PORT PHILLIP BAY ENVIRONMENTAL STUDY

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

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ACRONYMS

CSIRO	Commonwealth Scientific and Industrial Research Organisation
DOC	Dissolved Organic Carbon
EPAV	Environment Protection Authority of Victoria
HNO2	Nitrous Acid
HNO3	Nitric Acid
NDDN	National Dry Deposition Program (USA)
NH3	Ammonia
NH4 +	Ammonium Ion
NH4NO3	Ammonium Nitrate
(NH4)2SO4	Ammonium Sulfate
NOx	Oxides of Nitrogen (only NO and NO2 considered)
N2O	Nitrous Oxide
NO2-	Nitrite Ion
NO3-	Nitrate Ion
PM10	Particles having a diameter greater than 10 um
PPB	Port Phillip Bay
PPCR	Port Phillip Control Region
TOC	Total Organic Carbon
TSP	Total Suspended Particulates
VOC	Volatile Organic Compounds
umole	10-6 mole

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1. INTRODUCTION

Increased nutrient loadings from rivers and sewage treatment sites coupled with direct discharges from coastal sources have caused detrimental eutrophication effects in a number of coastal marine ecosystems (Hinge et al., 1991; Fischer and Oppenheimer, 1991).

The effects of growing nutrient loads in coastal waters may include an increase in the frequency of algal blooms, increased length and severity of low oxygen conditions, and episodic death and long-term decline of marine organisms (Fisher and Oppenheimer, 1991).

It is becoming increasingly evident that atmospheric inputs of nutrients to estuaries and coastal waters can contribute sign)ficantly to the total nutrient load of regions similar to Port Phillip Bay (PPB). Omission of the atmospheric inputs of nutrients to estuaries and directly over the Bay may not only lead to a sign)ficant underestimation of nutrient loads (particularly carbon and nitrogen) but also reduces the effectiveness of any subsequent Bay Management Plans which only consider point sources (e.g. sewage treatment plants, rivers and streams) and agricultural sources.

A clear example of the importance of the atmospheric inputs of nutrients to coastal waters has recently been reported by Fisher and Oppenheimer (1991). Fisher and Oppenheimer (1991) estimated that approximately 25% of anthropogenic nitrogen loading to Chesapeake Bay in north-eastern USA originates as atmospheric nitrate deposition (over Bay and water catchment area), arising almost entirely from anthropogenic emissions of oxides of nitrogen. Atmospheric ammonium deposition was estimated to contribute a further 14% of the total nitrogen load.

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 nutrients to PPB included:
* Port Phillip Bay

* Bay(s)

* Atmospheric

* Deposition

* Coasts/Coastal

* Nutrients

* Aeolian

* Wind-blown

* Dust

* Nitrogen

* Carbon

* Phosphorus

* Deposition velocities

* Combinations of above key words.

Our literature review on aeolian and atmospheric inputs of carbon, nitrogen and phosphorus to Port Phillip Bay has highlighted the general lack of data on the subject, particularly in regard to carbon and phosphorus. The atmospheric input of nitrogenous species directly over PPB was investigated by Carnovale from fundamental principles in 1987 and later revised by Carnovale and Saunders (1988) using rainfall data from five suburban sampling sites around Melbourne.

3. ATMOSPHERIC SOURCES OF NUTRIENTS

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, 1.7x103 tonnes of volatile organic compounds (VOC), 7.9x104 tonnes of oxides of nitrogen (NOX) and 2.7x104 tonnes of particulates were emitted by anthropogenic sources 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). For a typical summer day, motor vehicles were estimated to account for 50% of VOC, 78% of NOX and 46% of particulates. A summary of the percentage contribution to PPCR anthropogenic emissions is presented in Figure 1.

Phosphorus was not inventoried, but is known to have no significant gas-phase component. Phosphorus is found in dust, microbial debris (Fuller, 1972) and fertilised soil (Morelli et al., 1984) and in years of extreme meteorological conditions (hot, dry and windy) considerable amounts of aerosol or dust-bound phosphorus may be blown into PPB from as far away regions as Western Victoria (Mitchell, 1983).

The seasonal variation of VOC, NOX and particulate emissions to air in PPCR is reproduced in Table 1.

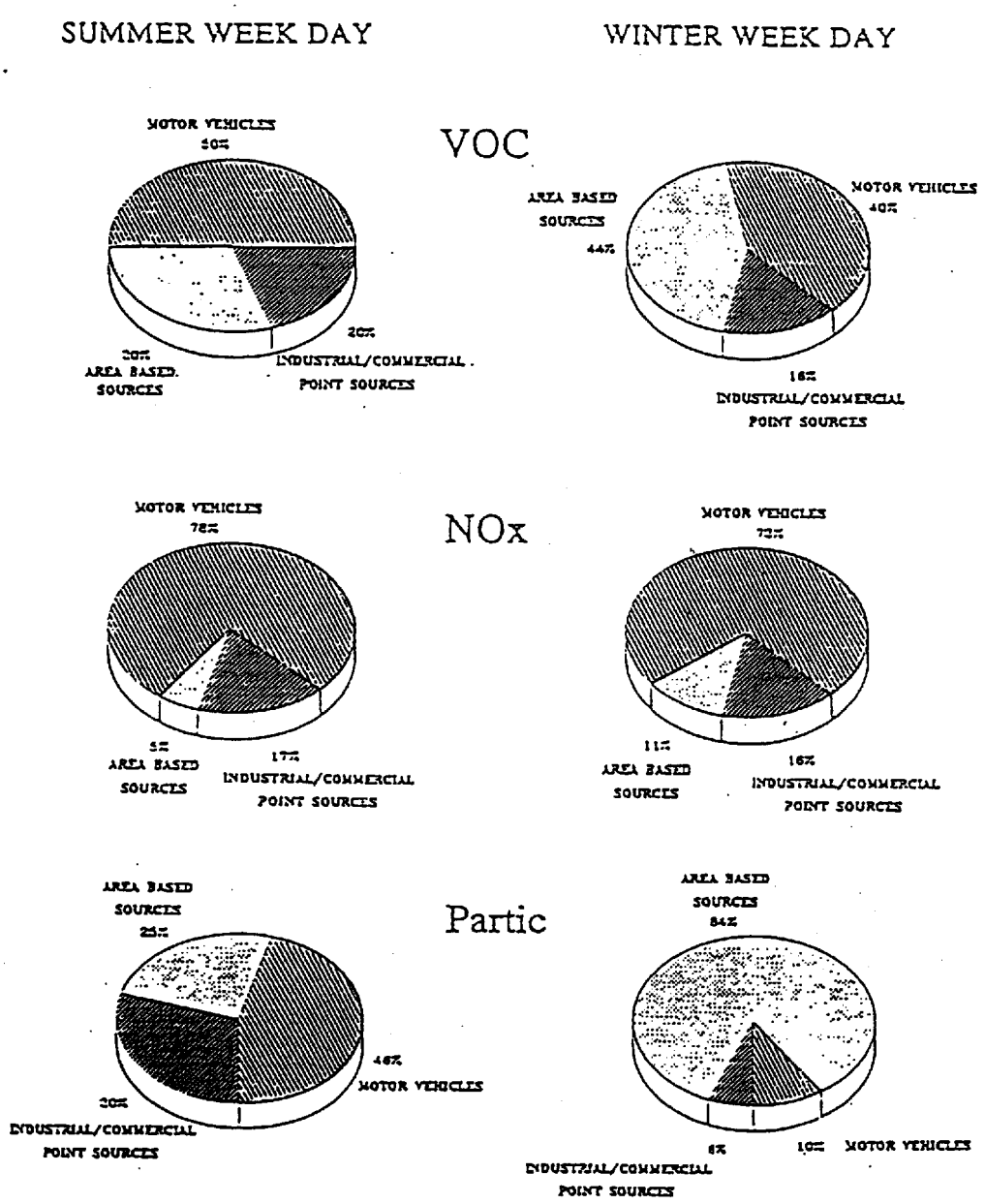

Figure 1. Percentage contribution to PPCR anthropogenic emissions to air, 1990. Reproduced from Carnovale et al., 1991.

Table 1. Summer and winter week day emissions of VOC, NOX and particulates for PPCR, 1990 (all anthropogenic sources considered). Reproduced from Carnovale et al., 1991.

	SUMMER WEEK DAY Tonnes	WINTER WEEK DAY Tonnes
VOC	430	537
NOx	220	234
PARTICULATES	26	125

4. DEPOSITION

All airborne matter may be removed from the atmosphere by wet or dry deposition mechanisms. Wet deposition involves the scavenging of gaseous or particulate matter by water droplets in clouds or by precipitation (rain, fog or snow). Dry deposition involves the uptake of gaseous or particulate 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 non-reactive 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.

Wet deposition represents an efficient atmospheric cleansing mechanism, particularly for water soluble gases, but occurs for a small fraction of the time. Dry deposition is generally less efficient than wet deposition (per unit time) but is continuous. On an annual basis, dry deposition may be comparable in importance to wet deposition.

Deposition of airborne matter to PPB is dependent on the emission and dispersal characteristics of the region. The synoptic and mesoscale winds around PPB in combination result in complex dispersal patterns varying with time and locality. The winds in Melbourne are indicative of the synoptic pattern over the whole Bay. Sea breezes and east-west funnelling effects however, lead to wind variations as one moves in an arc extending from the south west to south east of Melbourne. In the mornings, northerlies prevail for most of the year, except in summer when southerlies are the prevailing winds. In the afternoons, southerlies predominate, except in winter when the wind is mostly from the north (Beer, 1992).

Given the emission sources and wind patterns around PPB, most of the nutrients deposited to PPB are expected to originate from the PPCR.

5. CARBON

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

The atmospheric load of VOC around PPB has been considered in substantial detail by EPAV (Carnovale et al., 1991). Emissions of VOC from anthropogenic and natural sources have been estimated around the Bay in a manner that lends itself to the evaluation of atmospheric dry deposition using EPAV's emission inventory, atmospheric dispersion models and literature available data. An estimate of wet-deposited carbon may also be carried out using the methodology of Carnovale (1987). We will discuss the wet and dry deposition of toxic organic compounds in greater detail in our review of the atmospheric inputs of toxicants to PPB.

As an alternative to the calculation of deposition from first principles, total dissolved organic carbon (DOC) wet-deposited to PPB may be experimentally determined using a series of high quality, automated rain-activated samplers (Ayers et al., 1992) strategically located around the Bay. Collected samples may subsequently be analysed for DOC. On establishment of the wet deposited flux around PPCR, deposition to PPB may be estimated by regional interpolation.

The experimental measurement of dry deposited carbon to PPB is much more difficult technically than corresponding wet measurements (see Section 6). The feasibility and reliability of any dry deposition sampling program for the PPB Environmental Study would require serious consideration and analysis and is beyond the specifications of this Task.

Experimental measurements of DOC in rainwater and snow in the Dutch Delta area have recently been reported by Nguyen et al. (1990). Over a seven year period (1980-1986) the average DOC annual wet deposition was found to vary from 2.1-6.1 mg m~2 day-l. The corresponding wet deposition of nitrate and ammonium was approximately twice that of DOC. In other studies, the total organic carbon (TOC) deposition (wet deposition only) has been measured to be 3.8-6.6 mg C m~2 day-1 for E United States (Likens et al., 1983) and 15.6 mg C m~ day~ for Chesapeake Bay (North East USA, Velinsky et al., 1986).

Velinsky et al. also report the average dry deposited total carbon as 15.3 mg C m~2 day-1 for Chesapeake Bay. It was estimated that approximately four-five times as much TOC was brought into the Chesapeake Bay drainage basin by wet and dry deposition as was exported by rivers. Furthermore, atmospheric deposition of TOC directly to the Bay was calculated to be of the same order of magnitude as the lower limit of the range of estimates of phytoplankton carbon production (Velinsky et al., 1986).

5.1 SEASONAL ATMOSPHERIC INPUT OF CARBON

The carbon input from atmospheric sources 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. Furthermore, in a recent study on the chemistry and microphysics of aerosol particles during periods of reduced visibility in Melbourne, Gras et al. (1992) reported that 66% of the fine aerosol mass is comprised of carbon. This value is large by international standards, although in absolute concentration terms, the concentrations observed in Melbourne are typical of those observed in other comparably developed cities.

The seasonal variation in wet and dry deposition to PPB has not been considered to date. Although data on the seasonal emission of VOC to atmosphere in PPCR is available, the relative mass of carbon deposited to the Bay cannot be directly inferred due to the seasonal variation in rainfall scavenging efficiency, meteorology and photochemical aerosol formation.

In the absence of local data on the seasonal atmospheric input of carbon to PPB it is worth reviewing results found overseas. A seasonal variation in the wet deposited TOC to Chesapeake Bay was reported by Velinsky et al. (1986). The highest deposition of TOC was found to occur during summer, which was also the wettest season for the region. In summary, Velinsky et al. concluded that the atmospheric deposition of TOC can provide an important flux of organic material to coastal ecosystems and must be considered if biogeochemical cycles are to be fully understood.

6. NITROGEN

Although nutrients such as nitrogen and phosphorus are both likely to play a role in controlling primary phytoplankton growth, nitrogen is of primary concern in temperate zone estuaries and other coastal waters (Howarth, 1988; Ambio, 1990; Lancelot et al., 1987; D'Elia et al., 1986) because it is often the more limiting nutrient - particularly in the critical summer months when algal blooms and anoxia are most severe (Fisher and Oppenheimer, 1991).

6.1 WET DEPOSITION OF NITROGEN

In calculating the wet deposition of nitrogenous species to PPB, Carnovale (1987) considered the solubility and chemical reactivity of six inorganic compounds. These compounds were nitric oxide (NO), nitrogen dioxide (N02), nitrous oxide (N2O), nitric acid (HNO3), ammonia (NH3) and ammonium nitrate (NH4NO3). The six compounds were considered to account for the bulk of the atmospheric nitrogen load to PPB.

On the basis of physical solubility, Carnovale (1987) estimated that nitric acid made by far the largest mass input of nitrogen to PPB (0.1x10+3-1.7x10+3 tonnes N yr~1 compared to a total of 2 tonnes by the other five compounds). The calculations were severely limited by a lack of monitoring data over the Bay. In order to estimate the absolute upper limit mass deposition from nitric oxide and nitrogen dioxide to PPB, Carnovale employed the annual 1-fur average concentration of nitric oxide and nitrogen dioxide as measured at EPAV's Museum monitoring station in 1985. In the absence of local data on ammonia, nitrous oxide and nitric acid, a range of concentrations, typical of clean tropospheric averages (Demerjian, 1986) were used. The upper range value for nikic acid concentration reported by Demerjian (1986) coincided with the average nikic acid concentration measured in central Sydney over the 1980/81 and 1981/82 summers (Kanamori et al., 1983). The calculations of Carnovale clearly identified the importance of monitoring nitric acid concentrations around PPB.

The calculations of Carnovale (1987) based on Henry's law constants were severely limited by the lack of ambient data on nitric acid and ammonia. Sampling conducted by Cadle (1985) in a suburb of Dekoit, USA, over a one year period in 1981-1982, revealed an annual average nitric acid concenkation of 1.7 ppb, ranging from a seasonal minimum of 1.1 ppb in spring to a seasonal maximum of 2.7 ppb in summer. These concenkations are a factor of 5-10 greater than those employed by Carnovale (1987) for PPB. We must emphasise that sampling for low levels of nitric acid is difficult. A recent field comparison of eighteen instruments in Los Angeles revealed that side-by-side measurements of nitric acid varied by as much as a factor of four (Hering et al., 1988). Cadle (1985) also reported an annual concentration of ammonia of 0.4 ppb, ranging from a seasonal minimum of 0.1 ppb in winter to a seasonal maximum of 0.9 ppb in summer. The ammonia concenkation range used by Carnovale for PPB is comparable to that reported by Cadle for Dekoit.

Calculated results for nitric oxide, nikogen dioxide, nitrous oxide and ammonium nikate deposition were aufficiently low to indicate that more accurate ambient data on these species would not alter the overall nitrogen deposition results. In the case of ammonium nikate, it was assumed that the Henry's law constant was equal to that of nikous oxide. This assumption is questionable and consequently the sign)ficance of ammonium nikate aerosol may be greater than previously estimated.

Ambient measurements of nitrous acid (HNO2) in Los Angeles, USA, (Sickles et al., 1988) and Gothenburg, Sweden (Sjodin, 1988) have revealed that low ppb levels of this water soluble acid may be generated in night-time urban air. If similar levels of nitrous acid are generated in the PPB region, the atmospheric nitrogen input to the Bay from nitrous acid may be quite sign)ficant and consequently should be considered in future estimates of nutrient input to PPB.

In addition to physical solubility, Carnovale (1987) considered molecule/water reactions to estimate total wet deposition to PPB. In the case of nitric oxide and nitrogen dioxide, the aqueous phase formation of nitrate (NO3-) in rainwater was shown to be too slow to be of atmospheric importance. Nitric acid however, is reportedly scavenged efficiently by rainfall (Levine and Schwartz, 1982). Depending on the rainfall drop size distribution and rainfall rate, Levine and Schwartz (1982) estimated that 5-50% of nitric acid is removed from the atmosphere per millimetre of rainfall. In order to estimate the amount of nitric acid washout into PPB, Carnovale (1987) pointed out the need to consider the number of rain episodes, duration, volume of rain per episode and associated ambient nitric acid concentrations preceding each event.

In the absence of such data, literature values for ion precipitation fluxes were employed to estimate wet deposition of nitrogen to PPB. Based on extensive measurements on the abundance of major ions in precipitation over North America (MAP3S/RAINE Research Community, 1982; Munger, 1982; Raynor and Hayes, 1982) a range of nitrogen mass fluxes was estimated for PPB. For geographically comparable regions to PPB (semi-rural USA coastal areas experiencing 1-1.5 metres of rainfall annually), the nitrogen flux from nitrate, nitrite (NO2-) and ammonium ions (NH4+) was estimated to lie in the range 0.6-1.0 g m~2 yr-1. Taking the product of mass flux and PPB surface area, Carnovale (1987) estimated that 1.2x10+3-2.0x10+3 tonnes N yr-1 is washed into the Bay by direct overhead rainfall. Ammonia based products were estimated to contribute approximately 50-70% of the above mass load (see Table 2).

More recently, Carnovale and Saunders (1988) have analysed rainfall ion composition data of Ayers and Gillett (1988) in order to re-estimate the wet deposition of nitrogenous species to PPB. Rainfall samples from a total of five suburban sites (see Figure 2) in PPCR were collected and analysed over a two year period (1982-1984) by Ayers and Gillett. From a statistical analysis of data on approximately 280 samples, Carnovale and Saunders (1988) estimated the volume weighted ion flux of nitrate and ammonium ions to be 0.12 and 0.18 g N m~2 yr-l, respectively. The annual nitrate and ammonium ion flux calculated from the data of Ayers and Gillett is approximately one third that estimated from literature data. The direct, wet deposited nitrogen input to PPB was accordingly revised downwards to a value of approximately 600 tonnes N yr~1 (40% nitrate, 60% ammonium). From Figure 2 it is evident that the data of Ayers and Gillett is only strictly applicable to part of the Melbourne Metropolitan Area. Nutrient deposition to PPB may be significantly different to that in Melbourne's eastern suburbs.

Carnovale and Saunders (1988) commented that ammonium nitrate and ammonium sulfate are the most abundant nitrogenous compounds in urban aerosols. Both the nitrate and sulfate were accounted for in the deposition data of Ayers and Gillett (1988) whilst dry deposited ammonium nitrate was estimated to be negligible (Carnovale, 1987). Consequently, it was argued that the calculated atmospheric nitrogen load to PPB accounted for gaseous and aerosol deposition.

Gras et al. (1992) of CSIRO Division of Atmospheric Research have recently conducted a collaborative project with EPAV on the chemistry and microphysics of aerosol particles (less than 2.50 micron diameter) in Melbourne during periods of reduced visibility. Gras et al. reported that the sulfate plus nitrate aerosol mass contribution was small in autumn (3-lO%) but increased during spring and summer.

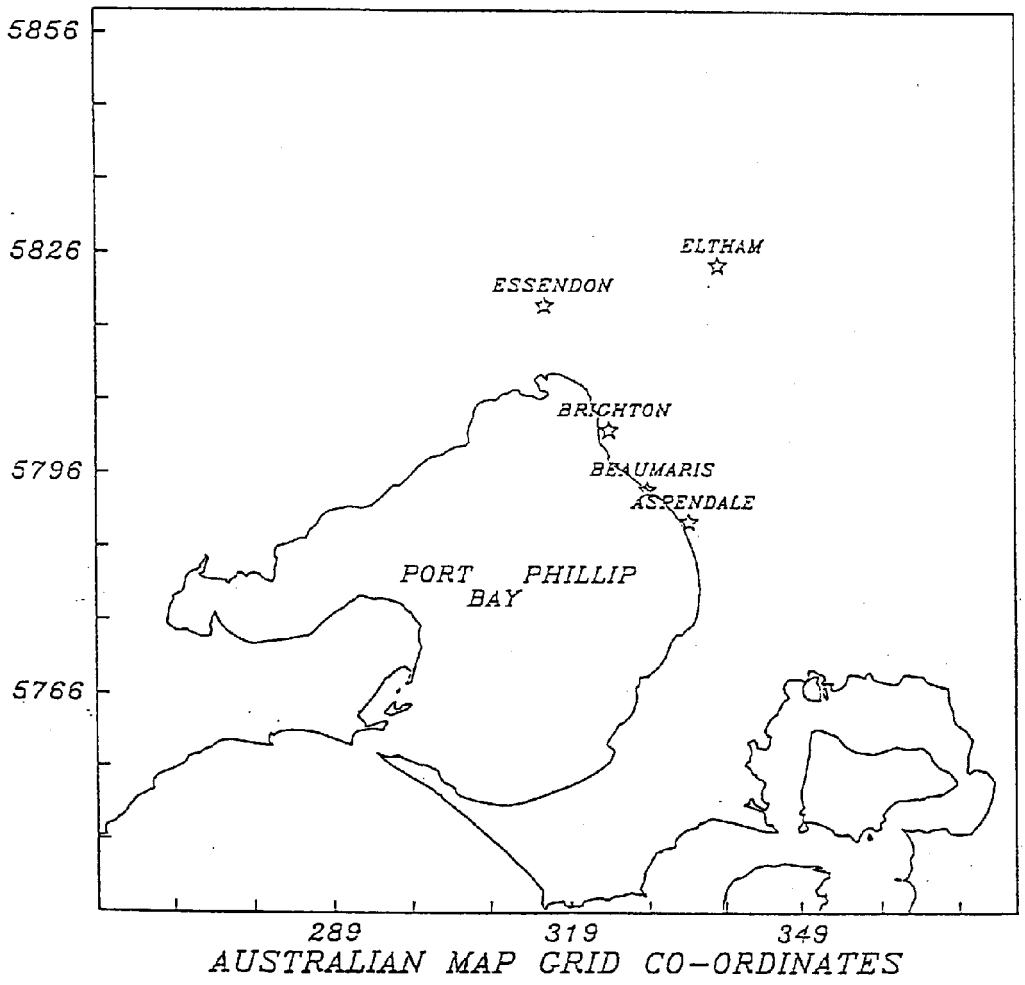

Figure 2. Location of rain sampling gauges used in study by Ayers and Gillett (1988).

Table 2. Estimated input of wet deposited nitrogen to PPB (from Carnovale, 1987).

Species	N (Tonnes) From Henry's Law	N (Tonnes) Literature Flux
NO	2.6x10-3	na
NO2	8.2x10-3	na
N2O	2.3x10-1	na
NH3	0.2-1.7	na
HNO3	(0.1-1.7)x10+3	na
NO2, NO3	na	(5.8-7.8)x10+2 a
NH4NO3	6.8x10-4	na
NH4+	na	(0.6-1.2)x10+3 b

a. Includes contribution from water soluble N-acids. b. Includes contribution from ammonia based compounds. na. Not available.

6.2 DRY DEPOSITION OF NITROGEN

The dry deposited, annual mass nitrogen input to PPB has been calculated by Carnovale (1987) based on literature deposition velocities (Voldner et al., 1986) and a combination of local and literature ambient concentration data (as discussed in Section 6.1). The results are presented in Table 3.

Table 3. Deposition velocities and calculated annual nitrogen mass input to PPB. Reproduced from Carnovale (1987).

Species	vd(m s-l)	N-input (Tonnes)
NO	7.1x10-6	1.2x10+ 1
NO2	1.5x10-4	1.6x10+2
N2O	1.0x10-4	1.2x10+3
HNO3	(0.05-5)x10-2	0-5x10+2
NH3	1.0x 10-4	0.4-4
NH4NO3	1.0x10-4	4

Deposition velocities for nitric oxide, nitrogen dioxide and nitric acid employed in the calculations of Carnovale (1987) were taken from Voldner et al. (1986). Lee and Schwartz (1981) provide independent estimates of deposition velocities for nitrogen dioxide and nitric acid to surface waters. On the basis of the limited solubility of nitrogen dioxide and its efficient oxidation in water, the deposition velocity was estimated to lie in the range 5x10-6-1x10-3 m s-l. The nitrogen dioxide deposition velocity used by Carnovale lies in the upper half of the range reported by Lee and Schwartz. In the case of nitric acid, Lee and Schwartz considered the deposition to surface waters to be limited by gas-phase mass transport (due to high solubility of acid) with a deposition velocity of the order of 1x10-2 m s-1. This value is within the range considered by Carnovale.

Dry deposition velocities for nitrate, ammonia and ammonium in remote marine areas have been reported by Galloway (1984). Values of 3x10-3, 8x10-3 and 3x10-4 m s-1 for nitrate, ammonia and ammonium respectively, were considered to be representative of literature reported data. The deposition velocity for ammonia and nitrate aerosol employed by Carnovale (1987) is one to two orders of magnitude less than that of Galloway (1984). Given the lack of data on ammonia and aerosol concentrations around PPB, any estimate of the associated deposition to PPB remains speculative - irrespective of deposition velocity employed.

Nitrous oxide is calculated to make the largest contribution to the dry deposited nitrogen input to PPB. However, nitrous oxide does not undergo significant chemical reaction in water and may be re-emitted to the atmosphere. Work conducted by Junge et al. (1971) and Junge and Hahn (1971) support the contention that the ocean is a major source of nitrous oxide to the atmosphere. This may be due to nitrification involving the oxidative regeneration of nitrate from organic nitrogen or ammonia via nitrous oxide (Cohen and Gordon, 1978; Cohen, 1978). Under anoxic environments however, nitrous oxide is reported to be consumed by denitrifying bacteria (Cohen, 1978; Carpenter and Capone, 1983).

There is considerable uncertainty in the literature regarding the relative importance of dry and wet deposition. Whereas experimental wet deposition measurement programs may now be carried out routinely, there are only few adequate measurements of dry deposited nitrogen reported in the literature (Vet et al., 1988; Riggan et al., 1985; Sirois and Vet, 1988). Where reasonable estimates of dry and wet deposition of nitrogenous species have been determined, dry deposition has been found to range from one half to four times wet deposition (Hinge et al., 1991).

Shannon and Sisterson (1992) have reported the use of extensive wet deposition monitoring data along with sparse dry deposition monitoring data and isolated field characterisation of droplet deposition to produce an annual deposition budget for each contiguous USA state and Canadian province. They report that for the entire USA and Canada, dry deposition accounts for 56% of anthropogenic nitrogen deposition, wet deposition accounts for 39% and drop deposition (i.e. cloud impaction on elevated terrain and fog droplet collection on lower terrain) 5 % . The North American data however, need not necessarily reflect the relative importance of wet and dry deposition to PPB.

Adsorption of nitric acid has been estimated to account for approximately 75 % of total dry deposition (Lovett and Lindberg, 1986). In a recent study of the vertical distribution of gases and aerosols in the lower atmosphere, Erisman et al. (1988) reported a deposition velocity for nitric acid which lies near the upper-end of the range listed in Table 3.

One recent approach for the routine estimation and reporting of dry deposition has been reported by Clarke et al. (1991). A National Dry Deposition Network (NDDN) was established in the USA in 1986 to document the magnitude, spatial variability, and trends in dry deposition of ozone, acidic particles and gases. In 1991 the network consisted of 50 stations. Dry deposition is not measured directly in the NDDN but is estimated from the product of measured ambient concentration and modelled deposition velocity. Chemical species considered include ozone, sulfate, nitrate, ammonium, sulfur dioxide and nitric acid.

6.3 TOTAL ATMOSPHERIC INPUT OF NITROGEN

Carnovale and Saunders (1988) estimated the total input of atmospheric nitrogen to PPB to be in the range 2.0x10+3-2.5x10+3 tonnes yr-1 "including nitrous oxide). Ammonium, nitrate and nitrite account for approximately 0.8x10+ -1.3x10+3 N tonnes yr-1.

A total of 1.2x10+3 tonnes of nitrous oxide was calculated to be dry deposited annually to PPB. However, nitrous oxide is relatively unreactive, being inert to the halogens, alkali metals and ozone at room temperature (Cotton and Wilkinson, 1972). Consequently, nitrogen bound in nitrous oxide may be considered to be unavailable to the Bay ecosystem.

It must be stressed that the surface area of PPB is approximately 15-20% (our estimate) of the PPB catchment area and consequently the total contribution of atmospheric nutrients to the Bay catchment area may be sign)ficantly greater than that estimated by Carnovale (1987) and Carnovale and Saunders (1988) for PPB alone. Berner and Berner (1987) estimate that on a global scale, of the total nitrogen deposited to land, 18% reaches rivers and marine waters.

6.4 SEASONAL ATMOSPHERIC INPUT OF NITROGEN

The nitrogen input from atmospheric sources is expected to vary with season. The seasonal variation in wet and dry deposition to PPB has not been considered to date. Although data on the seasonal emission of NOX to atmosphere in PPCR is available (Carnovale et al., 1991), the mass of nitrogen deposited to the Bay cannot be directly inferred due to the seasonal variation in rainfall scavenging efficiency, meteorology and photochemical oxidation of NOX to nitrogenous acids.

The seasonal nitrogen input to the Chesapeake Bay Estuary and its possible effect on algal growth has been discussed by Fisher and Oppenheimer (1991). They point out that although nitrogen input during various seasons of the year (particularly summer and autumn) may sign)ficantly effect phytoplankton growth, the persistence of nutrient inputs from winter and spring render annual data useful for understanding the consequences of nutrient loadings to the Bay.

6.5 FUTURE NITROGEN DEPOSITION SCENARIOS Carnovale and Saunders (1988) employed coarse projections of emissions of NOX (Martins, 1988) in the Melbourne Airshed (area within approximately 35 kilometres of Melbourne) to estimate the nitrogen input to PPB in 2005. On the assumption that wet and dry deposition flux to PPB is proportional to the ambient concentration of chemical species in the Melbourne Airshed, the total deposition of nitrogen in the form of nitrates was predicted to increase from 4x10+2-9x10+2 tonnes annually in 1985 to 0.6x10+3-1.2x10+3 tonnes in 2005. Ambient levels of ammonia were not expected to increase sign)ficantly. The total nitrogen input to PPB for 2005 was projected to be 0.9x10+3-1.6x10+3 tonnes (excluding nitrous oxide).

More recently, Carnovale (1992) has carried out detailed calculations of vehicle emissions in PPCR between 1980 and 2005. The calculations were based on an analysis of the motor vehicle fleet (light and heavy duty vehicles), vehicle age and emission deterioration. Carnovale used recent experimental test data on Australian on-road vehicles (available to EPAV) in order to estimate current emissions and projections to 2005. The results show that motor vehicle NOX emissions increased from a daily value of 146 tonnes in 1980 to 171 tonnes in 1990. NOX emissions are predicted to increase to 180-280 tonnes by 2005. Given the calculated increase in NOX emissions since 1980, a re-analysis of future nutrient atmospheric loadings to the Bay is warranted.

7. PHOSPHORUS

Phosphorus has not previously been considered to have any sign)ficant atmospheric sources (Fisher and Oppenheimer, 1991). Most investigators have assumed that riverine input is primarily responsible for phosphate in the sea and that vertical transport from deeper waters is responsible for the phosphate in the photic zone (Duce, 1986). Graham and Duce (1979) have estimated that, on a global scale, the net atmospheric transport of phosphorus to the ocean is approximately 5-7% of the riverine input (Lerman et al., 1975; Emery et al., 1955).

To the best of our knowledge, there are no sign)ficant vapour phase forms of phosphorus in the atmosphere although phosphine and methylated forms of phosphine may be produced in certain reducing environments, especially swamps and tidal flats (Duce, 1986). Some pesticides containing phosphorus are also undoubtedly present in the vapour phase, albeit at low concenkations.

Phosphorus however is found in dust particles and microbial debAs (Fuller, 1972). By far the largest contributor to wet and dry deposition of phosphorus is wind blown dust from deserts or dry land regions. Phosphorus is also found in fertilised soils (Morelli et al., 1984). The gross relative abundance of phosphorus on the earth's surface is approximately 0.10% (Ahl, 1988), which is comparable to the phosphorus content of dust.

We have not found any estimate of the atmospheAc input of phosphorus to PPB. Given that wind blown dust is the dominant source of wet and dry deposited phosphorus in PPB, the annual atmospheric input to the Bay may vary sign)ficantly from year to year (perhaps by orders of magnitude depending on meteorology).

AtmospheAc deposition fluxes of phosphorus have been reported for the USA (Ahl, 1988), Finland (Haapala, 1977), Canada (Schindler et al., 1974, 1976), England (Allen et al., 1968) and the USSR (Evdokimova et al., 1976). Phosphorus deposition ranging from 5-80 kg P km12 yr-1 have been reported in the Northern Hemisphere.

The annual wet deposition of total phosphorus has been estimated at three collector sites in the north-central USA by Munger (1982). Annual wet deposition of total phosphorus was measured to lie in the range 9-15 kg P km12 yr-1.

Graham and Duce (1982) attempted to evaluate the importance of atmospheAc phosphorus to a restActed area of the ocean off Bermuda. Over 30 aerosol samples were collected from ships in the region and on the island of Bermuda. The samples were analysed for total phosphorus and reactive (or water soluble) phosphorus. The difference between the two forms was assigned to organic plus acid soluble phosphorus (generally referred to as organic phosphorus). The total phosphorus concentration near the North AmeAcan coast was found to be 0.65 umol m~3. Approximately one third of the phosphorus enteAng the ocean was found to be soluble in seawater. Furthermore, Duce (1986) has estimated the net soluble phosphorus atmospheric input to the Pactfic region to lie in the range 0.005-0.012 umol m~ day-1 for the peAod sampled (equivalent to 0.6-1.4 g P ha-1 yr-1).

Walker et al. (1981) have measured the atmospheAc flux of phosphorus in rain for sub-tropical eastern Australia to be 250 g P ha-1 yr-1 (in vicinity of coastal sand dunes).

7.1 SEASONAL ATMOSPHERIC INPUT OF PHOSPHORUS Uematsu et al. (1983, 1985) have found that most of the mineral aerosol enteAng the North Pacific does so in three or four short duration bursts (3-5 days) associated with major dust storms originating over the deserts of China in spring. Reactive phosphorus (PO4 3-) in the marine aerosol is largely associated with mineral particles (Graham et al., 1979). A similar seasonal dependence of dust input has been documented for other regions (Chen and Duce, 1983).

For PPB, the annual phosphorus input is also expected to exhibit strong seasonal and yearly variation, with major inputs associated with dust storms over one or more days. As an example of this, we highlight the dust storm of 8 Feb. 1983 which is estimated to have resulted in approximately 140,000 tonnes (in range 26-260 thousand tonnes) of top soil from the western diskicts of Victoria being blown towards Melbourne (Mitchell, 1983).

8. AIRBORNE SUSPENDED PARTICULATES AND DUSTFALL Particles may be class)fied into several groups according to their size. Particles with diameters up to 10 um are normally referred to as PMlo or "suspended matter". Suspended particles up to about 25-50 um are referred to as "total suspended particulates" (TSP). Particles larger than 50-100 um are normally referred to as dust. Dust normally remains localised near its source (within 100 m) whilst PMlo and TSP may remain suspended in the air for long periods during which time it may be dispersed over a large area (e.g. PPB).

The deposition of particles directly to PPB has not been reported.

EPAV routinely measured dustfall at numerous sites in the Melbourne Mekopolitan area, Geelong and the Latrobe Valley until the late 1970's (annual Air Monitoring Results, 19721980). In 1978 approximately 64 dustfall gauges were in operation in the EPAV network after which the dustfall monitoring program was gradually scaled down to cover Geelong and the Latrobe Valley only. The yearly average insoluble dust deposition rate was reported to decrease by approximately a factor of 2 during the 1970's. By the late 1970's, the daily mean insoluble dust deposition rate in Melbourne was reported to lie in the 40-120 mg m~2 range. In country areas such as Geelong, Morwell and Traralgon, dust deposition rates were found to be at the lower end of the range reported for Melbourne. Total dust deposition was found to be approximately 1.5-2 times that of the insoluble mass fraction.

EPAV currently monitors PM10 and TSP on a regular basis at fixed monitoring sites in the Melbourne Metropolitan area. In 1990, the monthly mean PM10 concentrations near the Melbourne Central Business District were reported to lie in the 8-31 ug m~3 range with an annual average of 22 ug m~3 (EPAV, 1990) . Monthly average TSP concentrations were reported to lie in the 21-59 ug m~3 range (average of 42 ug m~3) at five inner city and suburban sites. PM10 and TSP measurements reported by EPAV between 1987 and 1989 are comparable to the 1990 values documented above (EPAV, Air Monitoring Data reports 19871990).

The elemental composition (by relative weight) of PM10, TSP and dust in the PPB region is not well known. Work conducted by Gras et al. (1992) on particles less than 2.5 um diameter, indicates that approximately 66% of the fine aerosol mass in Melbourne is due to carbon with sulfate plus nitrate accounting for approximately 3-10% of the mass. However, the elemental composition of PM10 need not be similar to that of fine particles.

TSP and dust are generally comprised of soil elements over the continents and sea salt elements over the oceans (Finlayson-Pitts and Pitts, 1986). Major elements found in TSP and dust over land include silicon, aluminium, iron and calcium in ratios similar to those in the earth's crustal material. TSP and dust in PPCR may also contain sign)ficant quantities of bitumen debris, vehicle tyre debris and fuel combustion products.

In order to estimate the atmospheric input of particles to PPB, the size distribution of particles needs to be considered. Dry deposition velocities over continents and oceans for particles less than about 2 um diameter are in the range 3x10-5-3.6x10-4 m s-1 (Giorgi, 1986). In the case of particles having diameters greater than 2 um, dry deposition velocities are reported to be in excess of 5x10-3 m s-1, increasing non-linearly with diameter (Giorgi, 1986).

The above information may be combined with the known surface area of PPB (1.95x109 m2) to make an "order-of-magnitude" estimate of TSP and dust deposited directly to PPB.

9. CALCULATION OF TOTAL NUTRENT DEPOSITION

Hinga et al. (1991) have recently outlined a procedure for assessing the total nutrient loads in coastal marine ecosystems. Briefly, the steps may be summarised as follows:

i. Take existing measurements of wet deposition in or near the watershed. If data is not available initiate field sampling program.

ii. Estimate the dry deposition based on the wet deposition. This makes the explicit assumption that dry deposition equals wet deposition.

iii. Multiply the total deposition by the fraction of nutrient which would pass through the terrestrial ecosystem and enter rivers. This may be done using different factors to characterise different land categories.

iv. Multiply the flux entering the rivers by a factor to account for denitrification in rivers.
v. Add nutrient deposited directly on the coastal ecosystem to the riverine flux.

Hinga et al. (1991) point out that it would not be particularly difficult to make the calculations appear more elegant by the subdivision of the watershed into more land-use categories, using a detailed database of land uses, assembling more detailed lists of point source and agricultural inputs in addition to using some technique for contouring deposition over the watershed. However, they argue that such procedures are not likely to improve calculations until sign)ficant advances are made in the understanding of the many processes responsible for the behaviour of nutrients, such as nitrogen, in terrestrial ecosystems, rivers and streams.

10. SUMMARY

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

i. Data on the atmospheric input of carbon to PPBis not currently available.

ii. Extensive data is available to EPAV which may be employed to estimate dry deposited carbon to PPB and its associated water catchment area. The data is in the form of emission rates spatially (3 km by 3 km grid cells) and temporally resolved (hourly and seasonally) which if coupled with atmospheric dispersion models, windfields for PPB and literature deposition rates will permit the estimation of total dry deposited carbon.

iii. Wet deposition data on carbon, nitrogen and phosphorus may be acquired by the strategic location of rain-activated field sampling equipment and subsequent analytical analysis.

iv. Wet and dry deposition data on the input of atmospheric nitrogen directly over PPB has been estimated by Carnovale and Saunders using limited ambient data on nitric oxide, nitrogen dioxide, nitrous oxide and ammonium nitrate. Local data on nitric acid, nitrous acid and ammonia was not available.

v. Given the lack of year-round ambient concentrations for such important nitrogen species as nitric acid, nitrous acid and ammonia, the available estimates of atmospheric input to PPB are speculative.

vi. Dry deposition of nitrogen to PPB and its associated water catchment area may be estimated by the implementation of a field sampling program to measure the ambient concentrations of water soluble nitrogenous species such as nitric acid, nitrous acid and ammonia. Ambient concentrations may be coupled with available deposition velocities to estimate dry deposition. Alternatively, experimental sampling programs may be designed to explicitly measure dry deposition for each of the water soluble nitrogenous species.

vii. Data on the atmospheric input of phosphorus to PPB is not currently available. Our review indicates that under normal meteorological conditions, atmospheric phosphorus deposition may only be of marginal importance to the total phosphorus loading of PPB. Under extreme meteorology (resulting in drought) however, wind blown dust may episodically sign)ficantly add to the phosphorus loading of PPB.

viii. Dry deposited aerosols may be a more sign)ficant source of nutrients to PPB than previously estimated.

ix. A methodology proposed by Hinga et al. (1991) for estimating total nutrient input to coastal systems is detailed for consideration.

x. The atmospheric input of nutrients to PPB and its associated water catchment area needs to be re-assessed in view of the sign)ficant deficiencies in previous estimates. Any reassessment may require experimental field sampling programs in conjunction with mathematical models of deposition and nutrient transport.

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