



A survey of emerging contaminants in the estuarine receiving environment around Auckland, New Zealand



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HIGHLIGHTS

- The distribution of emerging contaminants was investigated in the urban environment of Auckland.
- Environmental concentrations were largely similar to those reported world-wide.
- Increased concentrations were observed in the vicinity of wastewater discharge and sewage overflows.
- Measurement of multiple pharmaceuticals in estuarine sediments augments sparse literature.

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ABSTRACT

Increasing urbanisation in the future will put mounting stresses on the receiving environments around those urban centres due to increased sedimentation and contaminant runoff. Emerging contaminants (ECs) are an extensive array of chemicals and many are not under regulatory action. Within New Zealand likely future pressures from ECs will be in both urban centres and rural areas due to intensive agriculture, although at present there is a lack of information on the state of the environment in both sectors. This study was initiated to gauge the distribution of ECs in the urban environment by measuring concentrations of flame retardants, plasticisers, alkylphenols, herbicides and pesticides, steroid oestrogens, pharmaceuticals and heavy metals in sediment from 13 estuarine sites around Auckland, New Zealand's biggest city. Total polybrominated diphenyl ether (PBDE) flame retardant concentrations (\sum_{PBDE}) ranged from 0.55 to 573 ng/g (dw). The phthalate plasticiser di(2-ethylhexyl)phthalate (DEHP) was measured at up to 11,500 ng/g from one site. Nonylphenol (NP) was found at up to 32,000 ng/g at one site adjacent to the city's major wastewater treatment plant (WWTP). However, median concentrations of NP were 153 ng/g, suggesting this site was not representative of the region. Nonylphenol mono- and di-ethoxylates (NPEO_{1,2}) had highest concentrations (1600 ng/g) at a marina. Highest glyphosate concentrations (up to 950 ng/g) were observed at residential sites. Steroid oestrogens were detected at extremely low concentrations (maximum 2.2 ng/g), while all other pesticides or herbicides were not detected at any sites. Multi-residue analysis of 46 pharmaceuticals showed presence of 21 compounds at one or more sites, with average concentrations ranging from 0.16 to 7.66 ng/g. Generally, environmental concentrations of ECs were similar to those reported world-wide. However, comparisons for pharmaceuticals were problematic, due to very few studies on pharmaceutical concentrations in estuarine sediments, with most focussed on sewage and stream water phases.

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1. Introduction

The world is becoming increasingly urbanised. Currently around 50% of the world population resides in urban areas; however that is expected to increase to around 59% by 2030 and 69% by 2050. In the

next 40 years it is expected that around 2.8 billion more people will live in urban areas (United Nations, 2009). Increasing urbanisation of the world population will place additional stresses not only on infrastructure but also on the receiving environments around those urban centres.

A major consequence of increased urbanisation – due to associated habitat loss and erosion – is increased loads of sediment in storm water, which ultimately ends up in the marine receiving environment.

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These sediment loads can contain historic Priority Organic Pollutants (POPs) such as polychlorinated biphenyls (PCBs), and organochlorine pesticides (OCPs). Higher population density also leads to higher input of current industrial chemical contaminants including those known for some time to be of environmental concern; i.e., heavy metals (especially zinc, copper and lead) and polycyclic aromatic hydrocarbons (PAHs); and those which are only recently coming to prominence for environmental regulators, i.e., emerging contaminants (ECs).

ECs are those that are suspected to have harmful effects – e.g. endocrine disruption and antibiotic drug resistance – but are not routinely monitored or regulated against. There is already a growing worldwide concern about ECs, particularly in urban areas and in aquatic environments where there are pressures on limited water resources. ECs may not necessarily have the toxic effects of POPs, with their associated food-chain bioaccumulation and biomagnification; however, many POPs have been banned under the Stockholm Convention (UNEP, 2010) so their input has declined or stopped since that time. In contrast, many ECs are still being produced and are a current source to the receiving environment.

Research on ECs is further complicated as there is an extensive array of different chemicals entering the aquatic environment, their volume is increasing and new compounds are continually being introduced to the market. As many ECs are continually being introduced into the environment they can be considered to be “pseudo” persistent (Daughton, 2002).

Some ECs have recently been subjected to international regulatory attention. Four brominated diphenyl ether (BDE) flame retardants were added to Annex A of the Stockholm Convention (UNEP, 2010). Certain flame retardants, pesticides, and surfactants are now priority contaminants under the European Commission Water Framework Directive (European Commission, 2008). Selected pesticides and pharmaceuticals are included in the United States Environmental Protection Agency Contaminant Candidate List 3 for drinking water (US EPA, 2009). Furthermore, these lists are not static and are frequently updated. For example, in the EU, diclofenac, 17 β -estradiol (E2) and 17 α -ethynylestradiol (EE2) will soon to be included in a watchlist of emerging pollutants that could one day be placed on the priority list (European Parliament, 2013). However, New Zealand does not have any current regulatory policy on ECs. As such, State of the Environment monitoring programmes in New Zealand are specifically tailored around legacy POPs (e.g. DDTs; PCBs). There is a clear need to assess the state of the environment around New Zealand, particularly in heavily urbanised areas, with respect to ECs.

Auckland is New Zealand's biggest city and has a population density similar to New York or Sydney (CityMayors, 2013). The population in the Auckland region is currently around 1.5 million people and is expected to increase by a further 1 million over the next 30 years. Much of this growth is expected in the urban areas (Auckland Council, 2013a). As such, Auckland is the most relevant area within New Zealand to survey environmental concentrations of urban ECs and address a knowledge gap about the current state of the environment.

2. Materials and methods

2.1. Auckland region

The greater Auckland region stretches from Wellsford in the north to Pukekohe in the south. Of the 16,141 km² total area, 69% is made up of sea, while 3% consists of numerous islands. The mainland contains 1800 km of coastline. Most of the land consists of low but occasionally steep hills with patches of native bush, exotic forest and scrublands (Auckland Regional Council, 2010). The major urban area (Fig. 1: dark grey) is situated between Waitemata Harbour in the north and Manukau Harbour in the south.

2.2. Sampling sites

Thirteen sites were selected for sampling around the greater Auckland region (Fig. 1). All sites except marinas were intertidal and were selected to cover a range of land uses which are defined in Table 1. Using the classification system of Folk (1968) all sites were classified as mud, with the exception of Cocks Bay (muddy sand) and Hobson Bay (sandy mud). Total organic carbon (TOC) ranged from 0.6 to 7.5 g/100 g, with a median of 2.3 g/100 g (Table 1). Approximately half the sites – Meola, Motions, Milford Marina, Westhaven Marina, Hillcrest and Mahurangi – have mixed land use influence, with combined sewage and industrial overflows the main contributors. Within central Auckland, there are 100 known locations where wastewater overflows during wet weather, with planned future upgrades to wastewater tunnels underway to address this (Watercare, 2013). However, potential contributions from landfill leachate were noted at Meola, Motions and Hillcrest. In addition, three subtidal marinas (Milford, Westhaven and Halfmoon Bay) and agriculturally influenced sites (Taihiki River and Mahurangi) were sampled. The Mahurangi site (58 km north of central Auckland) was sampled downstream of Warkworth sewage treatment plant, while Puketutu Island site was situated in an open estuarine site of what is the now decommissioned and remediated oxidation ponds of Auckland's major Waste Water Treatment Plant (WWTP).

2.3. Sampling methods

To avoid potential contamination with plasticisers, only plastics that do not leach phthalates were used, and only for collection. All glassware, plastics and utensils used for sampling were washed with detergent and rinsed with deionised water and acetone prior to use.

To remove site's spatial variability from the final results, each site (with the exception of the marinas) was marked with a quadrat of 50 × 50 cm and 2 replicate samples taken randomly within that quadrat. Sediments were collected using three different protocols. Where sediment could hold its form without collapsing, polypropylene housings were used to take sediment samples. The top 3 cm was extruded through the corer and transferred immediately into clean solvent rinsed glass jars and stored, on ice. For sites with sediment that would not hold its form, or had a high density of mangroves, a plastic polypropylene scoop was used to scrape off the top 3 cm. A Jenkins Perspex (poly(methyl methacrylate)) corer was used to sample subtidal sediments in marinas, with the sediment extruded as described above. Replicate samples of approximately 2 kg were collected in March 2008.

2.4. Sediment processing

Sediments were stored at 4 °C until processing. Each replicate sample was transferred to a large foil tray and stirred with a stainless steel spoon to form a homogenised mixture. Large debris (stones, shellfish, plant material) were removed at this time by hand. Once a homogenised sample was obtained, sub-sampling was carried out into relevant pre-cleaned labelled foil trays. A subsample was freeze dried with a shelf temperature of –20 °C and dry sieved (<500 μ m) to obtain a sample for metal and pharmaceutical analysis. All organic analyses, with the exception of pharmaceuticals, were performed on whole wet sediment. Freeze dried sediment was archived at –20 °C in glass jars with foil liners on the lids to avoid potential for leaching.

2.5. Analysis of contaminants in sediment

The contaminants chosen for analysis were based on information from a review on ECs of potential environmental concern in Auckland (Ahrens, 2008) and the logistics in finding analytical laboratories that provided robust analytical measurements of these contaminants. All analyses were undertaken between April and October 2008, with the exception of pharmaceuticals which were undertaken in May 2010.

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