against which different scenarios of risks can be worked out based on different pollution profiles in specific districts / areas. The impact on health of projected increases or reductions of TAP emissions with respect to future development in and around Hong Kong is beyond the scope of the current study. However, it is an important issue that needs to be assessed with proper trend analyses. Such results shall also provide useful information on a health-based priority in the control of TAPs, which must be considered together with other environmental, technological, social and economic concerns.

1.7 Table for Chapter 1

Priority	Chemical name	CAS number	Additional cancer risk per million	Additional cancer case for HK population*	Ambient concentrations (ug/m3) *	
1	List of pollutants having ambient concentrations associated with cancer health risk larger than ten in a million (listed in order of decreasing health risk).	Diesel Particulate Matter	9902 #	1216.0	8158	4.053e+0
2		1,3-Butadiene	106-99-0	78.7	528	2.810e-1
3		Formaldehyde	50-00-0	73.5	493	5.654e+0
4		Benzene	71-43-2	57.6	387	1.987e+0
5		Carbon Tetrachloride	56-23-5	44.7	300	1.065e+0
6		Hexavalent Chromium	1333-82-0	27.7	186	1.847e-4
7		1,2-Dibromoethane	106-93-4	25.5	171	1.158e-1
8		BaP	50-32-8	20.8	139	2.386e-4
9		Chloroform	67-66-3	18.0	121	7.830e-1
10		Arsenic	7440-38-2	15.6	104	3.617e-3
11		Dibenzo(a,h)anthracene	53-70-3	14.3	96	5.585e-5
12		1,1,2,2-Tetrachloroethane	79-34-5	10.7	71	1.837e-1
13		Hexachlorobutadiene	87-68-3	10.5	71	4.782e-1
14		Tetrachloroethene	127-18-4	10.4	70	1.764e+0
	List of pollutants having ambient concentrations associated with cancer health risk between one and ten in a million (listed alphabetically).	1,1,2-Trichloroethane	79-00-5	1.4	9	8.700e-2
		1,1-Dichloroethene	75-35-4	5.5	37	1.100e-1
		1,2-Dichloroethane	107-06-2	4.6	31	1.758e-1
		1,2-Dichloropropane	78-87-5	2.9	19	1.496e-1
		Acetaldehyde	75-07-0	5.6	37	2.060e+0
		Benzo[b]fluoranthene	205-99-2	4.2	28	4.076e-4
		Benzyl chloride	100-44-7	4.8	32	9.875e-2
		Bromodichloromethane	75-27-4	7.5	51	2.039e-1
		Cadmium	7440-43-9	5.6	37	1.322e-3
		Chloroethene	75-01-4	5.1	34	6.590e-2
		Chrysene	218-01-9	2.6	17	5.828e-4
		Dibromochloromethane	124-48-1	4.0	27	1.484e-1
		Dioxins	EDF-350 #	2.0	14	5.363e-2 **
		Indeno[1,2,3-cd]pyrene	193-39-5	4.9	33	3.798e-4
		Methylene chloride	75-09-2	3.0	20	2.981e+0
		Nickel	7440-02-0	1.9	13	5.107e-3
		p,p'-DDD *	72-54-8	2.9	20	1.214e-5
	p,p'-DDE *	72-55-9	9.9	67	2.920e-5	
	p-Dichlorobenzene	106-46-7	7.7	51	6.972e-1	
	Trichloroethene	79-01-6	2.4	16	1.215e+0	

Table 1-1 Health-based Priority List of Toxic Air Pollutants from this study.

* Based on a lifetime of 70 years' exposure to the mean concentration of corresponding TAPs measured at Tsuen Wan and Central / Western from Nov 2000 to October 2001, and using the higher risk estimate from Cal EPA, USEPA and WHO. Hong Kong population in March 2001 was 6,708,389.

p,p'-DDD and p,p'-DDE are not monitored regularly at Tsuen Wan or Central / Western; the additional cancer cases for them are based on data obtained in an ad hoc POP study conducted at Tseung Kwan O, Sha Ling, and Nim Wan.

CAS number not available for diesel particulate matters and dioxins. Reference number here are adopted from Scorecard, available at <http://www.scorecard.org/chemical-profiles/index.tcl>.

** Unit for dioxins is pg-I-TEQ/m3

2 Background

Since mid-1997, six groups of Toxic Air Pollutants (TAPs) have been monitored at two stations, Central/Western and Tsuen Wan in the TAP monitoring program. In addition, a year-long study has recorded ambient concentrations for five of these six groups of TAPs (excluding hexavalent chromium) at two sampling locations representing roadside (Mongkok) and rural (Hok Tsui) environments for the period from November 2000 to October 2001. The data along with recent scientific progress in understanding TAPs needs to be systematically reviewed and statistically analyzed in order to better understand Hong Kong's TAP situation and provide recommendations for future monitoring efforts.

The objectives of this project are to review this data and help prioritize actions on future TAP measurements. The scope of this study consists of the following:

- (1) Review current strategy and protocol of the TAP monitoring program in Hong Kong and overseas
- (2) Review the available TAP data, including data in the open literature, for the current situation of TAPs in Hong Kong and for episodic and trend analyses, location and seasonal variations. Compounds or species names of TAPs to be reviewed were listed in the Special Conditions of Contracts for Tender Reference AS 01-285.
- (3) Conduct a health impact assessment based on the measured ambient / roadside TAP concentrations in Hong Kong with reference to overseas experience.
- (4) Review TAP concentration trends against control measures introduced by the HKEPD.
- (5) Review the TAP measurement data against the priority TAP list generated in the 1995 Eureka study, identifying measurement gaps and making recommendations for future monitoring efforts.

Results of our assessment for the above tasks will be described in the following chapters.

3 The Current Strategy and Protocol of the TAP Monitoring Program

The current strategy and protocol of the TAP monitoring programs in Hong Kong are reviewed with reference to those overseas. A brief overview of the major relevant overseas TAP monitoring programs is given below to facilitate evaluation of the TAP monitoring program in Hong Kong. Monitoring programs for POPs (e.g., dioxins, furans, and PCBs) are often separated from those of other air toxics. For this reason, the review of air toxic monitoring programs is grouped into non-POP and POP monitoring programs

3.1 Non-POP Air Toxics Monitoring Programs

3.1.1 The United States

The U.S. has so far the most extensive and intensive ambient monitoring programs for toxic air pollutants among developed countries and regions in the world. In its 1990 Clean Air Act Amendments, the US Congress established a list of 188 toxic chemicals and designated them as Hazardous Air Pollutants (HAPs). As part of its overall efforts to reduce air toxics, the US Environmental Protection Agency (USEPA) identifies a list of 33 air toxics that present the greatest threat to public health in the largest number of urban areas. More recently, the USEPA has published an "Air Toxics Monitoring Concept Paper". This concept paper outlines the design of a national air toxic monitoring network, which is to be established over a several year period (USEPA, 2000). The U.S. national air toxics program encompasses four areas, one of which is National Air Toxics Assessment. Its activities include (1) expanding air toxics monitoring, (2) improving and periodically updating emissions inventories, (3) improving national- and local-scale air quality and exposure modeling, and (4) continuing research on effects and assessment tools. Ambient monitoring is an essential component of the program. The ambient monitoring data serve multiple purposes, including

- (1) Characterization of ambient concentrations and deposition in representative monitoring areas,
- (2) Providing data to support and evaluate dispersion and deposition models,
- (3) Establishing trends and evaluating the effectiveness of control strategies, and
- (4) Providing data for risk assessment.

This monitoring network is still in its initial establishing phase and only limited monitoring data is available. Outside this national monitoring program, there are a number of air toxics monitoring activities at federal, state, and local levels. Some individual states, particularly California, have long-running monitoring programs for a selected number of TAPs. Brief descriptions of a number of federal and state air toxic monitoring programs are given below.

A Monitoring Activities at the National Level

Urban Air Toxics Monitoring Program (UATMP)

The USEPA initiated the UATMP in 1988 to characterize the magnitude and composition of potentially toxic air pollution in, or near, urban locations. The USEPA approved standard methods were adopted in this program. The number of participating sites as well as the number of monitored air toxics has changed from year to year. In 1997, the UATMP included 12 monitoring stations that collected 24-hour integrated air samples, typically on a biweekly basis (USEPA, 1999a). The monitoring efforts were limited to 47 VOCs and 16 carbonyls. Overall, nearly 28,000 ambient air concentrations were measured during the 1997 UATMP. The 1999/2000 UATMP included 15 monitoring stations on a 6 or 12-day schedule for 24-hour

integrated samples (USEPA, 2001). The number of air toxics monitored varied among the sites. Six sites analyzed 59 VOCs and 16 carbonyls; ten sites also analyzed for 80 VOCs; and one site analyzed 92 semi-volatile VOCs in addition to VOCs and carbonyls.

Photochemical Assessment Monitoring Stations (PAMS) Network

The PAMS network monitors ozone and its precursors in areas with persistently high ozone levels. As precursors of ozone, a number of VOCs are measured in the PAMS program. Some of the monitored VOC species are air toxics (e.g., benzene). Currently there are 22 operating PAMS network stations, mostly in large metropolitan areas (Atlanta, Houston, Los Angeles, New York, etc.). A list of current PAMS sites can be found at <http://www.epa.gov/oar/oaqps/pams/arealist.html>. Most PAMS sites measure 56 target hydrocarbons on either an hourly or 3-hourly basis during ozone seasons. The annual statistics (e.g., annual averages) of VOCs from the PAMS sites can be retrieved at http://www.epa.gov/aqspub1/annual_summary.html.

B Air Toxics Monitoring Programs in Selected States

California Air Toxics Program

The California Air Resources Board (CARB) established an air toxics monitoring network in its major urban areas in 1985. The objective of this monitoring network is to determine the average annual concentrations of toxic air contaminants for identification of air toxics of concern and to assess the effectiveness of control measures (CARB, 2002a). Twenty-four hour integrated samples are collected every twelve days at 21 sites throughout California. The list of monitored air toxics includes VOCs, carbonyls, PAHs, MTBE, toxic metals, and hexavalent chromium, adding up to about 60 different toxic air contaminants. Detailed standard operating procedures for the analyses have been specified (CARB, 2003), and monitoring data from 1990 onwards for these toxics is available. With over 40,000 measurements generated each year, this monitoring data set is used to evaluate emission trends, to establish background risk levels, and to prioritize identification and control actions.

New York Toxics Air Monitoring System

The New York State Bureau of Air Quality Surveillance initiated its toxics air monitoring system with seven monitoring sites in 1989 (New York State Department of Environmental Conservation, 2002). The network expanded in 1990 to a statewide network of ten monitoring sites covering industrial, urban, suburban, and rural areas. One more station at a rural location was added in 1996 to make a current total of eleven stations. This monitoring network focuses on measurements of 17 VOCs, with fifteen of the 17 VOCs on the 189 HAPs list identified in the 1990 Clean Air Act Amendments. The main objective is to monitor air quality related to toxics in the State's urban, industrial, residential, and rural areas. This network provides a long-term toxics air quality database for New York State. The data provides actual air quality measurements of the VOCs and has been used in the design and management of New York's air quality program, which includes risk assessment, modeling, planning, and trend analyses.

Minnesota

The Minnesota Pollution Control Agency (MPCA) monitors VOCs, carbonyls, and Cr (VI) at 25 sites across the State (MPCA, 1999). The monitoring program was started as early as 1991 at some of the 25 sites, of which fourteen are still in active operation. The sampling network was developed over several years to address multiple concerns. Some sites were established to measure concentrations in the vicinity of specific sources. Other sites were established to collect baseline data on air toxics concentrations in the State's largest urban area, the Minneapolis-St. Paul metropolitan area. A third group of sites was established for a one-year

study, with the objective of collecting 1-year snap shots of concentrations at selected sites throughout the state. The sites in this one-off study were selected with weighting for geographic coverage and population density (Pratt et al., 2000). Three types of samples were collected at each site: VOCs, carbonyls, and PM₁₀. The VOC samples and carbonyl samples were collected every six days over a 24-hour period using the USEPA air toxic reference methods TO-14A and TO-11A, respectively. Metal species in PM₁₀ samples were analyzed using energy dispersive X-ray fluorescence.

Texas Air Toxics Monitoring Program

The Texas Natural Resource Conservation Commission (TNRCC) began deployment of an ambient air toxics monitoring network in 1992 by a mandate of the state legislature (Pendelton, 1995). The network had expanded to 23 monitoring sites across Texas as of 1995, with the objective of monitoring the long-term population exposure to a number of selected VOCs. VOC samples were collected into canisters for 24 hour sampling duration and at a sampling frequency of once every six days, and analyzed in accordance with USEPA method TO-14. During 1995, a total of 69 VOCs were on the measurement list. The monitored compounds were selected based upon their expected frequency of occurrence, potential to cause health effects, potential to form ozone, and known applicability to the canister collection techniques in use (TNRCC, 1995).

Vermont Air Toxics Program

The air toxics monitoring program in Vermont began in 1993 with collection of data on VOCs and has since expanded to include carbonyls and semi-volatile organic compounds (i.e., PAHs, PCBs, and dioxins) (Vermont Air Pollution Control Division, 1998). The monitoring network consists of five sampling sites ranging from urban to rural. VOCs and carbonyls were collected every twelve days using USEPA methods TO-14 and TO-11, respectively. The three classes of semi-volatiles were collected simultaneously over 72 hours using modified USEPA standard methods. The semi-volatile samples were collected quarterly at four of the five sites. Results of the semi-volatile organics on two sampling dates, one in 1995 and the other in 1996, were reported in their air toxics report.

3.1.2 Canada

The Canadian National Air Pollution Surveillance (NAPS) Network was established in 1969 to monitor and assess the quality of ambient air in Canadian urban centers. In 1998, the network consisted of 239 stations in 136 cities, with 430 continuous analyzers for criteria pollutants and 219 samplers for criteria pollutants and VOCs. Monitoring of air toxics as part of NAPS began in 1989. Aromatics (e.g., benzene), aldehydes, ketones, PAHs, dioxins and furans are monitored at 40 urban and rural locations across Canada. Noting that many of the criteria pollutants also have significant health impact, the Canadian government has recently decided to include ozone, nitric oxide, nitrogen dioxide and sulfur dioxide in its list of toxic substances (Environment Canada 2003).

3.1.3 The United Kingdom

The UK toxic organic micro pollutants network monitors a range of toxic organic micro-pollutants including PAHs, dioxins, and PCBs at selected sites around the UK. There are a total of 17 sites for which data is available. The starting date of these monitoring sites varies from 1991 to 1999 (www.airquality.co.uk/archive/tomps.php). Seven sites are currently operating, including one roadside site, three urban sites, one semi-rural site, and two rural sites across the nation (www.defra.gov.uk/environment/airquality). The PAH samples in this monitoring network are collected with a filter and an adsorbent material over two weeks. They

are then bulked together and chemically analyzed every three months. Eighteen PAHs are measured in this network.

3.1.4 Denmark

The Danish Air Quality monitoring network has been monitoring air pollutants continuously since 1982. As of 2001 the air quality network consisted of four urban sites and two background sites (Kemp and Palmgren, 2001, 2002). The aim of the network is to follow the concentration levels of toxic pollutants in the urban atmosphere and to provide the necessary knowledge to assess trends, perform source apportionment, and evaluate the chemical reactions and dispersion of the pollutants in the atmosphere.

The air toxics on the monitoring lists are three aromatic VOCs, i.e., BTX (benzene, toluene, and xylenes), and heavy metals in PM₁₀. BTX was monitored in 1999 at a location close to traffic in Copenhagen at a sampling frequency of one sample every 30 min. The elemental composition of the aerosols in 24-hour integrated samples was measured at the four urban stations and one of two rural stations. The analytical method for measuring the heavy metals in aerosol samples is a Photon Induced X-ray Emission (PIXE) method.

3.1.5 Australia

The National Environment Protection Council in Australia has had a nationwide program, namely “Living Cities-Air Toxics Program”, since July 1999. This program aims to provide information to set up the National Environment Protection Measures for ambient air toxics, including PAHs, PCBs, PCDD/Fs, some VOCs, and other air toxics (Environment Australia, 2002). No sampling and measurement details are available as yet.

The New South Wales (NSW) Environmental Protection Authority has undertaken a study to obtain data on the concentrations of a number of air toxics in the Sydney-Newcastle-Wollongong area (NSW Environmental Protection Authority, 2002). The monitored air toxics included dioxins, 41 VOCs, 11 PAHs, and 12 heavy metals. The study ran for five and a half years from early 1996 to August 2001. More samples were taken during winter months when pollutant concentrations were expected to be highest.

The Victoria Environmental Protection Authority commissioned a study to assess particles and carbonyl pollutants close to the South Eastern Arterial Freeway (Victoria Environmental Protection Authority, Australia, 1999). A total of 17 carbonyl samples were taken on days with predicted poor air quality during the period of 30 April to 29 July 1994. The measurement results of five carbonyls (formaldehyde, acetaldehyde, propanal, acrolein, butanal, and methyl ethyl ketone) were reported. Common aromatic hydrocarbons (e.g., BTEX) and a few halogenated VOCs were measured in a number of studies. A comprehensive description of the studies and the results is given in the review report by the Victoria Environmental Protection Agency (1999).

3.1.6 Japan

In Japan, monitoring efforts of non-POP air toxics are mainly limited to formaldehyde, with biannual measurements of formaldehyde initiated in 1985/1986. The monitoring sites were classified into three types of residential areas (near industrial zones, in major cities, and in medium and small cities) and background areas. The number of sampling sites has been increasing over the time, from 24 sampling sites in 1990 to 40 sites in 1994. Some measurement results can be found in annual reports of Quality of the Environment in Japan (<http://www.env.go.jp/en/w-paper/index.html>).

3.2 POP Monitoring Programs

The Stockholm Convention targets twelve POPs. They are aldrin, endrin, dieldrin, toxaphene, chlordane, mirex, DDT, heptachlor, dioxins, furans, PCBs, and hexachlorobenzene (<http://www.pops.int>). Below is a brief summary of POP monitoring programs around the world.

3.2.1 United States of America

The USEPA has established a National Dioxin Air Monitoring Network (NDAMN) to serve three primary purposes (Cleverly et al., 1999):

- To provide measurements of background atmospheric levels of dioxin-like compounds in different geographic regions of the U.S.;
- To determine the atmospheric levels of dioxin-like compounds in agricultural areas where livestock, poultry and animal feed crops are grown; and
- To provide data to evaluate results from long-range transportation and deposition air models.

The network has been implemented in phases due to the complexity in operation and resource constraints. The first phase of the NDAMN consisted of nine monitoring stations, operating from June 1998 to June 1999. By the end of 1999, the NDAMN had expanded to 21 sampling stations (Cleverly et al., 2002), and at the end of 2001, 32 sites were operational (Riggs et al., 2002). The NDAMN sampling stations are located in rural and other non-impacted areas, and use standard USEPA method TO-9A modified in order to achieve congener-specific detection limits of 0.1 fg/m³ for 2,3,7,8-TCDD and 10 fg/m³ for OCDD (Ferrario et al., 2001; CARB, 2002b). The standard TO-9A method adopts a sampling duration of 24 hours per sample. In the modified sampling protocol, samples are taken every other month and four 6-day continuous samples are taken in each sampling month. On the 7th day, the day immediately after a 6-day sampling, the filter is replaced with a clean one but the polyurethane foam cartridge (PUF) is not changed until the end of the sampling month. The four 6-day sub-filter samples in each month are combined to produce one monthly composite sample. The composite filter sample, together with the 24-day PUF sample, represents a pair of monthly samples. Samples are shipped to the USEPA's Environmental Chemistry Laboratory for extraction, clean-up and analysis with High Resolution Gas Chromatography (HRGC) coupled with High Resolution Mass Spectrometry in accordance with a modification of USEPA method 1613 (Cleverly et al., 2002).

The sampler is operated at a flow rate of 0.24 m³/min. As a result, each pair of samples corresponds to a sampled air volume of 8300 m³. With a detection limit of 0.5 pg/sample for TCDD/CDF (Ferrario et al., 2001), dioxins air concentration as low as 0.06 fg/m³ can be detected. During the whole process, all meteorology parameters are simultaneously recorded, including ambient temperature, wind direction and speed.

California has established its own ambient dioxins air monitoring program. It is a two-year monitoring program with the objective of providing information on ambient levels of dioxins and dioxin-like compounds in California (CARB, 2002c). This program adopts the same sampling and analytical techniques as the NDAMN. Unlike the rural focus of the NDAMN, the California program has an urban focus and emphasizes sampling for ambient dioxin levels in populated areas. Selection of individual sites has been made in accordance with the objective of achieving 'population-oriented' sampling. Nine sampling sites, five in the San Francisco Bay area and four in the Los Angeles basin, have been deployed in this program, with ambient air monitoring conducted over a two-year period starting in December 2002. The monthly

sampling frequency allows the observation of any seasonal variations in dioxin concentrations due to changing emissions and/or meteorological effects.

In both the NDAMP and CADAMP, PCBs are among the target pollutants to be monitored in addition to dioxins.

3.2.2 The Japanese Persistent Organic Pollutants Monitoring Program

The Ministry of the Environment of Japan introduced a “System of Investigation of Chemical Substances in the Environment” in 1974. The POP monitoring program is regarded as an extension of the investigation of Chemicals in the Environment (Ministry of the Environment of Japan, 2002). Samples are taken from air, water, sediment, and wildlife matrices. The major objective of the Japanese POP monitoring program is to determine the current average concentrations and future trends in ambient air for the 12 POPs targeted by the Stockholm Convention. The survey of some POPs was started as early as 1986 for air, and 1982 for other environmental media.

In the POP monitoring network, sample collection points are set with an average interval of about 100 km. Sampling locations are selected from geological and land-use points of view, including cities (densely populated areas, business districts or industrial districts), agricultural areas, mountains, and coasts. Sample collections are conducted in summer and in winter, or in all four seasons if feasible, with the duration of one or two weeks. When feasible, daily changes are monitored during each sampling period. Detailed investigations are conducted at selected sampling points for optimization of sampling protocols as well as for obtaining information on the deposition of POPs.

A comprehensive national survey for dioxins was launched in 1998 to monitor dioxin levels in air, water, soil, and bottom sediments. A total of 387 sites were selected covering large and small cities (N=354), rural (N=7), roadside (N=6) and those closed to emission sources (N=20). This program aimed to determine: (i) the actual pollution status throughout Japan during 1998; (ii) the actual status of pollution in different environmental media during the same period.

Dioxins are measured at different frequencies, depending on the environmental matrices. In ambient air, dioxins are measured four times a year, one in each season. Dioxins in soot, particulate and public waters from the areas closed to the sources are measured twice a year, in summer and in winter. Once a year dioxin measurements are made for public waters and ground waters in summer, sediments in summer, soils in summer, and aquatic organisms in autumn. All analyses follow in-house manuals prepared by the Ministry of the Environment.

3.2.3 United Kingdom

Dioxins, furans, and PCBs are on the list of monitored compounds in the Toxic Organic Micro Pollutant (TOMPS) program in the UK. The Department of Environment launched this program in 1991. Sampling and analysis are carried out at 17 sites covering urban, rural, and semi-rural areas around the country, of which 3 are presently closed. Among the 14 sites that are operating, samples are collected for analysis of dioxins and PCBs. The duration of each sample is two weeks while chemical analysis is conducted every six months.

3.3 Hong Kong's TAP Monitoring Program

3.3.1 Monitoring Objectives

The objectives of monitoring selected toxic air pollutants (TAPs) in the ambient air of Hong Kong include (HKEPD, 2001a):

- (1) To determine representative concentrations in areas of high population densities, and hence the exposure of the broad mass of population to toxic air pollutants of concern and to assess the consequential risks to human health, as well as other environmental effects, resulting from this exposure.
- (2) To determine the general background as well as the highest concentrations expected to occur in the area covered by the monitoring network, and hence to determine the ranking of Hong Kong when compared to other metropolitan cities.
- (3) To provide data for policy formulation of environmental control strategies and legislation, air pollution control plans, etc.
- (4) To observe long term trends in ambient levels of toxic air pollutants.
- (5) To validate and provide input for atmospheric dispersion models.
- (6) To study and evaluate pollution interactions and patterns, including long-range atmospheric transport.

Under these objectives, Central/Western and Tsuen Wan, were selected to represent typical residential/commercial environments in Hong Kong. The Tsuen Wan site is located on top of a school building, therefore has the additional advantage of providing TAP exposure data relevant to a susceptible group of the population. The two TAP monitoring stations began operation in July 1997, with samples taken regularly for measurement of ambient TAP concentrations. The target TAPs fall into six categories according to their chemical properties, namely VOCs, dioxins / furans, PAHs, PCBs, carbonyls, and hexavalent chromium. The sampling schedules have varied among the TAPs. For example, VOC samples are collected every six days; carbonyl samples are collected every twelve days; dioxin, PCB, and PAH samples were collected roughly every month before July 1999 and roughly every twelve days after July 1999. More details are listed in Table 3.1.

Besides monitoring at the two regular TAP stations (Tsuen Wan and Central/Western), the TAP monitoring program in Hong Kong also includes a number of ad hoc monitoring exercises for specific purposes. These include the dioxin monitoring exercise commissioned by the HKEPD and carried out by the ENSR (HKEPD, 2002), the Tai Mo Shan POP study (Louie and Sin, 2003), and the 12-month PM study.

The latter involves the setting up of two additional TAP stations, one at a roadside location in Mongkok and one in a rural area in Hok Tsui from November 2000 to October 2001. The roadside (Mongkok) site was designed to obtain exposure levels of TAPs by pedestrians close to traffic; monitoring at this site allows quantification of TAPs from vehicular sources in close-to-worst-case scenarios. The rural (Hok Tsui) site was to provide background TAP levels in Hong Kong. Situated on the coastline of Southern China, Hok Tsui has distinct seasonality, receiving continental outflow in the winter and maritime inflow in the summer.

Furthermore, in addition to TAP-oriented monitoring programs, the HKEPD also collects high volume TSP and RSP samples every six days for chemical analyses in its general air quality monitoring network. Metal species and one PAH compound, benzo(a)pyrene (BaP), are among the aerosol constituents monitored. Some metal species, such as cadmium, lead, and nickel, are known air toxics. In 2001, high volume TSP samples were collected at one roadside and nine general stations while high volume RSP samples were collected at one roadside and six general

stations (HKEPD, 2001b). A list of the general air quality monitoring stations and characteristics of each station can be found in the annual air quality reports prepared by the HKEPD (e.g., HKEPD, 2001b).

The routine TAP monitoring program in Hong Kong is modeled after the California Air Toxics Program, which is one of the more established TAP monitoring programs in the world. The sampling frequency, data quality control procedures and analysis techniques of the two programs are similar. However, the number of TAPs analyzed and reported is much higher in Hong Kong. This may be related to the fact that the Hong Kong program has much shorter history, and the HKEPD is still gathering information through studies such as this to better understand the overall TAP situation in Hong Kong. In terms of network design, the current network (with two routine TAP monitoring stations at Central/Western and Tsuen Wan) was established to provide information on the ambient TAP levels to help assess the population exposure to those pollutants (HKEPD, 2000), which is similar to the objectives of most other monitoring programs.

However, the small geographical extent, the complex topography, and the high but localized population distribution in Hong Kong dictate that TAP distribution in Hong Kong must be quite different from the national or state-wide networks listed above. Considering the small geographic size of Hong Kong, having two routine TAP monitoring stations may sound a lot. Nevertheless, using data from the ad hoc studies, our analysis found systematic and substantial variations in TAP levels across Hong Kong that are not well-captured by the two station network. To address this and other monitoring issues, changes were recommended for future TAP monitoring efforts that will be described in further details in subsequent chapters.

3.3.2 Sampling and Analysis Methodologies

The sampling and analysis methodologies of the six types of TAPs are summarized in Table 3.2 and described below. Procedure manuals were prepared for field operation in accordance with the respective USEPA standard methods and CARB standard operation procedures to ensure data quality (HKEPD, 2001).

A Dioxins

Three dioxin monitoring efforts of different durations have been carried out in Hong Kong. The first monitoring effort commenced in July 1997 and is on-going as part of the routine TAP monitoring program. Two sites, Central/Western and Tsuen Wan, were selected to represent typical residential / commercial environments in Hong Kong (Table 3.1). In addition, two temporary TAP monitoring stations were established near the roadside at Mongkok and in a rural site (Hok Tsui) for a period of twelve months from November 2000 to October 2001.

The third effort was carried out by the Special Waste Group at the HKEPD (hereafter referred to as the SWG study). The sampling and analysis in the SWG study was handled by ENSR Environmental International Inc. This program includes monthly monitoring of ambient air at three designated locations (Nim Wan, Sha Ling, and Tseung Kwan O) from April 2001 to March 2002. Nim Wan and Tseung Kwan O are two landfill sites and Sha Ling is a site near a livestock waste composting plant. In addition, biannual collection (August 2001 and January 2002) of soil, vegetation and tree bark samples was carried out at the Chemical Waste Treatment Centre and at four other designated locations. This study also includes biannual sampling of leachate at closed and operational landfills (Jordan Valley, Tseung Kwan O Stage II/III, Shuen Wan, Gin Drinkers Bay, NENT, WENT and SENT landfills) (HKEPD, 2002).

These three dioxin monitoring efforts are different in their objectives. Monitoring at Central/Western and Tsuen Wan aims to provide data for assessing the population exposure to

selected TAPs and to provide a picture of long-term trends in concentration levels (HKEPD, 2000). The 12-month study at Mongkok and Hok Tsui serves two objectives: the roadside Mongkok site was designed to obtain the exposure levels of TAPs for pedestrians close to traffic, while the rural site at Hok Tsui was intended to provide background TAP levels in Hong Kong. The SWG study was designed to determine the dioxin concentrations in various media near known or suspected local dioxin sources.

The sampling and analysis protocol of dioxins / furans is in accordance with USEPA method TO-9A (USEPA, 1999b). Air samples were collected using a high-volume sampler equipped with an 8x10 in filter and a polyurethane foam cartridge. Each sample was collected at a flow rate of 230 l/min for duration of 24 hours. Two laboratories, the ENSR and the Government Laboratory (GL) participated in the dioxin analysis. ENSR analyzed samples collected prior to July 1998 in the regular TAP monitoring program, along with all the samples collected in the SWG study. The GL analyzed all samples collected after June 1998 at all the four TAP monitoring sites (Central/Western, Tsuen Wan, Mongkok and Hok Tsui).

B Polycyclic Biphenyls

PCBs are monitored in the routine TAP monitoring program at Central/Western and Tsuen Wan, and also in the 12-month study at Mongkok and Hok Tsui. Details of these two monitoring programs are given in Table 3.1. The GL is responsible for PCB analyses for samples from these two programs; only the total PCB concentrations are measured.

An additional monitoring study involving PCBs was the SWG study, which included monthly monitoring of ambient air for PCBs at three designated locations (Nim Wan, Sha Ling, and Tseung Kwan O) from April 2001 to March 2002. As well, biannual collection (August 2001 and January 2002) of soil, vegetation and tree bark samples was carried out at the Chemical Waste Treatment Centre and at four other designated locations. Measurements also included biannual sampling of leachate at closed and operational landfills (Jordan Valley, Tseung Kwan O Stage II/III, Shuen Wan, Gin Drinkers Bay, NENT, WENT and SENT landfills) (HKEPD, 2002). The chemical analyses from the SWG study were carried out by ENSR's contractor. Concentrations of 12 dioxin-like PCB congeners were measured.

The sampling and analysis protocol of the total PCBs was in accordance with the USEPA method TO-4 (USEPA, 1984). Samples were collected with a high-volume sampler consisting of a solvent cleaned quartz fiber filter and a polyurethane foam backup cartridge at a flow rate of about 230 l/min for 24 hours. The sampling schedule was twice a month after June 1999. Prior to June 1999, sampling was carried out on an irregular schedule with lesser sampling frequency. The filter and polyurethane foam cartridge were placed in clean, sealed containers and delivered to the GL, where analysis of the total PCBs were carried out by gas chromatography/electron capture detector. Due to the irregular sampling schedule, data before June 1999 should be used with caution when compared with data taken afterwards.

C Polycyclic Aromatic Hydrocarbon

Three PAH monitoring efforts of different scales have been carried out in Hong Kong. The first two include the regular TAP monitoring program at Central/Western and Tsuen Wan and the 12-month monitoring program at Hok Tsui and Mongkok (Table 3.1). Sixteen PAH compounds were measured in these two programs.

A third monitoring effort is the monitoring of BaP at ten stations as part of the general air quality monitoring network across the territory. The ten stations are Central/Western, Kwai Chung, Kwun Tong, Mongkok, Sham Shui Po, Tung Chung, Tai Po, Tsuen Wan, Shatin, and Yuen Long. Both TSP and RSP samples were collected for analysis of BaP once every six days

at most of the ten stations in this network.

The sampling and analysis of the sixteen PAH levels from the Central/Western and Tsuen Wan TAP stations were in accordance with USEPA method TO-13 (USEPA, 1993); similar analytical methods were used for TSP and RSP samples at other AQMSs. PAH samples were collected by high-volume air samplers equipped with a PUF/XAD-2 cartridge unit over a continuous 24-hour period at a designated flow rate of 230 l/min. Sampling at the Tsuen Wan and Central/Western stations in the regular TAP monitoring program was conducted concurrently. Collocated sampling was conducted quarterly at each station for evaluation of precision, with the precision of field replicates found to be $\pm 30\%$ or better. A High Pressure Liquid Chromatography/Ultraviolet (HPLC/UV) detection method is used to identify and quantify the individual PAH compounds.

D Volatile Organic Compounds

The canister-based USEPA method TO-14A (USEPA, 1999c) was adopted. This method allows the determination of more than 100 VOCs, including a wide range of recognized TAPs. Volatile organics are collected into 6-liter stainless steel, Summa passivated canisters or fused silica lined SilcoCan canisters and are subsequently analyzed at the GL using Gas Chromatography/Mass Spectrometry (GC/MS). Each sample is collected at a flow rate of 10 ml/min over a period of 24 hours starting and ending at midnight. The sampling frequency is once every six days at the two regular monitoring sites (Tsuen Wan and Central/Western) and once every twelve days at the two 12-month study sites (Hok Tsui and Mongkok). Such a sampling frequency should be sufficient for providing data to characterize annual average concentrations. Furthermore, the six day sampling schedule ensures that samples will be taken during all of the seven days of the week. Precision of sampling was assessed by duplicate samples on about 10% of the sampling days.

E Carbonyls

Carbonyls have been monitored at Tsuen Wan and Central/Western since July 1997, where a 24 hour integrated sample is collected every twelve days. The sampling and analysis protocol is in accordance with the USEPA standard method TO-11 (USEPA, 1987). Briefly, the method can be described as follows.

Sep-Pak cartridges impregnated with a known amount of 2,4-dinitrophenylhydrazine (DNPH) are used in conjunction with a XonTech Automated Carbonyl Sampler. Ambient air is pulled through the carbonyl sampler at a flow rate of 700 ml/min to give a total sampled air volume of ca. 1 m³. Carbonyls in the air react with the DNPH reagent to form hydrazone products. After sample collection, the DNPH cartridges are packed in ice and transported to the analytical laboratories for analysis. The hydrazone derivatives are eluted from the cartridges with acetonitrile and the resulting eluents are analyzed using HPLC with UV detection. The analytical method allows simultaneous determination of 17 carbonyl compounds.

Two analytical laboratories participated in the analysis of carbonyl samples. ENSR analyzed samples before July 1998 and the GL analyzed data afterwards. Unusually high carbonyl concentrations were reported by ENSR in some of the early 1997 samples when the TAP monitoring program was quite new. No inter-laboratory comparison was done to compare the results of these two analytical laboratories. Hence, only data from the GL will henceforth be used in subsequent discussions to ensure reliable analysis of the annual and seasonal trends as well as the spatial variation, to avoid the complications caused by systematic differences between the two laboratories.

F Elemental Carbon

Elemental carbon (EC) concentrations have been measured since 1998 using high-volume TSP and RSP samples collected in most of the ten general air quality monitoring stations. Small punches of the filters were removed and analyzed for EC and organic carbon (OC) using a thermal / optical method (Birch and Cary, 1996; Sin et al., 2002). The method uses programmed thermal vaporization and oxidation steps to evolve carbonaceous species into the gas phase for detection. The filter is first heated in a helium atmosphere in an oven, the temperature of which is programmed to rise stepwise from ambient to 350°C for 70s, 550°C for 70s, and 850°C for 110s. The oven is then cooled to 550°C and oxygen is introduced to the carrier gas to oxidize the residue carbon. The oven temperature is raised gradually from 550 to 850°C, with intermediate steps at 600, 700, 750, 800, and 850°C for a residence time of 50, 40, 30, 30, and 70s, respectively. The evolved carbon is oxidized to CO₂ on a heated catalyst and quantified as methane after an additional step to convert CO₂ to methane. An optical feature is incorporated in the method to correct for pyrolytically generated EC from OC parent materials by monitoring the filter transmittance of a laser light at 680 nm. A split line for EC and OC is set where the filter transmittance returns to the initial value at beginning of the analysis. Carbon that evolves before the split line is regarded as OC while the portion of carbon that evolves after the split line is regarded as EC.

G Hexavalent Chromium

Monitoring of Cr(VI) has been carried out at Tsuen Wan and Central/Western since July 1997. Sampling and analysis of Cr(VI) is in accordance with CARB method MLD 039 (CARB, 2002d). Samples are collected onto bicarbonate impregnated 47 mm ashless cellulose filters at a flow rate of 10 l/min for 24 hours. The sampling frequency is once every twelve days. The filters are then extracted in de-ionized water via sonication for three hours. The extract is analyzed by Ion Chromatography (IC) using a system comprised of a guard column, an analytical column, a post-derivatization module, and a UV-Vis detector. In the analysis procedure, Cr(VI) exists as chromate due to the near neutral pH value of the eluent. After eluting from the IC column, Cr(VI) forms a complex with diphenylcarbohydrazide (DPC), which can be detected at 540 nm (HKEPD, 2001).

H Toxic Elemental Species

Toxic elemental species exist mostly in the aerosol phase, and as a result can be determined by analyzing aerosols collected on filter substrates. High volume TSP and RSP samples are used for the determination of toxic elements in Hong Kong. The USEPA TO-3 coupled with two analysis techniques are adopted for such measurements (USEPA, 1999d). The inductively coupled plasma atomic emission spectroscopy (ICP-AES) analysis is used for aluminum, beryllium, calcium, cadmium, chromium, copper, iron, magnesium, manganese, nickel, lead, vanadium, and zinc, and the Flow Injection Analysis – Atomic Absorption (FIA-AA) technique is used for arsenic, mercury, and selenium. These techniques do not differentiate the various chemical forms of a given element, so as a result the total amount of an element is measured. For the determination of a specific chemical form, a special method needs to be used. Cr(VI) (CARB, 2002d) and vapor mercury (e.g., Temme et al., 2003) are such examples.

3.4 Tables for Chapter 3

Table 3-1 Summary Information for TAP Monitoring Programs

Table 3-2 TAP Monitoring Methods

4 Review of TAP Measurement Data

A search has been conducted to gather information and publications in open literature on TAP measurements⁴ in Hong Kong. The open literature data and the HKEPD data are combined to provide an overview of current TAP levels in the region. For the purpose of comparison, we have also searched open literature TAP data in selected major cities in other parts of the world, including web-based sources.

Initial inspection of the TAP data suggested that the characteristics of many parameters were quite different before and after July 1998, which coincided with the time the analytical labs were changed from ENSR to the Governmental Laboratory for many of the TAP species. It is possible that there were systematic differences in analytical results by different analytical laboratories. Since it is difficult to differentiate such differences with the actual trends in the data, after consulting with the HKEPD, it was agreed that data measured after June 1998 would be used for detailed analyses in this study.

Nevertheless, other problems in data quality were still apparent even after applying this cutoff criterion. For example, there were a number of TAPs (e.g., acrolein, p-tolualdehyde, crotonaldehyde) with reported concentrations dropping to a constant value rather abruptly. It was difficult for us to tell whether this was real (i.e., the concentration dropped below the detection limit) or there were some problems with the data or the analysis.

Moreover, on the high side, a number of very spiky measurements (e.g. formaldehyde, m-dichlorobenzene and 2,2,4-Trimethylpentane) occurred in the TAP database at one of the TAP sites but not both. Without collaborating measurements from the other TAP station, it is impossible to say whether these extremely high measurements are related to local episodic events or analytical problems. Inclusion of this data would skew the statistics unreasonably, particularly those numbers relating to trend analyses. Hence, a 95% percentile filter was applied to each of the TAP time series, retaining only data at or below the 95% percentile level for subsequent analyses.

Except for this 95% percentile filtering and a small number of TAPs excluded for clearly questionable data, we have assumed that the data record is accurate as is.

4.1 Diesel Particulate Matter

Diesel exhaust contains mixtures of gaseous and particulate constituents, resulting from incomplete combustion of diesel fuel, lubricating oil or other additives. Diesel particulate matter (diesel PM) has been classified as a probable human carcinogen (IARC, 1989). It is among the 33 priority hazardous air pollutants compiled by the USEPA. The California Air Resources Board has also identified diesel PM as a toxic air contaminant (CARB, 2000). The Eureka study ranks diesel PM as the most important toxic air pollutant in Hong Kong on the basis of a health risk assessment using emission data (Eureka, 1996).

Diesel PM is composed of a center core of EC and adsorbed organic compounds (e.g., PAHs), as well as small amounts of sulfate, nitrate, metals, and other trace elements (USEPA, 2002). EC is a major component of diesel PM, contributing approximately 50-85% of diesel PM mass, while the organics takes up ~20% (range: 7%-49%) of the diesel particle mass. The

⁴ Noting that they can also have significant health impact on the population, there is movement internationally in officially designating criteria pollutants (e.g. O₃, NO₂, SO₂) as TAPs (Environment Canada 2003a). Hence, we have also included the criteria pollutants in this review.

composition of diesel PM varies with fuel types, engine types, operating conditions (e.g., idle, accelerate, decelerate), fuel formulations (e.g., high/low sulfur fuel), and the availability of other installed control devices.

Despite considerable research efforts devoted to the chemical characterization of the solvent extractable fraction of diesel PM, a unique tracer has not been found (Birch and Cary, 1996). However, some measures of exposure must be selected. As EC constitutes a large portion of diesel PM mass and can be quantified at low levels, the use of EC as a surrogate for diesel PM has been proposed and widely used (USEPA, 2002). Diesel PM is not the only source for EC in ambient environments since other combustion processes of carbonaceous fuels also contribute to EC loading in the air. As a result, use of EC as a direct surrogate could overestimate the contribution of diesel PM. To complicate matter further, there is no consensus on the analytical methods for EC measurements, and different methods are known to give different results (e.g., Chow et al., 2001). All these contribute to uncertainties in the health risk assessment associated with diesel PM.

4.1.1 Air Quality Guidelines

Diesel PM has long been recognized as harmful to human health. The WHO has suggested an annual air quality guideline of $2.3 \mu\text{g}/\text{m}^3$ on the basis of its non-carcinogenic health endpoints (WHO, 2000). The WHO also estimates that diesel PM has a cancer risk of 16-71 per million for every $1 \mu\text{g}/\text{m}^3$ of diesel PM. The California EPA (CEPA) recommends using a cancer risk of 300 in a million per $\mu\text{g}/\text{m}^3$ for diesel particulate (STAPPA & ALAPCO, 2000), and also recommends a chronic reference exposure level of $5 \mu\text{g}/\text{m}^3$ (OEHHA, 2002).

4.1.2 Observations and Findings

A Distribution of EC Between RSP and TSP

Table 4.1-1 lists the distribution of aerosol mass, OC, and EC between RSP and TSP at the ten monitoring stations in Hong Kong. While the RSP aerosol mass accounts for on average 61-74% of the TSP mass, OC and EC in RSP account for on average 77-89% and 86-97% of those in TSP, respectively. In comparison with the bulk aerosol mass, the enrichment factors in the RSP fraction range from 1.2-1.3 for OC and 1.2-1.5 for EC. It is clear that both OC and EC are enriched in the RSP fraction. In particular, the majority of EC mass is associated with RSP particles, consistent with its combustion-derived sources. For this reason, the ensuing discussion of EC temporal and spatial distributions is based on the EC concentrations in RSP samples.

B Ambient Concentrations of EC in RSP

Table 4.1-2 summarizes the annual average EC concentrations in RSP samples at the 10 measurement sites. EC accounts for 6.4-16.7% of RSP mass, indicating EC is an important contributor to RSP mass loadings in Hong Kong. The order of abundance of EC by location is Mongkok > Kwai Chung > Kwun Tong > Tsuen Wan > Yuen Long ~ Sham Shui Po > Tai Po > Central/Western ~ Shatin > Tung Chung. The roadside station at Mongkok has the highest EC concentrations while the Tung Chung station in residential surroundings has the lowest EC concentrations. This order reflects the spatial distribution of combustion source intensity and the proximity of individual monitoring stations to major combustion sources.

Although a steady declining trend has been observed in TSP, RSP and NO_x at the roadside (HKEPD 2003), there is no discernable annual trend in EC concentrations during the four annual cycles (1998-2001) at nine of the ten monitoring sites. This suggests that other factors are also important in affecting the EC concentrations at the general stations. A rising trend in

EC concentrations is seen for three annual cycles from 1998 to 2000 at Kwai Chung, which is also a general site. This may be related to the proximity of the Kwai Chung AQMS to the nearby container terminals, as residual fuel combustion from marine shipping contributes significantly to variations in EC (Wu, 2003). It is impossible to assess whether this rising trend continues beyond 2000 as the EC measurements at Kwai Chung are only available up to 2000. We recommend that elemental analysis for RSP samples be resumed for Kwai Chung in the future to monitor the change in EC concentrations at this site, and to help study the impact of marine shipping on the overall EC spatial distribution in Hong Kong.

The annual average EC concentration ranged from 2.7 to 7.5 $\mu\text{gC}/\text{m}^3$ for the nine general air quality monitoring stations. Approximately 40% of annual means exceed the 5 $\mu\text{g}/\text{m}^3$ chronic reference exposure level recommended by the CEPA. Every annual EC mean at the roadside station exceeds this California exposure limit.

The EC levels in Hong Kong are compared with those of a few other locations in the Eastern Asian region (Table 4.1-3), noting that EC measurements by thermal methods are operationally defined and method-dependent (Chow et al., 2001). Differences as large as 29% have been reported for ambient aerosol samples in inter-laboratory comparison exercises (Schimid et al., 2001), and as a result, EC measurements using different thermal protocols are not directly comparable. Nevertheless, values in Table 4.1-3 can be regarded as a rough indicator for the relative abundance of EC in different locations. The EC levels at urban background stations in Hong Kong are similar to those found in agricultural areas and urban residential environments in the Yangtze River Delta region, lower than other urban cities in Asia, but a little higher than European cities such as Helsinki, London and Vienna.

C Seasonality

Figure 4.1-1 plots the time series of EC concentrations for all ten monitoring stations. No consistent seasonal pattern can be identified for EC. To the contrary, OC has a clear seasonality with higher concentrations observed in the winter months and lower concentrations observed in the summer months (Figure 4.1-2). A possible explanation for such a contrast is offered below.

The OC seasonality may be attributed to the seasonality of long-range transportation. During the summer months, aerosol carbon loading in Hong Kong is largely from local emission sources under the influence of clean marine air masses. During the winter months, Hong Kong receives aged polluted air masses originating from northern China in which OC is enriched relative to EC due to the formation of secondary OC (SOC) (Turpin and Huntzicker, 1991). SOC was estimated using EC as a tracer for primary emissions, and accounted for a significant fraction of total OC both in summer and in winter. However, SOC levels in winter were found to be three times those in the summer, thus the additional OC input through long-range transportation is mainly responsible for the higher OC concentrations in the winter. The elevated SOC levels in the winter in comparison with those in the summer indicate that long-range transportation is a major carbonaceous aerosol source in Hong Kong.

To the contrary, seasonality of EC is much weaker or not discernable, and is variable among the monitoring sites. EC concentrations in a cluster of five stations to the immediate north of the city's container port are higher in the summer than in the winter. EC concentrations at the three stations further north of the container terminal do not have discernable seasonality whereas EC at the two stations south of the terminal have the same seasonality as OC. This can be explained by seasonal influence of ship emissions at the port. The stations to the north of the container terminal are downwind of ocean-going vessel emissions in the summer, but upwind in the winter. The variable EC seasonality is a result of two counteracting factors, i.e., the

influence of ship emissions versus dirtier air masses transported from the North. Spatial and seasonal variation patterns of other pollutants characteristic of residual fuel oil combustion (e.g., sulphur dioxide, vanadium, and nickel) have further provided corroborative evidence for the influence of ship emissions (Wu, 2003).

4.1.3 Summary

EC levels in Hong Kong are generally high. Many locations exceed $5 \mu\text{g}/\text{m}^3$, the chronic reference exposure level set by California. The roadside environment has the highest EC levels compared with other monitoring sites, consistent with the knowledge that vehicular emissions are major sources of EC. For the general stations, Kwai Chung has the highest EC concentration, likely related to EC emissions from residual fuel oil combustion by marine vessels at or near the container terminal at Kwai Chung.

OC has clear seasonality with higher concentrations observed in the winter months and lower concentrations observed in the summer months. In particular, SOC in winter was found to be three times that in the summer. The elevated SOC levels in the winter in comparison with those in the summer indicate that long-range transportation is a major carbonaceous aerosol source in Hong Kong. In contrast, seasonality of EC is much weaker or not discernable and variable among the monitoring sites. Many of the AQMSs with higher EC concentrations (e.g. Kwai Chung and Tsuen Wan) are to the north of the container terminal and hence they are downwind of ocean-going vessel emissions in the summer, but upwind in the winter. The variable EC seasonality may be a result of two counteracting factors, i.e., the influence of ship emissions versus dirtier air masses transported from the North.

As EC is one of the most important parameters used in health risk assessment, further clarification of the role of marine contributions to the EC budget in Hong Kong is recommended in future studies.

4.1.4 Tables and Figures for Section 4.1

A Tables

Table 4.1-1 Distribution of Aerosol Mass, OC, and EC between RSP and TSP

Table 4.1-2 Annual Means of EC Concentrations in RSP Samples

Table 4.1-3 Summary of EC Concentrations in Urban Areas around the World

B Figures

Figure 4.1-1 Time Series of EC Concentrations in RSP Samples in Hong Kong

Figure 4.1-2 Time Series of OC Concentrations in RSP Samples in Hong Kong

4.2 Toxic Elemental Species

Twelve elemental species are listed as priority toxic air pollutants in Hong Kong in the Eureka study (Table 4.2-1). Eight of them are on the 33 priority HAP list compiled by the USEPA. Three elemental species (arsenic, hexavalent chromium, and nickel) have been identified as carcinogens by the WHO (WHO, 2000). Hexavalent chromium has also been identified as a toxic air contaminant by the CARB (CARB, 1987). Discussions below are limited to these twelve elemental species, and with the exception of chromium, only the total amount of a given elemental substance is available. Phosphorus, an element identified in the Eureka Report, was not available in the TAP database as it was found to be below its MDL in a 2001 study by the HKEPD. Hence, our analyses are focused on the other elemental species listed above.

4.2.1 Air Quality Guidelines

Air quality guidelines in various forms are made for a number of toxic elements. Table 4.2-2 lists the guidelines proposed by the state governments of California, Minnesota, and North Carolina in the US, the WHO, Australia, the UK, and European Communities. Among these toxic elements lead is a criteria air pollutant in Hong Kong with its air quality objective set to a three-month average of $1.5 \mu\text{g}/\text{m}^3$ in TSP samples.

4.2.2 Observations and Findings

A Comparison of Elemental Measurements by ICP-AES and by XRF

The most-often used analytical methods for measuring elements in aerosols are either direct analysis of the filter substrate using X-ray fluorescence (XRF) or digestion of filter materials followed by Induced Coupled Plasma – Atomic Emission Spectroscopy (ICP-AES) analysis of the resulting solution. Both methods can simultaneously determine a large number of elements in a single analysis.

The general air quality monitoring program uses the ICP-AES technique to determine elemental concentrations in TSP and RSP samples, with the GL undertaking the analysis. A recently completed ad-hoc study has collected Fine Suspended Particulate (FSP or $\text{PM}_{2.5}$) samples at Tsuen Wan, Mongkok, and Hok Tsui over a period of 12-months spanning Nov. 2000 to Oct. 2001. The Desert Research Institute (DRI) analyzed the elemental substance concentrations in these FSP samples using the XRF technique. The sampling dates at Tsuen Wan during this 12-month ad-hoc FSP study were the same as those of filter-based TSP and RSP sample collection in the general air quality monitoring program. This offers a unique opportunity for comparison of XRF and ICP-AES techniques. We note that the sizes of aerosols collected are different in the two data sets (FSP versus RSP). However, the two data sets can be compared to see if particular aerosol species exist mainly in the fine mode.

The two methods offered different MDLs for the same elements, as reported by the GL and DRI. For example, the MDL for lead by XRF was almost 10 times higher than that by ICP-AES, whereas the MDL for selenium by XRF is 185 times lower than that by ICP-AES.

Figure 4.2-1 compares the two sets of measurement for five elements, arsenic, lead, nickel, vanadium, and zinc. The two sets of data are highly correlated, with r^2 greater than 0.94. The slope values are within 1.0 ± 0.1 for lead, nickel, vanadium, and zinc, and the slope is 1.3 for arsenic. The two types of measurements have excellent agreement for four of the five elements, suggesting that these elemental species are mostly in the fine mode. Arsenic in FSP was measured by DRI to be higher than arsenic in RSP. This discrepancy suggests that there is either a systematic positive bias in arsenic measurements by DRI or negative bias in arsenic

measurements by GL. However, this systemic bias should not affect our estimation of the trend or seasonality of this element.

B Method Detection Limits

Table 4.2-3 lists the MDL for Cr(VI) and other elemental species in RSP samples. The MDL for Cr(VI) is reported to be 3.0 ng/filter, which corresponds to an air concentration of 0.21 ng/m³. This MDL is roughly the same as the chronic inhalation reference exposure level of Cr(VI) in California. The majority of Cr(VI) data is above its MDL. The MDLs for the RSP samples are clearly element specific, ranging from 0.10 ng/m³ for beryllium to 19 ng/m³ for aluminum and barium. The MDL values reported by the CARB (CARB, 2002a) are also included in Table 4.2-3 for reference. For the majority of elements, the MDLs from CARB are 2-9 times lower than those reported by GL (red rows in Table 4.2-3). Barium, chromium, Cr(VI), and vanadium have similar MDLs for both CARB and GL (black). There were two exceptions, mercury, and selenium, for which the GL reported MDLs 4-6 time lower than the CARB (blue).

C Ambient Concentrations and Annual Trends of Elements in Hong Kong RSP

The annual statistics of elemental species are computed using a time break-up of November to October to be consistent with the sampling period of the additional 12-month FSP monitoring carried out at Tsuen Wan, Mongkok and Hok Tsui from November 2000 to October 2001. The annual average concentrations are tabulated in Table 4.2-4. A brief description is given below for each of the twelve toxic elements identified in the Eureka study.

Aluminum

The annual mean concentrations of aluminum in RSP range from 169 to 466 ng/m³ at a total of 10 sampling sites in Hong Kong during the four annual cycles from Nov. 1997 to Oct. 2001. The annual average concentrations of all-sites vary from 194 to 355 ng/m³. No annual trend is discernable. Yuen Long consistently shows the highest aluminum annual average levels in RSP among all the sites, exceeding the all-site average concentrations by 24-37%. Mongkok also consistently exceeds the all-site average values, but at a reduced percentage (5-17%) in comparison with Yuen Long.

The aluminum levels in FSP samples at three sites (Hok Tsui, Mongkok, and Tsuen Wan) are generally a quarter of the all-site average level in RSP. Mongkok and Tsuen Wan have both RSP and FSP measurements for the period of Nov. 2000 – Oct. 2001. The results show that 23% and 32% of aluminum exists in FSP as compared with RSP at Mongkok and Tsuen Wan, respectively. These observations reflect that a large portion of aluminum in aerosols is in the coarse mode and probably crustal material in origin. The aluminum level in FSP at Mongkok is similar to those measured at Hok Tsui and Tsuen Wan, indicating that vehicular traffic is not a significant source for aluminum in fine particles. The observation that Mongkok has slightly higher aluminum in RSP than the majority of other sampling locations may be explained by enhanced dust re-suspension by passing vehicles.

Arsenic

The annual average arsenic concentrations in RSP range from 2.0 to 5.9 ng/m³ at the ten monitoring stations over the period of Nov. 1997- Oct. 2001. The all-site annual average concentrations vary from 2.6 to 4.1 ng/m³. No annual trend can be discerned. The levels are generally similar across the entire monitoring network while levels at Yuen Long are consistently among the highest.

The arsenic levels in FSP samples at the three sampling sites (Hok Tsui, Mongkok, and Tsuen

Wan) are similar to those in RSP, indicating that arsenic mainly resides in the fine particulate fraction of aerosols. This is consistent with its size distribution measured at two sites in the Helsinki area, which showed a predominant presence of arsenic in aerosols of less than 2.5 μm (European Communities, 2001). The lack of spatial gradients between the roadside station Mongkok and other general air quality monitoring stations indicate that the emission sources of arsenic are not related to local vehicular traffic. This is in agreement with arsenic measurements made in European countries, where similar arsenic concentrations were observed at traffic hot spots and at urban background sites (European Communities, 2001).

The arsenic levels in Hong Kong are 6 to 10 times lower than the 30 ng/m^3 chronic inhalation reference exposure level guideline set by California. However, they are higher than the 2 ng/m^3 guideline suggested by Minnesota, which has set the most stringent guideline of all (Table 4.2-2).

Beryllium

The majority of beryllium concentrations fall below the MDL of 0.10 ng/m^3 . Only a few measurements exceed the MDL. As a result, no annual statistics are tallied for this element. The large number of below-MDL measurements for beryllium indicates that the beryllium levels in Hong Kong are well-below the 7 ng/m^3 chronic inhalation reference exposure level guideline set by California (Table 4.2-2).

Cadmium

The annual average cadmium concentrations in RSP samples ranged from 0.92 to 2.06 ng/m^3 at the ten sampling stations for the period of Nov. 1997 – Oct. 2001. The all-site annual average concentrations varied from 1.12 to 1.47 ng/m^3 . Yuen Long is again consistently the highest among all the sites. The annual average concentrations at Yuen Long exceed the all-site average values by 31-40%, with the highest concentration occurring in the latest 2000/01 annual cycle. The Mongkok roadside station does not show elevated cadmium levels in comparison with other stations, indicating that cadmium is unlikely to be a contaminant related to vehicular traffic. This observation is in agreement with cadmium measurements made in Europe, which show concentrations at traffic-exposed sites were in the same range as urban background levels (European Communities, 2001).

No cadmium measurements in FSP were reported. Size distribution measurements of cadmium at an urban site and a rural site in the Helsinki area indicated that cadmium was predominantly concentrated in the fine particle mode (European Communities, 2001).

Chromium and Hexavalent Chromium

The annual average chromium concentrations in RSP samples range from 1.44 to 4.41 ng/m^3 at the ten monitoring sites for the period of Nov. 1997 – Oct. 2001. There is no discernable annual trend, nor is there spatial variation between sites. The all-site annual average concentrations range from 1.72 to 2.45 ng/m^3 . Only a small fraction of chromium, 19% at Mongkok and 37% at Tsuen Wan, is associated with the fine particles. This suggests that chromium is mostly in coarse mode and may be related to crustal materials. The finding that Mongkok has higher chromium in RSP than the majority of other sampling locations may be explained by enhanced dust re-suspension by passing vehicles.

Chromium is the only elemental substance of which a specific form of the element (i.e., hexavalent chromium) is also determined. The annual statistics of Cr(VI) are summarized in Table 4.2-4d, with levels similar throughout the years and across the two sampling locations. They are only slightly higher than California's chronic reference inhalation exposure level. No

discernable annual trend can be observed, although the 2000/01 annual average concentrations are the lowest.

The annual means of Cr(VI) range from 0.20 to 0.28 ng/m³. Note these levels are close to the MDL 0.21 ng/m³, and consequently a large measurement uncertainty is associated with these values. No annual trend could be discerned. These Cr(VI) levels are approximately double the annual mean concentrations observed in Los Angeles (0.13 ng/m³, n=31) and San Francisco (0.12 ng/m³, n=26) in 2000 (CARB, 2002A). The CARB adopted a statewide measure to control the emission of Cr(VI) in chrome plating and chromic acid anodizing operation in 1988, and prohibited the use of Cr(VI) in cooling towers and in heat exchangers in 1989 (CARB, 1988 and 1989). These control measures could be the reasons for the lower Cr(VI) levels in California.

Monitoring of both Cr(VI) and chromium at Central/Western and Tsuen Wan allows the assessment of the fraction of Cr(VI) in chromium. Cr(IV) accounts for approximately 13% of the total chromium at both sites. Pratt et al (2000) reported that Cr(VI) accounted for at least one-seventh of the total chromium in Minnesota, similar to the observations in Hong Kong. Cr(VI) can be easily reduced to Cr(III), although the detailed reaction mechanisms are not yet well understood (USEPA, 1998). As a result, it is anticipated that Cr(VI) accounts for a minor fraction of the total chromium.

Lead

The 3-month and annual running average concentrations of lead in TSP samples ranged from 0.01 to 0.21 µg/m³, and 0.04 to 0.12 µg/m³, respectively. The Air Quality Objective for lead in Hong Kong is a 3-month average of 1.5 µg/m³, and the annual UK standard for lead is 0.25 µg/m³. Hence, the measured lead concentrations in Hong Kong are well below these lead air quality criteria. The Mongkok roadside station had similar lead levels to other locations in Hong Kong, indicating that local vehicular traffic is not an important source of lead. Also, the measured 3-month and annual average concentration in RSP samples ranged from 0.009 to 0.185 µg/m³ and from 0.037 to 0.087 µg/m³, respectively. The close range of RSP and TSP mean values show that most of the lead occurs in aerosol with a size of less than 10 µm. In fact the lead levels in FSP samples at the three sampling sites (Hok Tsui, Mongkok, and Tsuen Wan) are similar to those in the RSP samples, indicating further that lead mainly resides in the fine particulate fraction of aerosols.

A weak decreasing trend is found in the lead concentrations, mainly in the drop of peak 3-month average concentration during winter (Figure 4.2-2f). This may be related to the ban in the sale and supply of leaded gasoline in Hong Kong starting from 1 April 1999. Due to the voluntary action by oil companies in reducing the use of lead in gasoline, the lead concentration was already at a rather low level when unleaded gasoline was introduced to Hong Kong in April 1992 (HKEPD, 1999), and the decreasing signal in the annual mean lead concentrations is not that clear.

Yuen Long sees the highest lead annual average concentrations among all the monitoring stations. Lead levels at Yuen Long exceed the all-site annual average by 21-46%. The consistently elevated lead concentration at Yuen Long in comparison with other stations may indicate that regional or long-range transportation has a significant impact on lead levels in Hong Kong, as compared to local lead sources.

Manganese

The annual average manganese concentrations range from 10.6 to 30.0 ng/m³ at the ten

monitoring stations in the four annual cycles (Nov. 1997 – Oct. 2001). The all-site average concentrations vary from 14.0 to 22.1 ng/m³, with no obvious annual trend. These levels are far below the 150 ng/m³ guideline suggested by the WHO (Table 4.2-2).

Yuen Long again is the highest among all the sites, with the exception of Kwun Tong in the 1997/98 cycle, which has slightly higher annual average than Yuen Long. The annual average levels at Yuen Long exceed the all-site average by 12-41%. The manganese levels at Mongkok are lower than those at Yuen Long, and similar to those at the other non-roadside stations, suggesting local vehicular traffic is not an important source for manganese in RSP.

Mercury

The majority of mercury concentrations in RSP fall below the MDL of 0.49 ng/m³. Only a few measurements exceed the MDL. As a result, no annual statistics are tallied for this element. The MDL of mercury is much lower than the California chronic inhalation reference exposure limit of 90 ng/m³ for mercury and mercury compounds. The large number of below-MDL measurements indicates that the particle-bound mercury levels in Hong Kong are low in comparison with the health-based exposure limit (Table 4.2-2).

Nickel

The annual average nickel concentrations in RSP ranged from 1.85 to 7.46 ng/m³ at the ten monitoring stations during the four annual cycles. The most recent annual cycle (2000/01) has the highest mean concentrations at each station, suggesting a weak rising trend. The all-site annual means vary from 3.65 to 4.90 ng/m³. The highest 24 hour concentration in the dataset was recorded in August 2000 at Kwai Chung at 31 ng/m³, yet this remains lower than the 50 ng/m³ California chronic inhalation exposure limit (Table 4.2-2).

Kwai Chung consistently shows elevated nickel concentrations in comparison with other sampling locations. The annual average levels at this site exceed the all-site annual average concentrations by 33-81%. The Kwai Chung AQMS is near the Kwai Chung container terminal, where a strong influence from ocean-going vessels is expected. Combustion of heavy fuel oil in these transportation trades is a known major source for nickel (European Communities, 2001). Mongkok, Sham Shui Po and Tsuen Wan also display higher nickel concentrations than the other stations, possibly reflecting that they are located in the same air shed affected by heavy fuel oil combustion activities (Wu, 2003).

Vanadium

The annual average vanadium concentrations vary from 4.5 to 14.6 ng/m³ for all ten sites in the four annual cycles. The all-site annual means ranged from 7.2 to 8.4 ng/m³, with a consistent rising trend. The highest 24-hour concentration was 86 ng/m³, measured at Kwai Chung, which is still much lower than the 1 µg/m³ 24-hr air quality guideline set by the WHO.

The vanadium levels at Kwai Chung are clearly higher than all the other monitoring stations, exceeding the all-site annual average values by 43-96%. This suggests that Kwai Chung has a local vanadium source, in line with the elevated activities in combustion of heavy fuel oil due to the nearby cargo terminals.

A comparison of vanadium measurements in RSP and FSP at Mongkok and Tsuen Wan indicates that nearly all the vanadium resides within the fine particles.

Zinc

The annual average zinc concentrations varied from 102 to 237 ng/m³ at the ten sites in the four annual cycles. The all-site annual means range from 119 to 189 ng/m³. These levels are much

lower than the $35 \mu\text{g}/\text{m}^3$ California chronic inhalation exposure limit, and no annual trend is discernable. Yuen Long has the highest zinc concentrations in the last three annual cycles (Nov. 1998 – Oct. 2001), exceeding the all-site annual average concentrations by 14-64 %. Roadside Mongkok has similar levels of zinc in RSP as the general air quality monitoring stations. This suggests that local vehicular traffic is unlikely a significant source for zinc in RSP.

A comparison of zinc measurements in RSP and FSP at Mongkok and Tsuen Wan indicates that nearly all the zinc resides within the fine particles.

D Seasonal Variation

A three-month running average is used to identify seasonal trends within our samples. Figure 4.2-2 plots the 3-month running average curves for aluminum, arsenic, cadmium, chromium, Cr(VI), lead, manganese, nickel, vanadium, and zinc using monitoring data at Tsuen Wan, Central/Western, Mongkok, and Kwun Tong as examples.

Six elemental species, aluminum, arsenic, cadmium, lead, manganese, and zinc, exhibit a clear seasonal pattern in that higher levels were observed in the winter and lower levels in the summer. The concentrations generally peaked around December and bottomed out in July. Such a seasonal pattern can be explained by the prominent influence of generally dirtier air masses from the north in combination with the lower mixing height in the winter as compared with the cleaner oceanic air and higher mixing height in the summer. Yuen Long recorded the highest concentrations for these elements among all the monitoring stations, which serves as evidence for the strong influence of polluted air masses of continental origin.

In contrast, seasonality of nickel and vanadium is much weaker among the monitoring sites, but their variations shown in Figure 4.2-2 are similar to one another. Noting that the highest levels for both elements were found at Kwai Chung, and these elements are typical markers for residual fuel oil combustion (European Communities, 2001), emissions from ocean-going vessels at or around the container port are likely to be an important local source for these elements. Geographically, the Kwai Chung AQMS is downwind of the container terminal in the summer, but upwind in the winter. The lack of seasonality may be related to two counteracting factors, i.e., the larger influence from marine vessels but higher boundary layer heights in the summer versus the weaker influence of marine vessels but lower boundary layer heights in the winter. This is somewhat similar to the lack of seasonality for EC mentioned earlier in section 4.1.

Cr(VI) shows seasonal variation, but a consistent pattern cannot be identified. Comparable Cr(VI) concentrations are found in Tsuen Wan and Central/Western. Since the Cr(VI) levels are close to the MDL, it is possible that the lack of a seasonal pattern can be attributed to the large measurement uncertainty.

Chromium does not show an obvious seasonal pattern, although the minimum tends to occur in the summer. Presently, there is not a clear explanation of this lack of seasonality.

4.2.3 Mercury

Mercury has received much global concern because it bioaccumulates in the aquatic ecosystem food chain in its most toxic form, methylmercury (Ebinghus et al. 2002). Mercury is emitted into the air predominantly by the combustion of fossil fuels (mostly coal) and the incineration of medical and municipal waste. Two mercury species, elemental mercury and dimethyl mercury, exist in vapor form in the troposphere and are chemically stable, which permits long-range transportation (Liu et al., 2002; Steding and Flegal, 2002). The residence time of elemental mercury is in the order of years (0.5-2 years) in atmosphere. As a result, mercury can

be dispersed over regions and entire countries. Both elemental mercury and dimethyl mercury are formed from other mercury species (mercury ion, methyl mercury ion) in sediment through bacterial actions. Elemental mercury can readily be alkylated to methyl-mercury and alkyl-mercury compounds, which exert toxicity to marine lives. Recent research also suggests there is evidence for the oxidation of elemental mercury to ionic mercury species by atmospheric oxidants (e.g., ozone) (Steding and Flegal, 2002).

Currently only particle-bound mercury levels are determined in Hong Kong. It is apparent that such a methodology is not suitable for capturing the various forms of mercury in the ambient air. As a result, the below MDL levels detected in RSP samples do not permit an assessment of the true concentrations of mercury in air. An ad-hoc study that uses proper sampling and analysis techniques is necessary before the health risk associated with mercury and the need for regular monitoring can be assessed.

4.2.4 Summary and Recommendations

Hexavalent chromium concentration levels are similar at Tsuen Wan and Central/Western and remain relatively unchanged. Their annual means range from 0.20 to 0.28 ng/m³, close to the MDL of 0.2 ng/m³. For this reason, measurement uncertainty of Cr(VI) is expected to be high. The level of Cr(VI) in Hong Kong is approximately two times that in California. No seasonality could be discerned, possibly reflecting the large measurement uncertainty and/or short lifetime in air.

Levels of all other toxic elemental species were determined from their total amounts in RSP samples at ten monitoring stations in Hong Kong. The two carcinogenic substances, arsenic and nickel, have concentrations at least six times lower than their California chronic exposure limits. The maximum 3-month averaged lead levels in Hong Kong are seven times lower than the current Hong Kong air quality objective for lead, and its maximum annual level is two times lower than the annual UK standard. Beryllium and mercury have their majority measurements below their respective MDLs, 0.10 ng/m³ and 0.49 ng/m³, indicating that their levels in RSP were low across all the monitoring stations and over the time of more than four years.

Six elemental species, aluminum, arsenic, cadmium, lead, manganese, and zinc, have a clear seasonality with higher levels in winter and lower levels in summer at all ten sites. In addition, Yuen Long sees the highest concentrations among all sites for these elements. Both observations point to more polluted air masses of continental origin transported to Hong Kong impacting the ambient concentration levels for these elements, and may be the controlling factor for their seasonal variabilities.

Using the roadside station at Mongkok to gauge the importance of local vehicular emissions, we found that arsenic, cadmium, lead, manganese, and zinc displayed similar levels at Mongkok to those at other stations. This indicates that local vehicular traffic is not an important source for the above elements. Aluminum and chromium have higher concentrations at the Mongkok roadside station but these two elements are found more in the coarse mode. As vehicular emissions are typically associated with the fine mode, these may be related to other coarse mode emissions (e.g. crustal material), their higher levels at the roadside is probably more related to re-suspension by passing vehicles rather than direct vehicular emissions.

Nickel and vanadium showed elevated levels at Kwai Chung in comparison with other stations, pointing to a source local to Kwai Chung. This source is most likely the cargo terminals in the vicinity, which had large numbers of ocean-going vessels that other locations do not have. Combustion of heavy oil, which is more prominent in these transportation trades than others, is

known to be a major source of vanadium and nickel. While the level of these elements are low in comparison to established guidelines (Table 4.2-2), weak rising trends are identified for both of them. Hence, further study is recommended for the identification of emission characteristics of marine vessels, and also for monitoring of the trends of their emissions and investigation of their overall impact on the nearby environment.

4.2.5 Tables and Figures for Section 4.2

A Tables

- Table 4.2-1 Important Toxic Elements
- Table 4.2-2 Air Quality Guidelines for Toxic Elemental Species
- Table 4.2-3 Method Detection Limits of Hexavalent Chromium and Elemental Species in RSP
- Table 4.2-4 Annual Average Concentrations of Toxic Elemental Species

B Figures

- Figure 4.2-1 Comparisons of Elemental Concentrations in RSP Determined by ICP and in FSP Determined by XRF.
- Figure 4.2-2 Three-month Running Averages of Selected Toxic Elemental Species.

4.3 Dioxins

Polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are collectively called “dioxins”. PCDDs and PCDFs are two of the 12 POPs targeted by the Stockholm Convention. Dioxins are chemically stable semi-volatile solids. There are 210 known congeners, of which 75 are PCDDs and 135 are PCDFs. Congeners with chlorine atoms in the 2,3,7, and 8 positions are of particular environmental concern due to their toxicity. There are 17 such congeners which the dioxin monitoring programs in Hong Kong monitor (Table 4.3-1). The exclusion of the other congener groupings (mono-, di-, and tri-chlorinated dibenzodioxins and dibenzofurans) does not mean that they are non-toxic. Their toxicity, as known at this time, is much less than the toxicity of the 17 monitored congeners.

The vapor pressures of dioxins range from 8.1×10^{-7} to 8.3×10^{-13} mmHg, putting them in the category of semivolatile organic compounds. The congeners are very different in toxicity due to their differences in chemical reactivity and their physical properties. Among all the congeners, 2,3,7,8-tetrachlorodibenzo-p-dioxin (2378-TCDD) is classified as the most toxic (UNEP, 1999). A series of toxic equivalency factors (TEFs) have been developed for individual congeners to allow quantitative conversion into a single Toxic Equivalent (TEQ) value. Today's most commonly applied TEFs were established by a NATO/CCMS Working Group on Dioxins and Related Compounds as International Toxicity Equivalency Factors (I-TEF) (NATO/CCMA, 1988a; Kutz et al., 1990). These factors were established relative to the most toxic congener, 2378-TCDD. The I-TEF scheme is also adopted by the HKEPD for reporting dioxin toxicity. By virtue of these I-TEFs, the measured ambient concentration (pg/m^3) of a congener can be converted into a toxic equivalent concentration (e.g., $\text{pg I-TEQ}/\text{m}^3$) so as to reflect its contribution to the overall toxicity of dioxins. The summation of the TEQ values calculated for the 17 congeners, expressed as “ $\text{pg I-TEQ}/\text{m}^3$ ”, gives a measure of the toxicity contributed by all dioxins in ambient air.

4.3.1 Air Quality Guidelines

Table 4.3-2 summarizes a number of air quality guidelines and source emission standards for dioxins by various government agencies around the world and the WHO. The guidelines applicable to the ambient environment vary from 0.02 to 40 $\text{pg I-TEQ}/\text{m}^3$. Japan⁵ has established that the annual average of dioxins, including PCDDs, PCDFs, and coplanar PCBs, shall not exceed 0.6 $\text{pg TEQ}/\text{m}^3$. The WHO does not specify the concentration guidelines for dioxins in air, but it has set a guideline of tolerable daily intake (TDI) to be 1 to 4 pg/kg per body weight per day, 7 to 28 pg/kg per week, and 30 to 120 pg/kg per month. The European Commission also recommends a guideline of TDI to be 2 pg/kg per body weight per day, 14 pg/kg per week, and 60 pg/kg per month. The most stringent guideline is set by the State Government of Vermont in the US to be 0.02 pg/m^3 as 2378-TCDD (the same as $\text{pg I-TEQ}/\text{m}^3$). Hong Kong currently has no ambient air standard for dioxins.

Dioxin emission standards are proposed by a few governmental agencies. The European Commission has set a guideline for dioxin emissions from municipal waste incinerators to be 0.1 $\text{ng I-TEQ}/\text{m}^3$ (EC, 1994). The dioxin emission standard in Japan is variable, with lower emission standard applied to facilities of higher emission quantities. The Japanese dioxin emission standard ranges from 0.5-5 $\text{ng I-TEQ}/\text{m}^3$. Municipal and clinical waste incineration

⁵ Japan use the WHO version of TEF scheme (Van den Berg et al., 1998), which also define TEFs for 12 coplanar congeners of PCBs within the overall TEQ scheme. Appendix 4 lists the I-TEF for dioxins and WHO-TEF values for dioxins and the 12 dioxin-like PCB congeners. Toxic equivalent concentrations derived using the WHO-TEFs are often expressed as $\text{pg TEQ}/\text{m}^3$.

facilities in Hong Kong currently operate to a PCDD/F emission limit of 0.1 ng I-TEQ/m³ while the PCDD/F emission limit for crematoria is set at 1 ng I-TEQ/m³ (HKEPD, 2002).

4.3.2 Observations and Findings

A Method Detection Limits

The method detection limits (MDLs) for the 17 individual congeners are listed in Table 4.3-1. Two analytical laboratories, ENSR and the GL, have analyzed the dioxin samples (see section 3.3.2.A). In general, the MDLs by ENSR are 2-8 times lower than those by GL. The only exception is 2378-TCDD, for which the two laboratories have similar MDL values. The MDLs reported by both laboratories are lower than the target MDLs specified by the CARB for the California Ambient Dioxin Air Monitoring Program (CARB, 2002b).

An examination of the dioxin concentrations reveals that a large proportion of dioxin congener measurements are less than their respective MDLs. Table 4.3-3 tabulates the number of below-MDL measurements for individual congeners at Central/Western and Tsuen Wan. Take the dioxin measurements at Central/Western in 2000 as an example: 17 dioxin/furan congeners were measured for each of the 25 samples taken, giving 325 individual congener concentrations. Of these 325 congener concentrations, 72% were reported to be below the MDLs. Furthermore, the most toxic congener 2378-TCDD was below its MDL in all the 25 samples.

The large number of below-MDL measurements indicates low levels of dioxins in the air over Hong Kong. However, having too many below-MDL samples leads to a large uncertainty in the annual average concentrations. This is because the annual average values are sensitive to the ways in which below-MDL data are treated and how analytical laboratories report the MDLs.

There are different ways of reporting “below-MDL” data. The below MDL data can be substituted a value of: (a) ½ MDL; (b) MDL; and (c) zero. The most common practice is to substitute below-MDL data with ½ MDL values, and this is also the current practice adopted by the HKEPD. Because of the large portion of below-MDL measurements, the method of reporting “below-MDL” data is expected to have a large influence on the descriptive statistics, such as annual total dioxin concentrations. Table 4.3-4 uses as examples the 2000 and 2001 dioxin measurements at Central/Western to demonstrate the influences of using different reporting methods on the annual mean values of the total dioxin equivalent concentrations in pg I-TEQ/m³. The respective equivalent concentrations are calculated for each sample assuming the three schemes of treating below-MDLs data. When all the below-MDL data are assigned a zero concentration, the resulting annual dioxin equivalent concentrations in 2000 and 2001 calendar years are 51% and 64%, respectively, lower than those calculated with the ½ MDL approach. On the other hand, when all the below-MDL data are substituted with the full MDL values, the resulting annual equivalent concentrations in 2000 and 2001 are 51% and 64%, respectively, higher than those obtained with the ½ MDL approach.

When most of the samples are below detection limits, the reported annual average concentrations are strongly dependent on the reported MDLs. The MDLs are not fixed; they vary from one analytical laboratory to another, as demonstrated by the values reported by ENSR and the GL (Table 4.3-1). They also vary within the same laboratory; the MDL variations could be related to change in instrument performance and in skill levels of analysts between laboratories and over time within one laboratory. Take one scenario as an example: If all the congeners are below detection limits, the calculated dioxin equivalent concentrations would be 0.036 and 0.015 pg I-TEQ/m³ using the GL and ENSR MDLs, respectively.

The above calculations show that the MDLs achieved are adequate if the monitoring objective is only to ensure that the dioxin levels are within a dioxin environmental quality standard similar to those set in Japan (0.6 pg I-TEQ/m^3). However, the MDLs are not adequate for other monitoring objectives such as the tracking of long-term trends, providing data for atmospheric deposition models, and investigation of atmospheric interactions and the impact of long-range transportation.

The problem of having too many samples below the MDLs can be avoided by either increasing the sampling duration and / or combining several 24-h samples before analysis. Compared with compositing multiple samples of 24-h durations, prolonging sampling has the advantage of reducing the time and resources needed in multiple sampling operations. The US NDAMN addressed the MDL issue by sampling for a longer duration and combining samples before analysis. Specifically, each filter sample is collected continuously for six days and four 6-day filter samples are combined to generate one monthly composite sample, corresponding to a total sampled air volume of 8300 m^3 .

However, prolonging sampling duration beyond 24-h is not a viable option for Hong Kong. The aerosol loadings in Hong Kong, especially during winter months, are much higher than the levels encountered in rural areas in the US. As a result, filter clogging could result from sampling beyond 24-h at a flow rate of 230 l/min . Hence, one measure to reduce the number of below-MDL measurements in Hong Kong is to combine multiple samples before analysis. Although this would reduce the temporal resolution of the data, the improved data quality will better serve objectives of providing more accurate data for health risk and trend assessments.

The generally low-levels of dioxin congeners highlight the need for improving MDLs if we are to achieve a better understanding of the sources and dynamics related to the dioxin issue. Take the dioxin measurements in 12 air samples at Tseung Kwan O as an example. Of 204 congener measurements, 31% are below MDLs reported by ENSR. The percentage of below-MDL measurements would increase to 62% using MDLs by GL. The resulting annual average would artificially increase from $0.051 \text{ pg I-TEQ/m}^3$ as reported by ENSR (HKEPD 2002) to $0.063 \text{ pg I-TEQ/m}^3$, if the MDLs by GL are adopted.

B Dioxin Levels in Hong Kong Ambient Air

Table 4.3-5 tabulates the annual average concentrations of dioxins at the four monitoring stations for periods where measurements are available. It is noted that both Tsuen Wan and Central/Western recorded an unusually high dioxin level on 2 February 1999. The dioxin level on this day exceeded the mean value of the remaining measurements in the 1998/99 annual cycle by approximately 6 and 16 times at Central/Western and Tsuen Wan, respectively. The reason for these unusually high dioxin levels is unclear. Moreover, this was the only event in the four and a half year measurement history that recorded such high levels of dioxins. The singularity led us to believe that this event was not representative of the average conditions and therefore should be excluded from calculation of annual statistics. When this date is excluded, the annual average concentrations range from 0.043 to $0.100 \text{ pg I-TEQ/m}^3$. The territory-wide annual average for the 2000/01 is $0.052 \text{ pg I-TEQ/m}^3$. This level is lower than the Japan's limit, 0.6 pg TEQ/m^3 , and the California chronic inhalation exposure limit, 40 pg I-TEQ/m^3 , (Table 4.3-2) by a factor of approximately 10 and 700, respectively.

Figure 4.3-1 compares the dioxin levels in Hong Kong with those measured in a number of urban locations around the world. The data is also summarized in Table 4.3-6. The urban ambient dioxin levels in Hong Kong generally fall within the lower end of the range of dioxin concentrations measured at other urban locations. Table 4.3-7 summarizes dioxin levels in a

number of rural or remote locations around the world, with the average dioxin levels at these background sites in the range of 4-25 fg I-TEQ/m³. The recent survey of dioxin levels in rural areas in nine states in the US reports an average dioxin level of 12 fg I-TEQ/m³ (Cleverly et al. 2000). The Hong Kong territory-wide dioxin average level in 2000/01 is therefore higher by about three times when compared with rural sites in the US.

Figure 4.3-2 hinted that there may be a weak decreasing trend in dioxin levels at both Central/Western and Tsuen Wan from 1998 to 2001, but noting the large uncertainty relating to the MDLs as discussed above, this result has to be taken as preliminary.

It is interesting to note that Hong Kong was found to be among areas of high dioxin deposition by Wagrowski and Hites (2000). In their study, 70 pairs of soil and tree bark samples were collected in locations all around the world between 1996 and 1998. Three pairs of soil and tree bark samples from Hong Kong were collected in Port Shelter, New Territories in 1996. The authors noted that the three soil samples from Hong Kong were among six samples in a total of seventy samples that show homologue profiles dominated by octa chlorinated dibenzo-p-dioxin (OCDD). The OCDD dominance was also present in the soil samples collected from four sites in Hong Kong in the SWG study (HKEPD, 2002). Two possible causes were put forward by Wagrowski and Hites (2000) to explain the OCDD dominance: there might be natural formation of OCDD, or the additional OCDD might come from pentachlorophenol, a once heavily used wood preservative that contained high levels of OCDD as an impurity.

C Seasonality and Spatial Variation

Figure 4.3-2 plots the time series of dioxin measurements at all four sites in Hong Kong in logarithmic scale for better visualization. The seasonal pattern can be clearly seen, with concentrations generally higher in the winter months and lower in the summer months.

OCDD is generally the most abundant congener, accounting for about 22% to 45% of the total congener mass concentrations in pg/m³. This individual dioxin compound also shows a clear seasonal pattern. Figure 4.3-3 plots the time series of OCDD at Central/Western, Tsuen Wan, Mongkok, and Hok Tsui, shown in both linear and logarithmic scales. The logarithmic scale is used to better visualize the variation pattern for the lower concentration measurements.

The seasonal pattern of high in winter months and low in summer months may be attributed to several factors: (1) the generally lower mixing heights in wintertime, (2) more frequent removal by wet deposition in summertime, and (3) the influence of dirtier air masses of continental origin in the winter versus cleaner air masses of marine origin in the summer.

Spatially, the 2000/01 annual data show that the dioxin levels at Tsuen Wan (0.058 pg I-TEQ/m³) > Mongkok (0.051 pg I-TEQ/m³) ~ Central / Western (0.050 pg I-TEQ/m³) > Hok Tsui (0.043 pg I-TEQ/m³), but the difference is quite small. From these measurements, a north to south gradient can be noted but the spatial variation appears not to be significant (considering the large uncertainty in MDLs). However, recent ad hoc studies such as the SWG study (HKEPD 2002) and an additional study by Sin et al. (2002) showed that when samples were taken further to the north or northwest (at Yuen Long, Nim Wan in Tuen Mun and Sha Ling in Sheung Shui), the concentrations were much higher and a clear northwest to southeast gradient can be seen. This is noted here first for completeness and its implication in the regional transport issue will be further elaborated in the next section.

Comparing dioxin levels at the roadside site in Mongkok with those found at the two urban sites within the same period (Figure 4.3-2 and Figure 4.3-3), we do not find elevated dioxin concentrations at Mongkok. This indicates that vehicular emission is not a significant

contributor to atmospheric dioxin loadings in Hong Kong. This observation is also consistent with the monitoring results in Japan (Japan MOE, 2001b), where in fiscal year 2000, the Ministry of the Environment in Japan surveyed the dioxin concentrations at 26 roadside sites (98 samples) and 705 general sites (2816 samples). The mean dioxin concentrations were 0.14 pg I-TEQ/m³ for general sites and 0.17 pg I-TEQ/m³ for roadside sites.

Hok Tsui is least influenced by local emission sources. A comparable level of total dioxins in pg I-TEQ/m³ was recorded at this site when compared with the other three sites for the period of Nov. 2000-Oct. 2001. However, the OCDD levels at Hok Tsui were much lower than the other three sites for the period from April to the early half of August in 2000. As a matter of fact, levels at Hok Tsui were below the MDLs whereas those at Mongkok were 6-10 times ½ MDL value during this period. Outside this period, the OCDD levels at Hok Tsui were similar to the other three sites. Presently we do not have a good explanation for this.

D Examination of Possible Sources of Dioxins against Monitoring Results

Table 4.3-8 summarizes the major local dioxin emission estimates reported in a recent HKEPD study (HKEPD, 2000b).

Vehicular Emission

As seen in Table 4.3-8, vehicles (cars, goods vehicles, buses, and motorcycles) are very minor sources of dioxins. This is verified by the monitoring data, i.e., the roadside site at Mongkok has lower dioxin levels than Tsuen Wan. Our observation is consistent with similar monitoring results in Japan (Japan MOE, 2001b), where in a 2000 study the Ministry of the Environment surveyed the dioxin concentrations at 26 roadside sites (98 samples) and 705 general sites (2816 samples), and found that dioxin levels were not particularly higher at the roadside. A recent study by Sin et al. (2002) noted consistently high dioxin concentrations at Yuen Long during the winter in 2000. The authors attributed the elevated dioxin concentrations to vehicular emissions and other combustion sources nearby. This explanation is not supported by monitoring data at Mongkok.

Municipal and Clinical Waste Combustion

Since April 1997, the old municipal waste and clinical waste incinerators have ceased operation. All the municipal and clinical waste requiring incineration is treated by the Chemical Waste Treatment Center (CWTC), with the Tsuen Wan monitoring site situated 3.5 km north of the CWTC. In summer, south / southwesterly monsoon winds bring in air from the South China Sea, and thus Tsuen Wan is a downwind receptor of CWTC emissions. If the CWTC had a significant impact on the concentration of dioxins at Tsuen Wan, the dioxin levels at Tsuen Wan would be elevated in the summer. However, the reverse is observed, i.e., the lowest concentrations were seen in the summer. Recent study of local dioxin emission sources estimates that the CWTC could contribute up to 0.1 to 0.4% of the existing ambient dioxin concentrations in Hong Kong (HKEPD, 2000b). These arguments are also in line with Rappe (2000) in his review of the same HKEPD report: the measurements suggest that the CWTC is unlikely to be an important contributor to dioxins measured at Tsuen Wan.

Landfill Emissions

Landfills are also possible dioxin emission sources, since ash from incinerators ends up in landfills. Dioxins, formed as one of the post-combustion products, are present in ash residues. In addition, spontaneous landfill fires also lead to emission of dioxins (Rappe, 2000). Dioxin measurements made at the three landfill / waste composting sites are compared with those at Tsuen Wan and Central/Western for the overlapping period, Apr. to Dec. 2001 (Figure 4.3-5). The three landfill / waste composting sites have average dioxin concentrations higher than the

two urban sites, particularly for Sha Ling and Nim Wan. This could be explained as either the landfill / waste composting sites providing local dioxin emission sources or a north to south spatial gradient resulting from regional or long-range transport. Analysis of the two possible hypotheses follows.

Figure 4.3-6 plots the 17 congener concentrations at Sha Ling versus Nim Wan for each of the twelve pairs of samples measured in the SWG study. Measurements for the same day are plotted with the same symbol, and measurements taken at different days are plotted with different symbols. With the exception of one or two congeners, the figure shows that measurements taken on the same day appear to follow a straight line, but there are different straight lines for different days. These findings suggest that the congener profiles are the same on the same day for the two sites. The different slope of the lines for different days shows that the total and relative dioxin loading received at the two sites are different for each measurement day.

A similar linear relationship can be seen between congener patterns plotted for Tseung Kwan O and Nim Wan in Figure 4.3-7. These results suggest that the congener profiles are the same for the same day for the three sites. Sha Lin is a livestock waste composting plant while Nim Wan and Tseung Kwan O are landfill sites. It is unlikely that the three facilities would emit dioxins of similar congener profiles on every sampling date. Hence, we conclude that the higher dioxin levels measured at the three sites are likely not related to emissions locally from landfill / composting sites, but are due to transport from single or multiple emission sources not related to these sites. The different slopes of the lines are likely related to different transport patterns or dynamics.

A comparison of the congener profiles at the landfill sites and the urban sites offer additional information on the possible contribution of landfill emissions to the dioxin levels in the air. There are three sampling days with concurrent sampling at the landfill sites and the urban monitoring sites, namely 8 August 2001, 7 September 2001, and 24 December 2001. Only the OCDD level was above the MDL in the 8 August 2001 samples at both Tsuen Wan and Central/Western. In the 7 September 2001 samples, Tsuen Wan again had only the OCDD level above the MDL and Central/Western had three congeners above their respective MDLs. The dioxin levels on 24 December 2001 were higher, with nine and four congeners detected above MDLs at Central/Western and Tsuen Wan, respectively. Six of the nine above-MDL congeners at Central/Western exhibit similar relative composition to the 24 December 2001 sample taken at Nim Wan. This again suggests that Central/Western and Nim Wan were affected by common dioxin sources.

In summary, the evidence does not support that local landfill / waste composting sites are significant dioxin sources. The hypothesis of transport is examined below.

Regional and Long Distance Transport

Dioxins have long lifetimes in air. The half-life times for TCDD and TCDF are at least two and five days respectively, and the half-life times of OCDD and OCDF are 60 and 400 days, respectively (Lohmann and Jones, 1999). The half-lifetime increases with the degree of chlorination in the congeners. Dioxins are degraded by sunlight in solution under laboratory conditions; however, the extent to which dioxins are degraded in the atmosphere is unknown. As a result of their long lifetimes in the environment and their airborne nature, regional and even long distance transport of these materials in the atmosphere is possible and has been documented (Tysklind et al., 1993; Lohmann et al., 1999).

There is some evidence suggesting that transport may be a dominant factor influencing the

ambient dioxin levels in Hong Kong. Firstly, the clear seasonality with high loadings in winter and low loadings in summer is consistent with the regional transport assumption. Hong Kong receives more air masses of continental origin in the winter while it is influenced more by air masses of marine origin in the summer. Secondly, the weak north to south spatial gradient in the four sampling sites from Tsuen Wan, Mongkok, Central/Western to Hok Tsui suggests an influence from continental air masses. Thirdly, the regular TAP monitoring program recorded almost identical congener compositions at Tsuen Wan and Central/Western on days with high dioxin levels. On these days, most congeners were above their MDLs, which permits a meaningful statistical comparison. Figure 4.3-8 shows five of these examples.

Lastly, a study by Sin et al. (2002) in which dioxin measurements were made simultaneously at five locations (Yuen Long, Tsuen Wan, Central/Western, Tap Mun and Central) revealed a much clearer northwest to southeast spatial gradient in dioxin concentration. In their study, samples were taken during two events in January 2000 for the five sites, and it was found that Yuen Long recorded the highest dioxin levels followed by Tsuen Wan, while Central/Western, Tap Mun, and Central had consistently lower dioxin concentrations. Moreover, dioxin levels in Yuen Long were 1.6 and 2.6 times those at Tsuen Wan in the two sampling events.

The congener profiles excluding OCDD were found to be remarkably uniform at different locations despite a significant spatial gradient. Figure 4.3-9 shows the correlation of congeners between Yuen Long and the other four sites during the two simultaneous sampling events. Excluding OCDD, the composition of the other congeners was almost identical at all the five sampling locations, as evidenced by the close to unity correlation coefficients. The most logical explanation for such a uniform congener profile is that the dioxins identified at all the sites came from the same source (an air mass). The spatial gradient with higher levels at Yuen Long is likely related to the dilution of the air mass as it traveled south/southeastward and mixed with the ambient air.

The most abundant congener OCDD, however, was an exception. Its concentration ratios for sites south of Yuen Long to those measured at Yuen Long were generally higher than the ratios for other congeners. The OCDD concentration at Tap Mun even exceeded that at Yuen Long during the 9 January 2000 sampling event. This suggests that the background air had higher OCDD concentrations than the air mass coming through Yuen Long. Finally, we note that the significantly higher concentrations measured at Yuen Long suggest the transport is more likely related to regional movement rather than long range transport. Long range transport is typically associated with stronger winds and the variations at different stations in Hong Kong would be much less. This is also supported by the circular pollution wind map at Tsuen Wan (Figure 4.3-10) which show higher levels of dioxins during weak to moderate north / northwesterly conditions (a circular pollution wind map cannot be plotted for Yuen Long due to the small number of samples).

In summary, the body of evidence collected so far suggests that the dioxin levels in Hong Kong are mostly associated with regional transport by weak to moderate winds from emission sources outside Hong Kong. Contributions from the CWTC, local vehicular sources or from landfill / composting sites are small.

E Comments in Relation with the Eureka Report

In the Eureka report, 2378-TCDF was classified as a Category I TAP and 2378-TCDD as a Category II TAP, on the basis of their toxicity and estimated local emission strengths. Category I consists of TAPs with relatively high emissions and / or high cancer potency and / or high health value weighting. Category II is similar to Category I but additional verification of

emissions is warranted (Eureka, 1996).

Figure 4.3-11 overlays the time series of the two congeners in fg/m^3 on the same graph. Although many measurements show that 2378-TCDF was present at higher concentrations than 2378-TCDD, the two have similar concentration levels in a significant portion of the measurements prior to 2000. 2378-TCDD is the most toxic among all congeners, and is defined to have a toxicity equivalent factor (TEF) of 1. 2378-TCDF is assigned a TEF of 0.1, ten times less than that of 2378-TCDD. As a result, 2378-TCDD is probably as important as, if not more than 2378-TCDF, in terms of their toxicity. As far as chemical analysis is concerned, one single injection allows the detection of all congeners. In addition, the same sources emit a series of congeners. Therefore, from the point of chemical analysis and elucidating dioxin sources, it is more appropriate to treat dioxins as an integrated group instead of treating the individual congeners separately.

It is interesting to note that the 2378-TCDD measurements since 2000 were below the MDL at both Tsuen Wan and Central/Western. This echoes the MDL issue discussed in section 4.3.2A.

4.3.3 Summary and Recommendations

Our examination of the dioxin monitoring effort showed that a large number of measurements were below the analytical method detection limits. This affects the accuracy of the mean dioxin level estimated, and this issue should be addressed when planning future monitoring programs.

The urban dioxin levels in Hong Kong generally fall within the lower end of the range of dioxin concentrations measured at other urban areas around the world. There may be a weak decreasing trend in dioxin levels at both Central/Western and Tsuen Wan from 1998 to 2001, but noting the large uncertainty relating to the MDLs as discussed above, this result has to be taken as preliminary.

A clear seasonal cycle is found in dioxin concentrations, with much higher levels in the winter when Hong Kong is mainly affected by continental air, and lower levels in summer when Hong Kong is under the influence of oceanic air. The circular pollution wind map also shows that higher levels of dioxins are associated with weak to moderate north / northwesterlies. Spatially, Tsuen Wan and Hok Tsui recorded the highest and lowest dioxin levels in 2000/01, respectively, suggesting a north to south dioxin gradient, but the differences amongst the four TAP monitoring stations were quite small. On the other hand, results from ad hoc studies showed more clearly a northwest to southeast dioxin gradient, with two to four times higher levels of dioxins found at Yuen Long, Nim Wan and Sha Ling (as compared to urban areas in Tsuen Wan and Central/Western). Correlation analysis shows that the congener profiles of dioxins (excluding OCDD) are the same for samples taken on the same day at different sites.

These results combine to suggest that the dioxin levels in Hong Kong are mostly associated with regional transport by weak to moderate winds from emission sources outside Hong Kong. Contributions from the CWTC, local vehicular sources or from landfill / composting sites are small.

Noting that dioxin levels are generally higher in the northwestern part of Hong Kong, monitoring resources for dioxins on a site identified to be a receptor location for regional transportation but upwind of local sources (e.g. Yuen Long) is recommended. Since vehicular emission is not an important contributor to dioxins, future dioxin monitoring in a roadside environment is probably not necessary.

4.3.4 Tables and Figures for Section 4.3

A Tables

- Table 4.3-1 List of 17 Monitored Dioxin Congeners and their Method Detection Limits by ENSR and GL.
- Table 4.3-2 Air Quality Guidelines and Source Emission Standards for Dioxins
- Table 4.3-3 Number of Below-MDL measurements for individual congeners recorded at Tsuen Wan and Central/Western.
- Table 4.3-4 Comparison of Dioxin Concentrations at Central/Western Using Different Reporting Methods for Below-MDL Data in 2000 and 2001.
- Table 4.3-5 Annual Average Concentrations of Total Dioxins in Hong Kong.
- Table 4.3-6 Summary of Ambient Dioxin Concentrations in Hong Kong and Other Urban Locations in the World
- Table 4.3-7 Summary of Ambient Dioxin Concentrations in Rural/Remote Areas.
- Table 4.3-8 Estimates of Dioxin Emission Sources in Hong Kong

B Figures

- Figure 4.3-1 Ambient Dioxin Levels Measured in Urban Areas in Hong Kong and Other Countries
- Figure 4.3-2 Time Series of Total Dioxin Concentrations at Tsuen Wan, Central/Western, Mongkok Roadside and Hok Tsui.
- Figure 4.3-3 Time Series of OCDD Concentrations at Central/Western, Tsuen Wan, Mongkok, and Hok Tsui. (a) Linear Scale, (b) Logarithmic Scale
- Figure 4.3-4 Correlation of OCDD Concentrations at Tsuen Wan and Central/Western
- Figure 4.3-5 Comparison of Average Dioxin Concentrations for the Period of Apr. – Dec. 2001 between the Landfill/Waste Composting Sites and the Urban Sites.
- Figure 4.3-6 Correlation of Dioxin Congener Concentrations at Sha Ling and Nim Wan.
- Figure 4.3-7 Correlation of Dioxin Congener Concentrations at Tseung Kwan O and Nim Wan.
- Figure 4.3-8 Correlation of Congener Concentrations between Tsuen Wan and Central/Western on High Dioxins Days.
- Figure 4.3-9 Correlation of Dioxin Congeners between Yuen Long and four other Sites during two Simultaneous Sampling Events.
- Figure 4.3-10 Circular Pollution Wind Map for Dioxins at Tsuen Wan
- Figure 4.3-11 Comparison of 2378-TCDD and 2378-TCDF concentrations at Tsuen Wan and Central/Western.

4.4 Polychlorinated Biphenyls

Polychlorinated biphenyls (PCBs) are organic compounds with two chlorinated benzene rings linked by a C-C bond (see Appendix-1 for the chemical structures). There are 209 PCB isomers, in which one to ten chlorine atoms can be attached to a biphenyl. About 130 isomers are likely to occur in commercial products. The WHO has identified twelve co-planar PCB congeners as “dioxin-like PCBs” on the basis of their similar toxicity to dioxins (UNEP, 1999; Dyke and Stratford, 2002). The WHO has assigned each of them a toxic equivalency factor (WHO-TEF) and these are listed in Table 4.4-1.

4.4.1 Air Quality Guidelines

Japan has set an air quality standard of 0.6 pg-TEQ/m³ for the sum of dioxins (PCDDs and PCDFs) and dioxin-like PCBs (Japan MOE, 1999). In the US, California suggests a chronic inhalation exposure limit for PCBs of 1.2 µg/m³ (OEHHA, 2002), while North Carolina recommends an annual average of 0.083 µg/m³ as the guideline for acceptable ambient levels (NCDENR, 2002). Hong Kong currently has no ambient air standard for PCBs.

4.4.2 Observations and Findings

A Method Detection Limit

The GL reports a method detection limit of 0.001 ng/PUF for total PCBs. This translates to an air concentration of 0.003 pg/m³ for a sampling volume of 330 m³. All of the total PCB levels detected during the studied period are well above the MDL of 0.003 pg/m³ by at least 33,000 times.

B Concentrations of Total PCBs in Hong Kong

Table 4.4-2 tabulates the annual average total PCB concentrations in three annual cycles from November 1998 to October 2001. An unusually high PCB concentration of 4.26 ng/m³ was reported at Hok Tsui on 19 June 2001, while the PCB level in Mongkok was only 0.39 ng/m³ on the same day. It appears that this high value at Hok Tsui likely arose from a singular event specific to Hok Tsui. Because of its singular nature, we have excluded it in the calculation of the annual average concentration for this site and for territory wide average.

The annual average concentrations show a decreasing trend at both Tsuen Wan and Central/Western. Hok Tsui has the highest PCB level among the four monitoring sites, while Tsuen Wan and Central/Western have similarly lowest levels of PCBs. The level at Mongkok is higher than Central/Western and Tsuen Wan, but lower than Hok Tsui. A more detailed discussion of spatial variation is given later in section 4.4.2D.

Efforts have been made to correlate the total PCBs with the sum of dioxin-like PCBs using a set of 12 measurements taken at Tsuen Wan in 2002. No correlation ($r^2=0.02$) is found between the two. Hence, we find it is impossible to estimate the dioxin-like PCB concentrations from existing total PCB measurements.

Table 4.4-3 lists the annual average concentrations of 12 PCB congeners measured in the SWG study for the period from April 2001 to March 2002 at three landfill / waste composting sites (HKEPD 2002). It is noted that the total PCB levels reported in Table 4.4-2 are more than two orders of magnitude larger than the sums of the 12 dioxin-like PCB congeners. This indicates that the dioxin-like PCB congeners account for a very small fraction of the total PCB loading in the air. The dioxin-like congener pattern at all three monitoring sites exhibits a very consistent profile with a strong dominance by the mono-ortho coplanar congener 118.

The simultaneous measurements of dioxins and dioxin-like PCBs in the SWG study allow the comparison of relative magnitudes of PCB and dioxin toxicity. Comparing the total TEQ for dioxins and dioxin-like PCBs for each sample (in total 36 samples, twelve samples at each of the three sites), we found that the dioxin-like PCBs contribute 1-73% of the total TEQ from dioxins and dioxin-like PCB. On the annual average basis, we found that the dioxin-like PCBs account for 10-14% of the total TEQ from dioxin-like PCBs and dioxins. This result is similar to recent measurements in rural and suburban areas in the US (Cleverly et al., 2002). Hence, the toxicity arising from dioxin-like PCBs is quite significant in comparison with dioxin compounds, suggesting that monitoring of dioxin-like PCB congeners is necessary for better assessment of health risk.

The PCB levels in Hong Kong are compared with other urban locations around the world in Table 4.4-4 and Figure 4.4-1. Note that the PCB concentrations reported in the open literature are often for measurement periods in the early 1990s whereas the Hong Kong measurements are more recent. PCB levels have undergone a globally declining trend as a result of banned use and regulated disposal in many countries since the 1970s (Spiro and Stigliani, 2003).

One urban location in Taiwan (Ping Tung) and a site in Bangkok have considerably higher PCB concentrations than others. The average level of PCBs in Ping Tung city in southern Taiwan was reported to be 4.51 ng/m³ for the period of 1993-1994 (Chen et al., 1996). The high PCB levels may be related to the sampling site being in proximity to the heavily industrialized district in Ping Tung City. There were a number of PCB emission sources in the city, including electric appliances, uncontrolled waste disposal, and self-power generation facilities (Chen et al., 1996).

The territory-wide PCB mean concentration in Hong Kong for the period from November 2000 to October 2001 is 0.58 ng/m³. This value is 5-6 times lower than those in Bangkok, Thailand and Ping Tung, Taiwan. It is also lower than those measurements made in the earlier 1990s in Tokyo, London and Manchester in UK, and a few urban areas in Germany. The levels in Hong Kong are similar to those observed in Chicago in 1995.

The PCB levels in rural and remote areas around the world are summarized in Table 4.4-5. The levels in the rural areas are typically in the range of a few tenths of a ng/m³ whereas those in remote areas, such as over Atlantic Ocean and the Arctic region, are less than 0.1 ng/m³. Hence, the current PCB levels in Hong Kong, while similar to levels in other urban cities (Table 4.4-4), are about ten times higher than the world background levels.

C Seasonal Variation

Figure 4.4-2 overlays the time series of the total PCB concentrations at the four sites between July 1997 and December 2001. At both Tsuen Wan and Central/Western, no obvious seasonal variation can be found. Data at Hok Tsui and Mongkok are available for only one year, making it difficult to identify any clear seasonal trends. Nevertheless, we note that the highest PCB levels occur mainly in the summer 2001, suggesting a possible seasonal trend of PCBs at the Hok Tsui site.

Figure 4.4-3 shows the time series of the sum of 12 dioxin-like PCB congeners and the most abundant congener PCB118, together with the temperature profile, at the three landfill / waste composting sites. Higher PCB concentration days are seen to be associated with days of higher temperatures: a pattern of high in the summer months and low in the winter months is fairly discernable at Tseung Kwan O and Sha Ling. This pattern, however, is less clear at Nim Wan.

Studies in a number of countries (e.g., Japan, Sweden, Norway, UK, Lake Michigan and the

Atlantic region in the U.S.) reported a seasonal trend for PCB concentrations in air. Higher PCB levels were generally seen in the summer and lower concentrations in the winter. The temperature dependence of volatilization rates from the soil was thought to contribute to this seasonality. However, this volatilization process could be offset by the opposite dependence of mixing height on temperature; the mixing height is usually higher in the summer and lower in the winter, leading to more dilution of the pollutants in the summer and less dilution in the winter.

At the landfill / waste composting sites, particularly at Tseung Kwan O and Sha Ling (Figure 4.4-3), the temperature effect of the volatilization process appear to dominate over that of the mixing height, leading to a discernable positive correlation of PCB concentrations with temperature. Such seasonality is also reported for two sites near municipal solid waste incinerators in Japan (Sugita et al., 1994). For places away from emission sources, with the volatilization and mixing height factors acting in opposition with each other, it can be more difficult to see a clear seasonal pattern. This may partly explain the lack of seasonality of the total PCB concentrations at Tsuen Wan, Central/Western, and Mongkok. On the other hand and as noted earlier, there is an indication of higher PCB concentrations at Hok Tsui in summer 2001 as compared to the other seasons, indicating that the volatilization process appears to dominate over the dilution effect of the increased mixing height in the summer. This in turn suggests the presence of a local PCB source(s) near the Hok Tsui site.

D Spatial Variation

The total PCB measurements from November 2000 to October 2001 at the four TAP sampling sites are examined for spatial variation patterns. Figure 4.4-4 compares the mean concentrations for this period, showing that Hok Tsui has the highest level of PCBs, with Mongkok the second, and Tsuen Wan and Central/Western with similarly low PCB levels. The PCB concentrations at Tsuen Wan and Central/Western are significantly and positively correlated ($r^2=0.46$, $n=65$). The higher PCB levels at Mongkok in comparison with Tsuen Wan and Central/Western may be related to enhanced dust re-suspension due to heavier vehicular traffic.

The high PCB levels at Hok Tsui were at first unexpected, but it is in agreement with the result in the last subsection suggesting that there may be a local PCB source(s) near the Hok Tsui site. Circular pollution wind maps of PCBs (Figure 4.4-5) show that higher PCB levels at Hok Tsui were associated with southwesterlies, which are more prevalent in the summer. At the southern rim of Hong Kong, there are a lot of local and international telecommunication facilities near the Hok Tsui site at Cape D'Aguilar. It is of interest to check whether (earlier) use of PCB-containing transformers at or near these facilities may contribute to elevated PCB levels observed at Hok Tsui.

4.4.3 Summary and Recommendations

The PCB levels in Hong Kong are a few times higher than the world background level, but they are lower in comparison with other major cities worldwide where measurements for the earlier 1990s are available. The annual mean PCB levels at Tsuen Wan and Central/Western appear to be lowering. Spatially, concentrations at Central/Western, Tsuen Wan, and Mongkok varied rather coherently, with Tsuen Wan and Central/Western having similarly low PCB levels and Mongkok slightly higher. Amongst the four TAP sites, the highest levels of PCBs are found at the rural Hok Tsui site. In addition, higher levels of PCBs at Hok Tsui are found in the summer, suggesting local volatilization may be significant near Hok Tsui. This in turn suggests the presence of a local PCB source near the Hok Tsui site in Cape D'Aguilar. Further ad hoc study to verify this hypothesis is recommended.

The current regular TAP monitoring program only monitors the concentration of total PCBs. However, measurements of individual PCB congeners, and more specifically, the dioxin-like PCB congeners, are necessary for assessment of health risks attributed by PCBs, and they cannot be estimated from total PCB concentrations. Limited measurements from the SWG study suggest that the toxicity equivalent attributable to dioxin-like PCBs could be significant (up to about 20%) in comparison with that attributable to dioxins. For this reason, analyses of dioxin-like PCB congeners are recommended for future PCB monitoring efforts.

4.4.4 Tables and Figures for Section 4.4

A Tables

- Table 4.4-1 Dioxin-like PCB Congeners and their Toxic Equivalency Factor
- Table 4.4-2 Annual Average Concentrations of Total PCBs in Hong Kong
- Table 4.4-3 Annual Average Concentrations of 12 Dioxin-like PCB Congeners at Three Landfill/Waste Composting Sites for the Period of April 2001-March 2002
- Table 4.4-4 Comparison of Ambient PCB Levels in Hong Kong and Other Urban Areas in the World.
- Table 4.4-5 Summary of Ambient PCB Levels in Rural/Remote Areas.

B Figures

- Figure 4.4-1 Comparison of PCB Levels in Hong Kong with Other Urban Areas around the World
- Figure 4.4-2 Time Series of the Total PCB Concentrations at Central/Western, Tsuen Wan, Mongkok, and Hok Tsui.
- Figure 4.4-3 Time Series of the Sum of 12 PCB Congeners and the Most Abundant Congener (PCB118) at the Three Landfill / Waste Composting Sites.
- Figure 4.4-4 Average Total PCB Concentrations for November 2000 to October 2001
- Figure 4.4-5 Circular Pollution Wind Maps for PCBs

4.5 Polycyclic Aromatic Hydrocarbons

Polycyclic aromatic hydrocarbons (PAHs) are defined as organic compounds of two or more fused benzene rings, in linear, angular or cluster conformations (see Appendix-1 for the chemical structures). A related term, Polycyclic Organic Matter (POM), refers to a broad class of compounds that includes the PAH compounds. POM is listed as one of the 33 priority HAPs by the USEPA (USEPA, 2002b). POM compounds are products of pyrolytic processes during incomplete combustion of carbonaceous substances. They can also be formed through carbonization processes, in which small hydrocarbon molecules and free radicals react to form PAHs (Seinfeld and Pandis, 1998). Because of limited emissions data, for assessment purposes POM data has been limited to either a group of 7 or 16 individual PAH species referred to as 7-PAH and 16-PAH, respectively (USEPA, 2002b). The 7-PAH compounds are classified as either probable or possible human carcinogens by the International Agency for Research on Cancer (IARC). The 16-PAH group includes the 7-PAH group. Table 4.5-1 lists the individual PAH compounds in the 16-PAH and the 7-PAH group. Five of 16-PAH compounds were listed as Category II air toxics in the Eureka study. These are anthracene, benzo[a]anthracene, chrysene, benzo[a]pyrene (BaP), and dibenzo[a,h]anthracene (Table 4.5-1).

4.5.1 Air Quality Guidelines

Among all the PAH compounds, BaP is the most studied. It is often selected as a marker for the PAH group due to its stability and relatively constant contribution to the carcinogenic activity of particle-bound PAH (European Communities, 2001b). The WHO suggested a cancer risk value of 87 for every ng/m^3 of BaP (WHO, 2000). This translates to 87 ng/m^3 BaP producing a cancer risk of one in a million. Generally, no ambient air quality standards have been set for PAHs, however there are proposals and guidelines developed for BaP by various countries (Table 4.5-2). The air quality guidelines set by states in the US are in the range of a few tens of ng/m^3 (MPCA, 1999; NC DENR, 2002). The UK proposed an annual average goal of 0.25 ng/m^3 to be achieved by 2010 (DEFRA, 2000). The European Commission is also considering a proposal for an annual averaged BaP guideline of between 0.5 and 1.0 ng/m^3 to be implemented by 2005 (European Communities, 2001b). A few other European countries have more stringent guidelines for BaP. The most stringent value, 0.1 ng/m^3 , is recommended by Sweden, France, and Croatia.

The relative potency of other PAHs compared to BaP has been established and listed in Table 4.5-1. This allows the comparison of the relative health risk of other PAH compounds.

4.5.2 Observations and Findings: BaP

A Benzo[a]pyrene (BaP) Levels in Hong Kong

BaP has the most comprehensive data sets among the 16-PAH compounds. Within the study period, BaP analyses were performed in both the TAP and the regular air quality monitoring program. The BaP analyzed in the TAP monitoring program included both gas and aerosol phases by using a filter followed by a PUF sample strategy. For the four TAP sites measured in 2000/01, a north to south gradient is apparent: the highest mean BaP concentration of 0.54 ng/m^3 is found at Tsuen Wan, with a slightly lower level of 0.47 ng/m^3 at Mongkok, followed by 0.37 ng/m^3 at Central/Western and the lowest level of 0.12 ng/m^3 at Hok Tsui (Table 4.5-3).

B Seasonality and Spatial Variabilities

Circular pollution wind maps of BaP for the four TAP stations are plotted in Figure 4.5-1, showing that elevated BaP levels are associated with moderate northerlies. Strong seasonality can also be noted for BaP with higher levels seen in the winter and lower levels in the summer. These results suggest that elevated BaP levels in the winter are possibly associated with regional transport by the northerlies.

Additional information about BaP can be obtained from data analyzed from RSP samples taken for the regular air quality monitoring program. Instead of capturing both the gas and the aerosol phases of BaP, the data analyzed from the RSP samples included only the aerosol phase BaP associated with small particulates (size < 10 μ m). Although incomplete, BaP levels derived from the RSP samples can also provide very useful information about the space-time distribution of BaP over Hong Kong.

We start by examining the validity of using BaP derived from RSP samples as a surrogate for the total BaP from TSP samples. BaP is a relatively non-volatile member of the PAH family, it should mainly be present in the aerosol phase. BaP measurements made in European countries indicated that the fraction of BaP in the vapor phase ranges from non-detectable to 10% (European Communities, 2001b). Measurements made in Hong Kong also indicate that gas-phase BaP accounts for less than 10% of the total BaP (Sin et al., 2003). Secondly, comparing BaP concentrations available from TSP and RSP samples collected for the regular air quality monitoring program, we found that the two sets of measurements are highly correlated at each site, with r^2 greater than 0.92, slopes close to unity and intercepts close to zero (Appendix-2). This confirms that BaP is mainly associated with smaller particles, and hence the BaP data from the RSP sample set should be able to tell us much about the total BaP variations over Hong Kong.

Table 4.5-3 tabulates the BaP annual average concentrations at eleven sites in Hong Kong for both the TAP and the regular air quality monitoring program. Overall, Yuen Long consistently had the highest BaP concentration for all the sites during the three years when analyses for BaP in RSP samples were carried out. Then it was followed by higher concentrations in Mongkok and Tsuen Wan, Tai Po, ... and the lowest BaP levels were found at Central/Western. The north-south gradient observed in the BaP measurements from the TAP monitoring sites can also be noted in the RSP samples.

Figure 4.5-2 plots the BaP monthly average concentrations at the RSP sampling sites between January 1997 and March 2000. A consistent seasonal trend with higher levels in the winter and lower levels in the summer can be clearly seen at all sites. For instance, the ambient level of BaP at Tsuen Wan was only 0.12 ng/m³ in August 1999 (mid-summer) but the value increased to 0.53 ng/m³ in January 2000 (mid-winter), which was more than a four-fold increase. This clear seasonal cycle, the apparent north-south gradient, and the circular pollution wind map (Figure 4.5-1) combine to suggest that the BaP variations in Hong Kong are controlled mainly by regional transport by north / northwesterlies.

It is interesting to note in Figure 4.5-2 that in these three years of monitoring, Mongkok actually recorded the highest BaP levels as compared to all other sites during the summer. This suggests that local vehicular traffic is also an important source contributing to BaP levels recorded in Hong Kong, although its overall contribution is not as large as that of regional transport during the winter. The importance of vehicular emission of BaP is also in agreement with earlier finding by a few studies on BaP and PAHs in Hong Kong (Zheng and Fang 2000, Lee et al. 2001).

Zheng and Fang (2000) measured 15 PAHs, including BaP, in TSP samples at six locations: Hok Tsui, HKUST, Kwun Tong, Kwai Chung, Central/Western, and Mongkok. Their sampling duration spanned from August 1993 to August 1995, with four to eight samples taken at each site. The BaP concentrations reported varied from 0.01 to 3.2 ng/m³. The highest BaP levels were observed at Mongkok whereas lower concentrations were measured at the rural sites (e.g., Hok Tsui and HKUST). The concentration range and the spatial variation pattern are in general agreement with the monitoring results by the HKEPD.

Lee et al. (2001) made PAH measurements at a site only eight meters away from a heavily trafficked road near the HKPU campus. Their study period was from April 1999 to April 2000, including a two-week intensive sampling program in December 1999 and January 2000; 33 samples were collected. The BaP concentration at this roadside location varied from 0.05 to 1.34 ng/m³, with an average value of 0.49 ng/m³. The HKPU and the Mongkok sites are both in roadside environments with heavy influence from vehicular traffic. The BaP annual average concentrations in Mongkok were 0.24 and 0.20 ng/m³ for the two years 1999 and 2000, which overlapped with the sampling period of Lee et al. (2001). The higher BaP average concentration reported by Lee et al. may attribute to the fact that the sampling schedule was not uniform and more samples were taken in the winter months. As noted earlier, Hong Kong has much higher levels of BaP in the winter.

C Comparison of BaP Levels with International Air Quality Guidelines

The annual average concentration for BaP at any monitoring site in Hong Kong is less than 0.54 ng/m³. Figure 4.5-3 shows a comparison with the three most stringent BaP air quality guidelines, set by EC, UK, and Sweden. It shows that the BaP levels at all sites exceeded the most stringent Sweden guideline of 0.1 ng/m³, but were all lower than the proposed EC guideline of 0.5 ng/m³, with the exception of Tsuen Wan in 2001 which was just above at 0.54 ng/m³. Compared with the 0.25 ng/m³ standard set by the Expert Panel on Air Quality Standards in UK (DEFRA, 2000), the BaP levels at Yuen Long exceeded this threshold for all years since 1997, while the BaP averages recorded for Kwai Chung, Kwun Tong, Tung Chung and Hok Tsui were all lower than this threshold. For the other stations (Tsuen Wan, Mongkok, Sham Shui Po, Tai Po and Central/Western), there was at least one value exceeding the proposed UK guideline of 0.25 ng/m³ since 1997. On the other hand, when compared with less stringent guidelines, the levels recorded in Hong Kong are lower than the proposed or adopted guidelines in Belgium, Germany, Italy, Netherlands, as well as Minnesota and North Carolina in the US (Table 4.5-2).

D Comparison of BaP Concentrations with Other Locations in the World

The ambient levels of BaP at eleven sampling sites in Hong Kong are compared with other urban areas around the world (Table 4.5-4). The levels in Hong Kong are lower than or similar to those at the urban background sites in the UK and Germany. The BaP levels in Hong Kong are about five times higher than those in Los Angeles and San Francisco. A recent study by Bi et al. (2003) took a few measurements of PAHs in an urban location in Guangzhou in April and July 2001. The BaP levels at this site are three to twenty times that of the annual average BaP levels in Hong Kong.

4.5.3 Observations and Findings: Total PAHs

A Method Detection Limits

The regular TAP monitoring program provided measurements for the 16-PAH group and Benzo[e]pyrene. The MDL is specific to individual PAHs, ranging from 0.008 to 0.332 ng/m³ for Benzo[b]fluoranthene and Acenaphthene, respectively (Table 4.5-5). The CARB has

reported MDLs for six PAHs to be a uniform value of 0.05 ng/m³ (CARB, 2002A). The MDLs by the GL are lower than those by the CARB with the exception of Indeno[1,2,3-c,d]pyrene, for which the GL reports slightly higher MDL.

B Levels of Total PAHs in Hong Kong

The most volatile PAH, naphthalene, mainly exists in the gas-phase. It is the most abundant PAH compound in air, with air concentrations typically two to three orders higher than other PAHs. For this reason, this compound is excluded when calculating the total PAH concentrations (Σ PAH). Otherwise, the total PAHs would be dominated by naphthalene and the spatial and temporal variation of other PAHs would be masked by that of naphthalene.

Central/Western and Tsuen Wan have measurements of four complete annual cycles whereas Hok Tsui and Mongkok have measurements of one complete cycle. Table 4.5-6 summarizes the annual mean levels for 16 individual PAHs as well as the total PAH level. Acenaphthene, acenaphthylene, fluorene, phenanthrene, and fluoranthene are the five most abundant PAH compounds, the sum of which add up to 83-95% of the Σ PAH mass. In comparison, BaP only accounts for a small percentage of the total PAH, making up 0.2-0.6% of the Σ PAH mass.

A rising annual trend in Σ PAH concentrations and in two of the most abundant PAHs (acenaphthene and acenaphthylene) is apparent. The Σ PAH levels in 1999/00 and 2000/01 are two to four times those in the previous two cycles. The increase in acenaphthene and acenaphthylene largely account for the rise in Σ PAH. The cause for the rise in acenaphthene and acenaphthylene is unclear, and needs to be studied.

The Σ PAH concentrations at Tsuen Wan were consistently higher than those at Central/Western in each of the four annual cycles. This may be related to the presence of more industrial and vehicular emission sources at Tsuen Wan than at Central/Western, and may also be related to regional transport from the north. For the 2000/01 annual cycle, the spatial coverage also includes Mongkok and Hok Tsui, two locations subject to local vehicular influences at the two extremes. Mongkok is under heavy influence of vehicular emission while Hok Tsui is virtually free of local vehicular traffic. The Σ PAH annual average concentration at the Mongkok roadside was approximately two, four, and eight times that at Tsuen Wan, Central/Western, and Hok Tsui, respectively. This spatial variation pattern shows that vehicular emission is an important source for PAHs. Zheng et al. (1997) measured PAHs in TSP at six locations in Hong Kong in December 1993. They reported a PAH spatial gradient with the order of Mongkok > Central/Western > Kwun Tong > Kwai Chung > HKUST > Hok Tsui, consistent with observations made from the HKEPD TAP monitoring programs.

PAH data in the literature includes a variable number of PAH compounds depending on the measurement objectives. In addition, the values for a given PAH compound may be reported as the amount in the particle phase or the sum of the amounts in the vapor and aerosol phases depending on the type of measurement methods used. As a result, total PAH values cannot always be compared. For this reason, no comparison is made for the total PAHs in Hong Kong with other locations around the world.

C Seasonality

Unlike BaP trends, no clear seasonality is discernable for the Σ PAH from its time series plot. A cursory analysis indicates that lighter and heavier PAHs show different seasonal patterns. Table 4.5-7 summarizes winter and summer seasonal average concentrations of a group of nine lighter PAHs consisting of three or four rings and a group of seven heavier PAHs consisting of five or more rings. The heavier PAH group exhibits a consistent seasonal trend with higher

average concentrations observed in winter than in summer. However, such a consistent seasonal trend is absent in the temporal profiles of the lighter PAH group. When each winter average of the lighter PAH group is compared with the previous summer average at Central/Western and Tsuen Wan, the winter-high-and-summer-low pattern is only present in two annual cycles from the summer of 1999 to the winter of 2000/01. For the 2000/01 annual cycle, this group has lower concentrations in the winter than the summer at all four monitoring sites. The summer-winter difference in this annual cycle is largest at Hok Tsui (4-fold) and smallest at Tsuen Wan (1.3-fold). The long monitoring record at Tsuen Wan and Central/Western allows the inspection of three more seasonal cycles. It is possible that a rising trend in the lighter PAHs in 2001 could have obscured the winter-summer difference for this annual cycle.

The seasonality of heavier PAHs in Hong Kong can be rationalized by northerly transport and lower mixing heights in the winter and enhanced removal through wet deposition of aerosols in the summer. PAH monitoring results in New South Wales in Australia and in European countries also show the seasonality of higher levels in the winter than in the summer (NSW Environmental Protection Agency, 2002, European Communities, 2001). In both locations, higher emissions from wood burned for use in domestic heating in winter are the principal reason for the higher concentrations in the cold season.

4.5.4 Summary and Recommendations

The PAH marker compound BaP was analyzed in both RSP and TSP samples collected at ten general air quality monitoring stations in Hong Kong. Comparison of the measurements indicates that BaP mostly resides in aerosol-phase small particulates, making the BaP in RSP samples statistically similar to the BaP in TSP samples. This implies that simultaneous monitoring of BaP in both TSP and RSP is redundant, and that the monitoring of BaP in TSP samples can be stopped.

The BaP annual average concentrations in Hong Kong range from 0.10 to 0.54 ng/m³. Compared with the 0.5 to 1.0 ng/m³ guideline that is being considered by European Communities, the Hong Kong levels are lower than or at the lower limit of this benchmark, and meet most other guidelines set by countries in Europe. On the other hand, the BaP levels at all monitoring sites exceed the most stringent BaP guideline (0.1 ng/m³) proposed by Sweden.

In the four complete annual cycles from November 1997 to October 2001, no rising or declining trend is seen for BaP, however, a rising annual trend in the total PAH is clearly present. The increase in the two most abundant PAHs, acenaphthene and acenaphthylene, largely account for the annual rising trend in the total PAH amount. The reason for this increase is still unclear.

BaP and other heavier PAHs of 5-rings or larger have a clear seasonal pattern with higher concentrations observed in the winter than in the summer. Such seasonality is anticipated as a result of lower mixing heights in the winter and higher wet removal rates in the summer. However, such seasonality could not be unambiguously identified for the lighter PAHs with 3- or 4-rings.

Spatially, Yuen Long consistently has the highest BaP concentrations among the ten monitoring stations in three annual cycles with available measurements. Pollution roses show that higher BaP levels are associated with northerly wind, suggesting regional transport of BaP from the north. During the summer, Mongkok has the highest BaP concentrations indicating the influence of local traffic emissions.

4.5.5 Tables and Figures for Section 4.5

A Tables

- Table 4.5-1 Priority PAH Compounds
- Table 4.5-2 Air Quality Standards for PAHs and BaP
- Table 4.5-3 Annual BaP Concentrations
- Table 4.5-4 Comparison of BaP Levels in Hong Kong with Other Urban Areas around the World
- Table 4.5-5 Method Detection Limits of PAH Compounds.
- Table 4.5-6 Annual Average Concentrations of 15 PAHs and Total PAHs.
- Table 4.5-7 Winter and Summer Average Concentrations for PAHs of 3- and 4-Rings and PAHs of 5-Rings or Larger.

B Figures

- Figure 4.5-1 Circular Pollution Wind Maps for BaP
- Figure 4.5-2 BaP Monthly Average Concentrations in RSP between January 1997 and March 2000 at Ten Sampling Sites
- Figure 4.5-3 Comparison of BaP Annual Average Concentrations in Hong Kong with the Three Most Stringent BaP Air Quality Guidelines.

4.6 Carbonyls

Carbonyls are a class of organic compounds containing a ketone or an aldehyde group (see Appendix-1 for the general chemical structure of a carbonyl compound and a few examples of carbonyl). They mainly exist in the gas phase because of their small molecular sizes and consequently higher volatility.

Many members of the carbonyl chemical class are known to be toxic, such as formaldehyde which is a probable human carcinogen (IACR, 1995). It causes eye irritation at levels as low as tens of ppbv and respiratory tract irritations at levels as low as 100 ppbv (NRC 1981). Acetaldehyde, furfural, and crotonaldehyde are known animal carcinogens (WHO 2000). Acrolein causes eye irritation and odor annoyance and exacerbates asthma.

Carbonyls in ambient air have direct emission sources such as combustion processes. They can also be formed through the oxidation of hydrocarbon precursors. The relative importance of primary and secondary sources depends on locations and the time of a day. The photooxidation processes that lead to the formation of carbonyls are also responsible for the accumulation of ozone in the air.

Carbonyls are monitored either for assessing their exposure risk to the public health or for better understanding the photochemical pollution problem. Formaldehyde and acetaldehyde are the two most abundant carbonyl compounds found in both urban and rural environments. Of all the carbonyl compounds, formaldehyde has received the most attention because of its established toxicity and abundance in the air.

4.6.1 Air Quality Guidelines

The WHO and government agencies in many countries have established exposure guidelines for carbonyls known to pose adverse health effects. The USEPA lists three carbonyls, formaldehyde, acetaldehyde, and acrolein, among the 33 priority air toxics (USEPA, 2002b). Canada has also recently proposed adding formaldehyde to its List of Toxic Substances under the Canadian Environmental Protection Act (CanEPA) (Environment Canada, 2002). Table 4.6-1 lists the air quality guidelines for the three toxic carbonyls. California has recommended the most stringent chronic exposure limits for the carbonyls listed above, which are $3 \mu\text{g}/\text{m}^3$ (2.4 ppbv) for formaldehyde, $9 \mu\text{g}/\text{m}^3$ (5.0 ppbv) for acetaldehyde, and $0.06 \mu\text{g}/\text{m}^3$ (0.03 ppbv) for acrolein.

4.6.2 Observations and Findings

A Method Detection Limits

Table 4.6-2 lists the carbonyls that can be analyzed using current methods and their respective MDLs as reported by the GL. The lighter carbonyls up to crotonaldehyde have a uniform MDL of $0.05 \mu\text{g}/\text{cartridge}$ while the heavier ones have a different but also uniform MDL at $0.1 \mu\text{g}/\text{cartridge}$. It appears that these MDL values are estimated rather than actually measured. In comparison with the MDLs for three carbonyls (formaldehyde, acetaldehyde, and methyl ethyl ketone) reported by the CARB, the MDLs from the GL are approximately two times lower. Table 4.6-2 also lists the MDLs reported for the 1997 UATMP (USEPA, 1999). The MDLs from the GL are 1.5 – 10 times higher than those reported for UATMP, with the exception of tolualdehyde.

It is noted that some carbonyls had blocks of recurrent low concentration values, which were close to $\frac{1}{2}$ MDL values but not necessarily equal to the reported $\frac{1}{2}$ MDL values. This is likely

due to slight shifts in MDLs over time and therefore a set of variable $\frac{1}{2}$ MDL values.

Table 4.6-3 compiles the percentages of above-MDL carbonyl measurements at Central/Western. Eight carbonyls were present in the air with concentrations above their MDLs in more than two-thirds of all the samples collected at Central/Western. They are formaldehyde, acetaldehyde, acetone, propionaldehyde, butyaldehyde / isobutyaldehyde (referred as n+i-butanal thereafter), benzaldehyde, methyl ethyl ketone and valeraldehyde. Three carbonyls, acrolein, 2,5-dimethylbenzaldehyde and isovaleraldehyde, were below their respective MDLs in more than 70% of the measurements. The percentages of the above-MDL measurements for crotonaldehyde, valeraldehyde, and hexanaldehyde seem to be dependent on the measurement year, i.e., the majority measurements were above the MDLs in 1998 and 1999, but the percentage of above-MDLs samples fell sharply in 2001 to approximately one-third or lower.

Two aromatic aldehydes, m- and p-tolualdehyde, have shown odd annual trends in the percentage of above-MDL measurements. In 1998, m-tolualdehyde was above its MDL in more samples than p-tolualdehyde (72% versus 50%). In 1999, the percentages (52% vs. 59%) were similar. In 2000 and 2001, the majority samples had p-tolualdehyde above its MDL, but m-tolualdehyde below its MDL. There were two possible explanations. If the measurements are to be trusted, the explanation is that sources for the two tolualdehydes had changed in such a proportion leading to the rise of p-tolualdehyde and the decline of m-tolualdehyde in the air. The known major sources for m- and p-tolualdehyde are photooxidation of m- and p-toluene, respectively. However, the temporal variation in the relative proportion of m- and p-toluene is difficult to verify, as current analytical methods cannot separate the two isomers. An alternative explanation is that the HPLC method did not separate the two carbonyls well, leading to large uncertainties in their quantification. The fact that the ENSR reported a combined concentration for m- & p- tolualdehyde in eight samples supports the latter explanation. Co-elution of all three tolualdehyde isomers was reported in the UATMP program in the US (USEPA, 1999) further substantiating the difficulty in separating the tolualdehyde isomers. If m- and p-tolualdehyde are to remain target analytes in the future, the analytical laboratory needs to clarify whether these two aldehydes can be resolved under its specific analysis conditions.

The subsequent discussion focuses only on the eight carbonyls that were above their MDLs in the majority samples. Acrolein will also be discussed because of its perceived potential impact in public health, although acrolein was below its MDL in the majority of samples.

B Annual Average Concentrations and Trends

The annual average values are calculated using an annual cycle from November to October. The time scale is divided this way to enable better use of the 12-month intensive measurements for TAP species at Mongkok and Hok Tsui in addition to those at Tsuen Wan and Central/Western. Although no carbonyl measurements were made at Mongkok and Hok Tsui, the annual averages are calculated using the same time break-up as other TAP species to ensure consistency in data analysis.

Table 4.6-4 lists the annual average values for the eight carbonyls that were measured to be above their MDLs in the majority of samples. The interannual variability of these carbonyls at Tsuen Wan and Central/Western is similar. The annual average levels of formaldehyde show an increasing trend while the other seven carbonyls do not. Figure 4.6-1 provides a more detailed trend analysis for formaldehyde. The rising trend is clearly seen in both the three-month and the twelve-month running average plots. The increase of formaldehyde in 2000/01 from 1999/00 is especially prominent, mounting to a 47% and 28% increase at Central/Western and Tsuen Wan, respectively. Formaldehyde being the most toxic among all the carbonyl species,

its increasing trend is noteworthy.

The rising trend of formaldehyde may be related to the increased presence of the fuel additive methyl tert-butyl ether (MTBE) as a result of the gradual phasing-out of leaded gasoline and reduction of benzene content in gasoline. Such a link was observed in California (Kirchstetter et al. 1999a,b and Koshland et. al. 1998). Take Los Angeles as an example, the annual average level of formaldehyde rose 150% from 2.30 ppbv in 1992 to 5.87 ppbv in 1996 (CARB, 2002A), subsequent to the introduction of oxygenated fuel additives. In Hong Kong, the phasing-in of unleaded petrol was first introduced on 1 April 1991 and targeted to achieve a penetration rate of 87% and 94% by the end of 1997 and 1998, respectively (HKEPD, 2002). Benzene has been targeted with a reduction from 5% to 1%, and an implementation date of 1 April 2000. MTBE is likely added into the gasoline as an octane enhancer substitute for the leaded compound and benzene which have been removed. The GL analyzed 26 unleaded gasoline samples of different brands sold in Hong Kong from August to December 2000 (Wong et al., 2002). MTBE was found to take up on average 6.8% of gasoline by weight, ranging from non-detectable to 15%. The benzene content was on average 0.8%, ranging from 0.1 to 1.3%. Unfortunately, no MTBE or benzene data is available for gasoline sold before 2000. As a result, the suggestion of increased MTBE in gasoline in recent years has yet to be verified.

C Seasonality

To examine the seasonality of the carbonyls, the three-month running averages for the eight carbonyls are plotted on Figure 4.6-2. Seven carbonyls show a clear seasonality with higher concentrations in the winter and lower concentrations in the summer. They include acetaldehyde, propanal, acetone, benzaldehyde, methyl ethyl ketone, valeraldehyde, and n+i-butanal. Formaldehyde does not have a clear seasonal cycle (Figure 4.6-2). Sin et al. (2001) reviewed a subset of the carbonyl measurements at Central/Western and Tsuen Wan (October 1997 to September 2000) and concluded that the seasonality of both formaldehyde and acetaldehyde were not obvious. With more monitoring data, the seasonality of acetaldehyde was clearer, while the lack of a clear seasonality for formaldehyde is likely attributable to its short lifetime in the ambient air. Formaldehyde is the most reactive and shortest living carbonyl compound. It has an estimated lifetime of 1.2 hr due to its reaction with OH and HO₂ radicals. The overall lifetime is even shorter when considering its photolysis loss. Moreover, frequency / time aliasing by the increasing trend in background levels also make it more difficult to identify the seasonal cycle in formaldehyde.

The carbonyl compounds can have both primary emission sources and in-situ photochemical formation. The photochemical formation pathway is closely linked to ozone formation. If the photochemical pathway were the dominant source for carbonyls, the seasonal variation of the carbonyls would resemble that of ozone. Figure 4.6-3 shows the three-month running average of ozone at Tsuen Wan and Central/Western. Ozone has a semi-annual cycle with peak levels in October and April and relative minima around January and July; the magnitude of the ozone seasonal variation was generally smaller than those of the carbonyls. The peak month of the carbonyls occurred two to three months later than the ozone peak. This indicates that the secondary source / photochemical formation pathway was unlikely to be the dominant contributor for those carbonyls. This conclusion is further supported by the match of peak months between the carbonyls and two known primary air pollutants, benzene and carbon monoxide. The three-month running averages for benzene and carbon monoxide (Figure 4.6-3) clearly show that the peak month for both pollutants was December / January. The months of minimum concentrations were also in agreement with those of the carbonyls. The seasonality characteristic of primary pollutants is typically explained by seasonal variations of mixing heights and the existing synoptic meteorological conditions.

The lack of seasonality in formaldehyde indicates that its dominating sources and sinks are different from those carbonyls with a clear seasonality. It is also generally more difficult to discern the seasonal characteristics of a parameter, such as formaldehyde, which has a clear annual trend (change) that is comparable to its seasonal variations.

Ho et al. (2002) measured concentrations of ten carbonyl compounds at a roadside location near HKPU during April 1999 – April 2000. 41 samples were collected, 31 of which were taken on a regular schedule of one sample every 12 days. Results indicated that the concentrations are greater in the summer (May to August) than in the winter (November to February) for formaldehyde, acetaldehyde, propanal, and hexanal, similar for m-tolualdehyde and benzaldehyde, and less for acetone, valeraldehyde, butyraldehyde, and methyl ethyl ketone. Lee et al. (2001) noted that the lowest monthly average for formaldehyde and acetaldehyde was observed in December and October 1999, respectively. The difference in seasonality reported in these studies and the present review may be partly related to the roadside nature of their monitoring site, which is different from the general urban sites of Tsuen Wan and Central/Western. Furthermore, even for the same season, large sample to sample and site to site difference can be noted in typical carbonyl time-series such as the one shown in Figure 4.6-1 for formaldehyde in Tsuen Wan and Central/Western, suggesting that sampling once every twelve-days for a year is probably not enough to identify a stable seasonal cycle for the carbonyls.

Ho and Lee (2002) studied the correlation between formaldehyde and PAH compounds. They found that formaldehyde had a high correlation coefficient with indeno[123-cd]pyrene and benzo[ghi]perylene during the 2-week intensive sampling period in the winter ($r=0.83$ and 0.86 , respectively, $P<0.01$). On the other hand, a poor correlation between formaldehyde and these two PAHs was found during the summer. This strong correlation in winter suggests that vehicular emission was the dominant source for formaldehyde during this period. The weak correlation in the summer may be a combination of the increasing contribution of secondary sources of formaldehyde, and a more prominent washout effect for PAHs during summertime.

Seasonal variation of carbonyl compounds, particularly formaldehyde and acetaldehyde, has been discussed in the literature, with the seasonality found to vary from one place to another. For example, formaldehyde reached its maximum level in autumn in south California (Grosjean, 1991), summer in Rome (Possanzini et al., 1996) and Eastern Finland (Viskari et al., 2000), and spring and summer in Long Island, New York (Tanner and Meng, 1984). In the same New York study, acetaldehyde levels were lowest in winter, highest in summer, and intermediate in fall and spring (Tanner and Meng, 1984). The location specific seasonal characteristics suggest that many factors could affect the levels of the carbonyl compounds.

D Spatial Variation

In general, the carbonyl levels at Tsuen Wan and Central/Western were similar in magnitude, with most carbonyls present at slightly higher concentrations at Tsuen Wan than Central/Western (Figure 4.6-2). Acetone was an exception. Prior to August 2000, its levels were higher at Central/Western than Tsuen Wan, yet after 8/2000, the levels were very similar at the two sites (Figure 4.6-2). The slightly higher levels observed at Tsuen Wan are not surprising in consideration of the higher traffic density at Tsuen Wan.

The 3-month running average curves show (Figure 4.6-2) that the two sites exhibit similar temporal variation patterns, a rough indicator of the similar air characteristics of the two sites. Linear correlation curves are plotted in Figure 4.6-4 for better visualization. The correlation coefficient r^2 values range from 0.40 to 0.83, with formaldehyde having the weakest correlation

and n+i-butanal the strongest correlation between the two sites. It is noted that the high correlation of n+i-butanal is largely due to two collocated samples taken on 16 December 1998 that were measured at higher concentrations. After exclusion of the two high concentration data points, the correlation coefficient falls to 0.23. Benzaldehyde has a correlation coefficient of 0.74, the highest if n+i-butanal is excluded. The degree of correlation is inversely related to the reactivity of the carbonyl compounds.

The HKPU roadside measurements showed an average formaldehyde concentration of $4.14 \mu\text{g}/\text{m}^3$ and an average acetaldehyde concentration of $2.00 \mu\text{g}/\text{m}^3$ for the period of April 1999 – April 2000. In comparison, Tsuen Wan had average formaldehyde and acetaldehyde concentrations of $4.64 \mu\text{g}/\text{m}^3$ and $2.34 \mu\text{g}/\text{m}^3$, respectively, for the same period. The slightly higher formaldehyde and acetaldehyde level at Tsuen Wan suggests that other non-vehicle related sources could also be important sources for the two carbonyls at this site.

E Relative Carbonyl Composition

Formaldehyde is by far the most abundant carbonyl, accounting for on average ~60% of the total carbonyls by mole at both Tsuen Wan and Central/Western. Its contribution to the total carbonyls ranges from 24% to 86% at Central/Western and 29% to 83% at Tsuen Wan, with the highest contribution in the summer and the lowest contribution in the winter. The strong seasonal dependence can be clearly seen in the time series plot in Figure 4.6-5.

Acetaldehyde is the next most abundant carbonyl, taking up approximately 20% of the total carbonyls by mole. The molar percentage of acetaldehyde ranges from 8% to 40% at Central/Western and from 5% to 36% at Tsuen Wan.

The molar ratio between formaldehyde and acetaldehyde has been reported to be usually larger in rural areas (~7) than urban areas (0.7-3) in the literature. The higher C1:C2 aldehyde ratio in rural areas is attributed to production of formaldehyde from oxidation of biogenic hydrocarbons (Shepson et al., 1991; Possanzini et al., 1996). Central/Western has a C1:C2 aldehyde ratio of 2.6 ± 1.3 and Tsuen Wan has a ratio of 2.3 ± 1.0 , in line with the ratios observed in other urban areas. A closer examination revealed that this ratio was clearly elevated in the summer months (June-August) at Central/Western, and the seasonal elevation pattern less clear, but still discernable at Tsuen Wan (Figure 4.6-6). This seasonal pattern in the C1:C2 aldehyde ratio may be an indicator of the enhanced photochemical formation of formaldehyde in the summer months.

F Comparison of Carbonyl Levels with Other Countries

Formaldehyde and acetaldehyde are the two most abundant carbonyl compounds in ambient air, and the most widely reported in the open literature. Table 4.6-5 compares the ambient concentration levels of the two carbonyls in Hong Kong and a few urban areas in the world.

Formaldehyde and acetaldehyde levels are similar to those found in urban locations in the US (e.g., Los Angeles and Atlanta), but much lower than those in Rome and the major cities in South American countries (e.g., Mexico City and Sao Paulo). The much higher levels of formaldehyde and acetaldehyde in the urban areas of Mexico and Brazil are likely due to higher content of oxygenated compounds (e.g., ethanol) in their fuels. The formaldehyde annual average concentrations in Hong Kong exceeded the California chronic health exposure limit of $3.0 \mu\text{g}/\text{m}^3$ (2.4 ppbv), but the acetaldehyde levels were well below the California chronic health exposure limit of $9.0 \mu\text{g}/\text{m}^3$ (5.0 ppbv).

In the US, because of concern over the contamination of surface and ground water, the use of MTBE as a fuel oxygenate is being phased out in California (Happel et al. 1998) and its use is

declining in other states. As a replacement, increased use of ethanol as a fuel oxygenate is expected, and acetaldehyde levels are expected to increase as a result of such changes in fuel additives (Connecticut Department of Environmental Protection 2003). As Hong Kong imports its fuel from the global market, it is of interest to see whether changes in long-term averaged acetaldehyde and formaldehyde levels can be observed in the next few years.

G Acrolein

Acrolein is on the list of the 33 priority HAPs compiled by the USEPA due to its recognized toxicity and known emission sources. Acrolein is primarily used as an intermediate in the manufacture of acrylic acid. It can also be emitted from combustion sources such as vehicle exhaust, kitchen fumes, and biomass burning. In addition, the breakdown of certain pollutants (e.g., 1,3-butadiene) in outdoor air leads to the formation of acrolein.

The current carbonyl monitoring program is not adequate to determine the ambient concentrations of acrolein. The below MDL samples account for 86% of all samples taken at Central/Western (Table 4.6-3). The high percentage of below MDL samples makes it highly uncertain as to the true ambient concentration of acrolein. The same problem has been reported by the Air Pollution Control Division in Vermont, USA in its monitoring of ambient acrolein (Vermont Air Pollution Control Division, 1998). Acrolein concentrations were below the detection limit in 90% of the samples collected in Vermont's four sampling stations in 1994 and 1995.

It is also recognized, although not very well-documented in the open literature, that the current sampling and analysis methodology is not suitable for determination of acrolein. The problem arises from that fact that DNPH-acrolein hydrazone formed on the sampling cartridges are susceptible to attacks by oxidants in the air, leading to degradation of the hydrazone during the sampling process.

4.6.3 Summary

A clear rising trend is found in the formaldehyde annual average concentrations, but such a trend is not found for other carbonyls. The rise of formaldehyde level in the ambient air is possibly related to the suspected increasing presence of MTBE in gasoline sold in Hong Kong (Kirchstetter et al. 1999a, b, and Koshland et. al. 1998).

For seasonal variations, seven carbonyls show a clear seasonality with higher concentrations in the winter and lower concentrations in the summer. They include acetaldehyde, propanal, acetone, benzaldehyde, methyl ethyl ketone, valeraldehyde, and n+i-butanal. On the other hand, formaldehyde does not have a clear seasonal cycle. The lack of a clear seasonality for formaldehyde is likely attributed to its short lifetime in the ambient air, as formaldehyde is the most reactive and shortest living carbonyl compound. It also suggests that the dominating sources and sinks for formaldehyde may be different from those for carbonyls with a clear seasonality. Moreover, frequency / time aliasing by the increasing trend in the background levels also makes it more difficult to identify the seasonal cycle in formaldehyde.

As no carbonyl measurements were made for Mongkok or Hok Tsui, detailed analysis of the spatial distribution of carbonyls over Hong Kong cannot be provided. We only noted the carbonyl variations at Tsuen Wan and Central/Western were similar in magnitude, with most carbonyls found at slightly higher concentrations at Tsuen Wan. This may be related to the fact that vehicular emission, a major source of carbonyls compounds, is contributing a little more at the Tsuen Wan station.

In comparison with other cities, the levels of the two most abundant carbonyls, formaldehyde

and acetaldehyde in Hong Kong, are similar to those found in a number of urban areas in the US. When compared with the California chronic health exposure limits, formaldehyde levels exceed the exposure limit whereas acetaldehyde is far below its suggested exposure limit. Nevertheless, with increased use of ethanol to replace MTBE as a fuel additive in the US, increased levels of acetaldehyde are expected. As Hong Kong imports its fuel from the global market, it is of interest to see whether changes in long-term averaged acetaldehyde and formaldehyde levels can be observed in the next few years.

Finally, we note that although acrolein is one of the 33 priority TAPs listed by the USEPA, there is still no good method for measuring its ambient concentrations. Problems with acrolein have been reported elsewhere (Vermont Air Pollution Control Division, 1998) and the USEPA recently issue an addendum stating that the TO-11 and TO-11A methods are not useful for the determination of acrolein (USEPA 2002). Due to the lack of ambient measurements, it is not possible to make a health assessment for this compound at present.

Also, while the analytical method itself allows simultaneous determination of 17 carbonyl compounds, nine carbonyl species were routinely below their MDLs due to their low concentrations in the ambient environment. As a result, if separate resources are needed for analyses and reporting of these nine carbonyls, we suggest eliminating them in future routine monitoring efforts; on the other hand, if no additional resources are required (which is possible since the analytical method can allow simultaneous determination of 17 carbonyl compounds), their reporting can certainly be continued to allow for monitoring of any unexpected increases.

4.6.4 Tables and Figures for Section 4.6

A Tables

- Table 4.6-1 Air Quality Guidelines for Formaldehyde, Acetaldehyde, and Acrolein
- Table 4.6-2 Carbonyl Compounds Monitored and Their Method Detection Limits
- Table 4.6-3 Percentages of Above-MDL Carbonyl Measurements at Central/Western
- Table 4.6-4 Annual Average Concentrations of Eight Carbonyls at Tsuen Wan and Central/Western
- Table 4.6-5 Summary of Ambient Air Measurements of Formaldehyde and Acetaldehyde in Hong Kong and a Few Other Cities in the World

B Figures

- Figure 4.6-1 Trend Analysis for Formaldehyde at Central/Western and Tsuen Wan.
- Figure 4.6-2 Three-month Running Average Curves of Eight Carbonyl Compounds.
- Figure 4.6-3 Three-month Running Average of Major Primary (Benzene and Carbon monoxide) and Secondary (Ozone) Air Pollutants.
- Figure 4.6-4 Linear Correlations of Carbonyl Concentrations at Tsuen Wan and Central/Western
- Figure 4.6-5 Molar Percentage of Formaldehyde at Tsuen Wan and Central/Western for the period of July 1998 – December 2001.
- Figure 4.6-6 Ratio of Formaldehyde to Acetaldehyde at Central/Western and Tsuen Wan

4.7 Volatile Organic Compounds

4.7.1 Background

Volatile organic compounds (VOCs) are defined in terms of their volatility, i.e., organic chemicals with vapor pressures greater than 0.01 kPa (Moschandreas and Gordon, 1991). Some VOCs are toxic, while a few VOCs (e.g., benzene) are even carcinogenic. Table 4.7-1 lists the VOCs that are either on the 33 priority HAP list compiled by the USEPA or on the priority list given in the Eureka study. Carbonyls (e.g., formaldehyde, acetaldehyde) are also VOCs by definition. This class of oxygenated VOCs is excluded from ensuing discussions whenever VOCs are referred to, mainly because different sampling and analysis techniques are used for the determination of their concentrations in the ambient environment.

VOCs are monitored either for assessing their exposure risk to the public health or for better understanding the photochemical pollution problem. Two approaches are most often used for their measurement. One is canister-based (e.g., USEPA method TO-14) and the other is solid sorbent-based (e.g., USEPA method TO-1 and TO-13). In the canister-based methods, VOC samples are collected in canisters and portions of the canister air are injected into a GC/MS for analysis. In the solid-sorbent based methods, VOC samples are collected by pulling air through a solid sorbent tube, which is thermally desorbed onto a GC column for separation and subsequent detection by various GC detectors.

Ethylene oxide, methyl alcohol and butanol, three VOCs identified in the Eureka report, were not available in the TAP database as they were found to be below MDL in 2001 study by the HKEPD.

4.7.2 Air Quality Guidelines

Air quality guidelines have been established for a number of VOCs of established toxicity by various government agencies and the WHO (Table 4.7-2). Some VOCs, e.g., 1,3-butadiene and benzene, are carcinogenic. The UK has set air quality goals of 5 ppbv ($16.2 \mu\text{g}/\text{m}^3$) for benzene and 1 ppbv ($2.25 \mu\text{g}/\text{m}^3$) for 1,3-butadiene (DEFRA, 2000). The European Commission set a more stringent annual average of $5 \mu\text{g}/\text{m}^3$ (1.5 ppbv) for benzene to be achieved by 2010 (European Communities, 2000). This EC goal for benzene is based on fuel control measures which came into effect in Europe in 2000 when a 1% benzene limit in petrol was put in place, allowing a 10-year lead time to achieve the goal.

In the US, Minnesota and Vermont typically have the most stringent air quality guidelines for VOCs with cancer-causing potential. Minnesota defines its guidelines in terms of health benchmarks, which are concentrations of the pollutants below which there is likely to be no public health concern. The cancer health benchmarks represent the concentrations that are associated with an upper-bound excess lifetime cancer risk of 1 in 100,000 (Wu and Pratt, 2001). Vermont sets its hazardous ambient air standards (HAAS) for carcinogenic toxics by a similar but stricter definition. It sets the HAASs for carcinogenic compounds at a concentration estimated to correspond to a one in one million (1×10^{-6}) increase in the probability of developing cancer (over and above the background rate) over a lifetime of exposure (Vermont Air Pollution Control Division, 1998). Each standard then represents the estimated level of that compound in ambient air to which one could be exposed to for 24 hours per day, 365 days per year for 70 years and experience only a one in one million increase in the risk of developing cancer from that exposure.

4.7.3 Observations and Findings

A Method Detection Limits

The VOC concentrations in the database are expressed in units of $\mu\text{g}/\text{m}^3$. To facilitate inter-conversion between ppbv and $\mu\text{g}/\text{m}^3$, we list in Table 4.7-3 the conversion factors between the two units for the VOC species that appear in the following discussion.

The MDLs are reported to be a uniform value of 0.1 ppbv for each VOC species. The estimated MDLs for benzene, toluene, ethylbenzene, and xylenes (BTEX) are generally similar to the MDLs reported by the CARB (Table 4.7-3). The MDLs of 1,3-butadiene and chlorinated VOCs, with the exception of dichloromethane, by the GL are 2.5-10 times that of the MDLs by the CARB. This may represent an area for improvement, since levels of many chlorinated compounds in Hong Kong were found to be below their MDL in a large number of samples. Carbon tetrachloride is such an example.

B Ambient Concentration Levels and Annual Trends

BTEX

Table 4.7-4 summarizes the annual average levels of BTEX and 1,3-butadiene from 1997/98 to 2001/02 (discussed in the next subsection), updated with recently acquired 2002 VOC data. Appendix 3 displays more annual statistics (i.e., annual median, 5% and 95% percentile values) in addition to the annual average values for 1998/99 to 2000/01. A weak decreasing trend is noted for benzene, with the 2001/02 concentrations lowered by 15% and 22% at the Central/Western and Tsuen Wan stations respectively, from their 1997/98 mean. Two control measures were introduced to reduce benzene concentrations in air (HKEPD, 2001). A regulation to reduce benzene emission through vapor recovery at petrol filling stations was implemented in April 1999, and the HKEPD reported that oil companies began retrofitting their facilities around 1997. Lack of measurements before 1997 makes it difficult to assess the effectiveness of this control measure. The second control measure was to reduce the benzene content in unleaded gasoline, which started on 1 April 2000. This control measure is consistent with the reduction of benzene observed in the last few years. However, the reduction may also be partly related to reduced emissions in association with the overall economic slowdown during the same period. The latter hypothesis is consistent with the observation that similar decreases are being seen in the levels of other aromatics (toluene and xylenes).

The benzene levels at Central/Western, Tsuen Wan, and Hok Tsui were about one-half of the $4.8 \mu\text{g}/\text{m}^3$ air quality guideline suggested by the EC (Table 4.7-2). The Mongkok roadside location had a level at the same value as the EU benzene guideline. Benzene concentrations at Tsuen Wan and Central/Western are highly correlated, with an r^2 value of 0.74. This again demonstrates that Tsuen Wan and Central/Western share common air pollution characteristics.

Figure 4.7-1 compares the levels of benzene, toluene, and xylenes in Hong Kong with a few selected urban areas around the world. The details of the studies for comparison are described in Table 4.7-5. The benzene levels in Hong Kong are similar to those in Los Angeles (US), Auckland (New Zealand), UK, and Helsinki (Finland), but are lower than those in Karachi (Pakistan), Melbourne, Sydney (Australia), Athens (Greece), Izmir (Turkey), Mexico City (Mexico), and Porto Alegre (Brazil). Toluene levels in Hong Kong are similar to those in Los Angeles and San Francisco whereas xylenes levels are lower than those in Los Angeles and San Francisco. In general, the Australian cities, Athens, and Izmir have higher toluene and xylenes levels than Hong Kong.

The BTEX levels at the Mongkok roadside location are about twice as high as the urban

background sites (i.e., Tsuen Wan and Central/Western) because of the closer proximity of Mongkok to vehicular traffic, which is the major emission source of BTEX. Table 4.7-6 compares the BTEX concentrations in Hong Kong's roadside environments with similar environments in other countries. The roadside BTEX levels in Mongkok are generally lower than other roadside measurements in the literature.

The BTEX levels in roadside environments in three cities (Guangzhou, Macao, and Nanhai) in the Pearl River Delta region were recently reported by Wang et al. (2002). Guangzhou had the highest benzene levels in its roadside environments, followed by Macao and Nanhai. Ethylbenzene and xylenes showed excellent correlations ($r^2 > 0.94$) in all three cities, indicating that the two have common vehicular sources. On the other hand, the correlation of benzene with ethylbenzene was much weaker, especially in Guangzhou and Nanhai. This might suggest that benzene had important sources other than vehicular emissions in Guangzhou and Nanhai.

Chan et al. (2002) measured BTEX concentrations in roadside environments at four locations in Hong Kong, including Mongkok. Their sampling took place during two-hour intervals at noon and late afternoon rush hours in January and February of 1998, representing high emission periods from vehicular sources. The BTEX levels in their study were higher than the annual or even seasonal averages in this study as the samples examined here were daily averages, while theirs were two hour averages during peak traffic conditions.

1,3-Butadiene

With recently updated VOC data, three complete annual cycles of 1,3-butadiene measurements (1999/00, 2000/01 and 2001/02) are available at Central/Western and Tsuen Wan. The annual average levels in 2000/01 and 2001/02 are higher than those identified for 1999/00 at both sites. This may be partly related to the gradual fleet change of taxis from diesel to LPG. By November 2001, it was estimated that over 75% of all taxis had been converted to LPG. Measurements at the four monitoring sites (Central/Western, Tsuen Wan, Mongkok, Hok Tsui) in 2000/01 showed a clear spatial gradient with Mongkok > Tsuen Wan > Central/Western > Hok Tsui, consistent with the perceived spatial variation of traffic density. This reflects that vehicular emissions are a significant source for 1,3-butadiene. The annual average values ranged from $0.07 \mu\text{g}/\text{m}^3$ at Hok Tsui to $1.14 \mu\text{g}/\text{m}^3$ at Mongkok (Table 4.7-4). The average annual level to which the population is exposed clearly depends on proximity to motor traffic. The very striking difference between the roadside environment and the rural site reflects not only differences in traffic density but also the fact that 1,3-butadiene is removed from the atmosphere in a matter of hours by chemical reactions.

The 1,3-butadiene levels at all sites, including the roadside, were below the $2.25 \mu\text{g}/\text{m}^3$ UK guideline value. The levels at Central/Western and Tsuen Wan were similar to those observed in San Francisco and Los Angeles and the nationwide average in UK, but approximately 3 times lower than in Sydney (Table 4.7-7).

We have also studied the variation of other LPG-related VOCs, including butane, iso-butane, 1-butene / iso-butylene, 1-butyne, pentane, 1-pentene and propane. Data for these VOCs is only available since the 1999/00 cycle. We found similar patterns to 1,3-butadiene, in that the 2000/01 cycle has concentrations of these VOCs higher than those measured for the 1999/00 cycle. This increase may be partly related to the increased use of LPG in traffic with the fleet change of taxis from diesel to LPG. However, we caution that just two years of data is not enough to establish any trend; further monitoring is needed to check if these changes are part of longer term trends.

Halogenated VOCs

Table 4.7-8 summarizes the annual average concentrations of the five halogenated VOCs with the most health concerns, namely, carbon tetrachloride, chloroform, methylene chloride, trichloroethene and tetrachloroethene. More statistics (i.e., annual median, 5% and 95% percentile values) are displayed in Appendix 3. No clear annual trend is noted for trichloroethene. The three chlorinated VOCs, carbon tetrachloride, chloroform, and methylene chloride, show an increase in concentrations in 2000/01 and 2001/02 from the previous two years. It is surprising to see such an increase, especially for carbon tetrachloride, considering that this compound has mostly been phased out of production under the Montreal Protocol on ozone-depleting substances. We note that the levels of these three halogenated VOCs show larger variation after December 2000 than prior to this time. For example, a number of very high carbon tetrachloride concentrations up to about $6 \mu\text{g}/\text{m}^3$ (five or six times the average concentration) were reported in December 2000. Chlorinated solvents are often used in chemical laboratories, and their use may be a source of contamination for the elevated levels in the samples. It is not clear whether the changes in these halogenated VOC concentrations are real changes in ambient levels or laboratory contamination related to chlorinated solvents. One way to check is to look at, if available, results for laboratory blanks to check for sign of possible contaminations.

Regulations to control tetrachloroethene emissions from dry cleaning facilities were implemented in November 2001 (HKEPD, 2001). Before the implementation of this regulation, 20% of the approximately 400 dry-cleaning facilities in Hong Kong were of the latest models that meet the required emission standard. It is targeted that all facilities meet the emission standard by 2008. Data up to the end of 2001 did not show any change in the level of tetrachloroethene, however using recently updated data for 2002, we found a substantial decrease of about 30% during 2002 with respect to earlier years (averaged from the three earlier cycles) at Central/Western. Even at Tsuen Wan, a decrease of 13% is noted. These decreases are consistent with the control measures introduced, and suggest that the tetrachloroethene control measures may have helped in the reduction of ambient tetrachloroethene concentrations. The larger decrease in Central/Western is consistent with our finding that emissions from dry cleaning facilities are more prominent near this site (see next section). Nevertheless, we caution that the available data is not long enough to establish any trend, and further monitoring is needed to check if these changes are part of a longer term decreasing trends.

The levels of halogenated VOCs are within the chronic reference exposure levels set by the CARB. However, the levels observed in Hong Kong have either exceeded or are close to the Vermont HAAS, which set the most stringent air quality standards for these compounds.

Comparisons of the halogenated VOC levels are made between Hong Kong and other urban locations in the world in Table 4.7-9. The annual average levels for the most recent year (2000/01) available in Hong Kong are used for the comparison. Carbon tetrachloride and chloroform levels are higher in Hong Kong than those in the US cities, whereas methylene oxide and tetrachloroethylene have similar levels to those found in the US.

C Seasonal and Spatial Variations

BTEX and 1,3-Butadiene

Three-month running averages are used for discerning seasonal variation (Figure 4.7-2), with benzene showing a clear seasonality, high in winter and low in summer. Levels peak in December/January and bottom out in June/July, and this seasonality is observed at all four sites

(Central/Western, Tsuen Wan, Hok Tsui, and Mongkok). Toluene has a weak seasonality, high in winter /spring and low in summer, with the difference between the peaks and the valleys much reduced compared with that of benzene. In addition, the peak timing for toluene can range from December to March. Ethylbenzene and xylenes do not exhibit any discernable seasonal pattern, and 1,3-butadiene does not show any discernable seasonality either.

The general lack of seasonality could be related to the short residence time of the compounds, which are mainly controlled by reaction with OH radicals. The reactivity towards OH radicals is in the order of benzene < toluene < ethylbenzene < xylenes. If one assumes an average OH radical concentration of 5×10^6 molecules/cm³, one can estimate the lifetimes for benzene, toluene, ethylbenzene, and o-xylene at approximately 45, 9, 8, and 4 hrs, respectively.

Halogenated VOCs

The three-month running averages for the halogenated VOCs are plotted in Figure 4.7-3. Carbon tetrachloride, chloroform, and methylene chloride do not have apparent seasonality. As indicated in the previous section, the levels after December 2000 are elevated and show much larger variation than those prior to this time. Chloroform and methylene chloride exhibit similar temporal and spatial variation in samples collected after December 2000, but carbon tetrachloride has temporal variations different from those identified for chloroform and methylene chloride, and lacks a spatial gradient among the four sampling sites. Chloroform and methylene chloride have their highest concentrations appear at Mongkok whereas the concentrations at the other three sites (Tsuen Wan, Central/Western, and Hok Tsui) are similar.

Tetrachloroethylene displayed a clear spatial variation, highest at Central/Western, Mongkok and Tsuen Wan in the middle, and lowest at Hok Tsui. This spatial variation pattern was reversed for trichloroethylene; highest at Tsuen Wan, Mongkok in the middle, and similar lower levels at Central/Western and Hok Tsui. The difference in spatial patterns indicates that Central/Western may have an enhanced area source of tetrachloroethylene and Tsuen Wan may have an enhanced source of trichloroethylene. Mongkok did not appear to have sources as strong as those at Central/Western or Tsuen Wan. Tetrachloroethylene is known to be emitted from industrial activities, most notably dry-cleaning. Our result suggests that, compared with the other TAP sites, there are more dry-cleaning activity near Central/Western.

Seasonally, trichloroethylene showed significant temporal variations but with no consistent seasonal pattern. The seasonal change of tetrachloroethylene was most prominent at Central/Western, with higher concentrations in winter months and lower concentrations in the summer months for the measurement period up to December 2000. However, this pattern did not continue into 2001. Instead, its concentration in 2001 peaked in June and did not rise again in the winter of 2001. The change in seasonal pattern for tetrachloroethylene may be related to the phasing-in of control measures designed to reduce tetrachloroethylene emissions from dry cleaners in November 2001 – it is generally more difficult to discern a seasonal trend for parameters with comparable background long-term trends.

4.7.4 Summary and Recommendations

Benzene levels at the two urban sites Tsuen Wan and Central/Western are similar to those observed in other urban areas in industrialized countries around the world, and are lower than those reported for a few cities in South America and in Mediterranean areas (e.g., Athens). Hong Kong levels are about one-half of the $4.8 \mu\text{g}/\text{m}^3$ benzene air quality guideline suggested by European Communities. In annual measurements, benzene shows a clear seasonality, but such a seasonal signal is not found for the other aromatic hydrocarbons.

As expected, the roadside environment has higher levels of benzene and other aromatic hydrocarbons, demonstrating that vehicular emissions are important sources for these compounds. When compared with the levels reported in roadside environments in other countries and other cities in the Pearl River Delta region, the roadside BTEX levels in Mongkok are generally lower.

The 1,3-butadiene levels at all sites, including the roadside, are below the 2.25 $\mu\text{g}/\text{m}^3$ UK guideline value. There is a clear spatial gradient with Mongkok > Tsuen Wan > Central/Western > Hok Tsui, consistent with the spatial variation of traffic density. There is also a large difference (sixteen fold) in the annual average concentration between the roadside environment and the rural environment. No clear seasonality is observed for 1,3-butadiene.

Tetrachloroethylene displayed a clear spatial variation, highest at Central/Western, Mongkok and Tsuen Wan in the middle, and lowest at Hok Tsui. This result suggests that there are more dry-cleaning activities occurring near Central/Western than other TAP sites.

With respect to long term trends, a weak decrease can be noted for benzene, with benzene levels decreased respectively by 15% and 22% at the Tsuen Wan and Central/Western stations, from 1997/98 to 2001/02. This may be related to control measures introduced in 2000 to reduce the benzene content in unleaded gasoline. Nevertheless, similar decreases are also noted for other aromatics (toluene and xylenes) not directly targeted by the control measures. The latter is also consistent with possible emissions reductions associated with the overall economic slow down in the same period.

The annual average levels of 1,3-butadiene in 2001 were higher than those in 2000 for both Tsuen Wan and Central/Western. Similar findings were made for other LPG-related VOCs, including butane, iso-butane, 1-butene / iso-butylene, 1-butyne, pentane, 1-pentene and propane. This may be partly related to the gradual fleet change of taxis from diesel to LPG. However, we caution that two years of data is insufficient to establish any trend, and further monitoring is needed to check if these changes are part of longer term trends.

A large decrease of tetrachloroethylene was found in 2002 at Central/Western, and a small decrease observed at Tsuen Wan. These decreases may be related to control measures to reduce tetrachloroethylene emissions from dry cleaning facilities since November 2001, and is consistent with our understanding that emissions from dry cleaning facilities are more prominent near this site. Again, we must caution that the data available is not long enough to establish any trend, and further monitoring is required to check if these reductions are long lasting.

Three halogenated VOCs, carbon tetrachloride, chloroform, and methylene chloride, show an unexpected surge in their concentrations since around December 2000. Such a sudden increase is noteworthy because of their potential health impacts. It is important to further investigate whether such an increase is an artifact of laboratory contamination or is real. This question aside, the levels of halogenated VOCs are within the chronic reference exposure levels established by the California Air Resources Board.

4.7.5 Tables and Figures for Section 4.7

A Tables

Table 4.7-1	Important Toxic Volatile Organic Compounds
Table 4.7-2	Air Quality Guidelines for a number of VOCs
Table 4.7-3	Conversion Factors between ppbv and $\mu\text{g}/\text{m}^3$ for Selected VOCs and their Method Detection Limits
Table 4.7-4	Annual Average Concentrations of BTEX in Hong Kong
Table 4.7-5	Summary of BTEX Concentrations in a Number of Urban Locations around the World
Table 4.7-6	Summary of BTEX Concentrations in Roadside Environments in a Number of Urban Locations in the World
Table 4.7-7	Summary of 1,3-Butadiene Concentrations in a Number of Urban Locations in the World
Table 4.7-8	Annual Average Concentrations of Selected Halogenated VOCs
Table 4.7-9	Summary of Selected Halogenated VOC Concentrations in a Number of Urban Locations in the World

B Figures

Figure 4.7-1	Comparison of Levels of Benzene, Toluene, and Xylenes in Hong Kong with Other Urban Locations.
Figure 4.7-2	Three-month Running Average Curves of BTEX and 1,3-Butadiene
Figure 4.7-3	Three-month Running Averages of Selected Halogenated VOCs

5 Health Impact Assessment

A health impact assessment is a combination of procedures, models and tools by which a policy, program or project may be judged as to its potential effects on the health of the population, and the distribution of these effects within the population. It is useful as a planning tool for integrated decision-making.

In this study, in addition to studying the compounds traditionally identified as TAPs, we will also examine the health risks associated with criteria pollutants (e.g., O₃, NO₂ and SO₂) although they are typically not considered as TAPs. As we shall show in this chapter, the current levels of criteria pollutants in Hong Kong are associated with substantial health risks for the public, and hence our study would be incomplete if these pollutants are excluded solely because they are not officially designed as TAPs. Furthermore, there is also a movement internationally in officially designating these criteria pollutants as TAPs. For example, the Canadian government has just included O₃, NO, NO₂ and SO₂ in its list of toxic substances (Environment Canada, 2003).

5.1 Methods

5.1.1 Preamble

The health impact assessment for the TAP program was conducted separately for the cancer and non-cancer risk of toxic air pollutants using different approaches. The cancer risk is measured as the increase in the number of cases of cancer per million population that is attributable to a toxic air pollutant. The inhalation dose-response estimate is usually assumed to be linear with no threshold, and the risk is expressed as a ‘unit risk’, defined as the risk of developing cancer if a person is continuously exposed to a unit concentration (usually presented as one µg/m³) for a lifetime of 70 years. For non-cancer risk, a threshold level of exposure can usually be identified, below which it is generally believed that significant health effects will not occur.

5.1.2 Source of Data on Health Risk of TAPs

The chemicals listed in the TAP monitoring program database were checked against well-known databases from the International Agency for Research on Cancer (IARC), the United States Environmental Protection Agency (USEPA) – Integrated Risk Information System (IRIS), and the California Environmental Protection Agency (CEPA) – Office of Environmental Health Hazard Assessment (OEHHA), and the California Air Resources Board (CARB) to determine the carcinogenic classifications by these authorities. Information on unit risk was traced for chemicals that were classified as carcinogens, and reference exposure levels (RELs) were checked from the databases for non-carcinogenic effects. In addition, information on health effects was also obtained from the World Health Organization (WHO) database. The following shows the sources of information used for the health impact assessment:

- Carcinogenic classification: [USEPA-IRIS, CEPA - OEHHA, California Air Resources Board (CARB), IARC]
- Unit risk for cancer: [USEPA – IRIS, Scorecard, CEPA - OEHHA, WHO - Air Quality Guidelines]
- Reference Exposure Levels (REL) for acute health effect: [CEPA - OEHHA]
- REL for chronic health effect: [CEPA - OEHHA]
- Non-carcinogenic risk: [USEPA - IRIS, Scorecard, WHO]

5.1.3 Population Exposure Assessment Assumptions

Population exposures to TAPs were estimated from data collected in the TAP monitoring program of the HKEPD. Annualized mean concentrations of all TAPs for the period November 2000 to October 2001 were calculated for each AQMS with available information. These mean exposure levels were then applied to TAPs that had unit risk information listed in at least one of the reference databases to provide estimates of the additional numbers of cancers attributable to the exposures in one million population. For the estimation of cancer risks for all TAPs (including diesel fumes, for which comprehensive data was available only for Central/Western and Tsuen Wan stations), we used the mean concentrations of all TAPs measured in these two stations to represent the average exposure by the Hong Kong population (6,708,389 in March 2001), and to calculate the cancer risk for each TAP.

5.1.4 Cancer Risks from Diesel Fume Exposure by Airshed Using Data from PM₁₀ and Estimated Concentrations of PM_{2.5}

Since data on EC concentrations in PM₁₀ were available at most AQMSs, we maximized the use of the data by performing an estimate of cancer risks attributable to diesel fumes by combining districts into a number of airsheds. The airshed boundaries were estimated using the topographic features and the background wind conditions. Two more assumptions were made. First, air pollutants in Hong Kong were distributed in seven airsheds, each airshed consisting of several districts. Second, the EC concentrations in PM₁₀ in each airshed were represented by the mean values measured at the stations situated within the respective airshed. (There were two airsheds that contained only one station each.) The population in each airshed was the sum of the populations of the districts that made up the airshed. The HKEPD provided the data on the constituent AQMSs in each airshed and the district populations (Table 5-1a).

The cancer risk for diesel fumes was calculated based on the assumption that diesel fume concentration = 1.04 x EC concentration, as used in the Report of a Study of Cancer Risks of Diesel Particulates in Hong Kong (Wong, 2000), based on a report by STAPPA and ALAPCO (2000). The factor of 1.04 was derived from a thorough study by Cass and Gray (1995), in which diesel PM in the Los Angeles area was measured in 1982. Findings from their studies indicated that diesel sources accounted for 67% of EC in the air. It was then assumed that EC constituted 64% of diesel PM by weight. Hence,

$$\begin{aligned} \text{Diesel particulate concentration} &= \text{EC concentration} \times 67\% \div 64\% \\ &= \text{EC concentration} \times 1.04 \end{aligned}$$

The majority of EC measurements were for PM₁₀; however, EC concentrations in PM_{2.5} are a better proxy for diesel fumes as diesel PM is predominantly associated with PM_{2.5}.

To compare the cancer risk estimate from diesel fumes based on PM₁₀ EC with that based on PM_{2.5} EC, we used a formula⁶ to convert between the two measurements. EC in PM₁₀ were measured using TOT while EC in PM_{2.5} were measured using TOR. The conversion formula took into account differences in the detection methods, and assumed a fixed ratio (0.9) of EC in PM_{2.5} to EC in PM₁₀.

⁶ The HKEPD developed a formula to convert the PM₁₀ EC (at ten Hong Kong ambient AQMSs, measured by TOT) to PM_{2.5} EC (measured by TOR). The formula, a linear regression, was based on the relationship of the two species obtained from analyses of samples by the GL and the Desert Research Institute for a six-month ad hoc project by EPD in Tsuen Wan in 1999/2000.

$$\text{Estimated PM}_{2.5} \text{ EC (TOR)} = \{1.04 \times \text{PM}_{10} \text{ EC(TOT)} + 0.4247\} \times 0.9$$

5.1.5 Non-Cancer Health Risks

The following levels/guidelines were referred to:

- CEPA-OEHHA Acute Reference Exposure Level (Acute REL)
- CEPA-OEHHA Chronic Reference Exposure Level (Chronic REL)
- USEPA non-cancer risk value
- WHO non-carcinogen air quality guidelines

Acute reference exposure levels (CEPA- OEHHA)

OEHHA developed acute RELs for assessing potential non-cancer health impacts for short-term, generally one-hour peak exposures to facility emissions. (A few RELs are for 4 to 7 hour peak exposures.) By definition, an acute REL is an exposure that is unlikely to cause adverse health effects in a human population, including sensitive subgroups, exposed to that concentration for the specified exposure duration on an intermittent basis. Many acute RELs are based on mild adverse effects, such as mild irritation of the eyes, nose, or throat, or may result in other mild adverse physiological changes. For most individuals, it is expected that the mild irritation and other adverse physiological changes will not persist after the exposure ceases. Some acute RELs are based on reproductive/developmental endpoints, such as teratogenicity or fetotoxicity, which are considered severe adverse effects.

Chronic reference exposure levels (CEPA- OEHHA)

OEHHA has developed chronic RELs for assessing non-cancer health impacts from long-term exposure. A chronic REL is a concentration level (that can be expressed in units of micrograms per cubic meter for inhalation exposure or in units of milligram per kilogram-day for oral exposures) at or below which no adverse health effects are anticipated due to long-term exposure. Long-term exposure for these purposes has been defined as 12% of a lifetime, or eight years for humans.

USEPA non-cancer risk value (USEPA-IRIS; Scorecard)

The non cancer risk values from the USEPA are reference doses or concentrations and are estimates of the daily exposure to the human population (including subgroups) that is likely without an appreciable risk of deleterious effects over a lifetime. The information from the Scorecard was based on EPA risk assessment reports, although the central EPA toxicity databases do not record these values. Scorecard ranks and compares the pollution situation in areas across the US. Scorecard also profiles 6,800 chemicals, making it easy to find out where they are used and how hazardous they are. Using authoritative scientific and government data, Scorecard provides the most up-to-date and extensive collection of environmental information available online.

WHO non-carcinogenic chemicals guidelines (WHO)

The air quality guidelines for non-carcinogenic pollutants can only be applied if the averaging times are specified. The averaging time associated with a guideline value depends on the type of effects that are caused by short-term exposure producing acute effects, or long-term exposure producing chronic effects. Typical averaging times are 30 minutes for odorous pollutants, 24 hours to one week for acute exposures and one year for chronic health effects.

Only chronic effects were assessed for most TAPs, as data from the TAP monitoring program were not adequate for assessing the acute health effects. Data with measurement frequency of hourly to 8-hourly or even shorter duration are necessary for assessing acute, short-term health

effects. Hourly data was available only for criteria air pollutants. Acute health effects due to criteria pollutants have been studied and reported in previous HKEPD commissioned studies. In this review study, we compared the hourly concentrations of the criteria air pollutants with the various hourly reference levels or guidelines and calculated the frequencies at which the reference levels were exceeded. For CO and O₃, 8-hour moving averages were also calculated for comparison with the 8-hour reference levels. For SO₂, both the 24-hour moving averages and the hourly means were used. The risk of chronic health effects was assessed by examining whether the annualized mean levels exceeded the threshold exposure levels for chronic health effects in the reference databases. For criteria pollutants, comparisons of the hourly or daily data were made against the respective acute RELs.

5.1.6 Non-Cancer Health Risks (from Inhalation Route)

Apart from the methods outlined above, an alternative approach was taken on non-cancer health risks (from inhalation) based on the Air Toxics Hot Spots Program Risk Assessment Guidelines prepared by the Office of Environmental Health Hazard Assessment and the Air Resources Board of the California Environmental Protection Agency.

Non-cancer chronic inhalation health impacts were calculated by dividing the substance-specific annual average air concentration ($\mu\text{g}/\text{m}^3$) by the chronic inhalation REL ($\mu\text{g}/\text{m}^3$). The resulting ratio is termed the Hazard Quotient (HQ) for a single substance. The health impacts of simultaneous exposures to a number of substances are measured by the Hazard Index (HI), which is calculated by summing the HQs from all of the substances that affected the same organ system.

Similarly, the acute inhalation health impact was calculated by dividing the substance-specific short-term maximum concentration ($\mu\text{g}/\text{m}^3$) by the acute REL ($\mu\text{g}/\text{m}^3$) to give the HQ for a single substance and the HI for each target organ was obtained by summing the HQs of all of the substances that affected the same organ system. The maximum hourly concentrations for the four criteria gaseous pollutants in the 12-month period were used to compare with the hourly reference levels or guidelines. For CO and O₃, the maximum 8-hour moving average concentrations were also calculated for comparison with the 8-hour reference levels, whereas the maximum 24-hour moving average concentration was used for SO₂ in addition to the hourly means. An HQ or HI of one or less indicates that adverse health effects are not expected to result from exposure to emissions of that substance. As the HQ (or HI) increases above one, the probability of human health effects increases by an undefined amount.

5.1.7 Health Risks of Populations Exposed to Roadside Concentrations of Air Pollutants

An estimated population of 220,000 was exposed to elevated TAP concentrations similar to the levels reported for the Mongkok roadside station. Data obtained from the Mongkok roadside station was used to calculate the health risks for this population only. A breakdown for the 'estimated population of 220,000' is shown in Table 5-1b.

5.1.8 Health Risk from Persistent Organic Pollutants (POPs)

A separate dataset (April 2001 to March 2002) on POPs in three stations (Sha Ling, Nim Wan and Tseung Kwan O) provided by the HKEPD was used to calculate the cancer and non-cancer health risks from exposure to these chemicals per million population exposed. As these sites are unique and not representative of the exposure by the general population, the data could not be used to estimate the overall health risks in the Hong Kong population.

5.2 Results

5.2.1 Unit Risk of Cancer

Of the 58 TAPs that have information on unit risk in at least one of the three databases (USEPA, CEPA and WHO), 45 had information listed in two databases (USEPA, either IRIS or Scorecard, and CEPA), and 17 were listed in all three. The unit risk values for eight TAPs: 1234678-HpCDD, 123678-HxCDF, 123789-HxCDD, bromoform, crotonaldehyde, hexachlorobutadiene, chloromethane (methyl chloride), vinylidene chloride (1,1-Dichloroethylene) were listed only in the USEPA database. Unit risks for five TAPs were only quoted in the CEPA database: Allyl chloride, lead, ethylidene dichloride (1,1-dichloroethane), OCDF, and total HpCDD. The unit risk of one TAP, fluoranthene, was only quoted by the WHO (this TAP was not classified as a carcinogen by any other agencies). The maximum and minimum values available for each chemical were used to estimate the upper and lower bounds of the risk in the health impact assessment.

Among the 45 chemicals with a carcinogenic classification of ‘possible human carcinogen’ or stronger, acrolein, naphthalene and ethyl benzene, have no unit risk information available from any of the sources listed above.

Table 5-2 shows the unit risk of cancer for the chemicals in different groups obtained from different agencies. CAS numbers, used to uniquely identify the TAP species, are also listed in Table 5-2. For species that do not have assigned CAS numbers (e.g. RSP or PM₁₀), numbers in the form 999-99-X have been used to help identify the species internally in this study.

5.2.2 Classification of Carcinogens

The carcinogenic potentials are classified by the USEPA and IARC under different categories. The numbers of chemicals classified under the two systems are shown in Table 5-3 and Table 5-4 below.

We could not find the carcinogenic classification of 100 chemicals under any of the databases reviewed, but 16 of these (all under the group ‘dioxins’) had a unit risk given by either the USEPA or CEPA-OEHHA. The carcinogenicity of 27 chemicals was classified as ‘possible human carcinogen’ or stronger by the IARC (‘2B’ or above), whereas 29 [OEHHA], 37 [IRIS], 28 [CARB] chemicals received a corresponding classification from the USEPA (‘C’ or above). Altogether, 41 chemicals had a classification of ‘possible human carcinogen’ or stronger.

5.2.3 Non-Cancer Health Risks

Non-cancer risk information was available for only 51 chemicals from all sources. Eight TAPs had available information from all three databases (WHO, USEPA, CEPA). Information on some chemicals was available from only one database – 16 TAPs from the USEPA, 7 from the CEPA and 4 from the WHO. Of the 21 chemicals having non-cancer risks listed in the IRIS, only 1,1-Dichloroethylene did not have a non-cancer risk value identified through Scorecard.

We could not identify any information regarding carcinogenicity and non-cancer risk for 98 chemicals after searching all databases. The majority of these were different isomers of dioxins and VOCs. Table 5-5 details the non-cancer risk factors of TAPs by chemical type from various sources.

5.2.4 Summary Findings of Cancer and Non-Cancer Health Risks of TAPs

A summary of information on TAPs from our search is shown in Table 5-6.

5.2.5 Mean Concentrations of TAPs by Station and Estimated Cancer Cases per Million Population Exposed

Table 5-7 shows the mean concentrations of TAPs for a one-year period from November 2000 to October 2001, by AQMSs. This table also lists the number of cancer cases in one million population, based on unit risks from the three sources – WHO, USEPA and CEPA (OEHHA).

5.2.6 Mean Concentrations of Taps in Central/Western and Tsuen Wan Stations and the Estimated Cancer Cases Per Million and for the Total Hong Kong Population

Table 5-8 shows the cancer risk estimates per million population and for the total Hong Kong population, assuming 70 years of exposure to the mean concentrations of TAPs in Central/Western and Tsuen Wan. Diesel fumes accounted for 8,158 cases of cancer, or about 80% of the total cancer risk when the USEPA and CEPA unit cancer risks were used for the calculation. The WHO had no unit cancer risk for diesels, but we assumed the same risk across all three databases to facilitate comparison. The proportion of cancers attributable to diesel became larger in the column showing WHO unit cancer risks. This is because the WHO provided unit cancer risks for much fewer chemicals in the TAP list than the other two databases. Other important contributors include a few metals (i.e., hexavalent chromium, arsenic and cadmium) and a few organic compounds (i.e., benzene, 1,3-butadiene, carbon tetrachloride, formaldehyde, 1,1,2,2-tetrachloromethane, and tetrachloroethane).

5.2.7 Estimated Cancer Cases attributed to Diesel Fumes based on Mean Elemental Carbon Concentrations (based on PM₁₀ and PM_{2.5}) in Seven Airsheds

Table 5-9 shows the estimated EC concentrations in PM_{2.5} from EC concentrations in PM₁₀, using the following formula.

$$PM_{2.5} \text{ EC} = \{1.043 \times PM_{10} \text{ EC} + 0.4247\} \times 0.9$$

Shaded boxes in Table 5-9 represent estimated EC concentrations determined from the above formula. At concentrations lower than 6.235 µg/m³, the PM_{2.5} EC exceeded the PM₁₀ EC. At concentrations greater than 6.235 µg/m³, the reverse was the case. The PM_{2.5} EC concentrations at ten AQMSs were estimated from the PM₁₀ EC values. The reverse was done for Hok Tsui. In Tsuen Wan and Mongkok, where data for both PM₁₀ EC and PM_{2.5} EC were available, estimates of both particulate species were undertaken to provide a comparison with the measured values. Risk estimates were made using the measured values wherever available.

Table 5-10 shows the cancer risks calculated from PM₁₀ and PM_{2.5} in the seven airsheds. Using PM₁₀ EC measurements, the district-based approach (based on 7 airsheds) yielded a similar risk estimate (8,100 cancer cases) to that based on the mean of two urban stations (Central/Western & Tsuen Wan) measurements (8,158 cancer cases). The number of estimated cancer cases based on PM_{2.5} EC was 8560, 6% higher than that found using PM₁₀ EC data.

5.2.8 Health Risks of Dioxins

The cancer risk of dioxins based on the ambient concentrations of 2,3,7,8-TCDD was comparatively small, at 1.8 (USEPA) and 2 (CEPA) cases per million respectively. The ratio of dioxin-like PCBs to dioxins was calculated using measurements in an ad hoc ASG/EPD study (see Table 5-11). This was done by dividing the sum of the mean monthly values of dioxin-like PCBs by the mean monthly values of total dioxins (i.e., 0.015 pg I-TEQ/m³ / 0.062 pg

I-TEQ/m³ = 0.24:1).

The ratio of dioxin-like PCBs to dioxins is based on the best data available and hence a best estimate. However, the correlations between the 11 measurements (monthly measurements from January to December 2002, except April 2002) were low ($r=0.073$) and statistically insignificant ($P=0.83$) (See Table 5-11 for the data and statistical results). Therefore, one has to exercise caution in estimating the total health effects of dioxins and dioxin-like PCBs based on measurements of dioxins only.

Another way to assess the health effect of dioxins and dioxin-like PCBs is to estimate the total daily intake in relation to the intake by inhalation. A rough estimate commonly adopted is that the latter constitutes 2% of the total intake (HKEPD, 2000b), while dietary intake contributes the remaining 98%. If we project according to this assumption:

$$\begin{aligned} \text{The mean daily dietary intake} &= 1.24 \times 0.054 \text{ pg I-TEQ /m}^3 \times 17.3 \text{ m}^3 / (0.02 \times 70 \text{ kg}) \\ &= 0.82 \text{ pg I-TEQ/kg/day.} \end{aligned}$$

(The factor 1.24 was derived by assuming that the ratio of dioxins: dioxin-like PCBs is 1:0.24. The daily inhalation of air at 17.3 m³ was based on a respiratory rate of 20 / min and a tidal volume of 600 ml; the factor 0.02 was based on the assumption that inhaled dioxins and dioxin-like PCBs constituted 2% of total intake, and the body weight of a “typical” person was estimated to be 70 kg.)

This level of total daily intake can be compared to a WHO recommended level of 1-4 pg-TEQ/kg/day (WHO, 1998; WHO 2000b). (See 5.3.4, under Discussion)

5.2.9 Estimated Cancer Cases for Population Living Near the Roadside

Table 5-12 shows the cancer risk of an estimated 220,000 residents exposed to roadside concentrations of air pollutants using PM₁₀ and PM_{2.5} data from Mongkok.

5.2.10 Cancer Risks of Persistent Organic Pollutants (POPs)

Another dataset from the SWG study was used to calculate the cancer risk of these thirteen chemicals. These chemicals were monitored at three stations: Sha Ling, Nim Wan and Tseung Kwan O from April 2001 to March 2002. This period was different from that used to estimate health risks from TAPs, and hence the total cancer burden from both TAPs and POPs could not be calculated by simply adding the numbers obtained from the two estimates. Nevertheless, the contribution of POPs to the cancer risk gave a general idea of their relative importance in the health impact assessment of all the pollutants (TAPs and POPs). The cancer unit risk is presented in Table 5-13, the non-cancer risk is in Table 5-14, and the estimated cancer risks based on data from the three stations are presented in Table 5-15. The USEPA unit risks for p,p'-DDD and p,p'-DDE were much higher than those from CEPA, and these two POPs together contributed to over 95% of all cancers attributable to the 11 POPs. p,p'-DDE is the predominant contributor of cancer, accounting for 60%-83% of all cancer cases, while p,p'-DDD accounts for 14%-31% of cancer cases. Non-cancer health risks of these POPs are not available in either dataset for assessment (Table 5-13, Table 5-14 and Table 5-15). The numbers of cancer cases as calculated from readings obtained in the three stations are shown in Figure 5-1.

5.2.11 Non-Cancer Health Risks of TAPs:

The following is a brief summary of some important non-cancer health risk of the TAPs:

Exposure to most TAPs (including most criteria pollutants and organic chemicals) cause irritation to the eye and respiratory tract as acute effects, while some organics, notably methyl bromide, chloroform, perchloroethylene, carbon tetrachloride, benzenes and its derivatives, and mercury cause acute and chronic effects on the nervous system. Acute exposure to formaldehyde may affect the immune system, while chronic exposure to acetaldehyde is associated with olfactory epithelium degeneration. Acrolein can cause squamous metaplasia and neutrophilic infiltration of the nasal mucosa. Carbon monoxide, a criteria pollutant, affects cardiovascular function by binding to hemoglobin. The heavy metals lead, manganese and mercury all cause various nervous system damage. Benzene is a well-known toxin of the bone marrow. Chloroform and carbon tetrachloride cause liver damage.

Table 5-16 provides data on acute and chronic non-cancer health risks.

Acute effects:

Tables 5-17 and 5-19 list the number and percentage of short-term mean (1-hour, 8-hour and 24-hour) concentrations of criteria pollutants that exceeded the reference levels from various sources, together with the number of days and the percentage of days in which the reference levels were exceeded in different AQMSs.

The HQ of NO₂ (1-hour) exceeded one at all stations except Tap Mun. The condition was worst in Causeway Bay with an HQ of 1.9. The highest HQ of SO₂ (1-hour) was found in Yuen Long⁷ (2.5). The HQ of O₃ (1-hour) exceeded one at eight stations and was less than one at four stations (Central, Eastern, Kwai Chung, Kwun Tong) with the highest value in Tung Chung (1.9). The same distribution was seen for HQ of O₃ (8-hour), with the highest value again in Tung Chung (2.0).

The HI for the respiratory system (1-hour) exceeded one at all stations with available information on the criteria air pollutants, the highest value being 4.3 in Causeway Bay. For eye irritation, the hourly HI exceeded one at stations where the HQ of O₃ exceeded one. The 8-hour HI for the respiratory system and eyes had the same pattern and corresponded to the stations where the HQ of O₃ exceeded one.

Chronic Effects:

Tables 5-18 and 5-19 list the annualized means of the criteria pollutants and TAPs that exceeded the reference levels from various sources for different AQMSs.

The reference levels for acute effects of two gaseous criteria pollutants (NO₂, O₃) were exceeded frequently at many AQMSs. These observations agreed with the results of the earlier time series studies of the criteria air pollutants on health effects commissioned by the HKEPD. The annualized mean concentrations of NO₂ exceeded the reference levels for chronic effects at most AQMSs. The reference levels for chronic effects of formaldehyde were exceeded at the two urban stations (Central/Western and Tsuen Wan) and those for diesel fumes were exceeded at three urban stations (Tsuen Wan, Kwai Chung, and Mongkok).

The HQ of NO₂ again exceeded one at all stations that had available information except Tap Mun. The condition was worst in Causeway Bay with an HQ of 2.6. Other pollutants that had

⁷ It should also be note that the typical concentration of SO₂ over Hong Kong is quite low, and the high HQs of SO₂ were related to a few episodic events.

an HQ exceeding one included: formaldehyde in Central / Western (1.9) and Tsuen Wan (1.8); naphthalene in Mongkok (1.7); and diesel in Kwun Tong (1.1), Tsuen Wan (1.1) and Mongkok (4.1)

The HI for the respiratory system exceeded one at all stations except Hok Tsui and Tap Mun, the highest value being 5.9 in Mongkok. The eyes and the nervous system were the other two organ systems that had an HI greater than one: the eyes in Central / Western (1.9) and Tsuen Wan (1.9), and the nervous system in Mongkok (1.1).

Health Effects of Long-Term Exposure to PM_{2.5}

The annual mean concentration of FSP at Tsuen Wan is 34 µg/m³. Based on a relative risk of 1.14 (total mortality) per 10 µg/m³ of PM_{2.5} by the WHO guidelines (WHO, Air Quality Guidelines for Europe Second Edition), and assuming an annual number of 32,827 deaths (average of 1995 – 1998), a total of 4,596 deaths per year could be attributed to an increase of 10 µg/m³ of PM_{2.5} concentrations. The calculation of 15,626 deaths attributable to PM_{2.5} was based on reductions of PM_{2.5} from 34 µg/m³ to zero. The mean annual number of deaths (for the period 1995-1998) was 32,827, suggesting that 47.5% of deaths could be attributed to PM_{2.5}. However, this is a theoretical number and not a realistic one because it would be impossible to achieve a zero concentration of PM_{2.5}, and it should also be noted that the proportions of deaths attributable to several risk factors could in theory add up to >100%.

5.3 Discussion

5.3.1 Main findings

Our findings of the cancer risks attributable to various TAPs are similar to results of the MATES II Report (2000). Diesel fumes are the major contributor to the cancer risk, accounting for 80% of the cancer cases caused by the TAPs, followed by those chemicals of common concerns to environmental health, namely, the metals - hexavalent chromium, arsenic and cadmium, and the organic compounds - benzene, 1,3 butadiene, carbon tetrachloride, formaldehyde, 1,1,2,2-tetrachloromethane and tetrachloroethene. This is within our expectations. The proportional contribution of TAPs towards health risk in Hong Kong is not expected to be too different from that in other major cities, because the sources of air pollutants in major cities are generally similar, with traffic and industries contributing most of the pollution.

5.3.2 Regulatory Decisions for Cancer Risks

It is generally agreed that an additional cancer risk of less than one in a million for a lifetime exposure (70 years) is a low level of risk that does not warrant regulatory actions. A risk between one to 100 in a million is sometimes regulated, based on the USEPA's past risk-based regulatory decisions. A cancer risk that is above 100 in a million is often regulated. Our cancer risk estimates for the TAPs serve as a guideline in the consideration for regulation and control of these pollutants by the HKEPD.

5.3.3 Cancer Risk of Diesel Fumes

Diesel engine exhaust was classified as a Class 2A substance (probable human carcinogen) by the IARC (1989). It consists of a mixture of chemicals including PAHs known to be carcinogenic in animals. The cancer risk for diesel fumes was over one thousand cases per million for most stations, and accounted for about 80% of cancer risks from all TAPs. This is clearly a priority area for control. In view of the substantial cancer risk posed by diesel fumes to the population, control measures must be undertaken to reduce the ambient concentrations of

diesel fumes. The district-based cancer risk estimate from EC concentrations in PM₁₀ (8100 cases) differs little from that based on the mean of Central/Western and Tsuen Wan (8158 cases). The cancer risk (8560 cases) based on EC concentrations in PM_{2.5}, which were estimated by the regression equation, was about 6% higher than the estimate using PM₁₀ EC. There are considerable district differences in PM₁₀ EC in the urban sites ranging from 3.5 µg/m³ in Central/Western to 5.4 µg/m³ in Kwun Tong. The risk estimate for the total Hong Kong population is greatly influenced by the urban station measurement results, because 57% of the total population lives in the “urban” airsheds. This is a plausible explanation for the similar risk estimates between these two approaches. The small discrepancy (about 6%) between these two estimates illustrates the robustness of the underlying assumptions.

There are, however, some differences between the concentrations of measured PM_{2.5} EC and estimated PM_{2.5} EC in Mongkok (19.8 µg/m³ vs. 10.7 µg/m³) and Tsuen Wan (5.15 µg/m³ vs. 4.5 µg/m³). The larger discrepancy in Mongkok may be due to larger differences for high EC loading samples between the two measurement techniques.

5.3.4 Cancer Risk of Dioxins

Dioxins have received much attention in recent years. Our data shows that the contribution to cancer risk is comparatively small, and of the magnitude of several cases per million. Although the health risk from dioxins is relatively low in the general population, it should be noted that there are localized areas (outside the current TAPs monitoring network) with sources of emissions that might give rise to much higher health risks. In a previous study, Sin et al. (2002) reported considerable spatial variations in ambient dioxin concentrations in different districts of Hong Kong. The highest concentration was recorded in Yuen Long in winter, which was about four times higher than in Central. This implies that the estimated total daily intake of dioxins, as reported in Section 5.2.8, might be four times higher in the northwestern New Territories under certain synoptic conditions. This would be at the higher end of the recommended limit by the WHO. Thus, any further increase in ambient dioxin concentrations in this part of Hong Kong should be thoroughly studied to avoid posing a health hazard including both cancer risk and non-cancer health effects to the local residents. Further monitoring of dioxin levels in the northwestern New Territories is recommended.

5.3.5 Non-Cancer Risks of TAPs

Several TAPs (diesel, NO₂, O₃, SO₂ and formaldehyde) were found to have exceeded the reference values in some measurements. Superficially, the effects appear to be relatively minor (respiratory tract and eye irritation). In reality, the health consequences on respiratory health are substantial, as the effect on health is at least additive.

The short- and long-term exposures to criteria pollutants (including PM₁₀) have been shown to be associated with increased mortalities and respiratory diseases in local studies (Wong et al, 1999, 2002a, 2002b, Yu et al, 2001). For the acute inhalation non-cancer risks, three criteria pollutants (NO₂, O₃, SO₂) contributed to HQs and HIs of greater than one at many of the AQMSs. The level of NO₂ is of particular concern. The effects may appear to be relatively minor (respiratory tract and eye irritation), but could contribute to increased mortalities and respiratory diseases found in local time-series studies (Wong et al, 1999, 2002a, 2002b). For the chronic inhalation non-cancer risks, NO₂ was again the major contributor to health risk. The relative risks of NO₂ and O₃ are especially high. Although overseas studies have focused on PM₁₀ and recently PM_{2.5}, NO₂ is highly correlated with the former, and might have accounted for much of the short-term health effect.

In addition, some TAPs have other long-term effects – diesel and formaldehyde are

carcinogenic. The respiratory system was the organ system most affected and the findings also corresponded well with the results from an earlier local study on respiratory health of school children (Yu et al, 2001). A high HI for eyes was noted in Central / Western and Tsuen Wan and could be attributed mainly to the level of formaldehyde at those two stations. The only other high HI was for effects on the nervous system in Mongkok, where three metals and nine VOCs contributed to an HI of 1.15. Manganese was the main metal pollutant (HQ = 0.52) and methylcyclohexane was the major VOC involved (HQ = 0.29).

Stringent control of these TAPs should bring about considerable benefits on the health of the population. We recommend that a high priority should be accorded to control those TAPs (in particular, NO₂, diesel fumes and formaldehyde), exceeding the acute and chronic reference exposure levels (RELS).

5.3.6 Health Effects of PM_{2.5}

The health impact of PM_{2.5}, like dioxins, has been a focus of epidemiological research. Most of the studies have been on short-term health outcomes, such as hospital admissions and mortalities. Epidemiologists use time-series studies to assess these health outcomes. In this study, short-term variations in the concentrations of PM_{2.5} are examined against the aforementioned short-term health outcomes, after adjusting for various cyclical (such as seasonal) factors. It is beyond the scope of this study to investigate the short-term health effects of PM_{2.5}. We have calculated an overall mortality risk of nearly 4,600 deaths based on a relative mortality risk of 1.14 per 10 µg/m³ of PM_{2.5}. The monitoring of PM_{2.5} should be accorded a high priority, as Hong Kong-wide data on daily concentrations of PM_{2.5} are only available at a few stations (Tsuen Wan, Tung Chung and Tap Mun) and are not representative as a measure of exposure by the population for which health outcome (daily hospital admission statistics and mortality statistics) data are available.

In a separate study on air pollution and health outcomes conducted by CUHK, problems with the quality of the health outcome data were identified (Wong TW and Yu TS, personal communications). Mortality data, obtained from Census and Statistics Department via the HKEPD, was incomplete in terms of the date of death, although they are an important estimate of health outcome. Similarly, missing data in TPUs was commonly found in the mortality data. As a result, daily time series cannot be performed. Data from the Hospital Authority could not be easily divided into districts according to the residential address of the patients, which hinders district-based (i.e., health outcome data matched with the corresponding air pollutant data) analysis.

Hence, we recommend the use of TPUs or at least, well-defined districts comparable to the air pollutant data, for district coding of hospital patients, and the mandatory entry of date of death and TPU of the deceased to facilitate time-series and spatial analysis. A better monitoring network for PM_{2.5} is important for improved definition of local population exposures. Moreover, the availability of high quality mortality and hospital data, through the establishment of an online health and environmental database, would enable a more accurate risk estimate using the time series and spatial approach. In particular, data (with personal information removed) must be routinely updated into this online health and environmental database, and be readily accessible by researchers. A stable and open access health information system similar to the current air quality database will go a long way to help improve our understanding of environmental and public health issues in Hong Kong.

Long-term health impacts of PM_{2.5} have not been well researched, but it is reasonable to assume that PM_{2.5} contributes to a substantial fraction of cancer risk, considering diesel fumes

are a major contributor of PM_{2.5}. The long-term effect of PM_{2.5} on health was based on data from a cohort study in the U.S. by Dockery et al (1993). The data may not be applicable to Hong Kong because of differences in the level of population exposure and the composition of PM_{2.5}, and other differences in environmental and climatic factors. Local data on the health effects of PM_{2.5}, both acute and chronic, are not available. It is recommended that studies on the short-term and long-term health effects of PM_{2.5} be conducted as a matter of priority.

5.3.7 Combined Effects of Different Chemicals

It should be noted that when a person is exposed to more than one carcinogenic substance, the cancer risk increases. It is possible that the combined effect of simultaneous exposure to different carcinogenic substances is greater than the sum of the individual effects. Hence, the combined risk may be unacceptably high even though the risk of exposure to a single chemical might stay within the “acceptable limit”. The same holds true for non-cancer risks.

5.3.8 Cancer Risks in Populations Exposed to Roadside Air Pollutant Concentrations

Part of the Hong Kong population is affected by the roadside air pollutant concentrations (for details of this population, see Table 5-1b-e). The estimated cancer risks in this subgroup are generally higher than in the general population. The number of cancer cases attributable to diesel fumes was much higher for PM_{2.5} than PM₁₀. This is because the EC concentrations in PM_{2.5} measured in Mongkok was about twice as high as the measured EC concentrations in PM₁₀. This discrepancy is due to the fact that two different analysis methods were used to obtain EC in PM_{2.5} and in PM₁₀. The risk estimate using PM₁₀ EC concentrations in Mongkok was nearly three times the corresponding risk for the entire Hong Kong population, because the latter was based on a much lower EC concentration (about one-third), which was the average EC at the Central/Western and Tsuen Wan stations. The concentrations of 1,3-butadiene and benzene in Mongkok were also much higher than the ambient levels, at about four times and 2.5 times the averaged concentrations at Central/Western and Tsuen Wan. These differences in risk estimate reflect the large differences between the roadside concentrations and ambient concentrations of these TAPs. Owing to the lack of data on some TAPs (e.g., hexavalent chromium and formaldehyde), it is not appropriate to compare the percentage contribution by the individual TAPs in the two tables (overall Hong Kong population and the roadside population).

5.3.9 Cancer Risks of POPs

The dataset of POPs was collected at different sites in a different time period. An assessment of their cancer risks, however, revealed a low to moderate risk, except for p,p'-DDD and p,p'-DDE. Among the list of eleven⁸ POPs monitored at the three stations (Tseung Kwan O, Sha Ling and Nim Wan), p,p'-DDE contributes to the majority of the cancer cases (ranging from 62 to 83 % at the three stations). The concentrations of p,p'-DDE at these stations range from 23.5 to 38.3 pg/m³, or about three times higher than the ambient levels reported in other countries (see Table 5-20). The concentrations of p,p'-DDD in Hong Kong are also much higher than in Japan. In view of their relatively high ambient concentrations, monitoring of p,p'-DDE and p,p'-DDD should be continued on a regular basis.

The major risk to health posed by this group of chemicals is through food intake, owing to their persistence in the environment and propensity to enter the food chain. DDD and DDE together

⁸ Of the eighteen POPs monitored in these stations, unit risk of cancer was available for eleven POPs, as shown in Table 5-13.

contribute most of the cancer cases attributable to POPs. In future routine monitoring work by the HKEPD, it is advisable to include those chemicals and possibly others such as organo-mercury compounds, brominated flame retardant (PBDE), pentachlorophenol, etc. that might soon be included in the list of Stockholm Convention chemicals. In a local study (Louie and Sin, 2003), DDT was detected in the highest point in Hong Kong (Tai Mo Shan, 957m), suggesting non-local sources. This finding provides further justification for additional monitoring of these chemicals through routine and/or focused ad hoc studies.

5.3.10 Assumptions and Limitations

In our estimation of health risks, the unit risks expressed by various regulatory agencies refer to the health risk by the inhalation of airborne chemicals only. Other routes of entry into the human body such as ingestion or skin absorption cannot be assessed. While the atmosphere can be a major source of exposure for some chemicals, some other chemicals enter the human body predominantly by ingestion or skin absorption. For these chemicals, the health risk posed by inhalation would be a poor indicator of the overall health risk.

The major limitation of a health impact assessment is the wide range of the unit risk quoted by various regulatory / advisory agencies. This is an inherent feature of health risk assessment – the uncertainty is wide, owing to the intrinsic difficulties in projecting data derived from laboratory animals and epidemiological studies of specific groups (not necessarily representative) to the general population. Which unit risk should be used in Hong Kong is a difficult question to address. It is a general principle in environmental health to build in safety factors, and to err on the side of caution. Yet, we need to balance this with the practicality of control measures and their impact on other aspects, like the socioeconomic. This is an area outside the scope of this Report. Hence, we present the risk as an upper limit and a lower limit. The upper limit is based on the most stringent unit risk from the literature, and the highest concentrations of the TAPs from the stations. The lower limit is based on the least stringent unit risk and the background level. The true risk probably lies somewhere in between.

The lack of a comprehensive exposure assessment is another major limitation in this study. It is costly to implement a monitoring system that is representative of the exposure by residents in different districts. The mean concentrations of a number of TAPs that are available at more stations (e.g., the metals and EC) have substantial spatial characteristics, suggesting that variations in the emission sources and/or transport of these emissions may play an important role in determining the differences in the measured concentrations in these localities. The varied topography of Hong Kong and diversity of the sources of emissions of pollutants, together with the mobility of the population, mean that our use of average levels of TAPs from two urban stations as an index of exposure is an approximation of the true exposure.

Ambient TAP data was available for Central/Western and Tsuen Wan that represented exposure by the urban populations (March 2001 population: 6,708,389). Roadside TAP data was available for Mongkok that represented the population exposed to roadside concentrations (220,000). Because of the limited number of stations with data on TAPs, it seemed appropriate to represent the average population exposure in Hong Kong as the average of TAP concentrations from the two urban stations. Health risks in more ‘rural’, or less polluted districts than Central/Western and Tsuen Wan might be over-estimated, while districts that are similar to or more polluted than Tsuen Wan would have the health risks of their populations under-estimated (there seems to be a significant northwest to southeast gradient in many TAP concentrations). Data obtained from roadside stations should be used as an extreme situation for certain groups of population only, whereas baseline levels of TAPs are well represented by Hok Tsui station.

As diesel fumes constitute the major cancer risk, it is important to recognize the limitations of our results. Two major uncertainties of the cancer risk estimates for diesel fumes exist: 1) that the unit risk (ranging from 150 – 1000 per million per $\mu\text{g}/\text{m}^3$) was arbitrarily (and conservatively) chosen by the California EPA as 300 per million per $\mu\text{g}/\text{m}^3$; and 2) that the concentration of diesel fumes was estimated from the EC concentrations based on a factor of 1.04, again derived from Californian data. There might be considerable differences in the risk estimate if the percentage contributions by diesel fumes to the total ambient EC concentration and the percentage of EC in diesel exhaust differ substantially from the Californian data. Local data on the relationship between elemental carbon and diesel are lacking. There are also differences in the analytical methods in the measurement of EC. This adds to yet another uncertainty.

The estimated total daily intake of dioxins, based on a 2% contribution from the airborne route, may vary widely because of uncertainties in estimating the amount of dioxins from food (assumed as 98% of total intake), for which data were not available. The estimated ratio between dioxins (expressed as TEQ of 2,3,7,8-TCDD) and the dioxin-like PCBs was an approximation and lacks statistical significance. A more reliable estimate should be made when data on individual congeners are available.

5.3.11 Importance of the TAP Monitoring Program and the Health Impact Study

This is the first systematic study in Hong Kong on TAP monitoring and health risk assessment. Notwithstanding the above limitations, the impacts of TAPs on community health in terms of cancer risk and non-cancer risk have been estimated.

Results of this study set a baseline health risk estimate for the general population, against which different scenarios of risks can be worked out based on different pollution profiles in specific districts / areas. Moreover, the impact on health can be assessed from the trend analyses, with a projected increase or reduction of emissions of various TAPs in Hong Kong. The results also provide useful information on a health-based priority in the control of TAPs, which must be considered together with other environmental, technological, social and economic concerns.

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6 Review of Concentration Trends of Individual TAPs against Control Measures Introduced by the HKEPD

6.1 Limitation of the Concentration Trend Review against Individual TAP Control Measures

A list of air toxics control measures introduced by the HKEPD since 1998 is shown in Table 6-1. There were also a number of air toxics control programs before 1998. However, since it is difficult for us to use the TAP dataset to review the impact of control measures implemented before the beginning of the TAP monitoring program, only measures enacted after 1997 are listed in Table 6-1 (except for cases where the control measures started before 1998 and were fully implemented in or after 1998).

Further, we must caution that the TAP monitoring record is too short for any conclusive statement on long-term impact of the control measures. Many factors affect the TAP levels measured at monitoring sites, including local meteorology and emission changes. Large fluctuations are known to exist in local or regional meteorology on the interannual time-scale (a well-known example is the El-Nino Southern Oscillation which has a time-scale of two to three years). In addition, even in the absence of control measures, emission characteristics will change as a result of the use of newer technologies, migration of industries, or change in local economic conditions, etc.

Hence, we must caution that a few years of monitoring data is really too short for supporting any conclusive statement about the long-term trends or impacts of particular control measures. Reviews and findings provided in subsequent sections of this chapter must be considered preliminary and provisional; longer term monitoring and evaluation must be carried out to verify and challenge these findings.

6.2 Diesel Particulate Matters

A weak but discernible decreasing trend is noted in the level of NO_x and particulate matter including TSP, RSP and FSP concentrations recorded at the roadside (HKEPD 2003). This is consistent with continual efforts by the HKEPD to reduce particulate emissions from the vehicular fleet, which include the introduction of Ultra Low Sulfur Diesel (ULSD), Euro I, II, III standards for large diesel vehicles, Euro Phase 1, 2 and 3 for small diesel vehicles, the transition of diesel taxis to LPG taxis and the particulate trap / diesel oxidation catalysts retrofitting program for light diesel vehicles. Noting the dominant role of diesel PM and NO₂ in our chronic and acute health risk estimates, these control measures move in the right direction and should be further enhanced.

No clear and consistent trend can be noted for the variation of EC and PM levels at the general stations. This suggests that there must be factor(s) / sources(s) other than local vehicular traffic that are important for controlling EC and PM levels at these AQMSs. Without a good understanding of the emission profile of the various sources, their relative contributions to PM variations are not clear. Notable sources such as marine shipping activities at or near the container terminal at Kwai Chung may be related to the apparent rise in EC (and PM) concentrations at the Kwai Chung AQMS during the past few years. It is recommended that the HKEPD study the emission profiles of various EC and PM sources (e.g. the CARB type of control measures targets), and if their contributions are found to be significant, consider additional control measures to reduce their emissions.

6.3 Benzene and Vapor Recovery

In the trend analysis of VOCs (section 4.7.3), we note that a weak decreasing trend is found for benzene; with the 2001/02 concentrations lowered by 15 and 22% at Central/Western and Tsuen Wan stations, respectively, from their 1997/98 mean. Two control measures were introduced to reduce benzene concentrations in air (Table 6-1). The first is a vapor recovery rule at petrol filling stations implemented in April 1999, and the second is a change in benzene levels in unleaded gasoline from 5% to 1% effective April 2000. Lack of measurements before 1997 makes it difficult to assess the effectiveness of the first control measure. Nevertheless, the second control measure is consistent with the reduction of benzene observed in the last few years. This may be partly related to reduce emissions in association with the overall economic slow down during the same period. The latter is consistent with the observation that similar decreases are being seen in the levels of other aromatics (toluene and xylenes). Further monitoring is needed to see if these changes are part of longer-term decrease or simply part of transient variations for air quality in Hong Kong.

6.4 Tetrachloroethene

A vapor recovery regulation has been in effect from November 2001 to reduce the level of tetrachloroethene in Hong Kong. Before implementation of this regulation, 20% of the approximately 400 dry-cleaning facilities in Hong Kong were of the latest models that meet required emission standards. It is targeted that all facilities meet the emission standards by 2008. Data up to the end of 2001 did not show any notable change in the level of tetrachloroethene, however data for 2002 shows a substantial decrease of about 30% at Central/Western when compared with the average concentration for the previous three years (section 4.7.3). Even at Tsuen Wan, a decrease of 13% is noted. These decreases are consistent with the tetrachloroethene control measures introduced, and suggest that the control measures may have helped in the reduction of ambient tetrachloroethene concentration. The larger decrease in Central/Western is also in line with our finding that emissions from dry cleaning facilities are more prominent near this site. Improvement for one single year must at best be labeled as a consistent observation, and should be corroborated with subsequent measurements to verify that tetrachloroethene concentrations have indeed decreased as a result of the aforementioned control measure.

6.5 Phasing Out of Leaded Petrol

The phase out of leaded petrol began in 1991 with the eventual ban of leaded petrol implemented in April 1999. By the beginning of the TAP monitoring program in 1997, the lead levels in the Hong Kong was already very low (when compared with the corresponding Air Quality Objective for lead), and there was no clear trend in the annual mean lead concentrations. Instead of being a negative result, this is a clear cut example showing that the control measures are having an impact, and lead levels in vehicular emissions have become minimal. The lead concentrations recorded at the Mongkok roadside station are similar to those recorded in the other urban AQMSs, indicating that local vehicular traffic is no longer an important source for the ambient lead levels in Hong Kong. In contrast, the consistently higher lead concentrations at Yuen Long in comparison with all other stations and the associated circular pollution wind map further suggest that regional transport has significant impact on lead levels in Hong Kong.

6.6 Reduction of Sulphur Content in Diesel

Reduction of sulphur content in diesel and introduction of ULSD were implemented mainly as part of the overall effort to reduce secondary PM formation in Hong Kong. In addition, they also have direct impact on ambient SO₂ levels, particularly at the roadside. For example, the average SO₂ concentration at the roadside in 2002 (18 µg/m³) dropped by 36% compared with the 2000 value (28 µg/m³), likely as a result of the introduction of ultra low sulphur diesel for the vehicle fleet in late 2000 (HKEPD 2003).

6.7 Fleet Change of Taxis from Diesel to LPG

The LPG taxi pilot scheme started with 30 taxis in November 1997. By the same time in 2001, about 75% (or 13,500 taxis) of the fleet have been converted to LPG. A review of LPG-related VOCs commonly found in LPG (including propane, pentane, butane, iso-butane, 1,3-butadiene, 1-butene / iso-butylene, 1-butyne and 1-pentene, etc.) showed that their concentrations in the 2000/01 cycle were higher than that of the 1999/00 cycle⁹. With recently updated VOC data, three complete annual cycles of 1,3-butadiene measurements (1999/00, 2000/01 and 2001/02) are available at Central/Western and Tsuen Wan. The annual average levels in 2000/01 and 2001/02 are higher than those identified for 1999/00 at both sites. These increases are consistent with the increased number of LPG taxis on the road. However, the increases are relatively gradual and two years of data are not enough to establish any long-term trends; further monitoring is needed to assess the longer term impact of the LPG conversion. Of particular interest in this group of LPG-related TAPs is 1,3-butadiene, which is amongst one of the most important TAP species identified in section 5.2.6 in terms of the overall health risk assessment. It is of interest to find out how much 1,3-butadiene is in the LPG fuel, and whether this can be reduced to help reduce its ambient levels in the atmosphere.

6.8 Others

Last but not the least, the clear rise in formaldehyde (one of the top air toxics in our revised priority list) is believed to be related to the increased presence of MTBE as a fuel additive as a result of gradual phasing-out of leaded gasoline. MTBE is often added to gasoline as an octane enhancer substitute when the leaded compound is removed. A similar rise in formaldehyde was also observed in California after the introduction of MTBE in their fuel supply (Kirchstetter et al. 1999a,b and Koshland et. al. 1998). In Hong Kong, with the phasing-in of unleaded petrol and the reduction of benzene levels from 5% to 1%, MTBE is likely added into the gasoline as an octane enhancer substitute for the leaded compound and benzene removed. Wong et al. (2002) analyzed 26 gasoline samples of different brands of unleaded gasoline sold in Hong Kong from August to December 2000, and found that MTBE take up on average 6.8% of gasoline by weight, ranging from non-detectable to 15%. Unfortunately, no MTBE data is available for gasoline sold before 2000. As a result, the suggestion of increased MTBE in gasoline in recent years has yet to be verified. If this is actually the case, it should be considered as an unintended impact of the control measures. To better understand this issue and similar ones in the future, it is recommended that the HKEPD obtain more information from fuel manufacturers about fuel additives / octane enhancers in their products.

Along the same lines, because of concern for contamination of surface and ground water, use of MTBE as a fuel oxygenate is being phased out in California (Happel et al. 1998) and its use

⁹ Most of these LPG-related VOCs (e.g., propane, pentane, butane, iso-butane, 1-butene / iso-butylene, 1-butyne and 1-pentene) were not analyzed or below MDLs before 1999.

is declining in other states. As a replacement, increased use of ethanol as a fuel oxygenate is expected, and acetaldehyde levels are expected to increase as a result of such changes in fuel additives (Connecticut Department of Environmental Protection 2003). Information about ethanol and other fuel additive(s) is important for a better understanding of the impact of fuel changes, and it is also important for the HKEPD to continue monitoring to see whether there are changes in long-term averaged acetaldehyde and formaldehyde levels in the next few years.

6.9 Examples of TAP Control Programs in California

California has traditionally been very aggressive in its control of air pollutants. A brief summary of their control measures are outlined here for reference. For more details, one is referred to the Air Toxics control plan by the South Coast Air Quality Management District at <http://www.aqmd.gov/aqmp/atcp.html> and the California Airborne Toxics Control Measures at <http://www.arb.ca.gov/toxics/atcm.htm>.

6.9.1 Diesel Particulate Matter

Current regulation restricts the content of sulphur and total aromatic to be less than 500 ppm by weight and 10% by volume, respectively, in the diesel fuel used by on-road vehicles. Details of the “California Diesel Fuel Regulations” can be found at <http://www.arb.ca.gov/fuels/diesel/diesregs.pdf>.

California also has an extensive off-road control program aiming at the reduction of diesel as well as other emission from off-road sources, including (i) small spark-ignition off-road engines and equipment less than 25 horsepower (e.g., lawn and garden equipment, and other small industrial equipment), (ii) off-road recreational vehicles, (iii) off-road diesel engines and equipment, (iv) large off-road spark ignition (gasoline and LPG) engines and equipment 25 horsepower and greater (e.g., industrial equipment, forklifts, and portable generators), (v) airport ground support equipment, (vi) locomotives, (vii) commercial marine vessels, and (viii) recreational marine (e.g., personal water craft, ski boats, inboards, and outboards). Please refer to the CARB site at <http://www.arb.ca.gov/msprog/offroad/offroad.htm> for more details of their “Off-Road Mobile Sources Emission Reduction Program”.

Noting the dominance of diesel particulate matter in the contribution to both chronic and acute health risks for the Hong Kong population, we recommend the HKEPD look further into these control measures to try to reduce the particulate content in the atmosphere. Following the example in California, the HKEPD should look into on-road as well as off-road control measures. Moreover, marine vessels appear to be a significant source in contributing to elemental carbon variations over much of the Kowloon Peninsula (section 4.2). Hence, the HKEPD should confirm this preliminary result through source identification studies, and consider follow-up measures to limit marine emissions.

6.9.2 Benzene and Vapor Recovery

Current legislation requires the installation of the CARB-certified Phase I and II vapor recovery system on the stationary storage tank at the service station and used during the transfer. Information about the “Benzene Airborne Toxic Control Measures – Retail Service Stations” is available at <http://www.arb.ca.gov/toxics/atcm/benzatcm.htm>.

6.9.3 Tetrachloroethene

Current regulations are related mainly to dry cleaning services, and include operation and maintenance requirements, such as refrigerated condenser temperatures, the concentration of tetrachloroethene recirculated at the end of the drying cycle should be less than or equal to

8,600 ppmv, vapor adsorption, desorption and ventilation. Details of the “Perchloroethylene Airborne Toxic Control Measures – Dry Cleaning Operations” are available at <http://www.arb.ca.gov/toxics/atcm/percatcm.htm>. In addition, a website is also set up at <http://www.arb.ca.gov/toxics/dryclean.htm> to help introduce the environmental training program on dry cleaning, and use of new facilities with dry cleaning machines equipped with secondary control systems.

6.9.4 Hexavalent Chromium

Current legislation regulates chromium electroplating or chromic acid anodizing tanks at facilities performing hard chromium electroplating, decorative chromium electroplating, or chromic acid anodizing. Details of their “Hexavalent Chromium Airborne Toxic Control measures for Chrome Plating and Chromic Acid Anodizing Operations” can be found at <http://www.arb.ca.gov/toxics/atcm/chroatcm.htm>.

6.9.5 Dioxins

Our recent web survey failed to find any active municipal waste incineration facility in California. For medical waste incinerators, different regulations apply to facilities based on the weight of waste incinerated per year. For example, for an incinerator processing more than 25 tons of waste in a year the regulation requires:

- The dioxin emissions have been reduced by 99% or more of the uncontrolled emissions; or have been reduced to 10ng or less per kilogram of waste burned.
- The operation conditions of the control equipment should attain the flue gas temperature at the outlet of the control equipment below 300°F. For a single chamber incinerator, the combustion chamber shall be maintained at more than 1800 (± 200) °F.

Details of the “Dioxins Airborne Toxic Control Measure--Medical Waste Incinerators” can be found at <http://www.arb.ca.gov/toxics/atcm/dioxatcm.htm>.

6.9.6 Chlorinated VOCs

The regulation prohibits the use by any car owner or automotive maintenance facility or repair facility of any automotive consumer products which contains more than 1% by weight of methylene chloride, perchloroethylene or trichloroethylene (which were often found in brake cleaners, carburetor or fuel-injection air intake cleaners, engine degreasers and general purpose degreasers.) Details of the “Chlorinated Toxic Air Contaminants Airborne Toxic Control Measures – Automotive Maintenance and Repair Activities” can be found at <http://www.arb.ca.gov/toxics/atcm/amratcm.htm>.

6.10 Tables for Chapter 6

Table 6-1 Air Pollution Control Programs in place related to air toxics after 1997

7 Review of the TAP Priority List and Recommendations for Future Monitoring Efforts

7.1 List of Priority Pollutants

Based on the screening level emissions inventory, the Eureka Report (1996) had categorized three groups of TAPs for priority consideration (Table 7-1). With the TAP measurements and subsequent risk analyses (Chapter 5), the list of priority TAPs has been revised as shown in Table 7-2. Different from the list in the Eureka Report, this list is a measurement-based priority list on the cancer risk estimates. This table is calculated based on a lifetime of 70 years' exposure to the mean concentration of corresponding TAPs measured at Tsuen Wan and Central / Western from Nov 2000 to October 2001. The exceptions are p,p'-DDD and p,p'-DDE, which were not monitored regularly at Tsuen Wan or Central / Western. The additional cancer cases for p,p'-DDD and p,p'-DDE are based on data obtained in an ad hoc POP study conducted at Tseung Kwan O, Sha Ling, and Nim Wan. For each TAP, the unit risk used to produce Table 7-2 is the highest unit risk from the California EPA, the US EPA and the WHO (for the WHO where a range of unit risk is given, the unit risk used is the mean of the upper and lower unit risks).

From our calculations, the increased cancer risks associated with first 34 TAPs in this list are estimated to be more than one in a million. In particular, the first 14 TAPs are associated with increased risks more than ten in a million, and the next 20 TAPs are associated with increased risks between one and ten in a million.

Further, ten out of the eleven TAPs (except lead) listed as Category I TAP in the Eureka Report are also identified in this new list of 34 priority TAPs, and an additional ten of them are originally identified as Category II TAPs in the Eureka Report (five of these ten are PAHs which were listed as a group in the Eureka Report; dioxins is listed as one TAP in the current study but 2,3,7,8-Tetrachlorodibenzofuran and 2,3,7,8-Tetrachlorodibenzodioxin were listed separately as Category I and II in the Eureka Report). In addition, fifteen new TAPs not listed in the Eureka Report are found to have ambient concentrations associated with more than one in a million risks for increased cancer cases. Within this group, the one with highest risk is carbon tetrachloride.

On the other hand, there are a number of TAPs listed in the Eureka report for which unit risk factors were unavailable from any of the databases, even though some were classified as possible human carcinogen or stronger (e.g. acrolein). Hence, they are also not included in our new risk-based priority list. These include many of the Category III compounds listed in the Eureka report.

7.2 Recommendations for Future Monitoring Efforts

The HKEPD has been monitoring many more TAPs than the ones listed in the Eureka Report (1996). This is prudent considering the report was compiled with emission estimates and not actual monitoring data. It is better to obtain as much information as possible to define the baseline TAP situation in Hong Kong. The fact that fifteen new TAPs not originally listed in the Eureka Report was identified as priority TAPs in this study show that the open-minded approach taken earlier was important to have the baseline conditions better defined.

Nevertheless, now that the baseline conditions are better understood through this review, it is an opportune time to consider redeployment of resources in more extensive and focused monitoring efforts for the TAPs found to be important in Hong Kong.

7.2.1 Monitoring Objectives – Routine Monitoring and Ad Hoc Studies

The objectives of monitoring selected toxic air pollutants (TAPs) in the ambient air of Hong Kong include (HKEPD 2001):

- (1) To determine representative concentrations in areas of high population densities, and hence the exposure of the broad mass of population to toxic air pollutants of concern and to assess the consequential risks to human health, as well as other environmental effects, resulting from this exposure.
- (2) To determine the general background as well as the highest concentrations expected to occur in the area covered by the monitoring network, and hence to determine the ranking of Hong Kong when compared to other metropolitan cities.
- (3) To provide data for policy formulation of environmental control strategies and legislation, air pollution control plans, etc.
- (4) To observe long term trends in ambient levels of toxic air pollutants.
- (5) To validate and provide input for atmospheric dispersion models.
- (6) To study and evaluate pollution interactions and patterns, including long-range atmospheric transport.

Currently, the TAP monitoring program in Hong Kong consists of regular monitoring at two urban sites (Tsuen Wan and Central/Western), as well as ad hoc studies like the dioxin monitoring exercise carried out by ENSR (HKEPD, 2002), the Tai Mo Shan POP study (Louie and Sin, 2003), and the 12-month PM study.

While ad hoc studies can be targeted for any particular focus of interest, it is useful to clearly define the monitoring objective of the regular monitoring program. This is important since the constraints and requirements for different monitoring objectives are quite different. For example, monitoring sites catered to the objective of tracking TAP movement (i.e., long-range transportation), are not necessary good choices for assessing population exposure.

With respect to the six objectives, the regular monitoring program with observation frequency of one daily samples once every 6 or 12 days is more suitable to cover the first four objectives, which are more related to the overall population exposure and longer term situation in Hong Kong. Moreover, for protection of public safety, we recommend that the regular monitoring network should still be designed mainly for population exposure and health-risk assessments studies. With a clearly defined set of objectives for the routine monitoring program, other monitoring objectives can be supplemented through ad hoc studies. In particular, such studies can also be conducted once every few years to help gauge the ambient concentrations of other emerging TAPs such as mutagens, endocrine disruptive agents, organo-mercury, etc. that can affect health. If and when the concentration levels of any of these pollutants are found high enough to warrant more frequent monitoring, they can then be moved into the routine monitoring program.

In addition, ad hoc studies can include items such as hot-spot identification, tracking of TAP movements, source profile identification, and monitoring of new TAP species of concern, etc. In particular, the current monitoring frequency is not suited for monitoring of episodic events, for detailed tracking of regional and long-range transport, or for identification of pollutant interactions which can often take place in time-scales much shorter than the 24-hour sampling period (i.e., the fifth and sixth objectives).

Moreover, we need to understand the contribution to the various TAP species by different sources for formulation of control policies (third objective). For such source identification work, the ambient TAP data from the regular monitoring program must be supplemented with

the detailed chemical profile of the various emission sources. Hence, ad hoc studies targeted on the getting of local and regional chemical signature profiles for various emission sources are recommended.

7.2.2 Addition of a TAP Monitoring Station at Yuen Long

The recommended focus of the regular monitoring program on population exposure and health-risk assessment (section 7.2.1) implies that the routine monitoring stations should be established in locations to help capture the mean and extreme TAP levels over Hong Kong. It appears that the current stations at Tsuen Wan and Central/Western may not satisfy this purpose.

Our analyses show that there are substantial variations for many TAP species over Hong Kong. In particular, concentrations of many TAPs are found to be significantly higher in the northwest and lower in the southeast (see Chapter 4). As a substantial portion of Hong Kong's population lives and/or works in the northwest region, it is prudent that the ambient TAP levels in that part of Hong Kong be better defined. Hence, we recommend the establishment of another routine TAP monitoring station with FSP measurements at Yuen Long.

7.2.3 TAP Monitoring in Other Airsheds

Even with another monitoring site at Yuen Long, analysis of the local topography and meteorology suggest that there would still be quite a number of areas in Hong Kong with substantial population not adequately covered. These include Sheung Shui / Tai Po (northern), Tung Chung (western), and Tseung Kwan O (eastern), where large pockets of the population are living in relatively isolated airsheds. The northerly and westerly transports of pollutants are particularly noteworthy at Sheung Sui and Tung Chung, respectively.

To accurately gauge the health risk associated with population exposure, a better definition of the spatial distribution of TAPs in these areas is very much needed. Instead of the establishment of routine monitoring stations, ad hoc and less sophisticated but still systematically and densely placed (i.e. level-1 saturation monitoring network, Chow et al. 2002) studies are recommended to help define the baseline TAP levels in these areas. This would help map out the TAP gradients across different airsheds over Hong Kong. This type of level-1 survey studies can be repeated once every few years to help track the gradual change of TAP levels over the different airsheds in Hong Kong as a result of changed emission characteristics of the Pearl River Delta region.

We understand that sizeable resources are needed to start a new routine TAP monitoring station at Yuen Long as well as additional ad hoc study in other airsheds. These may be difficult considering the current fiscal situation. We note that while TAP levels are typically slightly higher at Tsuen Wan than in Central / Western (except for tetrachloroethene), the variations for most TAPs are similar for these two sites, suggesting they are within the same airshed. It may be possible to reduce the sampling at one of these two sites without impacting our overall ability to monitor TAP levels over Hong Kong. In particular, the Tsuen Wan site has a much longer record of collocated FSP measurements, and should not be modified. Hence, we find that the Central/Western site is more suitable for sampling reduction or modification. For example, the monitoring at Central/Western could be reduced to monthly or quarterly samples, taken during dates with simultaneous measurements at Tsuen Wan. Such a monitoring schedule would allow continued comparison between TAP concentrations at the two sites, as well as the monitoring of the mean trends using TAP data at Tsuen Wan.

7.2.4 Sampling Parameters

The list of parameters monitored by the HKEPD is much longer than the ones recommended in the Eureka Report (1996). This is prudent considering the report was compiled with emission estimates and not actual monitoring data. However, with the baseline condition better understood through this review, it is a good time for the HKEPD to consider modifying the list of TAPs monitored in its routine monitoring effort – putting more focus on priority parameters and less effort on TAPs that are of low concentrations or of questionable uncertainties.

Diesel Particulate Matter, FSP and Priority TAPs Identified in This Study

Our review identified diesel particulate matter (PM) as the most important parameter contributing to the overall health risk of the population. As most of the diesel PM is in the fine mode, expansion of the current FSP network (e.g. extending it to Yuen Long) can aid in providing more detailed information for subsequent health risk-assessment in Hong Kong. Along the same lines, we recommend that for the list of priority TAPs identified in this review, monitoring efforts should be continued and even strengthened (if resources are available).

The establishment of a routine monitoring station at Yuen Long will help a lot in this respect. Further, our study finds that the concentration levels of some of the parameters on the list (e.g. dioxins) are often below the detection limit at Tsuen Wan and Central/Western (during the summer). To address this problem, we recommend that the HKEPD consider modification of the sampling strategy (see sampling frequency section below) or switching to an analytical laboratory with better detection limits.

Elemental Analyses of RSP

Although strictly speaking not part of the TAP program, the elemental analyses performed on RSP samples have proven very useful for the identification of marine-related pollution sources (see section 4.1). We recommend the continuation of these analyses for RSP samples, particularly for AQMSs such as Kwai Chung which has characteristics distinct from the other sites. On the other hand, considering the limited amount of resources available, it is probably not necessary to continue these elemental analyses for all of the AQMSs since not all of them have characteristics distinct from others (e.g. Sham Shui Po).

Acrolein, Acetone and Styrene

Coming back to the TAP parameters, we found that some of the analyses results seem to be questionable. These include acrolein, acetone and styrene. Problems with acrolein have been reported elsewhere (Vermont Air Pollution Control Division, 1998) and the USEPA recently issue an addendum stating that the TO-11 and TO-11A methods are not useful for determination of acrolein (USEPA 2002). Problems with acetone and styrene appear to be known to analytical chemists but are not well-documented in the literature (Moore M, personal communications). In particular, the problem with detecting acetone is related to a low collection efficiency of the standard method while the problem with styrene is related to its high reactivity. Both of these complications can lead to analytical results not representative of ambient levels. Analyses of these pollutants should be stopped or at least flagged in the data base until better detection methods can be developed here or elsewhere.

Other TAPs with Concentrations That Are Near or Below Detection Limits

In particular, our review finds that the concentration levels of a number of TAPs are most at or below method detection limits. For example, nine of the 17 carbonyl species reported were routinely below their MDLs due to low concentrations in the ambient environment. Whether or

not to continue the analyses for these TAPs that are of low ambient concentrations should depend on whether additional resources are needed for the analyses and reporting of these TAP species. If no additional cost / resources are required, then they should be continued to allow for monitoring of any unexpected increase in their concentration levels.

The no cost scenario is possible since current analytical method allow simultaneous determination of multiple compounds. For example, the process for determining the level of one carbonyl compound needed in the TAP program (e.g. formaldehyde) can, in the same run, provide concentration levels for the other 16 carbonyl compounds. Although these compounds can be measured simultaneously, different analytical laboratories can have different charging schemes – some may charge them all together as a group while some may charge by the number of individual species requested. For the former case, there is no advantage in not getting the concentrations for the other compounds, but there could be savings if the charging scheme is per TAP species rather than per analysis.

If additional cost is required for the measurement of TAPs that are often at or below method detection limits, we recommend that they should be eliminated from the routine monitoring effort. The resources saved can then be used within the TAP program to help with the establishment of a new routine monitoring station at Yuen Long (7.2.2), or for ad hoc studies as outlined in section 7.2.3. After the elimination of these parameters in the routine monitoring effort, it is prudent that their ambient levels can still be monitored once every few years (e.g., every 5 years and preferably using methods with lower detection limits) as part of an ad hoc baseline review study to make sure that any unexpected and systematic increase in their levels would not remain unnoticed.

Similarly, following the overall theme in our recommendation of using the routine monitoring network for exposure and health assessment, and ad hoc studies for other objectives, we also recommend that ad hoc studies be conducted once every few years to help gauge the ambient concentrations of other emerging TAPs such as mutagens, endocrine disruptive agents, organo-mercury, etc. that can affect health. If and when the concentration levels of any of these pollutants are found to be high enough to warrant more frequent monitoring, they can then be moved into the routine monitoring program.

7.2.5 Sampling Issues

Currently, the samples are collected over a 24 hour period once every six or twelve days. The concentration levels of many of the TAPs, even some on the priority list (e.g. dioxins), are below or near detection limits. As most of the risk assessments are based on long-term averages, ways to improve the detection limit (e.g. identification of laboratories which can perform the chemical analyses with lower method detection limits) will certainly help. However, that may not be easy and hence we recommend looking into a modification of sampling strategies to allow for detection at current method detection limits.

7.2.6 Management of Health Data and Establishment of a Health-Related Database

In a separate study on air pollution and health outcomes conducted by the CUHK, problems with the quality of the health outcome data were identified (Wong TW and Yu TS, personal communications). Mortality data, obtained from Census and Statistics Department via the HKEPD, was incomplete in terms of the date of death, although this is an important estimate of health outcome. Similarly, missing data in Tertiary Planning Units (TPUs) was commonly found in the mortality data. As a result, daily time series cannot be performed. Data from the Hospital Authority could not be easily divided into districts according to the residential address

of the patients, which hinders district-based (i.e., health outcome data matched with the corresponding air pollutant data) analysis.

We recommend the use of TPUs or at least, well-defined districts comparable to the air pollutant data, for district coding of hospital patients, and the mandatory entry of data of death and TPU of the deceased to facilitate time-series and spatial analysis. A better monitoring network for FSP is important for improved definition of local population exposures. Moreover, the availability of high quality mortality and hospital data, through the establishment of an online health and environmental database, would enable an accurate risk estimate using the time series and spatial approach. In particular, data (with personal information removed) must be routinely updated into this online health and environmental database, and be readily accessible by researchers in Hong Kong. A stable and open access health information system similar to the current air pollution database would go a long way in helping the understanding of environmental and public health issues in Hong Kong.

7.2.7 Control Policy

Several TAPs (diesel particulate matter, NO₂, O₃, and formaldehyde) were found to have exceeded the reference values in some measurements. Superficially, the effects appear to be relatively minor (respiratory tract and eye irritation). In reality, the health consequences on respiratory health are substantial, as damage to the respiratory system is documented in most of the TAPs – the criteria pollutants, carbonyls, and some VOCs and metals, and the effect on health is at least additive. (The effects of different TAPs may be synergistic, which is not unlikely.)

The level of NO₂ is of particular concern. The short- and long-term exposures to criteria pollutants (including RSP) have been shown to be associated with increased mortalities and respiratory diseases in local studies (Wong et al. 2002a, 2002b). The relative risks of NO₂ and O₃ are especially high. Although overseas studies have focused on RSP, and recently, FSP, NO₂ is highly correlated with the former, and might have accounted for much of the short-term health effects. Besides, these chemicals have other long-term effects – diesel and formaldehyde are carcinogenic. Stringent control of these TAPs should bring about considerable benefits to the health of the population.

We recommend that a high priority be accorded to control those TAPs (in particular, NO₂, diesel fumes and formaldehyde) exceeding the acute and chronic reference exposure levels (RELs).

7.2.8 Impact Related to Future Development

This is the first systematic study in Hong Kong on TAP monitoring and health risk assessment. Notwithstanding the above limitations, the impacts of TAPs on community health in terms of cancer risk and non-cancer risk have been estimated. Results of this study set a baseline health risk estimate for the general population, against which different scenarios of risks can be worked out based on different pollution profiles in specific districts / areas. The impact on health with projected increases or reductions of emissions of various TAPs with respect to future development in and around Hong Kong is beyond the scope of the current study. However, it is an important issue that needs to be assessed with proper trend analyses. Such results shall also provide useful information on a health-based priority in the control of TAPs, which must be considered together with other environmental, technological, social and economic concerns.

7.3 Tables for Chapter 7

Table 7-1 Emission based Priority List of Toxic Air Pollutants in Eureka Report

Table 7-2 Measurement based Priority List of Toxic Air Pollutants from this study

8 References

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