Evaluation of emerging waterborne contaminants in Ireland

Jenny Pender, Carolyn Read, John Egan and Theo De Waal

ABSTRACT

A group of potential emerging contaminants in two Irish drinking water supplies were investigated. The aim was to investigate the presence of emerging contaminants which are not currently routinely monitored or regulated as part of the European Communities Drinking Water Regulations SI 278 of 2007 but are known or anticipated to occur in drinking water supplies. A monthly sampling and analysis programme was carried out to examine the presence of seven groups of potential emerging contaminants in two Irish drinking water supplies. The seven groups selected were: herbicides, molluscides, endocrine disrupters, perfluorinated chemicals, disinfection by-products, personal care products, and heavy metals. The investigation showed that the majority of the seven selected groups of contaminants were not detected at either drinking water site. Results from the first site (water treatment plant (WTP) 1) showed elevated levels of musk xylenes, a member of the personal care products suite of tests. The specific compound detected was galaxolide, a polycyclic musk found in perfumes, soaps, cosmetics and detergents. Results from the second site (WTP 2) showed elevated levels of oestrone, a member of endocrine disrupter steroid suite of tests. Oestrone is one of three types of oestrogen made by the body and is produced by the ovaries as well as by adipose tissue and the adrenal glands. Results from both sites showed that of the seven groups of contaminants chosen for examination most were not detected. The musk xylene compound galaxolide was detected on one occasion at a level just above the guideline limit and oestrone a component of the endocrine disrupting chemicals steroid suite of tests was found on two occasions throughout the study.

Key words | disinfection by-products, drinking water, emerging contaminants, endocrine disrupters, Ireland, personal care products, pesticides

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INTRODUCTION

The availability of safe drinking water is essential for health and well-being of humans all over the world. Traditionally, the microbiological quality of drinking water has attracted most attention and is still the most important issue in large parts of the world. During the last decades however, attention to the chemical quality of drinking water has grown because the knowledge of chemical compounds and their possible toxic effects has increased (Houtman 2010). These ‘emerging contaminants’ can be broadly described as any chemical substances which have the potential to enter the environment and cause known or suspected adverse ecological and (or) human health effects (USGS 2015). The term itself more accurately refers to the emerging awareness and scientific knowledge of the real and potential impacts of existing and newer chemicals, and not strictly to ‘new’ chemicals per se.

Sources of contaminants to surface water, groundwater, sediments, and drinking water are varied and include

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pesticide applications to agricultural land, horticulture, parks, gardens, golf courses, urban infrastructure, and the transport network, discharges or leaks of domestic, hospital or industrial wastewater containing pharmaceutical or personal care compounds, sewage sludge application to land, pharmaceuticals and pesticides used to treat animals present in manure stores or applied to agricultural land and solid waste disposal. Developments in almost every aspect of modern life have only served to increase the number and complexity of these compounds. Owing to their physical and chemical properties as they are discharged into wastewater treatment plants (WWTPs), many are not metabolised or efficiently removed during conventional water treatment processes (Bolong *et al.* 2009), and are being re-released into surface waters with the treated outflow. Some of these have been identified as potential threats to public and environmental health and have become a focal point for discussion in water quality.

The occurrence of emerging or newly identified contaminants in Irish water supplies is of continued public concern. Studies have shown that some emerging contaminants found in water have potential adverse human health effects (USGS 2015). In addition, various studies have identified adverse effects on other species including aquatic vertebrates in the environment. For instance, compounds in the endocrine disrupting suite are known to mimic or block the action of hormones leading to adverse effects on growth, development, fertility and reproduction (Colborn *et al.* 1993; Farre *et al.* 2008). Recent attention in Ireland has been focussed on the intersexuality of male fish. A study conducted by McGee *et al.* (2012) on the effects of endocrine disrupting chemicals on Irish roach found evidence of intersex of male fish indicated by the presence of oocysts in their testicular tissue. These fish were sampled from selected Irish rivers downstream of WWTPs.

In Ireland, all drinking water must comply with the European Communities (Drinking Water) (No. 2) Regulations, 2007. This regulation sets standards for 48 individual microbiological, chemical and indicator parameters (Hayes *et al.* 2012), however no guidelines currently exist in Ireland in relation to emerging contaminants in drinking water. In the European Union (EU), the Water Framework Directive (WFD), in force since 2000, aims to achieve good chemical status in all surface waters by 2015. This requires managing the river basin so that the quality and quantity of water does not affect the ecological services of any specific water body. Annexe II of the WFD Directive on Environmental Quality Standards (Directive 2008/105/EC) (EQSD) also known as the Priority Substances Directive (CEP 2008) lists 33 priority substances as well as eight other pollutants and defines environmental quality standards (EQS) for these substances in surface water. To comply, EU member states are required to meet the quality standards in the EQSD by 2015 at the latest. Moreover, direct discharges of pollutants into surface water must cease by 2025 (EUC 2012). The list of priority substances and EQS is currently under review with a proposal to add a further 15 chemicals to the list, and with provisions to improve the functioning of the legislation (DEP 2012).

In 2011, a project was undertaken to investigate emerging contaminants in Ireland's drinking water supplies which are currently not routinely monitored or regulated as part of the EU Drinking Water Directive SI 278 but are known to occur in drinking water supplies elsewhere.

MATERIALS AND METHODS

Selection of compounds

Following a comprehensive literature review seven groups of emerging contaminants were selected for analysis at two drinking water treatment plants (WTPs) in Ireland. The research and selection of the emerging contaminants of interest and guideline limits selected focussed on United States Environmental Protection Agency (USEPA), Drinking Water Inspectorate (DWI), Water UK and World Health Organization (WHO) publications. They have carried out several research projects and produced numerous publications on the subject of emerging contaminants in drinking water with some producing interim guidelines on maximum contaminant levels. Of those chosen all were selected based on their applicability to Ireland's drinking water supplies and the likelihood of their occurrence due to agricultural practices, industrial activities or as a result of water treatment processes. Disinfection by-products, for example, have gained particular attention in Ireland with the EPA publishing an advice note on how these are formed and what measures should be introduced to reduce levels in finished drinking water (EPA 2012). The guideline limits applied were based on those set in other countries, following research on the health effects of emerging contaminants in drinking water. None of these guideline limits are currently in force in Ireland. The groups chosen for this study are presented in Table 1.

Site selection

WTP 1

This is a large drinking WTP, providing over 30% of the drinking water requirements for water requirements for an urban region in the east of Ireland. Its average current throughput is approximately 165 Ml (megalitre) per day with a maximum output of 175 Ml per day. The catchment area contains 18 urban centres which covers a total of 21% of the catchment with a total population of approx. 120,000. These urban areas are a source of urban storm water runoff and associated pollutant loads. There is one WWTP located within the catchment as well as a several combined sewered overflows. There are a number of unsewered areas, i.e. the presence of rural drainage (septic tanks), particularly to the mid and upper portions of the catchment. Agricultural and urban runoff is the most likely cause of poor water quality in the catchment area which features livestock farming, including cattle, sheep and forested areas.

WTP 2

WTP2, situated in the south-east of Ireland, extracts water from a river at the point before it enters the city which it serves. It currently treats 45 Ml of water per day using a number of treatment processes, including pre-treatment with caustic soda, coagulation with alum, and flocculation with polyelectrolyte. This is followed by sedimentation, fluoridation, filtration and addition of lime. The final step in the treatment is chlorination. The only additional treatment which occurs is chlorine boosting using chlorous acid at one of its three reservoirs. This plant was chosen as it is located downstream of another WWTP and the catchment is predominantly dominated by agriculture, including cattle, sheep and pig farming.

Sampling programme

The sampling programme involved a monthly sample from WTP1 and a bi-monthly sample from WTP2 for 10 months at random sampling times during the month. This allowed for trending of results across each of the four seasons to determine if certain contaminants occurred as a result of a rise/fall in temperature, increased rainfall and agricultural practices such as slurry spreading. The time between sample collection and receipt in the laboratory did not

Table 1 | Contaminants and guideline limits

Group	Contaminant/Chemical name	Guideline limits	Source of guideline limit
Herbicides	Glyphosate	0.7 mg/l (DW)	
	Pichloram	0.5 mg/l (DW)	USEPA (2015b)
Molluscides	Metaldehyde	0.1 μg/l (DW)	USEPA (2015c)
Endocrine disrupters	Oestrone	0.7 ng/l (DW)	DWI (2013)
	Oestradiol		Ying *et al.* (2002)
	Ethinyl oestradiol		
	Nonyl phenol		
Perfluorinated chemicals	Perfluorooctanoic acid (PFOA)	<10.0 μg/l (DW)	
	Perfluorooctanoic sulphonate (PFOS)	<1.0 μg/l (DW)	DEFRA (2008)
Disinfection by-products	Chlorite	1 mg/l (DW)	USEPA (2015a)
	Halo-acetic acid (HAA)	60 μg/l (DW)	USEPA (2015a)
	Trihalomethanes (THM)	100 μg/l (DW)	EUC (2007)
Personal care products	Musk xylenes	0.1 μg/l (SW)	USEPA (2015d)
Heavy metals	Uranium	0.015 mg/l (DW)	WHO (2004)

DW = drinking water, SW = surface water.

exceed three days on any occasion. Specialised bottles containing preservatives were provided by the laboratory for sampling. These included amber and clear glass, azlon and polyethylene terephthalate plastic bottles of varying volumes. Preservatives used included ascorbic acid and $CuNO_3$. Information and guidance on bottles and preservative required were provided by Severn Trent Services (Coventry, United Kingdom). All samples were taken using aseptic techniques by trained environmental technicians at both sites. This ensured that sample stability was not compromised during transportation and storage. All samples were then shipped to Severn Trent Services (Coventry, United Kingdom) for analysis. Analysis techniques used for the samples included liquid chromatography–mass spectrometry, gas chromatography–mass spectrometry, inductively coupled plasma mass spectrometry and ion chromatography.

RESULTS

WTP 1

A total of 10 samples were collected between October 2010 and July 2011. Results for the molluscicides, steroid suite, perfluorinated chemicals, disinfection by-products and heavy metals were all below existing set guideline limits for each sample taken (Table 2). Two musk xylene results were above the 0.1 μg/l surface water limit in February and April 2012 where values of 142 ng/l and 122 ng/l were recorded, respectively (Figure 1).

WTP 2

A total of five samples were collected between October 2010 and July 2011. All compound groups were below existing set guideline limits for each sample taken with the exception of one compound (Table 3). In the June 2012 sample, one of the steroid suite of components, oestrone was detected above the guide value of 7 ng/l which is set in the Netherlands with a recorded value was 33.2 ng/l (Figure 2).

Reviews of the treatment processes and weather conditions on the days when elevated levels of specific compounds were detected at the two WTPs showed nothing unusual with no other water pollutants detected. Overall there were no seasonal variations in relation to results, particularly the disinfection by products with all samples reporting results less than the method limits of detection.

Table 2 | Emerging contaminant results WTP1 (finished water quality)

Contaminant	Existing current limit	Units	Min	Max	Mean
Glyphosate	0.7 mg/l	μg/l	<0.006	<0.0006	<0.006
Metaldehyde	0.1 μg/l	μg/l	0.004	0.011	0.01
Oestrone	The Netherlands: guide value - 7 ng/l	ng/l	<0.05	0.256	0.15
17-Oestradiol	Germany: precautionary value - 0.1 μg/l	ng/l	<0.05	1.87	1.19
17-α-Ethinyl oestradiol		ng/l	0.949	1.33	1.14
Nonyl phenol		μg/l	<1	<1	<1
PFOA	< 1.0 μg/l = PFOS	ng/l	<10	<25	<25
PFOS	<10.0 μg/l = PFOA	ng/l	<10	<25	<25
Chlorite	1 mg/l	μg/l	<0.0025	<0.0025	<0.0025
HAA	60 μg/l	μg/l	<1	4.4	<1
THM	100 μg/l	μg/l	18.97	49.43	28.86
Musk xylenes	0.1 μg/l (surface water)	ng/l	16	146	73.22
Pichloram	0.5 mg/l	μg/l	<0.02	<0.02	<0.02
Uranium	0.015 mg/l	μg/l	0.34	2	1.03

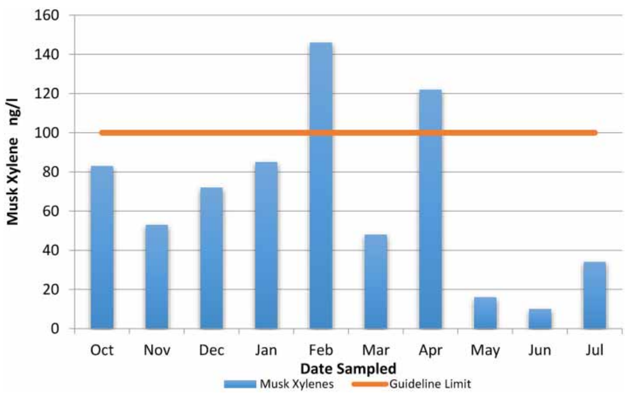

Figure 1 | Musk xylene results detected at WTP1.

DISCUSSION

The musk xylene component to have breached on two occasions the applied guideline limit at WTP1 was that of galaxolide (1,3,4,6,7,8-hexahydro-4,6,6,7,8,8-hexamethylcyclopenta(g)-2-benzopyrene (HHCB)) in February and April 2012. Both of these were just above the guideline limit of 0.1 μg/l; however, this limit is applicable only to surface water. In the USA, this limit has been applied to treated drinking water; however, there has been no evidence to date of the health effects of personal care products on the environment. This compound was detected in all samples taken at WTP1, though usually below our chosen cut-off limit, and was the only musk xylene compound identified. All other compounds examined as part of the musk xylene suite of tests were below the method limit of detection (<10 ng/l).

Galaxolide is currently listed on EC 2008/105/EC Annexe III as a substance subject to review for possible identification as a priority substance (CEP 2008). As a synthetic polycyclic musk fragrance introduced in the 1950s, galaxolide is found in almost all scented consumer products such as perfumes, soaps, cosmetics and detergents. The production of galaxolide use has increased continuously over the last number of years and today almost all of the musks used in cosmetics and fragrances products are of this synthetic origin (Rimkus 1999). Galaxolide together with tonalide accounts for about 95% of the EU market and 90% of the US market for polycyclic musks (Clara *et al.* 2011).

Owing to the widespread use of synthetic musks in consumer products, relatively large amounts of the compound are washed into wastewater after being applied to skin, hair, and clothing, and find their way into the aquatic environment via effluents, principally those from sewage treatment works (Ricking *et al.* 2003; Stevens *et al.* 2003).

Table 3 | Emerging contaminant results WTP2 (finished water quality)

Contaminant	Existing current limit	Units	Min	Max	Mean
Glyphosate	0.7 mg/l	μg/l	<0.006	<0.006	<0.006
Metaldehyde	0.1 μg/l	μg/l	<0.003	0.007	0.038
Oestrone	The Netherlands: guide value - 7 ng/l	ng/l	0.325	33.2	11.37
17-oestradiol	Germany: precautionary value - 0.1 μg/l	ng/l	3.54	4.18	2.59
17-α-ethinyl oestradiol		ng/l	<0.05	5.73	1.94
Nonyl phenol		μg/l	<1	<1	<1
PFOA	< 1.0 μg/l = PFOS	ng/l	<25	<25	<25
PFOS	<10.0 μg/l = PFOA	ng/l	<25	<25	<25
Chlorite	1 mg/l	μg/l	<0.0025	<0.0025	<0.0025
HAA	60 μg/l	μg/l	<1	9.2	2.68
THM	100 μg/l	μg/l	7.33	21.07	11.98
Musk xylenes	0.1 μg/l (surface water)	ng/l	<10	<10	<10
Pichloram	0.5 mg/l	μg/l	<0.02	0.05	0.10
Uranium	0.015 mg/l	μg/l	<0.06	10.47	2.14

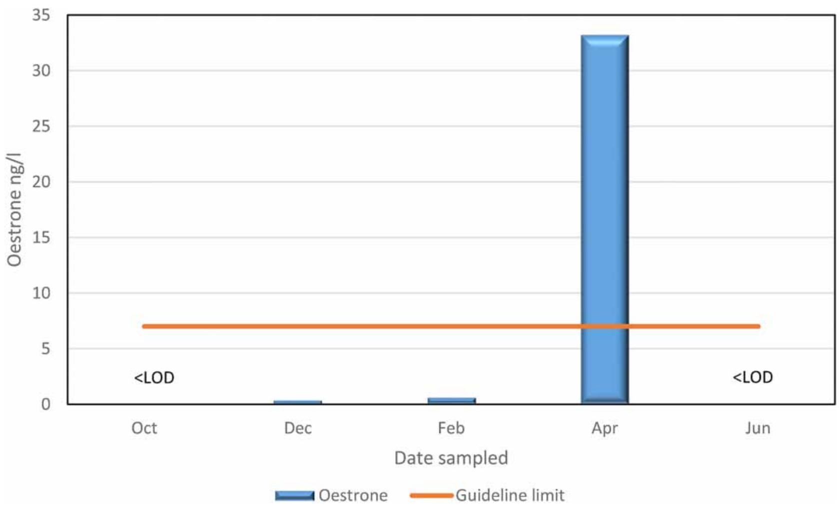

Figure 2 | Oestrone levels detected at WTP2 (<LOD: below level of detection).

Although removal efficiency of this compound is high in conventional activated sludge WWTP (and can be enhanced to >90% by ozonation and activated carbon treatment), the very significant concentrations in untreated wastewater still lead to occurrences in the μg/l range in treated effluents (Bester 2004; Ternes *et al.* 2007; Lv *et al.* 2010). Furthermore, synthetic musks that have not been degraded during digestion are introduced into the ecosystem as 'biosolids', digested and dewatered sewage sludge which is commonly applied to agricultural land (Stevens *et al.* 2003). Once in the aquatic environment, synthetic musks can enter the food chain, being taken up by wildlife such as fish and shellfish. Galaxolide is almost always present in low levels in finished drinking water. However, very little is known about the human health effects of these synthetic musks. They have been shown to have endocrine disrupting properties – that is, they are capable of interacting with hormone systems. Both tonalide (AHTN) and galaxolide (HHCB) can bind to oestrogen receptors in cells and can be both oestrogenic and anti-oestrogenic, depending on the type of cell and the type of oestrogen receptor involved (Schreurs *et al.* 2004). In spite of concerns due to their persistence and potential to bio-accumulate, the potential toxicity and environmental risks of these chemical and other synthetic musks have been usually regarded as low (Salvito 2005). Furthermore, a European Commission risk assessment concluded that at present there is no need for further information/testing and no need for risk reduction measures beyond those which are being applied already (EUC 2008).

In the samples taken at WTP2 the only compound found to have breached the guideline limits applied in the study was that of oestrone. This was analysed as part of the Endocrine Disrupting Chemicals Steroid Suite. In June 2012, a value of 32 ng/l was observed breaching the applied limit of 7 ng/l by almost 500%. Oestrone also known as estrone (E1), is one of the estrogenic hormones, together with estradiol and estriol. These are predominately female hormones although also naturally excreted by men and are important for maintaining the health of reproductive tissues, breasts, skin and brain. Oestrone as an estrogen together with contraceptives both natural and synthetic hormones are of concern to the aquatic environment due to their endocrine disruption potential (Ying *et al.* 2002). This is because they may interfere with the reproduction of humans, livestock and wildlife (Ying *et al.* 2002).

A European study of endocrine disrupters in drinking waters, found that oestrone was generally absent, and that endocrine disrupters via the consumption of drinking water are very unlikely to affect human health even if the highest concentrations of an individual compound reported for drinking water were considered (Wenzel *et al.* 2003). This finding is echoed for many of the other emerging contaminants selected as part of this study where the potential impact on human health had been either classified as low (Salvito 2005) or unknown. Contrary to this there has been known and reported adverse effects of endocrine disrupters in particular oestrogens on the reproduction of aquatic vertebrates as shown by McGee *et al.* (2012).

CONCLUSION

Results from the 10 month project examining the presence of seven groups of emerging contaminants at two drinking WTPs in Ireland found that the majority were absent. Only two contaminants were detected (galaxolide at WTP1 and oestrone at WTP2) at low levels. This study was a snapshot of two water treatment facilities and may not completely reflect the real presence of emerging contaminants in Irish drinking water. This will be an area of importance for Ireland over the coming years in terms of infrastructure and economic growth. The introduction of water charges in Ireland due in October 2014 will focus consumer interests on the quality of water provided and the presence of emerging contaminants will require future extensive research across all WTPs. This may include risk assessment of specific catchment areas known for high levels of runoff pollution and also reviewing and enhancing water treatment processes to deal with these unknown contaminants.

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