ELSEVIER

Water Research 39 (2005) 47–58

WATER RESEARCH

www.elsevier.com/locate/watres

Comparing steroid estrogen, and nonylphenol content across a range of European sewage plants with different treatment and management practices

A.C. Johnson[a,*], H.-R. Aerni[b], A. Gerritsen[c], M. Gibert[d], W. Giger[b], K. Hylland[e], M. Jürgens[a], T. Nakari[f], A. Pickering[g], M.J.-F. Suter[b], A. Svenson[h], F.E. Wettstein[b]

[a]*CEH Wallingford, Maclean Building, Crowmarsh Gifford, Wallingford, Oxfordshire OX10 8BB, UK*
[b]*EAWAG, Swiss Federal Institute for Environmental Science and Technology, Überlandstrasse 133, 8600 Dübendorf, Switzerland*
[c]*RIZA, Institute for Inland Water Management and Waste Water Treatment, P.O. Box 17, Maerland 16, Lelystad, 8200 AA, The Netherlands*
[d]*Companie Generale Des Eaux, Direction Recherche et Développement, 52 rue de Anjoux, 75008 Paris, France*
[e]*NIVA, Norwegian Institute for Water Research, P.O. Box 173, Kjelsas, 0411 Oslo, Norway*
[f]*Finnish Environment Institute, Hakuninmaantie 6, 00430 Helsinki, Finland*
[g]*CEH Windermere, Ferry House, Far Sawrey, Ambleside, Cumbria LA22 0LP, UK*
[h]*Swedish Environmental Research Institute, P.O. Box 21060, 10031 Stockholm, Sweden*

Received 15 October 2003; received in revised form 8 April 2004; accepted 16 July 2004

Abstract

The effluent of 17 sewage treatment works (STW) across Norway, Sweden, Finland, The Netherlands, Belgium, Germany, France and Switzerland was studied for the presence of estradiol (E2), estrone (E1), ethinylestradiol (EE2) and nonylphenol (NP). Treatment processes included primary and chemical treatment only, submerged aerated filter, oxidation ditch, activated sludge (AS) and combined trickling filter with activated sludge. The effluent strength ranged between 87 and 846 L/PE (population equivalent), the total hydraulic retention time (HRT) ranged between 4 and 120 h, sludge retention time (SRT) between 3 and 30 d, and water temperature ranged from 12 to 21 °C. The highest estrogen values were detected in the effluent of the STW which only used primary treatment (13 ng/L E2 and 35 ng/L E1) and on one occasion in one of the STW using the AS system (6.5 ng/L E2, 50.5 ng/L E1, but on three other occasions the concentrations in this STW were at least a factor of 6 lower). For the 16 STW employing secondary treatment E2 was only detected in the effluent of six works during the study period (average 0.7–5.7 ng/L). E1 was detected in the effluent of 13 of the same STW. The median value for E1 for the 16 STW with secondary treatment was 3.0 ng/L. EE2 was only detected in two STW (1.1, <0.8–2.8 ng/L). NP could be detected in the effluent of all 14 STW where this measurement was attempted, with a median of 0.31 μg/L and values ranging from 0.05 to 1.31 μg/L. A

*Corresponding author. Tel: +44 1491 69 23 67; fax: +44 1491 69 2424.
E-mail address: ajo@ceh.ac.uk (A.C. Johnson).

0043-1354/$ - see front matter
doi:10.1016/j.watres.2004.07.025

comparison of removal performance for E1 was carried out following prediction of the probable influent concentration. A weak but significant ($\alpha < 5\%$) correlation between E1 removal and HRT or SRT was observed.

Keywords: Sewage treatment works; Estrone; Estradiol; Endocrine disrupters; Nonylphenol; Ethinyl estradiol

1. Introduction

The issue of endocrine disruption of freshwater fish started with the observation of hermaphrodite fish in sewage treatment works (STW) lagoons by anglers in the UK, where it was later found that caged male fish placed in or near sewage effluent streams almost universally produced large quantities of the egg yolk protein vitellogenin (Purdom et al., 1994), normally only produced by mature females. Possible causative agents were the 4-tertiary isomers of nonyl- and octylphenol (OP) which were shown to be estrogenic to rainbow trout hepatocytes with an in vitro estrogenic potency around 10,000 times less than 17β-estradiol (E2) (Jobling and Sumpter, 1993). These compounds are breakdown products of two of the most important alkylphenol polyethoxylates (APE) which have been economically important as nonionic surfactants for decades and used in a variety of industrial and household applications. These have included such diverse uses as dispersing agents in paper and pulp production, emulsifying agents in latex paints and pesticide formulations, flotation agents, industrial cleaners (metal surfaces, textile processing, and food industry), cold cleaners for cars, and household cleaners (Thiele et al., 1997). The UK Department of the Environment (Uses, and fate and entry to the environment of nonylphenol ethoxylates, 1993) reported a UK consumption of 16–19,000 tonnes alkylphenol polyethoxylates/pa with approximately 6500 tonnes disposed to water for 1990. Clearly, a STW may receive little or a great deal of these compounds depending on the local industrial use. Whilst local high water concentrations, up to 180 μg/L, have been reported in the UK and Spain due to effluent emanating from textile and ceramic industries (Solé et al., 2000; Blackburn and Waldock, 1995) recent detections in receiving waters have generally been low but still surpassed the predicted no effect concentration for NP of 0.33 μg/L occasionally (Ahel et al., 2000). In a recent monitoring exercise of the rivers Exe, Dee, Mersey, Thames, and Aire, only in the Mersey and Aire were concentrations of NP found above 0.2 μg/L (Blackburn et al., 1999). In a recent survey of a range of streams in the US the median concentration for NP was 0.8 μg/L (Kolpin et al., 2002).

From the low μg/L effluent concentrations and the rather weak estrogenic potency of NP and OP, it was apparent that these compounds could not be responsible for all the endocrine disruption effects observed in caged fish studies. Further research in the UK, used a toxicity identification and evaluation approach based on the yeast estrogen screening assay (YES) (Routledge and Sumpter, 1996), to identify which compounds were responsible for the observed estrogenic effects in the effluent of seven separate STW (Desbrow et al., 1998). The steroid estrogens E2 and E1 and occasionally EE2 emerged as the most important in vitro estrogenic compounds present in the effluent, with concentrations between 2.7–48 ng/L E2 (median 6.3 ng/L), 1.4–76 ng/L E1 (median 9.4 ng/L) and up to 7 ng/L EE2. Similar studies in Japan (Matsui et al., 2000) and Germany (Körner et al., 2000) using the yeast assay and human breast cancer MCF-7 cells, respectively, have come to the same conclusion. It should be noted of course that in vitro assays may not perfectly reflect the true in vivo response of fish to the same compound. Nevertheless, overall, the steroid estrogens appear to be the most potent endocrine disrupters of sewage effluent, at least in vitro. The estrogenic activity of the steroid estrogens has been shown in a variety of in vivo studies conducted in fish and their potency demonstrated to be over a thousand times greater than any xenobiotic estrogen mimic (eg. Länge et al., 2001; Thorpe et al., 2001; Routledge et al., 1998; Jobling et al., 2002). Thus, the phenomenon of endocrine disruption in fish was seen in the UK as essentially a point source problem associated largely with sewage effluent (Jobling et al., 1998).

As the source of these steroid estrogens is the human population, there arose the possibility that these compounds would be found throughout the world where sewage treatment works collect human wastes before treatment and discharge into watercourses. However, across Europe a range of different sewage treatment approaches and management practices occur. Would estrogen mimics and steroid estrogens be present in the effluent across a range of European treatment works as had been observed in the UK? Some recent studies have indicated the presence of steroid estrogens in Italian (Baronti et al., 2000) and German (Spengler et al., 2001) sewage effluents. However, explicit details on the key sewage management process operating are lacking. Without these details, it is very difficult to extrapolate the results to other locations in Europe since, even with activated sludge treatment, very wide differences in key parameters such as sludge and hydraulic residence time can occur.