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The impact of variations of influent loading on the efficacy of an advanced tertiary sewage treatment plant to remove endocrine disrupting chemicals

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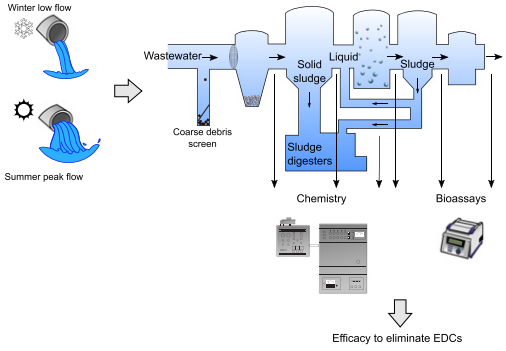
HIGHLIGHTS

- Efficacy of advanced tertiary treatment to remove endocrine disrupting chemicals
- Holiday season (High) and winter (low) flows tested using chemistry and bioassays
- Results show that the efficacy of the system was not reduced by large flow

GRAPHICAL ABSTRACT



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ABSTRACT

The impact of changes in influent load on the removal of endocrine disrupting chemicals (EDCs) by sewage treatment has not been fully characterised. This study assessed the efficacy of an advanced tertiary sewage treatment plant (STP) to remove EDCs during normal and peak flow events of sewage influent using trace chemical analysis of selected EDCs and four estrogenic in vitro bioassays. During the summer holiday season, influent volume increased by 68%, nutrient concentrations by at least 26% and hydraulic retention time was reduced by 40% compared with base flow conditions. Despite these pressures on the treatment system the concentrations and mass loading of estrone, 17β-estradiol, estriol, Bisphenol A, 4-*t*-octylphenol and technical nonylphenol were not significantly higher ($p > 0.05$) during the peak flow conditions compared with base flow conditions. Chemical analysis and in vitro bioassays showed that the efficacy of the STP in removing EDCs was not affected by the different loadings between baseline and peak flow regimes. This study demonstrates that large flow variations within the design capacity of advanced multi-stage STPs should not reduce the removal efficacy of EDCs.

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1. Introduction

Endocrine disrupting chemicals (EDCs) have been detected in sewage effluents worldwide (Baronti et al., 2000; Servos et al., 2005a; Vethaak et al., 2005b) including Australia (Braga et al., 2005a; Leusch et al., 2006a; Scott et al., 2014; Tan et al., 2007a). Estrogenic compounds detected in sewage treatment plant (STP) effluents can cause developmental and behavioural abnormalities in biota, particularly fish (Batty and Lim, 1999; Diniz et al., 2005; Jobling et al., 2006; Tetreault et al., 2011). Toxicity Identification Evaluation (TIE) investigations have demonstrated that steroidal estrogens (excreted by humans) and alkylphenol ethoxylate (APEO) degradation product nonylphenols are the major causative agents of endocrine disruption (Desbrow et al., 1998), the former being more important in STP effluent due to their potencies (Jarosova et al., 2014). The removal or reduction of EDCs to levels that minimise the risk to ecosystems and human health should be the objective of effective wastewater treatment. For example, treatment objectives for main estrogens like ethinylestradiol (EE_2) and 17β-estradiol (E_2) should be below their respective predicted no effect concentrations (PNECs) of 0.1 and 2 ng/L, derived from fish reproduction studies (Caldwell et al., 2012).

Many treatment technologies to remove EDCs from STP effluent have been investigated (Silva et al., 2012). Standard secondary wastewater methodologies such as activated sludge (AS) and trickling filter treatment provide limited (for trickling filters) or highly variable removal of EDCs from wastewater (for AS). In some cases, treatment can lead to an increase in estrogenic activity due to incomplete degradation and deconjugation of conjugated estrogens and conversion of non-estrogenic APEOs to their active metabolites - nonylphenol and octylphenol (Carballa et al., 2004; Servos et al., 2005a). Activated sludge treatment can achieve high levels of EDC removal (>90%) but is influenced by system design, influent quality and operational conditions. Advanced treatment technology can reduce the concentration of estrogenic EDCs in final STP effluent to near or below the analytical limits of detection (sub-ng to ng levels) (Gunnarsson et al., 2009; Leusch et al., 2005). There is limited information to assess EDC removal efficacy of advanced tertiary sewage treatment systems experiencing influent flow variations.

The aim of this study was to determine if an operational state-of-the-art STP continued to effectively remove EDCs while experiencing large fluctuations in flow. This study applied a combination of chemical analysis and in vitro bioassays to measure the concentrations of selected EDCs (including natural estrogens, Bisphenol A (BPA) and triclosan) and estrogenic activity as sewage effluent progressed through the treatment stages within a modern multi-stage advanced STP.

2. Materials and methods

2.1. Chemicals and materials

Granular anhydrous sodium sulphate 10–60 mesh (Mallinkrodt) and Mallinkrodt Nanograde organic solvents were from Biolab, Auckland, NZ. Hyflo Supercel filter aid, potassium carbonate and trifluoroacetic acid anhydride were purchased from Sigma-Aldrich NZ Ltd., Auckland NZ. Water Oasis HLB solid-phase extraction cartridges were purchased from Global Science, Auckland NZ. International Sorbent Technologies florisil solid-phase extraction cartridges and bulk aminopropyl adsorbent were purchased from John Morris Scientific, Auckland, NZ. Bond-Elut FL florisil SPE cartridges were purchased from Agilent Technologies, Australia.

Deuterated surrogate standards 17α-ethinylestradiol-d_4 (EE2-d_4), Estrone-d_4 (E1-d_4), 17β-estradiol-d_4 (E2-d_4) and 4-*n*-nonylphenol-d_8 (4-nNP-d_8) were purchased from C/D/N Isotopes Inc., Quebec, Canada. The deuterated surrogate standard bisphenol-A-d_{16} (BPA-d_{16}), the estrogenic steroid hormone estrone (E1), 17β-estradiol (E1), 17α-estradiol, estriol (E3), and 17α-ethinylestradiol (EE2); the industrial phenolic xenoestrogen 4-*tert*-amylphenol, 4-*n*-nonylphenol, 4-*n*-octylphenol, 4-*tert*-octylphenol, and technical nonylphenol; the faecal sterol coprostan, coprostan-3-ol, coprostan-3-one; the antimicrobial triclosan; and the plasticiser bisphenol-A were supplied by Sigma Aldrich NZ Ltd., Auckland, NZ. The internal standard chemicals 17β-estradiol diacetate, estriol triacetate, and coprostanol acetate were supplied by Steraloids Inc., New Hampshire, USA.

2.2. Study site

The Gerringong-Gerroa sewage treatment plant (GGSTP) located in coastal NSW, Australia, is an advanced tertiary STP with a capacity to treat 2.2 ML/day (11,000 Equivalent Population). The base population is around 4000 people but can double over summer resulting in increased influent flows and associated waste load.

The treatment train comprises screening, grit capture, BioDenipho phased isolation ditch system for the AS treatment comprising anaerobic, biological and post-denitrifying tanks. This is followed by clarification, continuous backwash sand filtration, ozone-enhanced biofiltration using biologically activated carbon (O_3-BAC), membrane microfiltration and UV disinfection (see Boake (2006) for description of the system). The resulting high quality effluent is stored in a 50 ML dam and is subsequently used to irrigate pasture. Fig. 1 diagrammatically shows the treatment train and sample collection points used in this study.

2.3. Monitoring of GGSTP performance parameters

As part of the operational license, the GGSTP monitors influent and effluent quality weekly for pH, chemical oxygen demand (COD), total suspended solids (TSSs), ammonia, total nitrogen and total phosphorus using National Association of Testing Authorities (NATA) accredited methodologies. The continuously recorded influent flow rate at the GGSTP was used to estimate hydraulic retention time and sludge age.

2.4. Sample collection periods

Whole liquid effluent samples were collected in November 2006 for baseline or "normal" operational in-flow conditions and in December 2006–January 2007 during peak operational in-flow conditions over the summer holiday season. Over this period, air temperature did not vary greatly; min-max 15.3–23.0 °C for November 2006, 16.3–22.6 °C December 2006, and, 18.0–24.3 °C January 2007 (Australian Government Bureau of Meteorology).

Process water samples from each treatment stage (Fig. 1) were collected within a one hour period on three separate days during the normal and peak flow periods. Samples were collected at 10 am, 2 pm and 5 pm to cover diurnal flow patterns on days separated by at least 48 h to minimise non-independence. Influent grab samples were collected after the screening and grit capture stages. Grab samples representing AS (biological treatment stage) treatment were collected from the post-denitrifying tank. Clarifier, sand filter, O_3/BAC, microfiltration, UV disinfection and storage dam grab samples were all obtained at outlet pipes from these treatment stages (Fig. 1).

2.5. Sample collection and extraction

Process water samples were collected in four-litre, solvent-rinsed, amber glass bottles, adjusted to pH 2 with concentrated H_2SO_4 for preservation and kept at 4 °C until extraction within 24 h. One litre of effluent was prefiltered through a 1.2 μm pore size filter (Advantec GC50, 47 mm or 90 mm diameter). The filtration of influent, post-DN tank, clarifier and sand filter samples required the addition of a 5–10 mm layer of filter-aid (Hyflo Supercel) on top of the glass fibre filter to reduce clogging (Gadd et al., 2010). After filtering, the samples destined for chemical analysis were spiked with 25 μL of a deuterated surrogate standard mix containing 1.0 ng/μL each of EE_2-d_4, estrone-d_4 (E_1-d_4), E_2-d_4, 4nNP-d_8, and BPA-d_{16} in acetone. Both spiked and unspiked samples were extracted