

**Comments of the
European Council for Alkylphenols and Derivatives
and
the Alkylphenols & Ethoxylates Research Council
On the Draft Background Document for 4-(1,1,3,3-tetramethylbutyl)phenol, ethoxylated (4-
tert-Octylphenol ethoxylates, 4-tert-OPnEO) Developed in the Context of ECHA's Fifth
Recommendation for the Inclusion of Substances in Annex XIV (June 24, 2013)**

**Submitted
September 23, 2013**

Executive Summary

The European Council for Alkylphenols and Derivatives (CEPAD) and the Alkylphenols & Ethoxylates Research Council (APEREC) jointly submit these comments in objection to the European Chemicals Agency (ECHA) proposal to include “4-(1,1,3,3-tetramethylbutyl) phenol, ethoxylated - covering well-defined substances and UVCB substances, polymers and homologues”, more commonly known as octylphenol ethoxylates (OPEs), under Annex XIV of REACH.

The Draft Background Document proposing the prioritization of OPEs for authorization provides rankings assigned by ECHA for the intrinsic properties, volumes in commerce in the EU, and dispersiveness of use of these compounds. As discussed below in these comments the background document overstates the priority assigned to the intrinsic properties and dispersiveness in the use of OPEs in the EU; therefore these assigned prioritization scores, as well as the total score, are not representative of this compound and overstate the need for its prioritization. In addition, the Draft Background Document for OPEs also does not adequately consider available environmental monitoring data that indicate that 4-*tert*-Octylphenol (4-tOP), a degradation intermediate of OPEs, which is the compound of actual interest, is not widely detected in EU water and when detected is generally below conservative Annual Average Environmental Quality Standards (AA-EQS) established for this compound under Directive 2000/60/EC (the Water Framework Directive). Furthermore, the Draft Background Document for OPEs does not consider that other existing regulatory instruments are already in place in the EU to control site specific emissions of OPEs and its degradation intermediate, 4-tOP.

The ECHA General Approach for Prioritisation of Substances of Very High Concern (SVHCs) for Inclusion in the List of Substances Subject to Authorisation states:

“Pursuant to Article 58(3) of the Regulation (EC) No 1907/20061 (REACH), whenever a decision is taken to include substances referred to in Article 57 of REACH in Annex XIV, priority shall normally be given to substances with PBT or vPvB properties, or wide dispersive use, or high volumes.

Article 58(3) indeed requires to take the mentioned 3 criteria ‘normally’ into account, but there is no provision that this needs to be done in all cases or how it should be done, e.g. with respect to evaluating, weighting or scoring of the criteria. Moreover, consideration

of further aspects and criteria for priority setting is not excluded. Hence, it can be assumed that Article 58(3) leaves discretion regarding the development and design of a prioritisation approach that in the end provides the Candidate Substances for which the recommendation to include them in Annex XIV is most relevant and appropriate (both in terms of potential risk and regulatory effectiveness) (ECHA, 2010, May 28).” (emphasis added)

OPEs do not themselves meet any of the inherent toxicity criteria for prioritization. OPEs are not persistent or bioaccumulative, nor are they carcinogenic (C), mutagenic (M) or reproductive (R) toxicants. OPEs were designated as a candidate chemical primarily on the basis that 4-tOP, one of its degradation intermediates, was previously designated as SVHC. The primary uses of OPEs in the EU are not widely dispersive applications and the monitoring data available for the EU supports this understanding.

The following comments provide further explanation to demonstrate that the intrinsic properties, volumes and uses of OPEs, along with available monitoring data in the EU do not support the addition of OPEs to Annex XIV. These comments also explain why authorization is not the most relevant and appropriate regulatory approach for addressing OPEs, both in terms of potential risk and regulatory effectiveness.

1.0 THE PRIORITIZATION SCORE IN THE BACKGROUND DOCUMENT FOR OPES OVERSTATES THE HAZARD FOR THE INTRINSIC PROPERTIES OF OPES.

OPEs were identified as a SVHC under Article 57(f) of Regulation (EC) 1907/2006 (REACH) “because (through their degradation) they are substances with endocrine disrupting properties for which there is scientific evidence of probable serious effects to the environment which give rise to an equivalent level of concern to those of other substances listed under Article 57(a) through (e) of REACH” (ECHA, 2013, June 24).

For prioritization, the hazard information that is available for a substance is scored (ranging from 0 to 4) and then the volume and dispersive use scores are added to obtain a total score. The total score can be seen as a proxy for potential risk to human health or the environment. Following are the scoring

criteria for inherent properties as listed in the ECHA General Approach for Prioritisation of SVHCs for Inclusion in the List of Substances Subject to Authorisation (ECHA, 2010, May 28).

Inherent properties	Score
PBT and vPvB or PBT with T non-threshold C or M	4
PBT or vPvB properties	3
C or M properties (without effect threshold)	1
C, M or R properties (with effect threshold)	0

The ECHA Background Document on OPEs gives a total inherent property score of 0 to 1 for these compounds, indicating that inherent properties of OPE are somewhere between a Carcinogenic (C), Mutagenic (M) or Reproductive Toxicant (R) with a threshold effect and a C or M toxicant without a threshold effect. The only listed inherent property given for OPEs in the ECHA Background Document is that of Art 57(f) "equivalent level of concern having probable serious effects to the environment". As discussed below, OPEs and 4-tOP are not Persistent, Bioaccumulative and Toxic (PBT), nor are they very Persistent or very Bioaccumulative (vPvB). OPEs and 4-tOP are also not C, M or R. Therefore, even based on inherent properties alone OPEs should not even be subject to prioritization

As described in companion papers by Staples et al (2008) and Klecka et al (2008) that review the persistence and bioaccumulation potentials for 4-tOP and their ethoxylates, neither of the parent compound nor any of its metabolites meet the various regulatory criteria for PBT or vPvB compounds, including those criteria listed in Annex XIII of REACH. In addition, neither OPEs nor 4-tOP meet the criteria for carcinogen, mutagen or reproductive toxicants category 1 or 2 in accordance with the DSD classification criteria or Cat 1A/1B in accordance with the CLP REGULATION (EC) No 1272/2008. It is important to also note that 4-tOP does not even meet the lesser criteria for Toxic to Reproduction Cat. 3 (DSD) or Cat 2 (GHS), which relates to "suspected human reproductive toxicants". 4-tOP is listed on the list of harmonised classification and labeling of hazardous substances based on its aquatic toxicity.

The fact that OPEs do not themselves meet any of the inherent toxicity criteria for prioritization should be basis enough not to prioritize these compounds for authorization.

2.0 THE USE AND EMISSION PROFILE OF OPEs DOES NOT SUPPORT PRIORITIZATION OF THESE COMPOUNDS UNDER ANNEX XIV, FURTHERMORE THEIR USE IS PROJECTED TO DECLINE.

The basis for the recommendation to prioritize OPEs for Authorization is that “these substances are used in high tonnage in products that can be assumed to lead to wide-dispersive emissions to the environment” (ECHA, 2013, June 24). The General Approach to Priority Setting for Authorization states “the extent to which a use is ‘wide-dispersive’ is roughly a function of the number of sites at which a substance is used and the magnitude of releases caused by those uses over all steps of the life-cycle” (ECHA, 2010, May 28). Therefore, the scoring of the ‘wide-dispersive use’ criterion is broken up in the two sub-criteria. The first is “Number of Sites”, which is basically the number of sites where the substance is used (i.e. the number of point sources or number of sites from which a substance is being released). The second is “Release”, which describes the releases in terms of pattern (where relevant) and amount versus anticipated risk.

2.1 The tonnage of OPEs used in the EU is declining

The Annex XIV Background Document for OPE acknowledges that since there are no registrants for OPEs under REACH, information on volumes, uses and the supply chain are lacking. Therefore, based on the estimated fraction of 4-tOP used to manufacture its ethoxylates and the estimated average contribution to the molecular weight of its ethoxylates, the volume of ethoxylates produced is assumed in the Background Document to be in the range of 1,000 – 10,000 t/y (ECHA, 2013, June 24). Based on this tonnage estimate the OPE Background document scores OPE as “high” or “7”.

The UK Risk Assessment on 4-tOP reported that 1,050 t/y OPE were used in 2001 (UK Environment Agency, 2005), based on 400 t/y of 4-tOP conversion to OPE. The UK Risk Assessment also recognized the agreement of the companies that supply OPEs in the EU to not promote OPEs as substitutes for nonylphenol ethoxylates (NPEs) as those surfactants were

subject to restrictions on their marketing and use in dispersive uses under EU Directive 2003/53/EC (European Parliament and the Council of the European Union, 2003, June 18). Due to antitrust regulations, APERC and CEPAD cannot share market and volume information directly. Current understanding of volumes for OPEs in the EU based on published reports indicate their tonnage to be in the lower half of the tonnage range estimated in the Annex XIV Background Document for OPEs with a decline in their use projected to be approximately 4.4% between 2009 and 2014 (Janshekar, H., 2010, July).

2.2 The primary uses of OPEs are not widely dispersive applications.

OPEs are used predominantly in the formulation of paint and coating products and are used at levels of generally 1% or less in those products. Due to their role in the emulsion polymerization process, OPEs are expected to be bound in the paint polymer and not widely dispersed to the environment. Waste from paint clean up are generally expected to be subject to treatment in wastewater treatment plants (WWTPs).

OPEs are not reported as being used in consumer applications with high potential for human exposure or environmental release i.e., household detergents and fabric softeners and personal care products (SRI, 2010). Furthermore, restrictions on the marketing and use of NPEs in dispersive uses under EU Directive 2003/53/EC is not resulting in replacement with OPEs, rather “other surfactants or blends of other surfactants are benefitting from the trend away from OPEs in these applications (Janshekar, H., 2010, July)”.

Some minor uses of OPEs (i.e., vitro diagnostic applications in the medical device sector) are also not expected to result in widespread dispersive emissions.

3.0 CONCENTRATIONS OF OPES AND 4-tOP IN EUROPEAN SURFACE WATERS DO NOT SUPPORT A NEED FOR AN EU-WIDE AUTHORIZATION PROCESS FOR OPES UNDER REACH.

When the UK Environment Agency conducted a risk evaluation on 4-tOP in 2005, information on the presence of 4-tOP and OPEs in the environment was limited; therefore the evaluation relied to a large extent on default assumptions and the Assessment Report acknowledges that its own “exposure assumptions may not be wholly realistic” (UK Environment Agency, 2005). That report also noted that at that time, surface water concentrations of 4-tOP in Europe and elsewhere were typically less than 1 µg/L, with “higher values detected on a few occasions” that may be “a consequence of high local discharges”. Since that time more environmental monitoring data are available for 4-tOP and to a lesser extent OPE; these data should be considered in the prioritization process for OPEs, as Article 58 (3) allows for “consideration of further aspects and criteria for priority setting” (ECHA, 2010, May 28).

The Water Framework Directive(WFD) established a framework for European Community (EC) water policy and strategies against water pollution, which requires Member States to take action for the progressive reduction of emissions of priority hazardous substances via the aquatic environment, through setting Environmental Quality Standards (EQS) and establishing emission control measures (European Parliament and Council 2000, 23 October Annual Average Environmental Quality Standards (AA-EQS) have been established for 4-tOP (European Parliament and Council, 2008, December 16). Monitoring for this compound has been conducted by the Member States under the WFD and additional monitoring has been published in the peer-reviewed literature.

3.1 Relevant Predicted No Effect Concentrations (PNECs) and Annual Average Environmental Quality Standard (AA-EQS) have been established for 4-tOP, which is the compound of interest.

3.1.1 PNECs for 4-tOP

There are reliable toxicity studies for fish, amphibians, and invertebrates for 4-tOP, which cover all parts of the test organisms’ life cycles from eggs to reproducing adults and cover life stages likely to be sensitive to an endocrine mode of action. Test procedures included screening tests, short-term reproduction tests, and full life-cycle tests. A consistent and treatment-related set of

No Observable Effect Concentration (NOEC) have been reported for 4-tOP and range from approximately 6 to 1,000 µg/L across relevant population-level endpoints related to survival, growth and development, and reproduction (CEPAD-APEREC, 2011, October 13, Coady et al, 2013, June 4, UK Environment Agency, 2005). Effects reported for endocrine sensitive endpoints occur at concentrations within the same range of NOECs and Lowest Observable Effect Concentrations (LOECs) that are also consistent with a narcotic mode of action (Coady et al, 2013).

The UK Risk Evaluation of 4-tOP calculated an intermittent exposure PNEC for 4-tOP of 0.13 µg/L based on the most sensitive acute toxicity value (EC50 for freshwater shrimp of 13.3 µg/L) and an assessment factor of 10 (UK Environment Agency, 2005). A chronic PNEC for surface water of 0.122 µg/L is calculated in the UK evaluation based on the most sensitive chronic study in fish (NOEC based on growth for rainbow trout) and an assessment factor of 50, which was applied with consideration for potentially more sensitive species (UK Environment Agency, 2005).

The REACH Chemical Safety Report (CSR) for 4-tOP utilized a species sensitivity distribution approach along with an assessment factor of five to calculate a freshwater PNEC of 0.632 µg/L for 4-tOP (CSR OP, 2010).

3.1.2 AA-EQS are established for 4-tOP under the WFD

Annual Average Environmental Quality Standards (AA-EQS) of 0.10 µg/L (inland waters) and 0.01 µg/L (other waters) have been established for 4-tOP under the WFD (European Parliament and Council (2008, December 16). AA-EQS values are considered protective against both chronic exposures and short-term pollution peaks in continuous discharges (European Parliament and Council (2008, December 16). While the PNEC of 0.632 µg/L calculated in the CSR for 4-tOP can be considered more reliable as it is based on a more robust data set, the AA-EQS developed under the WFD are the most conservative benchmarks for comparison to concentrations in water.

3.1.3 Established PNECs and AA-EQS for 4-tOP are protective of endocrine mediated effects.

OPEs were designated as SVHC primarily based on the argument that due to their degradation they are “an environmental source” of 4-tOP, which was previously designated SVHC due to concerns for environmental endocrine effects.

Based on the results of targeted *in vitro* studies, 4-tOP and nonylphenol (NP) have been shown to have a weak binding affinity for the nuclear estrogen receptor, and can, at sufficient concentrations, also cause subsequent estrogen-receptor dependent transactivation (Recchia *et al.*, 2004; Olsen *et al.*, 2005; Preuss *et al.*, 2006; Van den Belt *et al.*, 2004; Van Miller and Staples, 2005; USEPA, 2009). The estrogenic activity of both 4-tOP and NP varies, depending on the assay used, and is generally in the range of one thousand to one million-fold less potent than the endogenous estrogen, 17 β -estradiol (E2) (Coady *et al.*, 2010; Van Miller and Staples, 2005; Wenzel *et al.*, 2001).

Exposure to alkylphenols, specifically 4-tOP and NP exposure, can increase circulating levels of vitellogenin (VTG) in fish. VTG is a yolk-precursor protein normally expressed in female oviparous species that has been demonstrated to be a highly responsive biomarker for estrogen receptor agonists, especially in males who carry the VTG gene but do not ordinarily express it (Jobling and Sumpter, 1993; Harries *et al.*, 2000; Dussault *et al.*, 2005; Olsen *et al.*, 2005). VTG induction, which is not considered an adverse effect, occurs among various fish species at concentrations of 4-tOP and NP ranging from 1 to 100 $\mu\text{g/L}$ (Coady *et al.*, 2010; USEPA, 2007; Karels *et al.*, 2003; Jobling *et al.*, 1996; Rasmussen *et al.*, 2002; Seki *et al.*, 2003). In addition, reports of histopathological changes among gonadal tissues in fish exposed to either 4-tOP or NP have been reported in the range of 1.6 to 200 $\mu\text{g/L}$ (Miles-Richardson *et al.*, 1999; Gray and Metcalfe, 1997; Jobling *et al.*, 1996; Staples *et al.*, 2004; USEPA, 2007; Rasmussen *et al.*, 2005; Karels *et al.*, 2003; Rasmussen *et al.*, 2002; Gray *et al.*, 1999). While the observation of increased VTG in male fish and the occurrence of altered gonadal histopathology can inform upon one of the potential estrogenic modes of action of NP and 4-tOP, these biochemical and histopathological endpoints are not traditionally used as indicators of adverse effects in

ecological risk assessments. For 4-tOP and NP, the threshold for estrogenic activity (measured as induction of the yolk-precursor protein, VTG, and alterations in gonadal histomorphology) in fish is in the range of 1 to 200 µg/L. Therefore the previously described PNECs and AA-EQS are sufficiently protective of even these sensitive estrogenic responses in aquatic species.

3.2 OPEs were determined to be Substances of Very High Concern (SVHC) under REACH primarily based on the argument that due to their degradation they are “an environmental source” of 4-tOP, which was previously designated as SVHC: therefore the focus of environmental monitoring is most appropriately focused on 4-tOP.

Biodegradation has been shown to be the dominant mechanism responsible for removal of OPEs, 4-tOP and other alkylphenol (AP) and alkylphenol ethoxylates (APEs) during wastewater treatment and in the environment (Staples, 1999, Staples, 2001, Staples, 2008, Melcer, 2007). While OPEs are highly treatable in WWTPs, with removal rates commonly greater than 90%, low levels of their degradation metabolites have been reported in effluent and surface waters (Melcer, 2007). These intermediates continue to degrade in the environment, including mineralization of the phenolic ring, to carbon dioxide (Ahel, 1994, Staples, 1999, Staples, 2001, Staples, 2008, Naylor, 2006).

Considering that 4-tOP is the most toxic of the OPE degradation intermediates, and that degradation to 4-tOP is the primary reason that OPEs were proposed to be SVHC and are now proposed for prioritization for Authorization, the focus of environmental monitoring is most appropriately focused on 4-tOP.

3.3 Results from recent monitoring in the EU indicate that the majority of surface water samples do not contain detectable concentrations of 4-tOP; furthermore when detected, 4-tOP concentrations are generally below the AA-EQS.

Results of recent monitoring conducted in the EU are available through governmental monitoring programs and in the published literature. It should also be noted that all environmental monitoring results for 4-tOP represent emissions from all uses of 4-tOP, not just from the use of OPEs.

3.3.1 Results for 4-tOP from Monitoring Reported under the Water Information System of Europe (WISE)

As required under the WFD, surface water concentrations of 4-tOP and other substances have been measured in various European waterways. Monitoring data on 4-tOP from Fact Sheets published by the Environment Directorate-General, European Commission (DG ENV) under WISE were reviewed for the following names and CAS numbers for 4-tOP [CAS # 11081-15-5], 4-n-Octylphenol [CAS # 1806-26-4], Octylphenol [CAS # 140-66-9], Octylphenol [CAS # 67554-50-1], and Octylphenol [No specified CAS number]. Data from nine countries (Belgium, Czech Republic, Spain, France, Ireland, Luxembourg, Poland, Sweden, and United Kingdom) are summarized in the fact sheets, which covered the period from 2000 to 2008. The data were representative of a range of water categories including rivers (2497 samples from 354 stations), lakes (406 samples from 100 stations), coastal waters (22 samples from 18 stations) and estuaries (3 samples from 3 stations) (DG ENV, 2013a, DG-ENV, 2013b, DG-ENV, 2013c, DG-ENV, 2013d, DG-ENV, 2013e). Results for 4-tOP concentrations detected in whole water samples (liquid and suspended particulate matter) as part of this monitoring are summarized in Table 1 below along with a comparison to the AA-EQS for 4-tOP (0.10 µg/L).

Table 1: Summary of OP Concentration in Water Samples from Monitoring Data Listed in DG-ENV WISE Fact Sheets for 4-tOP (2000-2008)

Number of Analyses	N = 2795
Range	0 to 1.08 µg/L.
Mean ± SD	0.03 ± 0.05 µg/L
Median	0.03 µg/L
90 th Centile	0.05 µg/L
% samples < 0.10 µg/L	96%
0.10 µg/L < % samples < 1.0 µg/L	3.3 %
1.0 µg/L < % samples ≤ 1.08 µg/L	0.7 %

The concentrations of 4-tOP reported under the WFD monitoring are taken at discreet moments in time; therefore there were not sufficient data in the Fact Sheets to calculate average values over time in a particular location. Rather than relying on individual sample results, the median and upper 90th percentile concentrations better represent concentrations of 4-t-OP in these waters.

3.3.2 Results for 4-tOP Monitoring Conducted by the Member States under Water Framework Directive

Austria

Monitoring of 4-tOP concentrations was conducted under the Water Framework Directive by the Austrian Federal Agency for Water Management for the year 2004 (Federal Agency for Water Management, Austria, 2005). Of 403 samples taken from Austrian Waters in 2004, none exceeded the AA-EQS for 4-tOP (0.1 µg/L) and 226 samples (56%) are reported as non-detectable.

Switzerland

A report on monitoring data from the State of St. Gallen in Switzerland during a 2012 monitoring program where WWTP effluents were measured before dilution in the receiving waters indicates that 4-tOP was found above the detection limit of 0.025 µg/L in only one of 44 WWTP effluents. The one effluent sample where 4-tOP was detected contained 0.14 µg/L 4-tOP. After dilution, this corresponds to a concentration of 0.0001µg/L at this particular waste water treatment plant location (Office of Environment and Energy of the State of St. Gallen, Switzerland, 2013).

United Kingdom

The Department for Environment, Food and Rural Affairs (DEFRA) in the UK provided data tables with results of monitoring conducted for 4-tOP and OPE in the UK. Only 6 of 4143 samples tested for 4-tOP, or 0.1%, are reported at above the method Limit of Detection (LOD), which are listed as 0.1µg/L or 0.05 µg/L depending on the sampling location (UK DEFRA, 2013). Said differently, 99.9% of the UK samples were non-detectable at limits of detection that are less than or equal to the AA-EQS for 4-tOP. Of those samples that were non-detectable, 55% are reported as < 0.05 µg/L.

As expected, there are significantly less data reported for OPE. In the UK data tables for OPE, only 15 sample results are reported; however all are reported as non-detectable at LODs of 0.05 µg/L (6 samples) , 0.1 µg/L (5 samples) and 0.2 µg/L (4 samples) (UK DEFRA, 2013).

3.4 Monitoring results for 4-tOP reported in the published literature indicate that the majority of surface water samples in the EU contain non-detectable concentrations and those detected are generally below the AA-EQS (0.1 µg/L), which is protective even in chronic exposure situations.

Monitoring results for 4-tOP reported in the published, peer-reviewed literature indicate that the majority of surface water samples report non-detectable concentrations of 4-tOP and those detected are generally below the AA-EQS of 0.1 µg/L, which is protective even in chronic exposure situations. Following are summaries of the published monitoring data for 4-tOP in EU waters.

Ribeiro *et al.* (2008) reported monitoring results for 4-tOP in the Mondego River estuary on the west coast of Portugal. Samples were taken at high and low tides and in shallow and deep water at 8 locations. There were 12 sample results reported for 4-tOP. All results were reported at less than the detection limit of 2.0 ng/L, which is 50 time less than the AA-EQS for 4-tOP.

Jonkers *et al.* (2010) reported on the occurrence and concentrations of 4-tOP in Ria de Aveiro, a shallow coastal lagoon area in Portugal from a monitoring campaign that was conducted in 2006. Results (range, median, average) are provided for lagoons, harbors, sea water, sea water near WWTP outfall, city, rivers and WWTP effluent. With the exception of the rivers Caster and Antuã and WWTP effluent, the average and median concentrations of 4-tOP are reported at less than 1 ng/L. For all analytes, including 4-tOP, the highest concentrations were found in the river samples of Rio Caster and Rio Antuã, which the authors explain as being related to flow rates in those rivers. Nevertheless, all median and average results reported for 4-tOP, including

in undiluted WWTP effluent, are below the relevant AA-EQS for inland water (0.1 µg/L) and other wateri.e., marine (0.01 µg/L).

Colin et al (2013) reported the occurrence of 4-tOP and OPE2 in raw water and treated water samples from public water systems in a sampling campaign that was performed from October 2011 to May 2012. Sampling was equally distributed across 100 French departments. In total, 291 raw water samples and 291 treated water samples were analyzed in this study, which the authors state represents approximately 20% of the national water supply. Octylphenol monethoxylate (OPE1) and octylphenol ether carboxylate (OPEC) were not detected in any samples. 4-tOP was not detected in any surface water samples. 4-tOP was detected in only one ground water sample at a LOD of 17 ng/L, which is 6 times less than the AA-EQS for 4-tOP. 4-tOP, OPE and OPEC were not detected at all in any treated drinking water samples.

Esteban et al , 2013 analyzed a total of 30 compounds with endocrine activity, including natural and synthetic estrogens in the Jarama and Manzanares rivers, the main rivers in the Madrid Region (central Spain), which is the most densely populated area in Spain and also one of the most densely populated areas in Europe. There were 7 samples taken from the Mananares River and 7 samples taken from the Jarama River. Of the 7 samples taken from the Mananares River concentrations of 4-tOP exceeded the AA-EQS of 0.01 µg/L for “other” waters in 5 samples. Of the 7 samples from the Jarama River, 1 sample exceeded this AA-EQS. .

While there appears to be high contamination of all pollutants in the Mananares River, the sampling in this study was conducted in a limited time frame. The authors note that while 4-tOP was detected at concentrations exceeding the AA-EQS, there were insufficient data to calculate an average over time. The authors suggest that there is a need for further monitoring of this compound in both of these rivers.

The authors further note that the total estrogenicity in these two rivers did not exceed 1 ng/L Estradiol Equivalents Quotient (EEQ), which is the lowest level that may cause estrogenic effects in aquatic organisms, in any of the samples - even considering that 30 estrogenically

active compounds were monitored. The authors conclude that “the potential estrogenic risk to aquatic organisms in both rivers is low” (Esteban *et al.*, 2013).

Kotowska *et al.* (2013) monitored for phenols and pharmaceuticals in effluent from WWTPs in 9 cities in Poland. The study found that the removal efficiency for 4-tOP was 96% from wastewater. The range of undiluted effluent concentrations for 4-tOP was reported as non-detected to a maximum of 4.02 µg/L. The authors report that in 3 samples out of 172 samples the concentration of 4-tOP was above 1µg/L. More relevant is that the overall mean effluent concentration for 4-tOP is 0.02 µg/L, which is 5 times less than the AA-EQS of 0.1 µg/L and these concentrations will be diluted further in the receiving surface water.

Salgueiro-González *et al.* (2013) analyzed for alkylphenols in surface water, seawater and drinking water in the Coruna area in the northwest of Spain. Concentrations of 4-tOP in surface water were all less than the detection limit of 0.005 µg/L (n=5), which is 20 times less than the AA-EQS of 0.1 µg/L for inland surface waters. Concentrations of 4-tOP in seawater was 0.019 µg/L for one sample and less than the detection limit of 0.007 µg/L for 7 samples; therefore all but one seawater sample was less than the AA-EQS of 0.01µg/L for “other surface water”. Concentrations of 4-tOP in six drinking water samples were all below the level of detection for the method, which was 0.020 µg/L.

Stalter *et al.* (2013) reported monitoring for 26 sites impacted by wastewater effluent in several small rivers or streams and one mid-sized river, all in the Hessian Ried close to Frankfurt, Germany. Average concentrations of 4-tOP in water reported in this German study ranged between 12 and 147 ng/L, with an average result of 38 ng/L 4-tOP. With the exception of one sample (147 ng/L) all of the results in this river were below the AA-EQS for 4-tOP (0.1 µg/L).

Rocha *et al.* (2013) report concentrations of 4-tOP and other estrogenically active compounds in the Ria Formosa Lagoon in Portugal, which the authors state is highly impacted by discharge from 28 domestic and industrial WWTPs. The authors also note that these WWTPs have functional problems and, along with direct discharges from recreational boats and non-treated sewage, contribute to the pollution in this area. The authors state that this area is impacted by

metallurgic industries, which they note is associated with the use of Alkylphenol Ethoxylates (APEs) and represents 25% of the industrial production in the Ria Formosa area (Rocha *et al*, 2013).

This study found that APEs reached their maximal values in summer, which the authors attribute to “the scarcity of water from several riversides that usually supply the lagoon with fresh water and thus possibly dilute these chemicals in the channels” (Rocha, 2013). Concentrations of 4-tOP ranged from 5.9 to 43 ng/L, with 8 of the 10 samples slightly exceeding the AA-EQS (0.01µg/L) for 4-tOP in “other water” but none exceeding the AA-EQS (0.1µg/L) for inland waters.

Rocha *et al* (2013) report that the hormones estone (E1), 17β-estradiol (E2), 17α-ethynylestradiol (EE2), and a phytoestrogen sitosterol (SITO) were measured in considerable amounts in the Ria Formosa Lagoon. The authors also express concern for the total amounts of phosphorous and organophosphorus pesticides, which are present at up to ten fold higher than maximal concentrations recommended for rivers and streams..

These results indicate that discharge conditions in the Ria Formosa Lagoon can result in concentrations of 4-tOP that slightly exceed the AA-EQS for coastal waters. Considering the general pollution, presence of WWTPs “with functional problems”, and heavy industrial discharge in this area, it appears that efforts to improve municipal and industry wastewater treatment would benefit this water body. In addition, considering that other compounds appear to pose more risk to this area, prioritizing 4-tOP for authorization on an EU level does not appear to be the most relevant and appropriate approach for 4-tOP or OPEs, both in terms of potential risk and regulatory effectiveness.

4.0 THE PRIORITIZATION PROCESS FOR OPES SHOULD ALSO CONSIDER THAT 4-tOP IS NOT WIDELY DETECTED IN EU WATERS AND, WHEN DETECTED, IS GENERALLY BELOW THE CONSERVATIVE AA-EQS FOR THIS COMPOUND.

The Background Document recommending OPEs for prioritization for Annex XIV of REACH calculates a “relatively high” to “high” priority for inclusion in Annex XIV based scores of 0-1 for inherent properties (IP); 7 for high volume (V) and 9 for wide dispersive uses (WDU). However, as noted in section 2.0 of these comments, most uses of OPE are industrial, not consumer applications; therefore the number of sites and scope of dispersiveness is not as great as estimated in the Background Document prioritization. Also, the available environmental monitoring data for waters in the EU indicate that most samples of surface water tested did not detect 4-tOP at the method LOD and, when detected, most measured concentrations are less than the AA-EQS for this compound.

Article 58(3) provides for discretion regarding the development and design of a prioritisation approach that in the end provides the Candidate Substances for which the recommendation to include them in Annex XIV is most relevant and appropriate (both in terms of potential risk and regulatory effectiveness) (ECHA, 2010, May 28). Therefore, the prioritization process for OPEs should consider the available monitoring data and the score for dispersiveness should be subject to modification to reflect a lesser degree of dispersiveness and potential risk.

5.0 THERE ARE OTHER REGULATORY INSTRUMENTS IN PLACE IN THE EU TO CONTROL EMISSIONS OF OPES AND 4-tOP .

Recent monitoring studies in the EU show that concentrations of 4-tOP that exceed the AA-EQS are associated with specific locations and points in time, which are otherwise polluted or subject to intense or uncontrolled discharges. The following regulations are already in place in the EU to control emissions and environmental risks from OPEs and/or 4-tOP.

The Water Framework Directive (European Parliament and Council, 2000, 23 October Directive 2000/60/EC) established a framework for Community action in the field of water policy, which requires the Members States to measure aquatic concentrations relative to established Environmental Quality Standards (EQS) and to take action in case this value is exceeded. The monitoring data described in section 3.0 above notes specific locations and moments in time where concentrations of 4-tOP slightly exceed its AA-EQS. For the most part, these locations

have generalized problems with contamination that are most appropriately addressed under the WFD.

A UK voluntary industry agreement for the reduction in risk from NP, NPEs and 4-tOP and OPEs was finalized in 2004 (CSI, 2004, April). This agreement, which has impacted the EU market more generally, was taken to reduce the risks from NP/NPEs and 4-tOP/OPEs with the following objectives:

- Rapidly reduce the risk from NP/NPE to the environment by making early progress in replacing NP/E in a number of uses and to minimise discharges into the environment in order to reduce existing risks to the environment;
- Prevent the development of new risks from 4-tOP/E by preventing the use of 4-tOP/OPEs as substitutes for NP/E for those uses to be phased out; and
- Reduce the risks from 4-tOP/OPE by phasing out any dispersive uses of 4-tOP/OPE in sectors targeted by the M&U Directive for NP/NPE

The Integrated Pollution Prevention and Control (IPPC) Directive (96/61/EC) lays down measures designed to prevent or, where that is not practicable, to reduce emissions to air, water and land from the activities mentioned in Annex I to the Directive (European Parliament and Council, 1996, September 24).

Annex I of the IPPC Directive lists categories of industrial activities subject to regulation by the Directive. Surfactants and surface active chemicals are specifically covered under Annex I. Since OPEs are surfactants they are specifically covered by the IPPC Directive. Other categories of industrial activities that are subject to the IPPC Directive that are relevant to the major use of OPEs in paint and coatings include the chemical industry, including basic polymers and dyes and pigments. Other industrial activities subject to the IPPC directive that may be relevant to other minor uses of OPE include: energy industries, the production and processing of metals, chemical installations for the production of basic plant health products and biocides, installations using a chemical or biological process for the production of basic pharmaceutical products, waste management installations, and landfills. Industrial activities subject to IPPC where OPE use is not expected due to the voluntary agreement mentioned earlier in these comments include

industrial plants that process pulp and paper, plants for the pre-treatment or dyeing of fibers and textiles and tanning facilities.

In addition, Annex III to the IPPC Directive is a list including the main polluting substances in water to be taken into account, which includes “Substances and preparations which have been proved to possess carcinogenic or mutagenic properties or properties which may affect reproduction in or via the aquatic environment”. As noted in section 1.0, neither OPEs nor 4-tOP are C, M or R toxicants; however, if there is concern about the environmental impact of either the IPPC Directive provides an existing regulatory mechanism for addressing these compounds.

6.0 APERC AND CEPAD RECOMMEND THAT OPES DO NOT WARRANT PRIORITIZATION FOR AUTHORIZATION UNDER ANNEX XIV OF REACH BECAUSE THEY DO NOT THEMSELVES MEET THE PRIORITIZATION CRITERIA FOR INHERENT TOXICITY, ARE NOT USED IN WIDELY DISPERSIVE CONSUMER APPLICATIONS AND ARE NOT DETECTABLE WIDELY IN THE WATERS OF THE EU; FURTHERMORE, LOCATIONS WITH EXCEEDANCES OF AA-EQS CAN BE ADEQUATELY CONTROLLED THROUGH EXISTING REGULATIONS

OPE themselves do not meet any of the inherent toxicity criteria for prioritization for authorization, therefore on this basis alone should not be subject to prioritization for authorization. Furthermore, uses of these OPEs are generally not dispersive and the focus on OPEs for prioritization over other SVHC compounds is inappropriate. This is confirmed by recent environmental monitoring in the EU, which should be considered in the priority setting process for OPEs. Monitoring indicates that 4-tOP, the compound of actual interest in this case, does not have widespread occurrence in EU waters.

Existing regulatory instruments exist in the EU, which are better suited to address specific locations where concentrations of 4-tOP are detectable and of concern relative to the conservative AA-EQS for 4-tOP. 4-tOP, the degradation intermediate of OPE that is the stated

concern for prioritization, is already regulated under the Water Framework Directive 2000/60/EC. In addition, 4-tOP and OPE are regulated under the IPPC Directive (96/61/EC) and are subject to a voluntary agreement among manufacturers not to promote the use of OPEs in dispersive uses that lead to entry in the aquatic environment (CSR, 2004, April). These existing regulations provide grounds for an exemption for OPEs from prioritization under Art. 58(2) of Regulation 1907/2006/EEC.

As Rocha *et al* (2013) found in the Ria Formosa Lagoon, concentrations of 4-tOP that slightly exceed the AA-EQS are generally associated with areas impacted by general pollution, i.e., due to WWTPs “with functional problems”, and heavy industrial discharge. It appears that efforts to improve municipal and industry wastewater treatment in categories already regulated under the WFD and IPPC Directive would benefit water bodies such as this more effectively than an authorization process for OPE under REACH. Also, considering that other compounds appear to pose more risk to these areas, prioritizing 4-tOP for authorization under REACH is not the most relevant and appropriate approach, both in terms of potential risk and regulatory effectiveness.

The basis for given for prioritizing OPE for authorization is a concern for the environmental estrogenic activity of the degradant 4-tOP. Esteban *et al*, 2013 found that the total estrogenicity in the two rivers with the highest reported concentrations of 4-tOP – as well as 29 other estrogenically active hormones, phytoestrogens and industrial compounds - did not exceed 1 ng/L Estradiol Equivalents Quotient (EEQ). This is the lowest level that may cause estrogenic effects in aquatic organisms, in any of the samples. The authors conclude that “the potential estrogenic risk to aquatic organisms in both rivers is low.” Considering this, prioritizing OPE for authorization does not appear to be necessary to address concerns of environmental estrogenicity from 4-tOP.

For these reasons, APERC and CEPAD recommend OPE should not be prioritized for authorization under REACH and inclusion in Annex XIV.

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