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해안가 매립으로 인한 지하수의 수리화학적 특성

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Hydrochemical characteristics in groundwater affected by reclamation

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본 연구는 2000년 시드니올림픽사이트의 해안가를 따라 인위적인 매립으로 인한 지하수 내 수리화학적인 특징을 규명하는데 그 목적이 있다. 올림픽게임지역을 3개의 지역, 즉 간척지역(reclaimed area, 과거에는 강하구였으나 폐기물로 매립되어 있으며 현재는 조수간만 보다 높은 지역), 매립지역(landfill area, 해수면 위에 폐기물을 매립한 지역), 그리고 자연 상태지역(non-infilled area, 폐기물의 매립이 전혀 없었던 지역))으로 나누어 조사하였다. 또한 시추공 심도별로 심부시추공, 천부시추공 및 스텐드파이프로 구분하여 상대적인 농도들의 거동을 상오 비교하였다. 그 결과 간척지역 내 지하수는 Na, K 그리고 Mg 이온이 지배적이지만 매립지역은 Na 그리고 K가 지배적인 이온이다. 또한 간척지역 및 자연 상태지역의 지하수는 Mg 및 Ca 농축이 특징이지만, 매립지역은 K 및 NO_3의 농축이 특징이다. 그러나 시추공심도별로 살펴보면 심부시추공 지하수는 Na와 Mg가 지배적이지만 천부시추공 및 스텐드파이프 지하수는 Na와 K가 지배적인 원소로 나타났다. 간척지역, 매립지역 그리고 자연 상태지역 지하수 내 전기전도도와 중금속 농도와는 명확한 상관관계를 보여 주지 않는다. 간척지역 및 자연 상태지역과 비교시 매립지역 지하수의 Fe 및 Mn의 농도는 pH가 감소함에 따라 현저히 증가하는 양상을 보여준다. 심부시추공과 천부시추공 지하수 내 평균 전기전도도 값은 스텐드파이프보다 높지만 스텐드파이프 지하수 내 전기전도도 값의 최대 및 최소값은 현저한 차이를 보이는데 이는 국부적인 공동의 영향으로 사료된다. 심부시추공, 천부시추공, 그리고 스텐드파이프 지하수 내 pH 대비 중금속(Cu, Pb, Zn, Cr) 농도사이의 상관관계는 없었지만 이들 지하수 내 Fe와 Mn은 pH가 감소하면 농도가 증가하는 양상을 보여준다. 지하수 내 농축된 중금속 농도는 매립된 폐기물의 성상과 밀접한 관계를 보이며 해안가를 따라 매립되어 있는 지역의 토양 내 중금속의 용출은 주변 환경에 대한 잠재적인 환경위험을 결정시 중요한 역할을 한다.

주요어: 지하수, 토지이용별, 시추공 심도별, 매립, pH,

This study focuses on the hydrochemical characteristics in groundwater affected by reclamation at 2000 Sydney Olympic Games site, Sydney, Australia. The Olympic Games site can be divided into three areas, i.e. reclaimed areas; landfill areas and non-infilled areas. In the current work, 'reclaimed areas' were previously estuarine, and were filled with waste materials and are now above present high tide level, whereas

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'landfill areas' are areas where deposition of waste materials occurred above sea level. No deposition of waste took place in 'non-infilled areas'. This study was also evaluated by three different types such as deep boreholes, shallow boreholes and standpipes. The hydrochemistry of groundwaters in reclaimed and non-infilled areas is characterized by Mg- and Ca-enrichment, whereas groundwaters in landfill areas are elevated in K and NO_3. Na, K and Mg are the dominant cations in groundwater from reclaimed areas and Na and K are the dominant cations in groundwater in landfill areas. Na and Mg are the dominant cations in groundwater in deep boreholes, whereas Na and K are the dominant cations in groundwater in shallow boreholes and standpipes.

There is no distinct trend in heavy metals with electrical conductivity in the groundwater between the reclaimed, landfill and non-infilled areas. Fe and Mn in landfill areas with respect to reclaimed areas and non-infilled areas show a distinct increase in concentration with declining pH. Mean electrical conductivity values in the deep and shallow boreholes are higher than that of standpipes, but the minimum and maximum value of electrical conductivity in groundwater in standpipes shows remarkably different value, probably due to perched pond. There is no correlation between Cu, Pb, Zn, Cr concentrations in groundwater with pH, from deep boreholes, shallow boreholes and standpipes, except for Fe and Mn, which demonstrate increasing concentrations with declining pH. The results revealed a close association between elevated concentrations in groundwater and the presence of fill materials at the site. Trace metals leachability from reclaimed soils adjacent to estuary plays a significant role in determining their potential environmental risk to surrounding environment.

Key words: groundwater, land use type, borehole type, reclamation.

1. Introduction

The coastal zone, in particular, is undergoing extensive urbanization worldwide (Stuyfzand, 1995; Vale et al., 1998; Adams et al., 2001; Suh, 2003). The disposed of waste by an affluent society has resulted in a growing number of reclamation sites adjacent to estuaries in many countries (Goldberg, 1998; Bell et al., 2000).

Management and assessment of contaminated soils in reclaimed areas requires the recognition of complex chemical processes and a knowledge of the heterogeneity of such soils. The main sources of contamination in reclaimed areas may be characterized according to several broad descriptions, e.g. construction and demolition materials, household waste and disposal from transport, industry, power stations, agriculture and forestry sectors (Thornton, 1990). Many sites contaminated by past waste disposal continue to pose a serious threat to the environment. Due to a growth of industry, housing, construction, mining, and other changes in land-use, detrimental effects to groundwater quality are commonly detected. (Melloul and Goldenberg, 1998; Ansari et al., 1999). However, groundwater contamination often extends beyond the boundaries of the contaminated site and is therefore frequently more difficult to investigate than localized soil contamination (Appleyard, 1993; Suh et al., 2003a).

Management of estuaries must take into account natural hydrological, biological and chemical processes, as well as the development of anthropogenic stresses such as point/non-point sources, hydrological deficits, and interrelated hydrogeochemical and hydrogeological processes (Goldberg and Melloul, 1994). An important problem in estuarine environments is reclamation of wetlands and mudflats in adjacent embayments. For example, in Hong Kong, which has one of the highest population densities in the world, several million people live in a coastal strip in an area of about 30 km^2. More reclamation, which will be at the expense of nearby coastal fishing grounds and ecologically important wetlands, is planned in the near future (Nancy and Smith, 1987). Other examples of major land

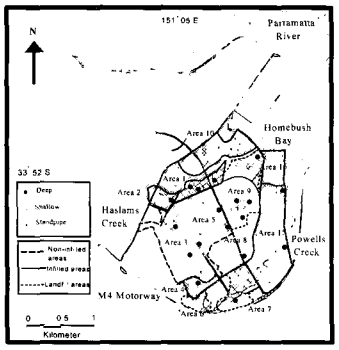


Fig. 1. Reclaimed and dredged areas in Port Jackson.

reclamation schemes are for the expansion of Tokyo Bay, and for the new Osaka airport between the Japanese islands of Honshu and Shikoku (Furukawa, 2000). Modification of harbour foreshores and fluvial environments has taken place throughout the USA, e.g. Boston Harbour, where many hills have been removed and the debris used as landfill to reclaim wetlands for construction. Such expansion of urban areas not only leads to the degradation of the ecosystems by the removal of vegetation and soil, but also increases the cover of the land surface by impermeable materials, thereby altering the hydrological characteristics of the region (Pickering and Owen, 1997).

Reclamation has increased substantially in estuarine environments around the world (Stuyfzand, 1995; Vale et al., 1998). The state of the estuary varies in response to forcing functions at different time-scales, e.g. seasonal variations, spring/neap tide cycles, synoptic scale variations (wind, precipitation, sea temperature, sea state, insolation), diurnal cycles (temperature, wind), tidal cycles and wave action force estuarine waters over a range of periods from months to seconds (Kitheka, 1997; Radhakrishna, 2001; Suh, 2003).

Hydrogeochemical interactions between saline water and groundwater in reclaimed land infilled with contaminated waste adjacent to an estuary may be sensitive to small changes in waste material compositions, topography and associated tidal regime (Suh et al., 2003a,b). Hughes et al. (1998) suggested that tidal fluxes, vegetation, rainfall, seasonal variations in evapo-transpiration, tidal or flood events and variations in regional groundwater flow play an important role in the hydrology of reclaimed land. The current study focuses on the Olympic Games site on the southern shores of Homebush Bay in upper Port Jackson to assess the impact of waste materials on groundwater based on both past land use type and borehole type.

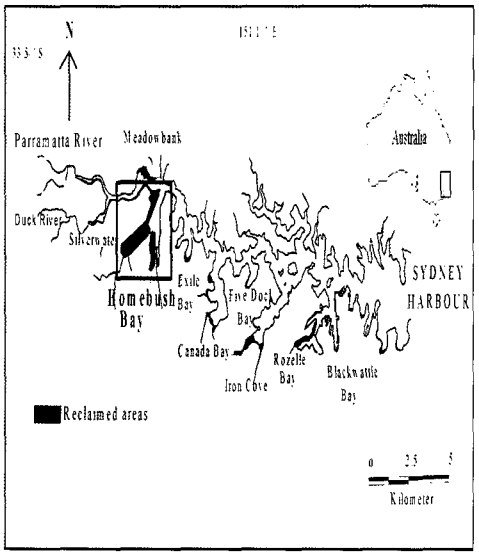


Fig. 2. Study area and sampling locations.

2. Human impact on the Olympic Games site

The Olympic Games site is bordered by the Parramatta River, the M4 motorway and Powells Creek (Fig. 2). The Olympic Games site can be divided into three areas, i.e. reclaimed areas (Areas 1, 2, 10, 15, 16); landfill areas (Areas 4, 6, 7, 9) and non-infilled areas (Areas 3, 5, 8). In the current work, 'reclaimed areas' were previously estuarine, and were filled with waste materials and are now above present high tide level, whereas 'landfill areas' are areas where deposition of waste materials occurred above sea level. No deposition of waste took place in 'non-infilled areas'. Table 1 shows the summary of past land use in the study area. The shores of Homebush Bay has been reclaimed using waste materials comprising an estimated 9 million tonnes of domestic, commercial and industrial waste

Table 1. Summary of past land use in the study area. R and L mean reclamation and landfill, respectively.

Areas	Location	Active period	Fill area(m^2)	Waste composition	Fill types	Fill depth(m)
1	Abattoir Haslams Creek (south-east)	1965-1982	133,200	Municipal garbage, putrescible, , building & chemical wastes	R	0-17
2	Abattoir Haslams Creek (south-west)	1951-1978	78,100	Municipal garbage, putrescible, , building & chemical wastes	R	0-3.5
4	Former State Rail Authority tip site	1960-1970	12,000	Railway & putrescible wastes	L	1.5-4
3 & 5	Homebush Abattoirs	-	-	-	No filling	-
6	West of State Sports Centre	1965-1981	20,000	Municipal garbage	L	0-5.5
7	State Sports Centre	1965-1981	70,500	Municipal garbage & putrescible wastes	L	0-9
8	Australia Centre	-	-	-	-	-
9	State Brick Works	1979-1982	25,000	Putrescible wastes & municipal garbage	L	0-14
10	Electricity Ash Ponds (Haslams Creek north)	1961-1990	28,000	Household garbage, construction & ash fill	R	3.5-6.7
15	Bicentennial Park	1950-1982	160,300	Dredged sediment	R	0-1.2
16	Bicentennial Park (north part)	1951-1982	32,000	Dredged sediment	R	0-2.4

over several decades (OCA, 1996). Past activities, such as reclamation of wetland areas, land clearing, shoreline remodelling, industrial activities and landfilling have caused an adverse environmental impact on the Olympic Games site.

The Olympic Co-ordination Authority (OCA) announced that about 20% (160 ha) of Homebush Bay (total 760 ha) has been defined as a contaminated site, based on dioxin concentrations (OCA, 1995) and has been characterized by heavy industry and medium-density housing for most of the 20th century (Homebush Bay Corp., 1994; Suh, 2003).

3. Materials and methods

3.1. Field work

Representative groundwater samples (Fig. 2) were collected from all parts of the study area during November 1990 to March 1991, using rotary drilling rigs by Coffey and Partners (1991). Two closely-spaced boreholes were drilled at each sampling site, a deeper hole was completed in the underlying hard rock (A series; n=19) and the other hole was completed as a piezometer in fill materials and/or shallow weathered bedrock (B series, n=19). The depth of the A series boreholes varied from 15.2 to 36.3 m beneath the surface, and was terminated after at least 8 m of competent hard rock was penetrated. The depth of B series boreholes terminated at between 3.6 and 23.6 m. All standpipes were constructed in fill materials and range from 1.6 to 26 m deep. Unfortunately, during constructing and monitoring, one deep borehole and 11 shallow boreholes did not collect water samples because they were dry, or damaged. Four leachate samples were collected in Areas 1 and 2 adjacent to Haslams Creek during February 1991 because these areas contained a substantial amount of domestic and industrial waste materials.

Slotted standpipes were constructed of 50 mm diameter, class 9 UPVC pressure pipe. The pipes were slotted at, and below, the water table and equipped with bottom caps and locked top caps. Sections were joined without solvents, or cements. A sand filter pack of 35 mm nominal thickness was installed around the slotted section, with a backfill cement and bentonite slurry seal around the non-slotted section. Standpipes were developed by pumping until the discharge was visually clear after purging. Standing water levels in the standpipes were measured and reduced to AHD. The standpipes

were purged using a gas-operated sampling pump, or bailer samples were obtained from the standpipes using a teflon bailer, or stainless steel and teflon environmental water sampling pump. Samples were dispatched to the appropriate laboratory in ice-chests; and duplicate samples were stored at 5℃.

3.2. Chemical analyses

One hundred and one groundwater and four leachate samples were collected for chemical analyses throughout the study area (Coffey and Partners, 1991). All samples were stored in polyethylene containers, labeled and preserved in a refrigerator until processing in the laboratory. Electrical conductivity and pH were determined in the field. Bicarbonate was measured by titration with methyl orange and chloride was determined by AgCl titration. Sulfate was determined by precipitation of DMR-$BaCrU_4$ and measurement of the light absorption by a spectrophotometer. All methods used for the various determinations followed Standard Methods (APHA, 1985). Major elements (Na, K, Ca, Mg) were analysed by a Perkin Elmer (model 3000) atomic absorption spectrophotometer (AAS). Heavy metals (Cu, Pb, Zn, Cr, Fe, Mn) were analysed by a Perkin Elmer (model 3000) atomic absorption spectrophotometer (AAS). A quality assurance and quality control (QA/QC) program was carried out during the study. The objectives of this program were to assess laboratory performance, identify deficiencies in data quality and classify the reliability and useability of the laboratory data. Analytical work was divided between two laboratories (Australian Analytical Laboratory (AAL) and Sydney Analytical Laboratory (SAL)) and duplicate samples were sent to both laboratories.

All concentrations in Standard Reference Materials (SLRS-1) analysed were within 10% of the recommended concentrations and the analyses are reproducible within 6% relative standard deviation (RSD) (Coffey and Partners, 1991).

4. Results and discussion

4.1. Physico-chemical characteristics in relation to past land use

The mean, range and standard deviation of physico-chemical parameters of groundwaters in relation to past land use are shown in Table 2. Mean pH values for groundwater in reclaimed, landfill and non-infilled areas at the Olympic site range only from pH 7.02 to pH 7.38, i.e. from neutral to slightly alkaline. It was calcu-

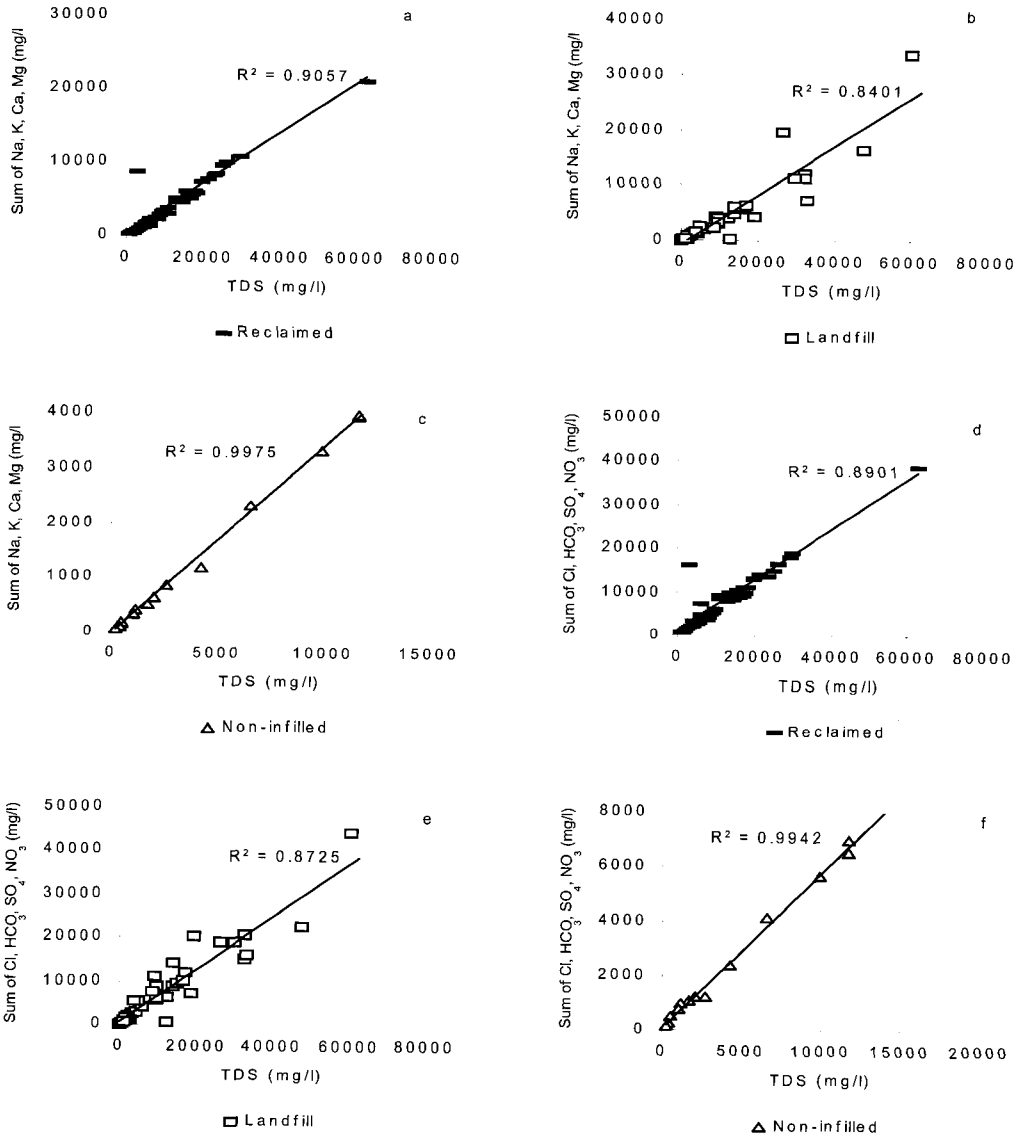
Table 2. Mean of physico-chemical parameters of groundwater samples in reclaimed areas, landfill areas and non-infilled areas. All values are mg/l, except for EC (μS/cm).

Parameter	Reclaimed area (n= 49)	Landfill area (n= 37)	Non-infilled area (n= 15)
pH	7.02 (0.44)	7.38 (1.53)	7.03 (1.42)
EC	18768 (16243)	19872 (21187)	5489 (6394)
Na	3392 (3034)	2132 (4043)	977 (1258)
K	341 (417)	1406 (2665)	39 (42)
Ca	344 (329)	320 (605)	72 (52)
Mg	406 (599)	132 (188)	118 (166)
Cl	6047 (6151)	5577 (7950)	1512 (2158)
HCO_3	1507 (1326)	2292 (3126)	405 (221)
SO_4	459 (544)	366 (636)	231 (204)
NO_3	226 (398)	369 (671)	24 (31)

lated by averaging all borehole data. Although electrical conductivity is influenced by saline intrusion, mean electrical conductivity for groundwaters in reclaimed, landfill and non-infilled areas are 18768 μS/cm, 19,872 μS/cm and 5,489 μS/cm, respectively. Boreholes adjacent to Haslams and Powell Creeks and Homebush Bay have elevated electrical conductivity levels between 30,000 and 45,000 μS/cm with gradients declining towards the brick pit where the conductivity is about 4,000 μS/cm. High electrical conductivity values are mostly associated with waste materials in reclaimed and landfill areas and it is probably influenced by saline intrusion. Low electrical conductivity values occur in groundwater which are reclaimed types, probably due to recharge from rainwater and irrigation.

Mean Na, K, Ca and Mg concentrations in groundwater are markedly higher in reclaimed and landfill areas than in non-infilled areas and leachate concentrations of these elements are similar to groundwater from reclaimed and landfill areas (Table 2). Leaching, or dissolution of readily solubilized constituents in the refuse and degradation of the organic matter are important factors, but are not the only reactions that affect the composition of leachates (Vadillo et al., 1999). Most chloride and sulfate salts have moderately high solubilities and high concentrations of these materials in the leachate are usually attributed to the direct leaching of Na, K, Ca and Mg salts of these cations from the waste materials (Goldenberg and Melloul, 1994). Soluble salts, i.e. sodium chloride in domestic refuse readily dissolves in infiltrating water (Appleyard, 1996). Other major cations such as Ca, Mg and K can originate from refuse, but dissolution of minerals in the geologic cover materials may also occur (Nicholson et al., 1983). Upon entering the groundwater system, the leachate is subjected to physical, as well as chemical processes as it moves along flow paths. Leachate concentrations decrease with hydrodynamic dispersion due to chemical reactions, i.e. cation exchange, mineral dissolution and precipitation (Ford et al., 1992; Ekpo et al., 2000; Sanchez Navarro et al., 2000).

Na, K and Mg are the dominant cations in groundwater from reclaimed areas and Na and K are the dominant cations in groundwater in landfill areas. Mean concentrations of K, HCO_3, NO_3 and electrical conductivity in groundwater are highest in landfill areas, followed by reclaimed areas and non-infilled areas because these elements are related to anthropogenic contamination by waste materials. Cation contents in three types of groundwaters investigated have different hydrochemical characteristics. The mean abundance of cations in groundwater from non-infilled areas is Na>Mg>Ca>K, whereas the mean abundance of cations in groundwater in reclaimed and landfill areas is Na>K>>Ca>Mg, suggesting that K in landfill areas may be derived from anthropogenic waste materials. The order of mean abundances of anions in groundwater in reclaimed, landfill and non-infilled areas are similar i.e. $Cl>HCO_3>SO_4>NO_3$. The maximum electrical conductivity values determined at the Olympic site are higher than those found in landfill leachates described from other locations, e.g. 132 to 22,700 μS/cm (Kuajara et al., 1997); 7,600 to 7,700 μS/cm (Vadillo et al., 1999); 4,050 to 40,900 μS/cm (Ekpo et al., 2000). Groundwater in landfill and reclaimed areas has considerably higher TDS than the groundwater in non-infilled areas due to the varied nature of waste materials at the disposal site. The sum of cations in groundwater in reclaimed (4,483 mg/l) and landfill areas (4,981 mg/l) are markedly higher than in non-infilled areas (1,206 mg/l). The correlation of total cations in groundwater and TDS in reclaimed, landfill and non-infilled areas are closely related, i. e. r^2= 0.9 (n= 49), r^2= 0.84 (n= 37) and r^2= 0.99 (n= 15), respectively (Figs. 3a-c). Although the sum of total anions (Cl, SO_4, HCO_3, NO_3) in groundwater in reclaimed, landfill and non-infilled areas varies widely (mean= 8,239, 8,605 and 2,173 mg/l, respectively), TDS is closely related to the sum of anions in groundwater in relation to past land use. The correlation of total anions and TDS in groundwater in reclaimed, landfill and non-infilled areas is r^2= 0.89 (n= 49), r^2= 0.87 (n= 37) and r^2= 0.99 (n= 15), respectively (Figs. 3d-f). This close relationship suggests that waste materials could be a factor affecting the chemical characteristics of groundwater because the correlation of cations and anions in non-infilled areas and TDS is



Figs. 3a-f. Correlation between TDS and sum of cations and anions in groundwater in relation to past land use types.

^{2}= 0.99, compared to reclaimed areas and landfill reas which are markedly lower (r^2= 0.88; n= 86; <0.001).

4.2. Physico-chemical characteristics in relation to borehole types

The mean, range and standard deviation of physico-chemical parameters of groundwaters and leachates from the deep boreholes, shallow boreholes and standpipes in the study area are given in Table 3. Mean pH values in groundwater in deep boreholes, shallow boreholes and standpipes are similar with

Table 3. Mean of physico-chemical parameters of groundwater samples in deep boreholes, shallow bore holes and standpipes. All values are mg/l, except for EC (μS/cm).

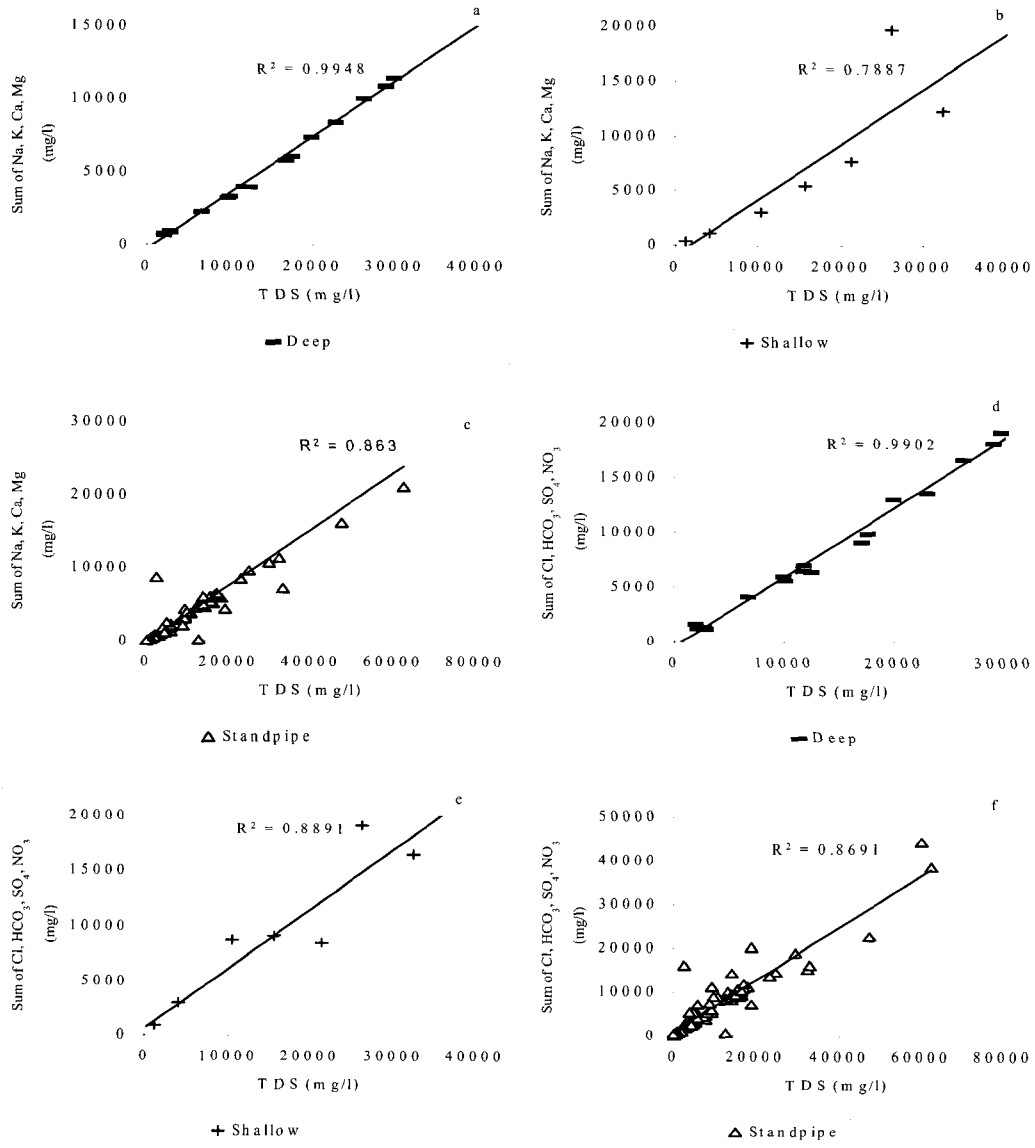
Parameter	Deep boreholes (n= 18)	Shallow boreholes (n= 9)	Standpipes (n= 74)
pH	7.4 (1.2)	7.12 (2.1)	7.10 (0.95)
EC	19753 (14383)	19336 (17941)	16320 (18702)
Na	3793 (2855)	3172 (3117)	2697 (3486)
K	107 (89)	1699 (2987)	704 (1690)
Ca	254 (266)	555 (1024)	273 (351)
Mg	535 (501)	143 (230)	211 (445)
Cl	6633 (5556)	5531 (5918)	4813 (6971)
HCO3	474 (218)	1612 (1803)	1915 (2397)
SO4	719 (568)	949 (1100)	244 (340)
NO3	59 (40)	469 (1141)	267 (432)

n= number of samples; ( )= standard deviation.

neutral to slightly alkaline characteristics, whereas in leachates mean pH value (mean: 7.78) is slightly higher. This is possibly due to rainwater infiltration of anthropogenic waste materials deposited at the disposal site. Mean electrical conductivity in leachates and in groundwater in deep boreholes, shallow boreholes and standpipes in the study area are very similar and all exceeded WHO (1984) permissible limits (1,400 μS/cm). However, the minimum and maximum values of electrical conductivity of groundwater in standpipes vary remarkably, possibility indicating the presence of perched ponds in the subsurface.

Na and Mg are the dominant cations in groundwater in deep boreholes, whereas Na and K are the dominant cations in groundwater in shallow boreholes and standpipes. Mean Cl concentrations in groundwater in deep boreholes, shallow boreholes and standpipes are 6,633 mg/l, 5,556 mg/l and 4,813 mg/l, respectively, whereas the mean Cl concentration in leachate is 15,590 mg/l. The highly variable natures of these concentrations are probably due to the proximity of Homebush Bay, Haslams and Powell Creeks. Most values exceeded WHO (1984) permissible limit (250 mg/l). Mean HCO_3 concentration in groundwater in deep boreholes is 474 mg/l, whereas mean values of HCO_3 for shallow boreholes, standpipes and leachates are 1,612 and 1,915 mg/l, respectively. Mean NO_3 concentration in groundwater in deep boreholes is 59 mg/l, whereas for shallow boreholes and standpipes and leachates mean values are >250 mg/l. All mean values exceed WHO standard (1984) limits of 45 mg/l, especially for groundwater in shallow boreholes, standpipes and leachates which exceed WHO limits by factors of 10, 5 and 9, respectively. Mean SO_4 concentrations in groundwater in deep and shallow boreholes and in leachates is >700 mg/l, whereas for standpipes the mean value is 244 mg/l. Mean SO_4 concentration in leachates exceeded the WHO standard (1984) limit of 500mg/l by a factor of two. These concentrations are an indication of the level of anthropogenic impact by waste materials at the disposal site.

Cation and anion contents of groundwater in the three types of boreholes investigated showed different hydrochemical characteristics. The order of mean abundance of major ions in groundwater in deep boreholes is Na>>Mg>Ca>K, while for anions it is $Cl>SO_4>HCO_3>NO_3$. The order of mean abundance of major ions in groundwater in shallow boreholes and standpipes is Na>K>Ca>Mg, while for anions it is $Cl>HCO_3>SO_4>NO_3$ and $Cl>HCO_3>NO_3>SO_4$, respectively. Elements such as K, HCO_3 and NO_3 are closely related to anthropogenic impact by waste materials and distributions are consistent with the depth of



Figs. 4a-f. Correlation between TDS and sum of cations and anions in groundwater in relation to borehole type.

fill materials in the study area. The sum of cations and anions in groundwater was highest in shallow boreholes, followed by deep boreholes and standpipes, while the sum of TDS in groundwater was highest in deep boreholes (13,037 mg/l), followed by shallow boreholes (12,761 mg/l) and standpipes (10,771 mg/l). The correlation of total cations in groundwater and TDS in deep boreholes, shallow boreholes and standpipes are closely related, i. e. r^2= 0.99 (n= 18), r^2= 0.79 (n= 9) and r^2= 0.86 (n= 74), respectively (Figs. 4a-c). The correlation of total anions and TDS in groundwater in deep boreholes, shallow boreholes and standpipes is r^2= 0.99 (n= 18), r^2= 0.89 (n= 9) and r^2= 0.87 (n= 74), respectively (Figs. 4d-f), probably due to the variety of

soils and waste materials penetrated by the different installation depths of these devices.

Figures 5a-b represent the relationship between Na and Cl in the groundwater and leachate with respect to rainwater and seawater, based on landuse and borehole type. The Na content of groundwater is directly related to Cl and the data fit the freshwater-seawater composition line except for leachates. The Na content is probably related to a salinity gradient in groundwater for both landuse type and borehole type.

4.3. Heavy metal concentrations in groundwater in relation to past land use and possible sources

Leachate is produced in landfills as a result of a number of chemical and biological processes. Leaching, or dissolution of the readily solubilized constituents in the refuse and degradation of the organic matter are important factors, but many other reactions affect the composition of leachates (Nicholson et al., 1983; Suh et al., 2003b). Nicholson et al. (1983) suggested that leachate is subjected to physical, as well as chemical processes as it is transported along flow paths. As hydrodynamic dispersion causes decreased concentrations and spreading of the plume, chemical reactions, such as cation exchange, mineral dissolution and precipitation occur throughout the zone of contamination. Heavy metals in leachate can be present in the form of free and exchangeable ions and organic and inorganic complexes, and can be precipitated with, or sorbed to, oxides of Fe, Al and Mn. They can also be bound within the crystalline lattice of primary minerals (Gambrell, 1994). The mobility of heavy metals is controlled primarily by a combination of dissolution/precipitation, sorption (and/or ion exchange) and complexation reactions. Iron and Mn are subject to valence state transformation in the range of pH and Eh conditions commonly found in bottom sediments of rivers, lakes and estuaries (Johnson et al., 1996). Suh et al. (2003b) suggested that there are a number of controlling the concentration, and hence migration, in the groundwaters associated with fill materials of the Bicentennial Park site at Rozelle Bay, Australia.

Table 4. Statistical results of the heavy metal concentrations, pH and electrical conductivity in groundwatersand leachate based on past land use.

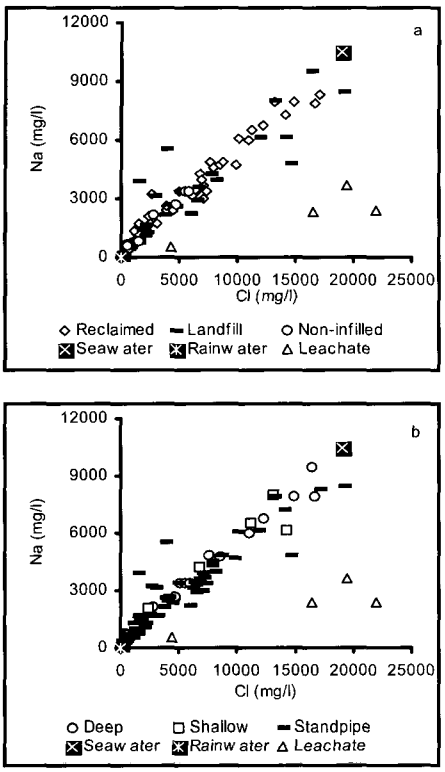
		pH	EC	Cu	Pb	Zn	Cr	Fe	Mn
Reclaimed area	Mean	7.02	18768	180	60	358	47	21900	1446
	Range	5.7-7.9	976-94900	5-1600	5-1300	30-1900	5-950	70-200000	20-10000
	SD	1.13	4513	280	180	380	150	39500	2000
	n= 49								
Landfill area	Mean	7.38	19872	1254	41	1074	222	28410	890
	Range	2.5-11.4	360-91000	5-17000	5-320	5-11500	5-3300	60-390000	10-13600
	SD	1.53	21187	3370	70	2200	660	87000	2285
	n= 37								
Non-infilled area	Mean	7.03	5489	177	17	562	6	12188	2240
	Range	5.2-11.7	320-17640	5-2300	5-110	20-3700	5-20	50-89000	5-6500
	SD	1.42	6394	590	30	1130	6	23990	1862
	n= 15								
Leachates	Mean	7.78	19625	57	13	79	12	2073	1950
	Range	7.4-8	6200-27500	20-85	0.5-50	4.5-170	3.2-29	173-6500	310-5300
	SD	0.26	9396	28	25	76	12	2994	2290
	n= 4								

SD= standard deviation, n= number of samples. All metal values in μg/l.
EC=Electrical conductivity in μS/cm.

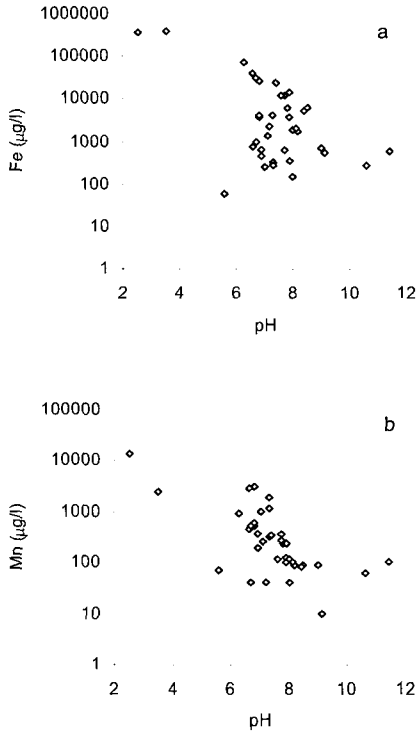
First, the redox behaviour of both the groundwater and its associated soil profiles can influence the behaviour of redox-sensitive elements, such as Mn and Fe. Second, during dry periods (Cu, Pb, Zn, As and Cr), where the water table recedes, oxygen ingress can lead to biologically catalyzed oxidation reactions resulting in a reduction in pH and an associated increase in the concentration of some trace elements. Finally, dilution of seawater by the ingress of freshwater from up gradient of the filled area decreases the solubility of some elements (Fe and Mn), relative to that of chloride (As, Cr and Cu), whilst increasing others (Pb, Zn, Ni, Co and Al).

The mean Cu, Zn, Cr and Fe concentrations in groundwater are highest in landfill areas, whereas the highest mean Pb concentration occurs in reclaimed areas (Table 4). Mean pH values for groundwater in reclaimed, landfill and non-infilled areas at the Olympic site range only from pH 7.02 to pH 7.38, i.e. from neutral to slightly alkaline. pH values for leachates in reclaimed and landfill areas range from 7.4 to 8.0 (mean = 7.78). It was calculated by averaging all borehole data. Heavy metal (Cu, Pb and Zn) concentrations in leachate are lower than in groundwater in other areas, possibly due to the influence of hydrous oxides of Fe and Mn and absorption by insoluble organic matter and clay minerals. Iron concentrations in groundwater in reclaimed, landfill, non-infilled areas are substantially higher than those found in leachates, whereas the mean Mn concentration in groundwater is highest in non-infilled areas and lowest in landfill areas. There is very little variance in Cu, Pb, Zn, Cr with pH in the groundwater between the reclaimed, landfill and non-infilled areas and in the leachates.

The cycling of Mn and Fe in aquatic systems with variable redox conditions is complex. Davison (1993) summarized the behaviour of these elements in lakes and pointed out that the distributions of Mn and Fe are influenced by redox conditions, the unique chemical characteristics of the metals, and physical mixing processes. The physical and chemical form of Mn and Fe differs across oxic-anoxic interfaces. Within the oxic regions, both elements exist in their oxidized forms (Mn(IV) and Fe(III)) as insoluble metal oxyhydroxides. These solid phases tend to strongly adsorb trace metals (Davis and Leckie, 1978; Catts and Langmuir, 1986). In anoxic environments, the elements are in their reduced forms (Mn(II) and Fe(II)) and exist as mobile metal ions, Mn^{2+} and Fe^{2+}. Random mixing processes influence the distributions of both dissolved and particulate phases while coagulation and gravitational settling also affects particulate phases. The settling of Mn and Fe oxyhydroxides through an oxic-anoxic interface results in the reductive dissolution of the solid phases while the diffusion of dissolved Mn(II) or Fe(II) from reducing to oxidizing conditions can result in the precipitation of oxyhydroxides phases (Balistrieri et al. 1994). Trace elements associated with metal oxyhydroxide phases should be released to the dissolved phase during reductive dissolution of the oxides, while precipitation of metal oxyhydroxides can potentially remove trace elements from solution by adsorption, or coprecipitation (Catts and Langmuir, 1986). There are two important differences in the behaviour of Mn and Fe. First, Fe(II) has a faster rate of oxidation than Mn(II). This difference can result in a physical separation of Mn and Fe oxyhydroxides because dissolved Mn(II) can be transported farther from the redox boundary than dissolved Fe(II) before oxidation. Second, Fe tends to be more involved in the sulfur cycle than Mn. Iron sulfide phases can form by direct precipitation of dissolved ferrous iron and sulfide or, possibly, by the reaction of dissolved sulfide with iron oxyhydroxides (Davison, 1993). Figures 6a-b show the relationship between pH and Fe and Mn in groundwater in landfill areas only. Fe concentrations in groundwater in landfill areas exhibit a different behaviour compared to other areas and leachates, in that Fe concentrations and pH in groundwater in landfill areas are highly variable, whereas groundwater in other areas and in leachates fall within a narrow pH range. In landfill areas, Fe and Mn show a distinct increase in concentration with declining pH (Figs.6a-b), a trend not observed for other metals (Cu, Pb, Zn and Cr). The mean heavy metal (Cr, Cu, Pb, Zn) concentrations in soils of the study area revealed the order of reclaimed



Figs. 5a-b. Relationship between the concentration of Na and Cl in groundwater with respect to past land use and borehole type.



Figs. 6a-b. Comparison between pH and Fe and Mn concentrations in groundwater in landfill areas.

(greatest), landfill and non-infilled area (smallest), whereas in groundwater it is all shown the order of landfill, reclaimed and non-infilled area. The correlations between heavy metals (Cr, Cu, Pb, Zn) in groundwater and soil are all very low, having r^2 values between 0.001 and 0.1. The inconsistent relationship between metal concentrations in soils and groundwater is due to the varied nature of the materials deposited at the Olympic Games site and the movement of groundwater in the study area. As water moves slowly through the subsurface, its composition gradually changes, reflecting an increasing saturation of some ions, or the results of various rock-water interactions. The current study has the limitation of not having the interpretation of mobility. The reason for the wide pH range in landfill groundwater is unclear, but probably lies with the heterogeneity of these materials. An important consequence of metal oxide dissolution in contaminated environments is the release of other heavy metals that may have been scavenged by Fe and Mn (Nicholson et al., 1983). These results are consistent with other research, which demonstrate a strong binding capacity of organic matter (Christensen et al., 1996) and a high affinity of heavy metals for colloidal material (Gounaris et al., 1993).

Although electrical conductivity is influenced by saline intrusion, mean electrical conductivity for groundwaters in reclaimed, landfill and non-infilled areas are 18,768 μS/cm, 19,872 μS/cm and 5,489 μS/cm, respectively, whereas mean electrical conductivity for leachates is 19,625 μS/cm (Table 4). The mean electrical conductivity values in the reclaimed areas, landfill areas and leachates are similar (18,768-19,872 μS/cm), but are much higher than that of non-infilled area (5,489 μS/cm), probably due to the proximity of saline bay and creek environments. These values exceeded WHO (1984) permissible limits (1,400 μS/cm). These reflect high dissolved solid waste in the groundwater and leachate samples. There is no distinct trend in heavy metals with electrical conductivity in the groundwater between the reclaimed, landfill and non-infilled areas and in the leachates. Boreholes adjacent to the Haslams and Powell creeks and Homebush Bay show elevated levels between 30,000 and 45,000 μS/cm with gradients developed towards the brick pit where much lower conductivities of 4,000 μS/cm are measured. Anomalous high electrical conductivity values are mostly associated with waste materials and saline intrusion in Areas 1, 2, 4, 7, 9 and 10. Low electrical conductivity values have also been measured in the south/ east within Areas 15 and 16, probably due to recent recharge from rainwater and irrigation.

Kaoser et al. (2000) reported that heavy metals can be categorized in three groups according to their mobility in clay soils, e.g. low mobility (Cu, Cr3+, Pb, Zn), moderate mobility (As, Cd, Fe, Ni) and high mobility (Hg, Cr^{6+}). High heavy metal concentrations in groundwater and leachates depend upon the nature and chemical composition of the waste deposited at the disposal site. Many of these materials are available at the Olympic Game site and may have acted as sources for the groundwater metals. The overall assessment of water quality of the groundwater indicates that although the maximum concentrations of these heavy metals in groundwater at the Olympic site are substantially elevated, the mean Cu, Pb and Zn concentrations are lower than WHO (1984) permissible limit, except for Cr. The current study revealed some strong correlations of heavy metals in combined groundwater samples from landfill, reclaimed and non-infilled areas. Strong heavy metals relationships exist for Cu and Zn 0.79; Pb and Zn 0.74 ($n = 37$, $p < 0.01$), Cu and Zn 0.59 ($n = 49$, $p < 0.01$) and Cu and Pb 0.94; Cu and Zn 0.57 ($n = 15$, $p < 0.05$). These relationships suggest that these metals in groundwater in the Olympic Games site are either sourced from the same materials or are being transported by similar processes, or both.

4.4. Heavy metal concentrations in groundwater in relation to borehole types

The mean, range and standard deviation of the levels of physicochemical parameters of deep borehole, shallow borehole, standpipe groundwater and leachate in the study area are presented in Table 5. Mean Cu, Pb, Zn and Cr concentrations are highest in groundwater taken from standpipes and lowest from leachates, except for Cr which is very close to being the lowest in leachate. Iron concentrations in groundwater are highest in shallow boreholes and lowest in leachates, whereas Mn shows the reverse distribution (Table 5). Mean Cu, Zn and Cr concentrations decrease markedly with depth, i.e. standpipe-shallow borehole-deep borehole, a trend that is similar for Pb, but not as strong. As expected, the mean concentrations of these metals are highest in the upper parts of the subsurface in close proximity to the contaminated material. The stability of these metals may be explained by coprecipitation of metals with hydrous oxides of Fe, Al and Mn which has been shown to play an important role in some waste areas, e.g. Cu, Mn and Zn are coprecipitated in Fe oxides, while Fe, Pb and Zn are coprecipitated in Mn oxides at the deposition site (Bagchi, 1990).

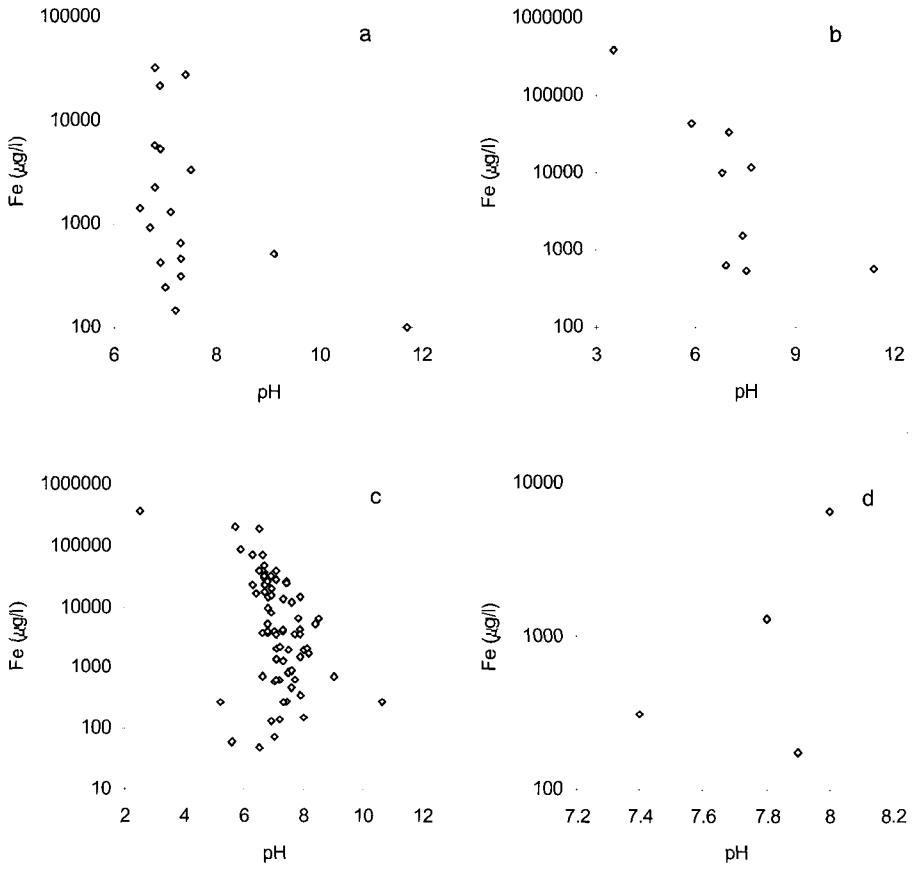
There is no correlation between Cu, Pb, Zn, Cr concentrations in groundwater with pH, from deep boreholes, shallow boreholes and standpipes and in leachates, except for Fe and Mn, which demonstrate increasing concentrations with declining pH. It is apparent from a study of coring devices that the varied

Table 5. Statistical results of the heavy metal concentrations, pH, and electrical conductivity in ground waters and leachate based on borehole types.

		pH	EC	Cu	Pb	Zn	Cr	Fe	Mn
Deep boreholes	Mean	7.4	19753	89	19	464	11	5851	1477
	Range	6.5-11.7	2810-45000	5-910	5-50	5-3700	5-40	100-32000	5-5100
	SD	1.2	14383	0.21	0.01	0.9	0.01	1018	1420
	n= 18								
Shallow boreholes	Mean	7.12	19336	104	14	489	64	54691	1273
	Range	3.5-11.4	1720-49300	10-410	5-30	100-1600	5-420	540-390000	100-7100
	SD	2.1	17941	0.14	0.01	0.63	0.14	126700	2310
	n= 9								
Standpipes	Mean	7.10	16320	748	57	716	673	23106	1341
	Range	2.5-10.6	320-94900	5-17000	5-1300	5-11500	5-13600	50-375000	20-13600
	SD	0.95	18702	2.44	0.16	1.62	1.87	54200	2250
	n= 74								
Leachates	Mean	7.78	19625	57	13	79	12	2073	1950
	Range	7.4-8	6200-27500	20-85	0.5-50	4.5-170	3.2-29	173-6500	310-5300
	SD	0.26	9396	28	25	76	12	2994	2290
	n= 4								

nature of pH in the landfill areas occurs mainly in the upper subsurface (groundwater from standpipes). The relationship between pH and Fe and Mn in groundwater in deep boreholes, shallow boreholes, standpipes, and leachate is presented in figures 7a-d, 8a-d, respectively. The Fe concentration trends in groundwater in shallow boreholes and standpipes generally exhibit a more varied pH distribution, whereas deep boreholes and leachates have a very narrow pH range (Figs. 7a-d). The Mn concentration trends in groundwater in shallow boreholes and standpipes generally exhibit a more varied pH distribution, whereas deep boreholes and leachates have a very narrow pH range (Fig. 8a-d). The varied nature of pH distributions in the shallower devices probably reflects the heterogeneous character of materials in the upper parts of the subsurface. The wide range in metal concentrations displayed by the other elements i.e. Cu, Pb, Zn and Cr may be explained by the influence of metal oxide coatings. Nicholson et al. (1983) reported that because reducing conditions exist in refuse, the leachate is capable of reducing Fe(III) and Mn(IV) which promotes the dissolution of metal oxides. At slightly acid pH and moderately reducing conditions, Fe^{2+} and Mn^{2+} are reasonably mobile because their concentrations are not generally controlled by the solubility of mineral phases (Hem, 1972).

Electrical conductivity in groundwater in the deep boreholes, shallow boreholes and standpipes in the study area range from 2,810 to 45,000 μS/cm (mean = 19,753 μS/cm), 1,720 to 49,300 μS/cm (mean = 19,336 μS/cm) and 320 to 94,900 μS/cm (mean = 16,320 μS/cm), respectively. Electrical conductivity values in leachates range from 6,200 to 27,500 μS/cm (mean = 19,625 μS/cm) (Table 5). These values exceeded WHO (1984) permissible limits (1,400 μS/cm). Mean electrical conductivity values in the deep and shallow boreholes are higher than that of standpipes, but the minimum and maximum value of electrical conductivity in groundwater in standpipes shows remarkably different value, probably due to perched pond. The Mn concentration trends in groundwater in deep boreholes and shallow boreholes generally exhibit a more positive electrical conductivity distribution, whereas standpipes and leachates show



Figs. 7a-d. Comparison between pH and Fe concentrations in groundwater in relation to borehole types, a) deep boreholes; b) shallow boreholes; c) standpipes; d) leachates, respectively

poor relationship. The Fe concentration trends in groundwater in deep boreholes, shallow boreholes, standpipes and leachates exhibit no distinct distribution.

4.5. Hydrochemical characteristics

Geochemical results reveal that groundwaters in deep shale and in shallower saline (>10 ppt) reclaimed/infilled areas are dominated by sodium chloride, whereas non-infilled areas (<10 ppt) exhibit high sodium bicarbonate to bicarbonate and chloride compositions. The groundwater types of the study area are characterized by their position on a Piper diagram (Piper, 1944; Ophori and Toth, 1988). Based on the major cations and anions in the deep boreholes, shallow boreholes and standpipes, four groundwater types are evident in the study area. These are: Na-Cl, Na-HCO_3, Na-SO_4 and Ca-HCO_3 (see Suh et al., 2003a). The deep aquifer system suggests a uniform class of water with predominant Na-Cl characteristics and indicates minimal impact from shallow aquifers, except through dilution by precipitation. The formation of NaCl water in the deep borehole is a result of progressive salinization.

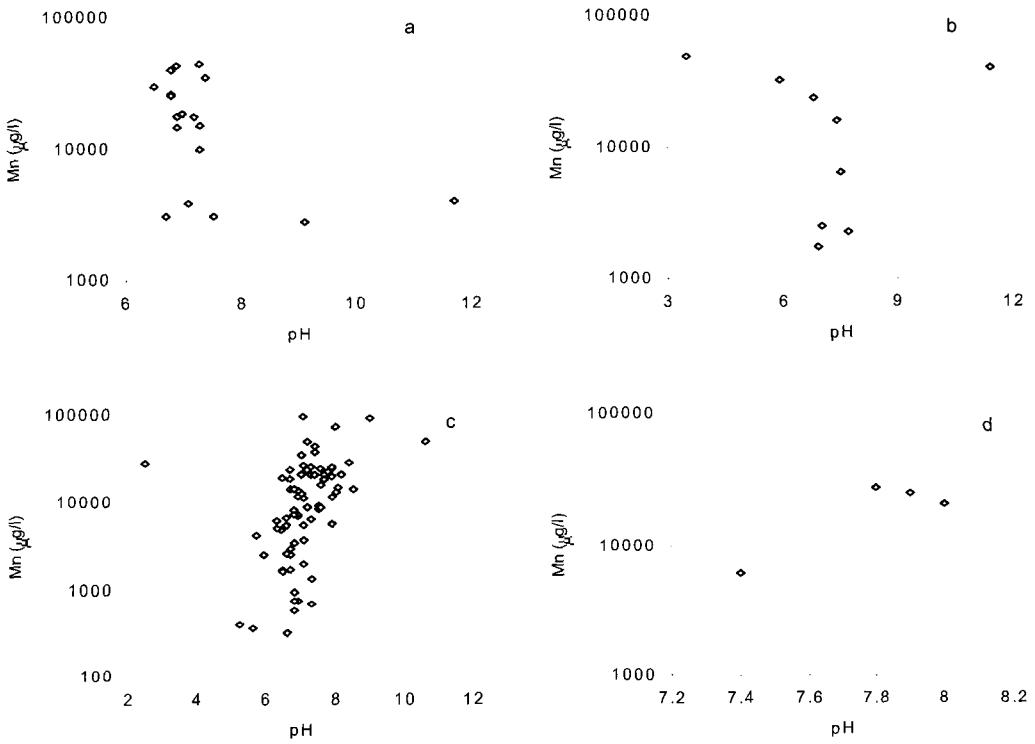
At higher salinities, the progress of reverse cation exchange may create $CaCl_2$ waters due to the removal of Na out of solution for bound Ca. The formation of water in both shallow boreholes and standpipes appeared to be various types of waters due to differently installed depth and varied waste materials and seawater intrusion. Processes that may be responsible for the formation of the various water types in the shallow and standpipes are salinization, ion exchange and SO_4 reduction (Adams et al., 2001).

Freeze and Cherry (1979) reported that the replacement of Ca by Na through cation exchange results in the formation of $Na(HCO_3)_2$ waters. Removal of Ca through the precipitation of carbonate minerals may cause a disturbance in the chemical equilibrium resulting in the dissolution of minerals containing Ca such as gypsum. In contrast, the shallow aquifer system, where present, indicates a different groundwater regime/or mixing processes. Groundwaters in the shallow system have migrated through numerous different materials including fills, waste materials, etc. Due to the various types of industry that have opportunity to impact the quality of water in this zone, water samples tend to exhibit widely varying signatures. Unlike the deeper samples, Na-Cl characteristics are no more dominant than other ionic species.

The following significant correlation between different chemical pairs was observed in the study area (n= 101, p> 0.001). Na/Cl (r = 0.98), Cl/Conductivity (r = 0.94), Na/Conductivity (r = 0.92), K/Conductivity (r = 0.65), K/Cl (r = 0.64), Na/K (r = 0.62), Mg/Cl (r = 0.62), Na/Mg (r = 0.61), Mg/Conductivity (r = 0.58), K/NO_3 (r = 0.54), Na/NO_3 (r = 0.51), Cl/NO_3 (r = 0.51). The strong relation between conductivity and Na was interpreted as saline water intrusion into the wells. Na concentration directly depends on the seawater intrusion into the groundwater horizons. NO_3 ion shows a strong correlation with major elements such as Na, K and Cl, resulting in the amount of waste materials. On the other hand, a very weak inverse correlation was observed pH/Mg (r = -0.07), pH/SO_4 (r = -0.24), pH/NO_3(r = -0.05), K/Mg (r = -0.07), Ca/NO_3 (r = -0.05), Mg/HCO_3 (r = -0.06), and HCO_3/SO_4 (r = -0.13). The correlation matrix indicates that groundwater compositions in the study area may fall into two main groups, i.e. industrial/domestic waste source (K and NO_3) and materials related mainly to saline intrusion (Na and Cl).

The characteristics of groundwater from the study area can therefore be summarized as follows: The cation contents in three types of landuse areas investigated and in three types of boreholes examined showed different hydrochemical characteristics. Groundwaters in reclaimed and landfill areas were characterized by high concentrations of K, Mg and Ca relative to groundwater in other landuse areas, whereas groundwaters in non-infilled areas contained minor concentrations of all cations analyzed. Groundwaters in the deep boreholes were characterized by Mg and Na concentrations which were consistent with seawater dilution, whereas groundwaters in the shallow boreholes and standpipes were enriched in K. Anion contents in three types of areas investigated and in three types of boreholes studied showed different hydrochemical characteristics. Groundwaters in reclaimed and landfill areas were characterized by high concentrations of Cl, HCO_3, SO_4 and NO_3 relative to groundwater in non-infilled areas, whereas groundwater in non-infilled areas contained relatively low concentrations of all anions analysed. Groundwaters in the deep boreholes were characterized by Cl and SO_4 concentrations, whereas groundwaters in the shallow boreholes and standpipes were enriched in HCO_3 and NO_3.

Mean concentrations of Cu, Zn, Cr and Fe in groundwater are highest in landfill areas, whereas the highest mean Pb groundwater concentrations are in reclaimed areas. The lowest concentrations of all metals are in groundwater from non-infiiled areas of the Olympics site. Leachate concentrations of Cu, Pb, Zn and Fe are lower than in groundwater from all three areas. Mean Mn concentrations in groundwater exhibits a different trend in thatit is highest in non-infilled areas and lowest in landfill areas. The low groundwater concentrations in non-infilled areas are probably due to the absence of waste material in



Figs. 8a-d. Comparison between pH and Mn concentrations in groundwater in relation to borehole types, a) deep boreholes; b) shallow boreholes; c) standpipes; d) leachates, respectively.

these parts of the site. The concentrations of metals in leachates are low compared to groundwater, possibly because the metals are bound by hydrous oxides, insoluble organic matter and clay minerals. No relationship could be established between soil and groundwater concentrations of heavy metals (Cu, Pb, Zn, Cr) in the landfill, reclaimed and non-infilled areas of the Olympic site. The lack of a consistent relationship between metal concentrations in soils and groundwater is probably due to the varied nature of the materials deposited at the Olympic site and movement of groundwater in the study area. Two distinct groundwater systems operate at the Olympic Games site; a discontinuous group of perched aquifers formed essentially by fill and other superficial materials, and a deeper system within fractures in the underlying shale and sandstone. Unfortunately, the current study did not monitor the potential flow of contaminants to the estuary based on the various groundwater flows resulting from the measured borehole hydraulic gradients.

5. Conclusions

The results obtained from the current study reveal a close association between waste materials and the concentration of major elements, based on past land use and borehole types. Cation contents in three types of groundwaters investigated have different hydrochemical characteristics. The mean abundance

of cations in groundwater from non-infilled areas is Na>Mg>Ca>K, whereas the mean abundance of cations in groundwater in reclaimed and landfill areas is Na>K>>Ca>Mg, suggesting that K in landfill areas may be derived from anthropogenic waste materials. The order of mean abundances of anions in groundwater in reclaimed, landfill and non-infilled areas are similar i.e. $Cl>HCO_3>SO_4>NO_3$. The hydrochemistry of the groundwaters in reclaimed and non-infilled areas are characterized by Mg and Ca enrichment, whereas groundwaters in landfill areas are elevated in K and NO_3. Groundwater in landfill and reclaimed areas has considerably higher TDS than the groundwater in non-infilled areas due to the varied nature of waste materials at the disposal site. However, the order of mean abundance of major ions in groundwater in deep boreholes is Na>>Mg>Ca>K, while for anions it is $Cl>SO_4>HCO_3>NO_3$. The order of mean abundance of major ions in groundwater in shallow boreholes and standpipes is Na>K>Ca>Mg, while for anions it is $Cl>HCO_3>SO_4>NO_3$ and $Cl>HCO_3>NO_3>SO_4$, respectively. Based on the major cations and anions in the deep boreholes, shallow boreholes and standpipes, four groundwater types are evident in the study area. These are: Na-Cl, Na-HCO_3, Na-SO_4 and Ca-HCO_3.

There is no distinct trend in heavy metals with electrical conductivity in the groundwater between the reclaimed, landfill and non-infilled areas and in the leachates. Anomalous high electrical conductivity values are mostly associated with waste materials in Areas 1, 2, 4, 7, 9 and 10. Low electrical conductivity values have also been measured in the south/ east within Areas 15 and 16, probably due to recent recharge from rainwater and irrigation. Heavy metal (Cu, Pb and Zn) concentrations in leachate are lower than in groundwater in other areas, possibly due to the influence of hydrous oxides of Fe and Mn and absorption by insoluble organic matter and clay minerals. There is very little variance in Cu, Pb, Zn, Cr with pH in the groundwater between the reclaimed, landfill and non-infilled areas and in the leachates. Mean electrical conductivity values in the deep and shallow boreholes are higher than that of standpipes, but the minimum and maximum value of electrical conductivity in groundwater in standpipes shows remarkably different value, probably due to perched pond. There is no correlation between Cu, Pb, Zn, Cr concentrations in groundwater with pH, from deep boreholes, shallow boreholes and standpipes and in leachates, except for Fe and Mn, which demonstrate increasing concentrations with declining pH. It is apparent from a study of coring devices that the varied nature of pH in the landfill areas occurs mainly in the upper subsurface (groundwater from standpipes).

Understanding hydrogeochemical processes in reclaimed lands bordering the estuary is critical for predicting contaminant transport and the interaction between groundwater and saline water. However, to determine the total amount of elements entering from the reclaimed lands, the current study requires further study both spatially hydrogeological movement and interaction between salt water and freshwater with large number of boreholes across the entire area.

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