

**An investigation of selected Watch List chemicals: their levels, toxicity and regulation status as part of the EU Sullied Sediments project**

**Authors:** Sullied Sediments project (<https://northsearegion.eu/sullied-sediments/>)

**Acknowledgment**

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**Overview**

The Sullied Sediments project partners have developed and tested innovative tools to better assess, treat and prevent contamination from new, emerging Watch List chemicals (WLCs) that have been found in the sediment in our inland waterways. The project has three aims:

- To provide regulators and water managers with new data, tools and guidance to help them make better decisions regarding the management, removal and disposal of sediment. In so doing, we seek to reduce the costs of disposing of dredged material to a range of private and public sector organisations and lessen the impact on the environment.
- To reduce the levels of selected WLCs entering the water system from a waste water treatment facility using an innovative spore technology.
- To reduce the levels of selected WLCs by raising awareness about what we, as consumers, are releasing into the environment through the use of common drugs and household products.

In addition, we are focusing on emerging chemical contaminants, their presence across a range of nine EU sites, and a review of the potential impacts without

**Introduction**

The Watch List Chemicals (WLCs) are a class of substances that have been highlighted under the EU Water Framework Directive as potential pollutants that require investigation to determine the risk they may pose to the aquatic environment and organisms living within it. Specifically, these chemicals are highlighted within this EU Sullied Sediments project to raise the discussion as to whether Environmental Quality Standards (EQS) should be set for them and their presence in waterbodies routinely monitored. This represents a measured way of looking at such chemicals that sits in between ignoring them and rushing in and developing legislation based on historically limited data.

As research accumulates on a particular WLC, decisions are made every two years whether they merit continued inclusion in the WL or removal. The estrogenic hormones are an example of a chemical type that persist on the WL, and diclofenac, the pain killer, is an example where it made an appearance but was then subsequently removed as a result of accumulating toxicity evidence to suggest it was less of a threat than previously envisaged. Occasionally there is debate surrounding the inclusion of a new chemical within the WL, which has been the case for the antimicrobial agent triclosan. This POSTNote focuses on the three pharmaceuticals: oestradiol (E2), diclofenac (DIC) and triclosan (TCS) as examples that are all excreted by humans, ineffectively removed at waste-water treatment plants (WWTPs), ultimately ending up in waterbodies where they have negative impacts on aquatic organisms, even at very low levels of ng/L.

This report only concerns human contribution and not agriculture, which also plays a role in discharging to watercourses. WWTPs are not designed to treat or remove these compounds, which can be highly diverse and subject to WL addition or deletion. The final effluents produced are the indication of the direct level of human impact because, for the most part, the treatment plants do not treat surface water, road run-off or agricultural discharges. There are additional untreated waste waters discharged into waterways that can account for



16-25% in some countries such as Belgium, and the Flanders region. In the UK there are thousands of Combined Sewer Outflows (CSOs) that are largely uncharacterised in terms of their composition and impacts on receiving waters: two studies report pharmaceuticals in such sources in the Thames tidal region and Aire/Calder river catchment (Munro *et al.* 2019; Kay *et al.* 2017) which must also be taken into account.

### **Why are such chemicals considered harmful?**

The WLCs differ in their harmful impacts, though all have been evidenced to have some level of detrimental effect on organisms inhabiting aquatic ecosystems. WLCs include many pharmaceuticals (and non-pharmaceuticals); compounds that are designed to be biologically active (even at low levels) and their presence in the aquatic environment has led to concern over their potential biological effects. Many pharmaceuticals have been found to elicit a negative response on biota at concentrations similar to those found in the aquatic environment (Eades and Waring 2010; Franzellitti *et al.* 2013; Minguez *et al.* 2016) and the following examples explain that 'cause and effect' relationship between these specific chemicals and serious biological damage.

### ***They act as 'hormone disruptors':***

Endocrine disrupting chemicals (EDCs) disrupt the reproductive endocrine system and may cause various biological impacts such as a skew in sex ratio (Gagné *et al.* 2003), delayed egg/sperm development (Gauthier-Clerc *et al.* 2002), a condition called imposex where a penis grows on the head of marine snails (Strand & Asmund 2003), and intersex disorders in fish (Kidd *et al.* 2007; Jobling *et al.* 2002). Intersex, where male and female gametes (egg and sperm) develop in the same individual, is a well-documented phenomenon in aquatic organisms including fish, crustaceans and molluscs (Jobling *et al.* 2002; Ford 2012; Ciocan *et al.* 2012). The estrogenic chemicals have specifically been evidenced to cause intersex

in both fish and invertebrates, which consequently have serious long-term impacts on their reproduction success. This is evidenced by large scale experiments in lake systems where fish have been exposed to estrogenic chemicals resulting in population declines due to the exposure (Kidd *et al.* 2007).

***They cause developmental and cell damage:***

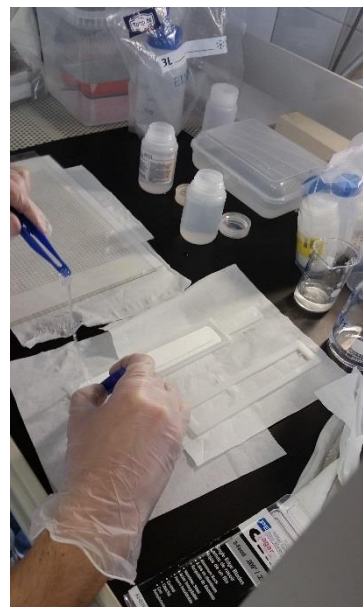


DIC has been in use as a pain killer since 1979 and the gradual cumulative data for humans has shown that a continuous DIC daily intake, of low levels at 1 µg/L, can cause kidney and liver damage (Bort *et al.* 1990) as well as cardiovascular side effects (McGettigan & Henry 2011), which has led to its declining (but not yet discontinued) usage. DIC was recently removed from the WL as a result. In 2019, 5%

of patients requiring a non-steroidal anti-inflammatory drug (NSAID) were prescribed DIC, ranking it as fourth in the prescription table, compared with 59% receiving the most prescribed as naproxen (NHS, 2019). Studies on the exposure of fish (*Oryzias latipes*) and daphnia (*Daphnia magna*) confirm toxicity at levels of 10 mg/L (Lee *et al.* 2011), whereas studies with a longer period of exposure using rainbow trout (*Oncorhynchus mykiss*) at low concentrations (of 1 µg/L) showed kidney and intestinal damage (Mehinto *et al.* 2010). Studies in amphibians have highlighted that levels of 250-1000 µg/L also affect larval development and swimming performance (Peltzer *et al.* 2019). Another concern relates to its bioaccumulation in fish body tissues, leading to a potentially secondary poisoning risk along the food chain for fish-eating birds (Green *et al.* 2004) or humans.

***Mixed impacts: developmental problems and endocrine disruptors combined***

TCS is a broad-spectrum bactericidal agent that has been in use for more than 50 years. It can also affect aquatic species (Ricart *et al.* 2010). Studies have identified TCS presence in both aquatic and terrestrial ecosystems, with variable bioaccumulation properties across species (Arnot *et al.* 2018). TCS has shown toxicity to humans associated with liver damage (Zhang *et al.* 2019) as well as in other organisms from fish (Falisse *et al.* 2017) to various algal species (Xin *et al.* 2019) all at µg/L levels. Examples of impacts include embryo and larval toxicity and hatching delay at 500 µg/L in zebrafish (*Danio rerio*) (Oliveira *et al.* 2009) as well as affecting the developmental stages of rainbow trout at concentrations above 700 µg/L. TCS acts as an endocrine disruptor in vertebrates (Stoker *et al.* 2010; Christen *et al.* 2010; Ha *et al.* 2018). TCS can also act as a thyroid-disrupting compound including an association with certain diabetes conditions (Xie *et al.* 2020), resulting in a number of large companies already banning its use.





## Usage levels: past and current trends

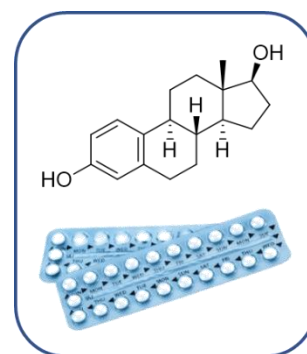
### Pharmaceutical oestrogens incl. estradiol (E2):

**What:** Solubility in water = 3.6 mg/L;  $pK_a$  = 10.46

**Where:** Waste-water treatment plant, agriculture effluent

**Why:** Toxic to aquatic organisms and birds. Accumulation in fish and mussels.

**Human E2 production:** F (15–59 yrs) 3–19 µg/day (170–330 µg/day if pregnant) & M 1.5–7 µg/day, UK ~38 million adults → ~167 kg/year oestrogens arriving at WWTPs. **Consumed sources:** 100 kg/year/5 million inhabitants<sup>2</sup> → 1000 kg/year. **Farming sources:** 1 g/animal/year for ~900 million farmed animals<sup>3</sup> → 900,000 kg/year. **UK total:** 901,167 kg (~900 tonnes) of oestrogens/year. **Watercourse pharmaceutical oestrogen load:** (from farmed sources via WWTPs or run off): unknown kg / year.



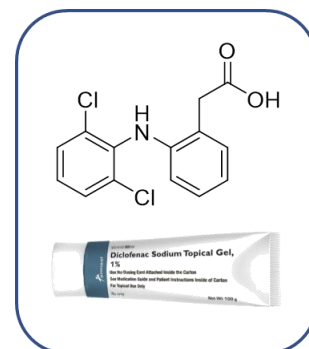
### Diclofenac (DIC):

**What:** Non-steroidal anti-inflammatory drug, available OTC since 2015. Solubility in water = 4.82 mg/L;  $pK_a$  = 4.08

**Where:** Tablets and creams for treatment of inflammation and pain.

**Why:** Toxic to aquatic organisms and birds. Accumulation in fish and mussels.

**England usage:** 26 tonnes / 49 million (pop) in 2000<sup>4</sup> with fewer than 5000 patients prescribed DIC in England (NHS, 2019), so most via OTC sales. **Germany usage:** 82 tonnes / 82 million (pop) in 1999.<sup>5</sup>

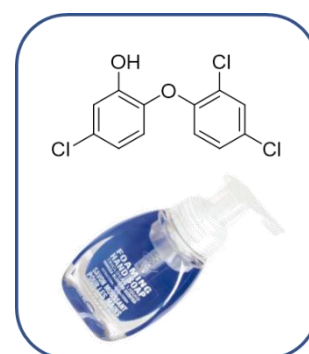


### Triclosan (TCS):

**What:** Preservative and antiseptic agent. Solubility in water = 10 mg/L;  $pK_a$  = 7.9

**Where:** Hand soaps, skin creams, toothpastes, deodorants, household cleaners, fabrics.

**Why:** Forms highly toxic dioxin-type derivatives under solar irradiation in surface water. Bioaccumulation in fatty tissues of organisms.



**Detected in breast milk:** 0–2100 µg/kg of milk lipid.<sup>9</sup>

**Baseline urine excretion:** (US citizens) 0.1–3790 µg/day (varies with age and socioeconomic status). **Usage UK:** ~60–90 tonnes / year in personal care products<sup>6</sup> (maximum allowed 0.3% w/w (EU 2007)). **Usage Europe:** 37–350 tonnes / year.<sup>7</sup>

**Est. personal use:** 0.01–2 g / year / person.<sup>8</sup>

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1. Combalbert & Hernandez-Raquet, 2010; 2. Stuer-Lauridsen and Kjolholt 2000; 3. RSPCA, 2020; 4. Jones et al. 2002; 5. BLAC, 2003; 6. unreferenced internet source; 7. Warming et al. 2016; van Wijnen et al. 2018; 8. von der Ohe et al. 2012; 9. Dayan 2007; 10. Sandborgh-Englund et al. 2006; Calafat et al. 2008.

## Environmental levels

At present, there are wide ranging spatial and temporal variations in the levels of such chemicals in the aquatic environment.

Tables 1a-1c. Examples of WLC levels reported in UK relative to international waters and sediments. \*modelled data, ND not detected.

Compound	Estuaries	Max water (ng/L)	Sediment (ng/g)	Reference
<b>Estradiol/ Ethinyl Estradiol (EE2)</b>	Humber, UK		Trace	Sullied Sediments 2020
	Douro, Portugal	113(EE2/E1)		Ribeiro <i>et al.</i> (2009)
	Sado, Portugal	10.8		Rocha <i>et al.</i> (2013)
	<b>Rivers</b>			
	Aire, UK		Trace	Sullied Sediments
	Pocklington Canal, UK		0.009	
	Scheldt, Belgium		Trace	
	Elbe, Germany		Trace	
	Aire, UK	2-20*		Sumpter <i>et al.</i> (2006)
	Ave, Portugal	ND		Sousa <i>et al.</i> (2019)
	Sousa, Portugal	ND		
	Leca, Portugal	10.4		Rocha <i>et al.</i> (2012)
	Douro, Portugal	5.7		
	Yangste, China	5.9 (EE2)		Liu <i>et al.</i> (2015)

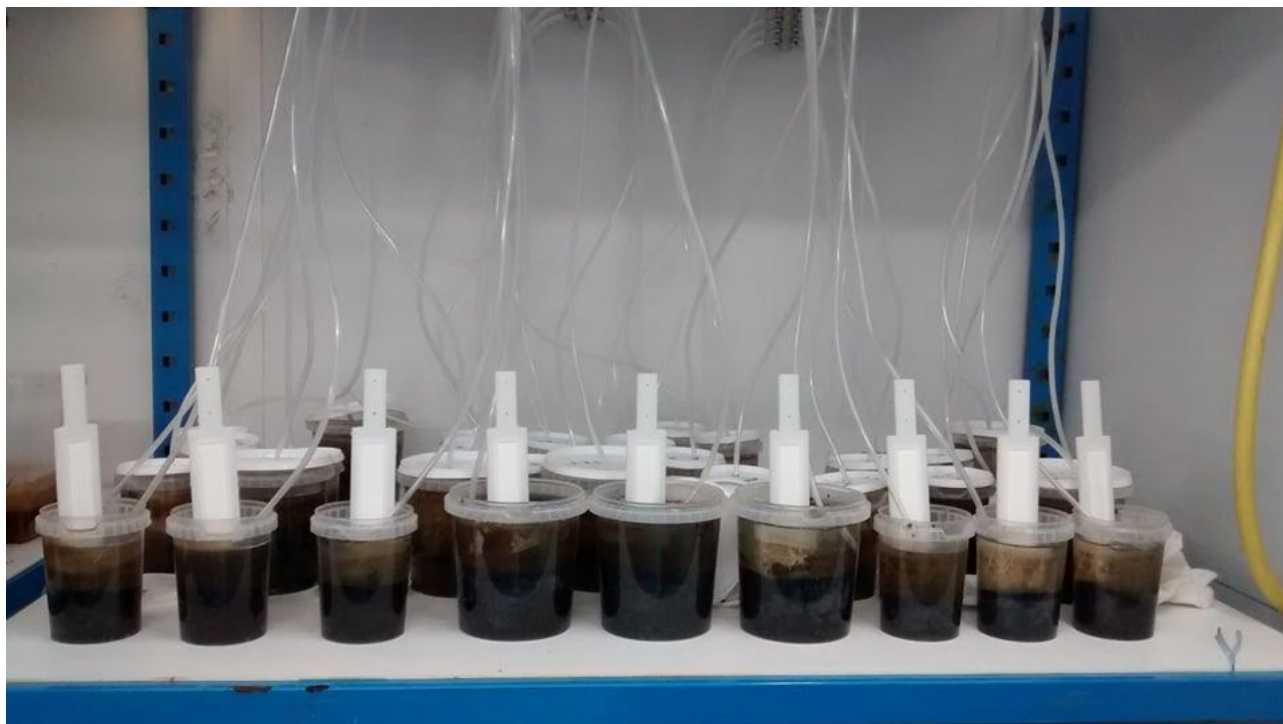
Table 1a. Estradiol/Ethinyl Estradiol (EE2)

Compound	Estuaries	Max water (ng/L)	Sediment (ng/g)	Reference
<b>Diclofenac</b>	Belfast Lough, UK	195		Thomas and Hilton (2004)
	Mersey, UK	191		
	Tees, UK	125		
	Thames, UK	90		
	Tyne, UK	251		
	Humber, UK			
	Cromarty, UK			Letsinger <i>et al.</i> (2019)
	Thames, UK	ND		
	Yangtze, China	31		
	Arade, Portugal	195		Yang <i>et al.</i> (2011)
	Jiulong, China	11.0		Sun <i>et al.</i> (2016)
	Bilbao, Spain	650		Mijangos <i>et al.</i> (2018)
	Plentzia, Spain	22		
	Urdaibai, Spain	35		
	Qinzhou Bay, China	7.17		Cui <i>et al.</i> (2019)
	Tejo, Portugal	51.8		Reis-Santos <i>et al.</i> (2018)
	Elbe, Germany	ND		Wiegel <i>et al.</i> (2002)
	<b>Rivers</b>			
	Aire, UK		0.160	Sullied Sediments
	Pocklington Canal, UK		0.109	
	Scheldt, Belgium		0.111	
	Elbe, Germany		0.103	
	Aire, UK	2830		Kay <i>et al.</i> (2017)
	Elbe, Germany (Hamburg site)	140		Wiegel <i>et al.</i> (2004)
	Ave, Portugal	30		Sousa <i>et al.</i> (2019)
	Sousa, Portugal	400		
	Yangste, China	3250		Liu <i>et al.</i> (2015)

Table 1b. Diclofenac

Compound	Estuaries	Max water (ng/L)	Sediment (ng/g)	Reference
Triclosan	German Bight	6.9		Xie <i>et al.</i> (2008)
	Victoria Harbour, HK	10.8		Chau <i>et al.</i> (2008)
	Almeria, SE Spain		131	Aguera <i>et al.</i> (2003)
	Boston Harbor, USA		100	Cantwell <i>et al.</i> (2010)
	<b>Rivers</b>			
	Aire, UK		0.576	Sullied Sediments
	Pocklington Canal, UK		0.032	
	Scheldt, Belgium		0.703	
	Elbe, Germany		0.104	
	Aire, UK	482		Sabaliunas <i>et al.</i> (2003)
	Thames/Midlands, UK*	36		Price <i>et al.</i> (2010)
	Mortsel/Scheldt	98		Covaci <i>et al.</i> Pers. Commun.
	Aa Uster, Switzerland	482		Singer <i>et al.</i> (2002)
	Greifensee, Switz.		~80	
	Elbe, Germany	1100		Von der Ohe <i>et al.</i> (2011)
	Ruhr, Germany	10		Bester (2005)
	Itter, Germany	90		Wind <i>et al.</i> (2004)*
	Pearl, China	478		Zhao <i>et al.</i> (2010)
	Pearl, China		1329	
	Pearl, China	100		Yu <i>et al.</i> (2011)

Table 1c. Triclosan



### Box 1: Oestradiol (E2)

Stable lipophilic (fat-loving) compound

E2 female hormone  $\log K_{ow} = 3.94$

EE2 oral contraceptive  $\log K_{ow} = 4.2$

Pharmaceutical oestrogens are found in formulations prescribed for hormone replacement therapies, contraception, menopause and hypoeestrogenism. The active ingredients of these formulations usually include ethinyl oestradiol (EE2) used in the contraceptive pill,  $17\beta$ -oestradiol (E2), which is a natural endogenous hormone and also used for hormone therapy, and other esterified or conjugated oestrogens. The main metabolites of these oestrogens found in urine and faeces include compounds such as estrone (E1), E2, EE2 and estriol (E3). E2 has a  $\log K_{ow}$  of 3.94 and  $S_w$  13 mg/L at 20 °C which indicates its low volatility and its hydrophobic nature, which increases its potential to bind onto sediments, sludge and soil (Lai *et al.* 2000).

Oestrogens enter the aquatic environment mainly via discharged domestic effluents from waste-water treatment plants, with larger point sources from the most densely populated areas. The most commonly found are the natural compounds, E1, E2 and E3 followed by synthetic EE2, none of which are significantly removed as part of the WWTP clean-up processes (Racz and Goel, 2010). Of these, EE2 has the highest estrogenic potency, followed by E2 (Thorpe *et al.* 2003). In addition to WWTP effluents, animal faeces and urine represent another source of oestrogens (Combalbert & Hernandez-Raquet 2010). For E2, females of reproductive age (15– 59 years) excrete 3–19  $\mu\text{g/day}$ , during pregnancy increases to 170–330,  $\mu\text{g/day}$  and out with these periods, it is similar to that of a man, with levels about 1.5–7,  $\mu\text{g/day}$  (reviewed in Combalbert & Hernandez-Raquet, 2010). If taking a pill, the daily EE2 intake is around 20–60  $\mu\text{g}$  for contraception and  $\sim 10$   $\mu\text{g}$  to control menopausal disorders; whereby approximately 30–90% is excreted (Johnson and Williams 2004; Webb *et al.* 2003).

The total amount of excreted endogenous oestrogens discharged by humans (both sexes combined) into the environment has been estimated at some 4.4 kg/year/million (reviewed in Combalbert & Hernandez-Raquet, 2010). The current UK population has  $\sim 38$  million adults (not including pensioners), amounting to  $\sim 167$  kg of oestrogens arriving at WWTPs each year. A further 100 kg of consumed pharmaceutical oestrogens/year/five million inhabitants must be added on top (Stuer-Lauridsen and Kjolholt 2000). In farmed animals, hormones are produced in different quantities by each species, with E2 and E1 the main ones excreted by cattle. Farmed animal levels of excreted oestrogens vary from 20-2300  $\mu\text{g/day}$ , leading to as much as 1 g per animal per year (reviewed in Combalbert & Hernandez-Raquet, 2010). There are  $\sim 900$  million farmed animals reared in the UK per year (RSPCA, 2020). Setting these levels into context, these pharmaceutical oestrogens are deemed to pose a human and ecological risk in their EDC capacity at levels of 1 ng/L – 0.35 ng/L once in the environment (Laurenson *et al.* 2014; Thorpe *et al.* 2003; EU, 2012).

### Box 2: Diclofenac (DIC)

DIC is a synthetic, non-steroidal anti-inflammatory drug (NSAID), introduced in the 1970s, in pills and creams. It has anti-arthritic, analgesic, anti-pyretic and anti-rheumatic properties used for the treatment of inflammations and painful conditions like arthritis, migraines and gout. From 2015, the pill form of DIC became only available by prescription in the UK. It can, however, still be bought without prescription for human use over the pharmacy counter within gels/medicated plasters, or veterinary use. It has weak acidic properties, with a pKa at ~4, and it presents a relatively medium solubility in water, 2.37 mg/L and a log K<sub>ow</sub> of 4.5 at pH 7 (Vieno and Silanpaa 2014).

In 2000, 26 tonnes of DIC were used by the population of England (@49 million people at that time) (Jones *et al.* 2002). DIC use has dropped significantly to less than 5000 prescriptions for humans per year in the UK, due to its various serious health complications (Bort *et al.* 1990; McGettigan & Henry, 2011), yet it continues to be used in over-the-counter products and veterinary applications. Voltarol™, containing DIC, is currently the eighth most common product purchased over the counter in pharmacies in the UK for example. Based on studies of toxicities in various organisms, Ferrari *et al.* (2003) calculated a predicted no-effect concentration (PNEC) for DIC as 116 µg/L, which also represents a level 1000-fold higher than normally measured in the environment. The PNEC has become increasingly more conservative as evidence of toxicities in different species accumulates in the literature.

### Box 3: Triclosan (TCS)

TCS is a halogenated aromatic hydrocarbon used as a general-purpose antimicrobial agent (Dhillon *et al.* 2015) that is added to more than 2000 products for personal care, clothing and cooking utensils. The effectiveness of its use is debated (Halden *et al.* 2017). It is a stable lipophilic (fat-loving) compound and non-volatile with a log K<sub>ow</sub>=4.8 at pH 7 which indicates that it has a high adsorption potential (Dhillon *et al.* 2015). While TCS has a low solubility in water, it has however been measured in waste water and surface water at concentrations ranging from 9 ng/L to 6.7 µg/L (Cho *et al.* 2011). While WWTPs are the main source of TCS, low volume and non-point sources, such as sewage leaks, storm events and biosolids added to agricultural fields represent other significant sources of 0.5 – 1000 ng/L (Goldsmith *et al.* 2020).

### Environmental Case Study: Sullied Sediments Project

Sediments in three major river systems in the North Sea Region (shown in Figure 1) were sampled over the period 2018-2019. These systems were chosen as they represent a range of potential pollution pressures present in industrial, urban and rural settings in the region. This project focussed on sediments because there is relatively little information available





Figure 1. Sullied Sediments sampling sites.

compared to the overlying water column. Contaminated sediments can have a considerable economic bearing in these locations given the importance of the sites for navigation, while environmental risks can be particularly prominent after flood events which can remobilise pollutants previously trapped in sediments. Moreover, sediment dwelling organisms can be exposed to these pollutants as can species higher up the food chain. Monitoring is a key step in managing these polluted sediments.

A key source of WLCs, particularly those associated with personal care products or pharmaceuticals, can be waste-water treatment plants (WWTP). To reflect this, the sediment sampling strategy incorporated upstream and downstream of WWTPs to assess if WLCs were more prominent in sediments downstream of such sites.

In contrast to E2 levels reported for the water column (Table 1), E2 was detected above the limit of quantification at only one of the nine European sites monitored, with a concentration of 9.1 pg/g from the sediments sampled at the UK Pocklington Canal location. Trace amounts of E2 (<8 pg/g) were detected at six of the sites sampled on occasion.

DIC sediment concentrations ranged from less than 1 pg/g to over 160 pg/g and was detected in all but one sample during the sampling campaign ( $n = 54$ ) (Figure 2). There were no significant differences in average concentrations between any of the nine European sites monitored, though concentrations were variable across this range at most sites. Given that DIC is not used in veterinary settings in the UK, the values in rural locations (e.g. Pocklington Canal) consistent with those in urban settings suggest a source associated with small rural WWTP that serves an approximate population of ~10,000 (and growing rapidly).

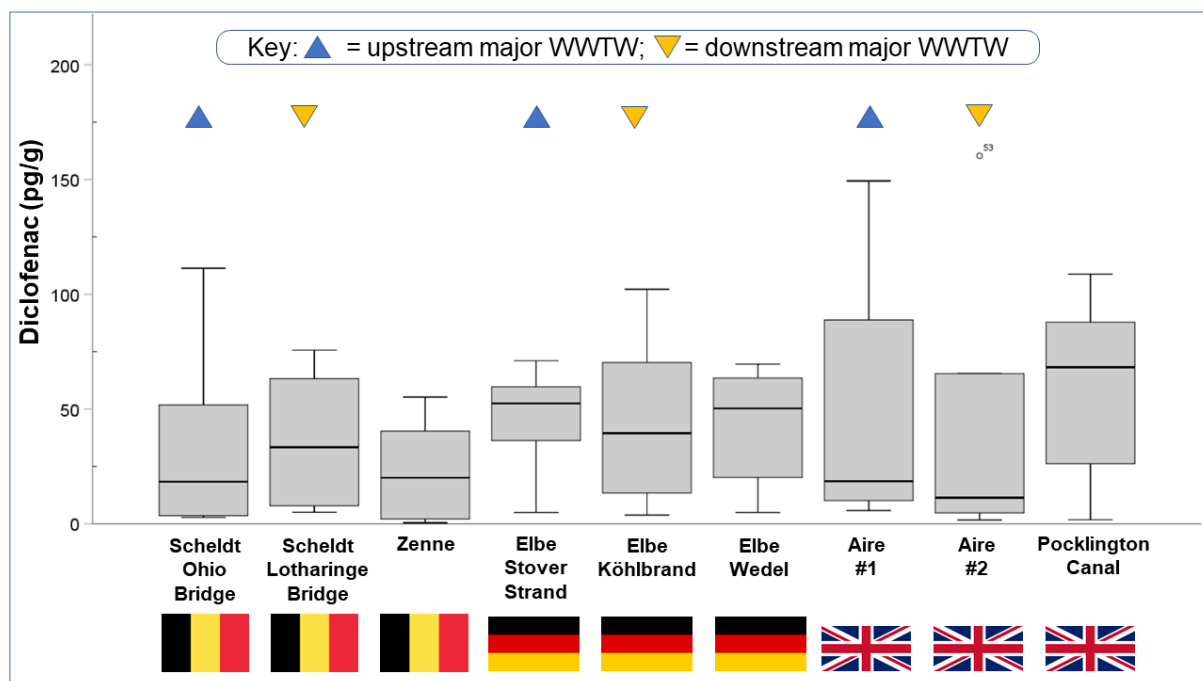


Figure 2. Diclofenac concentrations in river and estuary sediments across sample sites in the North Sea Region. Data show median (horizontal line), inter-quartile range (grey box) and range (extreme whiskers). Sample size = 9 for each site.

TCS sediment concentrations across the nine sampling sites show greater variability in absolute concentration than DIC and was present above detection limits in all but three samples (Figure 3). Significantly higher concentrations of TCS were present in River Aire sediments than any other sample location. There is however, no significant difference in concentration upstream and downstream of the major Knostrop WWTP which serves around 1 million people, suggesting that primary sources lie upstream, which could include other WWTP and storm sewer overflows. The higher concentrations in the River Aire relative to other sites may reflect the higher population density in this catchment compared to other systems. A further source may be attributed to biosolids application in agriculture settings in that region. Stutt *et al.* (2019) report that WWTP produced biosolids can contain anything from 200-1200  $\mu\text{g/kg}$  of TCS (in anaerobic digester biosolids at the top end) which may be applied to fields as manure. Efforts to phase out TCS use in personal care products have been apparent in recent years which should see declining concentrations in environmental settings in the future. However, the presence of TCS in modest concentrations in river sediments could delay the rate of decline environmental concentrations given the scope for storage and subsequent re-working and remobilisation of the sediment-bound TCS after high flow events.

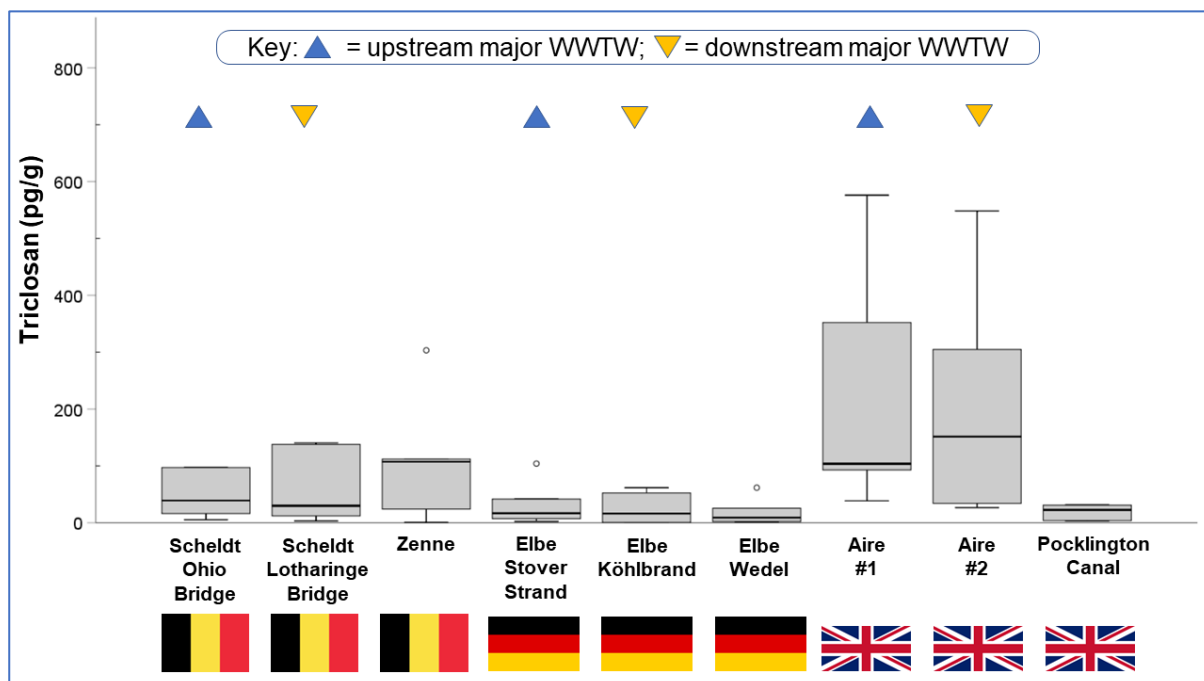


Figure 3. Triclosan concentrations in river and estuary sediments across sample sites in the North Sea Region. Data show median (horizontal line), inter-quartile range (grey box) and range (extreme whiskers). Sample size = 9 for each site.

### Can Environmental Quality Standards (EQS) be set and implemented?

There are EQSs for many chemicals, including proposed values for various WLCs, for both surface water and sediment (Table 2 below) including Predicted No Effect Concentration (PNEC). Occasionally the PNEC might be set at the Limit of Detection due to analytic constraints and a lack of knowledge, where it could cause a problem (according to the scientific literature) and it has been deemed sensible by regulators to set a limit than not.

Chemical	Type	Surface Water PNECs	'Proposed' EQS	Levels detected	Reference
E2	Natural hormone	None set	0.4 ng/L	pg/L – ng/L	SCHER, 2011
EE2	Synthetic estradiol hormone	0.35 ng/L 0.1 ng/L	None proposed	pg/L – ng/L	Caldwell <i>et al.</i> (2008) Laurenson <i>et al.</i> (2014)
DIC	Non-steroidal pain killer	116 $\mu$ g/L 0.1 $\mu$ g/L 0.05 $\mu$ g/L	0.1 $\mu$ g/L		Ferri <i>et al.</i> (2003) EU (2012) Carvalho <i>et al.</i> (2016) SCHER (2011)
TCL	Anti-microbial agent	4.7 ng/L 26.2 ng/L	0.02 $\mu$ g/L		Von der Ohe <i>et al.</i> (2012) Van Wijnen <i>et al.</i> (2018) LAWA (2010)

Table 2. Key information on possible WLC level ranges and standards for regulatory purposes.

### Cost benefits: is there a case for interventions and/or sediment remediation?

The assessment of the risks associated with sediment contamination is based on the evaluation whether this contamination constitutes a human, ecotoxicological or dispersal risk. If the contamination gives rise to unacceptable risks, a remediation must be carried out.

A framework for risk analysis and risk management (both *in situ* and *ex situ*) with background information can be used to assess a framework of remediation standards, allowing managers to make better informed decisions, and this is provided by OVAM (Public Waste Agency, Belgium), using the Sullied Sediments dataset as an example available from the Sullied Sediments website.

### **Relevance to sediment re-use**

The Sullied Sediments sampling sites are all subject to dredging and the costly disposal of contaminated sediments. The assessment whether dredged sediments can be reused on land is dependent on land use (human risks in different land-use scenarios) and potential leaching towards the groundwater or transport to surface waters. Whether a dredged sediment will be reused or be disposed is further dependent on the overall policy for beneficial reuse of waste and material. It should be stressed here that there is currently no requirement (in the UK) to take into account levels of E2, DIC and TCS when determining whether sediments are contaminated.



The way in which the Waste Framework Directive is implemented in the legislation differs in different member states. This results in different approaches towards reuse of non-contaminated sediments and removal and disposal of contaminated sediments. End-of-waste criteria can be developed to encourage reuse. Sediment that does not meet contamination thresholds that allow reuse, will require treatment unless this treatment is technically and/or chemically not feasible. The BATNEEC-principle is a factor in this feasibility assessment.

The contamination of the waterbed leads to higher dredging costs. Given that there is only a limited budget available, certain dredging activities may be temporarily postponed. This increases the risk that the pollution will spread over a larger area. The cost of keeping the waterways at depth varies with the both the environmental and technical quality of the sediment. The quality influences the price of storage, treatment, reuse and disposal.

It is useful to note that the Master Plan for inland navigation on Flemish waterways - Horizon 2020 speaks of an average cost of 20 to 45 euro/m<sup>3</sup> for the dredging of sediments. The cost for the reuse of non-contaminated dredged sediments is about 20 euros per m<sup>3</sup>. If sediments are dredged with the aim of being disposed, the cost increases to about 45 euros per m<sup>3</sup>. First results of the ongoing study in which a social cost benefit model for riverbed sediments is analyzed indicate that in a worst-case scenario, costs may rise up to approximately 120 euro/m<sup>3</sup> for the treatment or disposal of contaminated sediments.

Application of dredged sediments to agricultural land does not seem to represent an increased risk to human and environmental health. The concentrations of WLC we have measured in sediments are lower than those reported in sewage sludge which is currently applied to land (Ivanova *et al.* 2018; Radjenovic *et al.* 2009) and agricultural soils receiving organic amendments (Boxall *et al.* 2004).

Whether waterbed sediments should be remediated is based on the risk of the contaminant in the waterbed-water system and any ecological risks, using these can also be used to determine intervention levels.



### **Relevance to population-level effects using OMEGA**

Using the OMEGA approach (Wang et al. 2021), we calculated the Potentially Affected Fractions (PAF) attributed to E2, DIC and TCS. Using the Dutch National Institute for Public Health and the Environment (RIVM) toxicity database and EC50/NOEC values for DIC and TCS available in the literature, we can estimate that the PAFs would be high enough to exert toxicity, regardless of species sensitivity if detected in the sediments at such levels. PAF is the fraction of species affected by substances, based on traditional endpoints in lab assays. For the whole of Flanders, the average fraction of species affected was about 35%, with PAHs and metals contributing 23% and 9% (Wang *et al.* 2021). For an individual substance like cadmium, the fraction was 0.1% and 0.03 for the highest level detected and the water quality standard, respectively.

So, it should not be a surprise that the levels detected for individual pharmaceuticals as DIC and TCS are too low to contribute substantially to the overall risk. The relevant EC50/NOEC values are on average  $10^8$  higher than the calculated sediment pore-water levels. The situation is similar for E2 values: Using the highest E2 concentration of 0.009 ng/g sediment gives a very low PAF. Concentrations need to be more than  $10^6$  higher to achieve 5%. This is unsurprising in that Oldenkamp (2016), has previously reported that pharmaceutical drugs do not on average yield high risks according to these kinds of assessments. Sensitive endpoints such as feminisation caused by EDCs are not typically taken into account in such databases and quality standards that focussing on "population"-relevant endpoints growth, reproduction and survival. Yet, one cannot exclude that those subtle effects ultimately influence populations of some sensitive species substantially.

#### **Box 4: Keywords**

**Watch List** is a list of potential water pollutants that should be carefully monitored by the EU member states to decide if they pose a risk to the aquatic environment and whether EQS should be set for them.

**Predicted No Effect Concentration (PNEC)** is the concentration of a chemical at which no observed adverse effect can be measured. PNECs are designed to be both conservative and allow a prediction of the concentration at which a chemical is unlikely to have any toxic impact.

**Estrogens** are natural steroid hormones produced by vertebrate animals including humans.

**EQS** are levels that are set for chemicals, which are to be monitored in the environment. Intersex is a condition where an organism displays both male and female characteristics when they should be one or the other.

**Imposex condition** is a morphological disorder in marine snails where the female grows a penis on the head and their reproduction is impaired.

**Limit of quantification (LOQ)** - The analytical chemistry methods are robust enough to detect to these levels in a reliable, reproducible manner.

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