PORT PHILLIP BAY ENVIRONMENTAL STUDY

LITERATURE REVIEW OF AEOLIAN AND ATMOSPHERIC INPUTS OF TOXICANTS TO PORT PHILLIP BAY

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ACRONYMS

BaP	Benzo(a)Pyrene
CSIRO	Commonwealth Scientific and Industrial Research Organisation
EPAV	Environment Protection Authority of Victoria
GPO	General Post Office
NMHC	Non-Methane Hydrocarbons
PAHs	Polycyclic Aromatic Hydrocarbons
Pb	Lead
PPB	Port Phillip Bay
PPCR	Port Phillip Control Region
μm	10^{-6} metre
ng	10^{-9} gram
VOC	Volatile Organic Compounds

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1. INTRODUCTION

The atmosphere is a major pathway for the transport and deposition of natural and anthropogenic toxicants. Toxicants such as polycyclic aromatic hydrocarbons (PAHs), lead compounds and petroleum based hydrocarbons emitted to air in the urban and semi-rural areas around Port Phillip Bay (PPB) may be deposited via wet and dry deposition processes.

Aerial fluxes of toxicants have been reported to contribute a major share of pollutant loadings to the Great Lakes basin and marine waters (Bidleman, 1988). The sign)ficance of the atmospheric input of toxicants to PPB is not known.

The purpose of this Task is to critically review the available data on the atmospheric input of toxicants such as PAHs, trace metals such as lead (Pb) and petroleum based hydrocarbons to PPB. In the absence of local data, our approach will be to consider scientific work carried out elsewhere to address the general issue of toxicant deposition to aquatic systems. Given the complexity of determining the deposition of airborne matter to surface waters, we shall refrain from making "back-of-the-envelope" calculations in this work. However, the availability of local data potentially useful for future estimates of toxicant inputs will be identified.

2. LITERATURE SEARCH

Initial contact was made with various members of staff within EPAV, CSIRO Division of Atmospheric Research, Melbourne Water and Marine Science Laboratories in order to ascertain the current state of knowledge and availability of data (internally or otherwise) for this Task. Staff contacted include the following:

EPAV
John Kowarsky
Graham Rooney
David May
Trevor Bardsley

CSIRO
Ian Galbally
Greg Ayers
Rob Gillett
John Gras

MELBOURNE WATER
Tony Chiffings
Richard Peck
Malcolm Downes

MARINE SCIENCE LABORATORIES
Andrew Longmore
Rob Caddell

A formal literature search was subsequently conducted through the EPAV and Planning Library using *Dialog* and *Australis* Online Services. References to journal articles and reports were reviewed from *Chemical Abstracts, Enviroline,* and *National Technical Information Services (NTIS)* via the international system - *Dialog*. *Streamline* was accessed via the Australian equivalent -*Australis*.

The Victorian Institute of Marine Sciences has recently conducted a literature search on Port Phillip Bay on behalf of Melbourne Water. Access to the literature database may be obtained from Dr. Tony Chiffings of Melbourne Water.

The key words used in the computer literature search to obtain data on aeolian and atmospheric sources of toxicants to PPB included:

* Port Phillip Bay

* Bay(s)

* Atmospheric

* Deposition

* Rainfall

* Coasts/Coastal

* Toxicants

* Aeolian

* Wind-blown

* Dust

* Aerosols

* Lead

* PAHs

* Petroleum

* Mercury

* Asbestos

* Deposition velocities

* Combinations of above key words.

Our literature review on aeolian and atmospheric inputs of toxicants such as PAHs, lead and petroleum based hydrocarbons to PPB has highlighted the general lack of data on the subject.

3. ATMOSPHERIC SOURCES

In a recent inventory of emissions to air for the Port Phillip Control Region (PPCR), Carnovale et al. (1991) have estimated that on an annual basis, $1.7x10^5$ tonnes of volatile organic compounds (VOC), $5.0x10^2$ tonnes of lead (motor vehicle contribution only, representing approximately 90% of total PPCR lead emission) and $2.7x10^4$ tonnes of particulates were emitted in 1990.

The PPCR inventory considered emissions from motor vehicles, industrial, commercial and domestic sources of air pollution on an hourly, seasonal and regional basis (3 km by 3 km grid resolution).

PAHs are primary products of incomplete combustion of organic material such as fossil fuels and vegetation. PAHs are also emitted from industries involved in coal gas)fication and liquefaction processes, and the production of coke, asphalt and carbon black. PAHs are ubiquitous in the environment (Hites et al., 1980) with elevated concentrations being reported near urban areas (Wakeham et al., 1980). PAHs in the atmosphere are associated with fine particles, with most of the PAHs mass residing in submicron particles (Miguel and Rubenich, 1979). Benzo(a)pyrene (BaP) is one of the more studied PAHs because of its carcinogenicity (Kaufman, 1988). PAHs were not inventoried specifically but are included in estimates of VOC emitted to PPCR.

Lead is known to exist in air as vapour phase tetraalkyllead compounds derived from gasoline additives, typically accounting for about 2-8% of total lead in urban areas (Birch et al., 1980; Harrison, 1986). Approximately 90% of lead emitted in Melbourne originates from motor vehicle exhaust. The major lead compound emitted in vehicle exhaust is PbBrC1 with lesser amounts of 2PbBrCl.NH_4Cl and PbBrCl.2NH_4C1 (Habib), 1973). In the atmosphere, atmospheric reaction of PbBrCl with sulphates leads to the formation of $PbSO_4(NH_4)2SO_4$, the most commonly observed lead compound in the work of Biggins and Harrison (1979). In future analysis of lead emitted to PPB it is important to consider the inorganic versus organic components because the former are generally more toxic (Won" et al., 1978).

Individual petroleum hydrocarbons were identified in EPAV's inventory but are not reported as such (nor collectively under this category) in the report by Carnovale et al. (1991). Petroleum hydrocarbons were found to make the overwhelming contribution to VOC emission in PPCR.

4. **DEPOSITION**

Gaseous and particulate matter may be removed from the atmosphere by wet or dry deposition mechanisms. Wet deposition involves the scavenging of matter by water droplets in clouds or precipitation (rain, fog or snow). Dry deposition involves the uptake of matter at the earth's surface (water, soil or vegetation) and may include such processes as gaseous adsorption, particulate fallout and aerosol impaction.

Dry deposition is usually referred to in terms of a dry deposition velocity, Vd:

$$Vd = Fluxi / Ci$$

where Fluxi is the deposition flux of species i and
Ci is the corresponding atmospheric concentration.

Deposition velocities are very sensitive to the type and state of surfaces and are also dependent on meteorological conditions. Rates of dry deposition of gases to surface water vary by orders of magnitude depending on the concentration of the gas in the surface water and on the solubility and aqueous-phase reaction kinetics of the depositing gas (Schwartz, 1992). For nonreactive gases the key property is the Henry's law solubility. For reactive gases the deposition flux can be quantitatively related to the chemical reaction rate for known mass-transport parameters and solubility and kinetic coefficients for the depositing gas.

In considering the atmospheric input of toxicants which may form aerosols under normal conditions, the size distribution of particles needs to be considered. Dry (and wet) removal of particles is strongly influenced by size. Theoretical calculations and wind tunnel investigations of aerosols show that Vd is minimal for particles between approximately 0.1 μm (10^{-6} metre) and 2 μm (so called accumulation mode particles produced by gas-to-particle conversion and by coagulation of Aitken nuclei). Deposition velocities are higher for smaller particles because of Brownian motion and for larger particles because of gravitational settling and turbulent impaction (Bidleman, 1988; Slinn, 1983). Giorgi (1986) predicted mass-average Vd over the continents and oceans ranging between $3x10^{-5}$ and $3.6x10^{-4}$ m/s for accumulation-mode particles. The range for coarse-mode particles (diameter > 2 μm) was reported to be two orders of magnitude higher, $5x10^{-3}$-$2.5x10^{-2}$ m/s.

5. POLYCYCLIC AROMATIC HYDROCARBONS

We have not found any estimate of the atmospheric input of PAHs to PPB.

In the absence of local data, our approach is to identify the relevant literature data that may assist in firstly obtaining a rough indication of the likely significance of atmospheric inputs of PAHs to PPB, and secondly provide an overview of scientific programs carried out elsewhere in order to quantify the atmospheric input of PAHs.

A number of studies have been undertaken in Australia to experimentally determine ambient concentrations of PAHs over the last twenty years. Pradhan (1986) has reported ambient measurements of PAHs in Sydney and Port Kembla. Average monthly BaP concentrations of up to 4 ng/m^3 and 5 ng/m^3 respectively, were reported for commercial and residential areas of Sydney. Levels were found to be approximately a factor of two lower in Port Kembla.

Freeman (1987) conducted monthly monitoring of PAHs within 6 kilometres of the Sydney GPO during 1985. The annual average of total PAHs was found to lie in the range 11-16 ng/m^3, with levels being highest during winter (up to 34 ng/m^3 in June). Work conducted in Melbourne in 1974 (Mainwaring and Stirling, 1981) revealed winter BaP concentrations of around 1 ng/m^3. More recently, Lyall and Hooper (1988) have conducted measurements of PAHs in the Latrobe Valley. BaP concentrations were found to reflect the domestic and commercial activity in the immediate vicinity of the sampling site. At urban locations in Moe, Morwell and Taralgon, the peak winter averages of BaP ranged from 0.35 to 0.87 ng/m^3. Large seasonal variation in BaP was recorded by Lyall and Hooper, with winter levels being 210 times higher than summer levels.

EPAV has recently undertaken a monitoring study of respirable particles, lead and PAHs in Flemington, approximately 4 kilometres from Melbourne GPO (Bardsley, 1992). Levels of PAHs in particulate matter were found to average 2.1 ng/m^3 (annual average). Ten PAHs were individually reported by Bardsley, these include benzo(ghi)perylene (annual average 0.58 ng/m^3), indeno(l,2,3-cd)pyrene (0.32 ng/m^3) and benzo(a)pyrene (0.17 ng/m^3). Non particulate PAHs were not monitored.

Average winter BaP concentrations at urban and industrial locations in the USA and Canada typically have been found to be in the range 1-4 ng/m^3 (Gordon, 1976; Katz and Pierce, 1975), i.e. levels comparable to those reported for Sydney but higher than levels found in Melbourne in 1990-1991.

PAHs emitted from traffic related sources (e.g. exhaust, asphalt and tyre debris, oils) have been reported to essentially remain localised within the immediate neighbourhood of their emission (Gjessing et al., 1984; Johnston and Harrison, 1984; Harrison and Johnston, 1985; Hewitt and Rashed, 1990). For example, Hewitt and Rashed report that PAH deposition, 1990). For example, Hewitt and Rashed report that PAH deposition rates at distances beyond approximately 50 metres from major roads is comparable to the regional background.

The atmospheric fallout rate of PAHs has been measured at a remote site approximately 30-40 km from Tokyo. Deposition of 28-56 μg /m^2 per month PAHs was measured over a three month sampling period (Takada et al., 1991). PAHs in street dust were found to lie in the range 1.44.9 μg/g for urban roads carrying typically 30-60,000 vehicles per day$^-$ (Takada et al., 1991). Considerably higher PAHs concentrations were found in dust within a Tokyo expressway tunnel (up to 26 μg/g; traffic volume of 18,000 vehicles per day).

Particulate and gas phase sampling of PAHs has been conducted by Masclet et al. (1988). Particulate concentrations at a remote site in the Mediterranean Sea were reported to average 1.6 ng/m^3 whilst gaseous concentrations averaged 45.5 ng/m^3. The particulate phase consisted mainly of compounds with four rings or more. The principal gas phase PAHs were found to be naphthalenes and anthracenes.

Mc Veety (1986) employed a mass balance method to estimate dry deposition of PAHs to Lake Siskewit. When the measured precipitation flux of PAHs was balanced against sedimentation and advective flow from the water column, the shortfall was assumed to be caused by dry deposition. A dry deposition velocity of $8x10^{-3}$-$1.0x10^{-2}$ m/s was estimated for several PAHs.

5.1 SEASONAL ATMOSPHERIC INPUT OF POLYCYCLIC AROMATIC HYDROCARBONS

The atmospheric input of PAHs to PPB is expected to vary with season. Emission of VOC into the atmosphere of PPCR (from anthropogenic sources in 1990) has been estimated to increase from approximately 430 tonnes in a typical summer week day (Carnovale et al., 1991) to 537 tonnes in a typical winter week day. The higher winter week day emission of VOC is primarily due to wood combustion in residential stoves and fireplaces. Wood combustion is a well known source of PAHs.

The seasonal variation in wet and dry deposition of PAHs to PPB cannot be reliably inferred directly from available data.

6. LEAD AND OTHER TRACE METALS

We have not found any published estimate of the atmospheric input of lead or other trace metals to PPB.

EPAV is currently reviewing the toxicant status of PPB (Roberts, 1991). The atmospheric input of lead, zinc, cadmium, copper, chromium and nickel to PPB is being considered on the basis of literature data on the ratio of atmospheric to stormwater contamination for an literature data on the ratio of atmospheric to stormwater contamination for an urban area in Sweden (Malmquist, 1978). Malmquist reported that 7-40% of the metal contamination in stormwater is due to atmospheric deposition.

EPAV currently monitors the 3-monthly moving average concentration of lead at five sites around Melbourne (EPAV, 1990). In 1990, the highest 3-monthly average concentration of lead (1 μg/m^3) was recorded at EPAV's Collingwood monitoring site (within 5 km of Melbourne GPO). The emission of lead from motor vehicle exhaust in PPCR is predicted to decrease from approximately 1.4 tonnes per day in 1990 to 0.8 and 0.2 tonne per day by 1995 and 2000, respectively (Carnovale et al., 1991).

Toxic contaminants in the Great Lakes of North America have received considerable scientific attention over the last thirty years. A wealth of information on the atmospheric input of toxic metals and organics to aquatic environments has come from numerous investigations of the Great Lakes. In relation to atmospheric inputs of trace metals, Gatz (1975) used estimates of metal concentrations in the atmosphere of Chicago and northwest Indiana to assess wet and dry deposition to Lake Michigan. The atmospheric flux of metals was calculated using air quality data from shoreline locations in conjunction with a dispersion-deposition model which incorporated windspeed, mixing heights and elemental particle size effects on deposition rates (Schmidt and Andren, 1984).

Other studies (ACS, 1975; ACS 1977) used similar methodology to Gatz to estimate the atmospheric loading of metals to all the Great Lakes. ACS however, used an overall deposition velocity for all particles without allowance for the variation in particle size affiliation for metals. The effect of using a single deposition velocity for all metals is to underestimate deposition for elements having a high mass median diameter (e.g. aluminium) whilst overestimating deposition for metals having lower mass median diameter (e.g. lead).

Wet and dry deposition collection vessels have probably been most widely used in assessing trace metal atmospheric deposition rates (Schmidt and Andren, 1984). Several large and smallscale deposition networks have been designed and implemented for the purpose of evaluating deposition rates on each of the Great Lakes (Shiomi and Kuntz, 1973; ACS, 1975, 1977; Allen and Halley, 1980). Most stations used in these studies were land based, however several buoy stations have been used. Buoys were generally located within 10 kilometres from shore and their numbers were low compared to the total number of sites. The atmospheric deposition of cadmium, chromium, cobalt, copper, nickel, zinc and lead to the Great Lakes has been reported, with lead receiving significant attention (Schmidt and Andren, 1984).

The atmospheric metal input rates to the Great Lakes have been reviewed by Schmidt and Andren (1984). They conclude that wide disparity exists in estimated deposition rates, even for the same deposition regions, however the following ranges should bracket actual metal inputs (in kg/ha per year): cadmium, 0.005-0.01; cobalt and chromium, 0.002-0.7; copper, 0.05-0.15; nickel, 0.01-0.05; lead, 0.1-0.25; and zinc, 0.5-1.0. Schmidt and Andren further note that given the difficulties in sampling logistics, a variety of approaches should the difficulties in sampling logistics, a variety of approaches should be employed to estimate atmospheric loadings. The chemical composition of precipitation and aerosols should be measured with as fine a time resolution as possible, at standard heights, with standardised equipment and with supporting meteorological data (windspeed, direction and atmospheric stability). Schmidt and Andren further recommend that mass balance calculations, using elemental data on river and shoreline erosion inputs in combination with sedimentation patterns, also be vigorously pursued.

An assessment of the relevant importance of the atmospheric input of metals to Lake Michigan has revealed that the atmosphere is the source of 60% of total lead input, 33% of zinc, 13% of copper and 11 % of cadmium and manganese. The atmosphere was found to be a minor source of aluminium, iron and cobalt to T eke Michigan (Salomons, 1986). Sievering et al. (1981) compared the dry deposition loading with wet precipitation and runoff for Lake Michigan. They found that the atmospheric input by dry deposition contributed 75 % of the total loading for lead and 50% of the total loading for zinc to the lake ecosystem.

The relative importance of atmospheric trace metal inputs to coastal zones such as those bounding the North Sea has been reported (Salomons, 1986; Van Alst et al., 1983). For the North Sea, a direct comparison of riverine inputs shows that the atmospheric flux is equal to the amount of trace metals transported by rivers. However, if one takes into account the amounts of trace metals retained in estuaries, the atmospheric input is actually higher than the riverine input.

In a review of trace metal inputs to the hydrosphere, Salomons (1986) concludes that although the atmosphere is recognised as an important source for contaminants in coastal waters, its relative importance is difficult to assess because retention processes in estuaries are not well understood and the role of bottom deposits as a source of trace metals has only recently been recognised.

6.1 SEASONAL ATMOSPHERIC INPUT OF LEAD AND OTHER TRACE METALS

The emission of lead to air in PPCR is approximately constant over the four seasons. However, deposition of lead to PPB is expected to vary with season.

The seasonal variation in the emission of other trace metals to PPCR and their subsequent deposition to PPB is unknown.

7. PETROLEUM BASED HYDROCARBONS

We have not found any estimate of the atmospheric input of petroleum based hydrocarbons to PPB.

Petroleum based hydrocarbons emitted (from fuel combustion and fuel handling) around PPB consist of approximately 50% C2-C10 alkanes, 12% C2-C6 alkenes, 10% toluene, 9% miscellaneous organics and alkanes, 7% xylene, 5% benzene, 2% carbonyls (consisting of 40% formaldehyde, 15 % acetaldehyde and 45 % other aldehydes and ketones) and 5 % acetylene (Carnovale et al., 1991; Galbally et al., 1989). The above compounds have low water solubilities (with the exception of carbonyls) and high volatility and consequently are expected to exist in the gas phase at normal temperatures.

EPAV routinely monitors the total non-methane hydrocarbon (NMHC) concentration at four sites around Melbourne (EPAV, 1990). In 1990, the average 1-hour NMHC concentration in Melbourne was measured to be 0.4 parts per million Carbon.

The dissolution of organic gases into rain and cloud droplets can be determined by the Henry's law constant (H) which represents an air-water partition coefficient calculated from the ratio of vapour pressure to water solubility (Bidleman, 1988). If H is sufficiently high, gas dissolution in droplets is negligible. This has been reported to be the case for n-alkanes (Ligocki et al., 1985). Furthermore, Velinsky et al. (1986) report that total hydrocarbons in wet deposition in the Chesapeake Bay area account for less than 0.5 % of the total organic carbon concentration.

The dry deposition of petroleum based hydrocarbons to PPB may be determined from data and mathematical models available to EPAV. The atmospheric flux of individual petroleum based hydrocarbons, e.g. benzene, toluene and xylene, to PPB may be calculated using EPAV's emissions inventory in conjunction with a dispersion-deposition model which incorporates windspeed, mixing heights and deposition velocities.

7.1 SEASONAL ATMOSPHERIC INPUT OF PETROLEUM BASED HYDROCARBONS

The emission of petroleum based hydrocarbons to air in PPCR is approximately constant over the four seasons, however deposition to PPB is expected to vary with season.

The seasonal variation in the wet and dry deposition of petroleum based hydrocarbons to PPB is unknown.

8. SUMMARY

We present a summary of the main points of the above critical review on the aeolian and atmospheric sources of toxicants to PPB:

i. Data on the atmospheric input of toxicants such as PAHs, lead, other trace metals and petroleum based hydrocarbons to PPB is not currently available.

ii. From work conducted overseas, the atmosphere has been found to contribute significantly to the toxicant loading of lakes and marine zones. In some cases, the atmosphere has been estimated to contribute more to the toxicant loading of lakes and marine zones than rivers and streams.

iii. The data currently available on PPB and surrounding region is not sufficient to allow the satisfactory estimate of the atmospheric inputs of toxicants to PPB.

iv. In order to acquire the data necessary to calculate the atmospheric input of toxicants to PPB there is a clear need to initiate experimental sampling programs on the composition of ambient air (including aerosols) in the Bay region in addition to the collection and analysis of precipitation.

v. Data is available to EPAV which may be employed to estimate the dry deposition of petroleum based hydrocarbons to PPB. The data is in the form of emission rates spatially and temporally resolved which if coupled with atmospheric dispersion models, windfields and literature deposition rates will permit the estimation of dry deposited hydrocarbons to the Bay and its associated water catchment area. Similar data is available for lead and PAHs.

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