A P E RESEARCH COUNCIL

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March 16, 2010

Ms. Traci Iott
CT Department of Environmental Protection
Bureau of Water Protection and Land Reuse
Planning & Standards Division
79 Elm Street
Hartford, Connecticut 06106-5127

SUBJECT: COMMENTS ON CRITERIA FOR NONYLPHENOL IN PROPOSED REVISIONS TO CONNECTICUT WATER QUALITY STANDARDS (DECEMBER 22, 2009)

Dear Ms. Iott:

The Alkylphenols & Ethoxylates Research Council (APERC)[1] appreciates this opportunity to provide these comments as a to supplement to the statement I made at the public hearing on February 3, 2010 on the Connecticut Department of Environmental Quality (DEP) proposed revisions to the Connecticut Water Quality Standards (CT WQS).[2] APERC's mission is to promote the safe use of alkylphenol-based products, such as nonylphenol (NP) and nonylphenol ethoxylates (NPE), within the framework of responsible chemical management. As such, we support DEP's proposal to adopt the US EPA Ambient Aquatic Life Water Quality Criteria (WQC) for NP as the numeric water criteria for this compound in Appendix D of the CT WQS.[3]

US EPA conducted a review of the hundreds of available ecotoxicity studies for NP when they developed the WQC for NP. The Agency used data from a wide range of taxa and species to develop WQC that are, as EPA states, "an estimate of the highest concentration to which an aquatic community can be exposed indefinitely without unacceptable effects."[4]

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[1] Members of the Alkylphenols & Ethoxylates Research Council include: Dover Chemical Corporation, The Dow Chemical Company, SI Group, and TPC Group.
[2] Connecticut Department of Environmental Protection (CTDEP). (2009, December 22). Proposed Revisions to Connecticut Water Quality Standards.
http://www.ct.gov/dep/lib/dep/water/water_quality_standards/water_quality_standards_proposed_12_22_09.pdf.
[3] US Environmental Protection Agency (US EPA). (2006, February 23). Notice of availability of final aquatic life ambient water quality criteria for nonylphenol. Federal Register, 71 (36), 9337-9339.
http://www.epa.gov/fedrgstr/EPA-WATER/2006/February/Day-23/w2558.htm.
[4] US Environmental Protection Agency (US EPA). (2005). Aquatic life ambient water quality criteria - nonylphenol. Report 822-R-05-005. US Environmental Protection Agency, Washington, DC, USA.
http://www.epa.gov/waterscience/criteria/nonylphenol/final-doc.pdf

US EPA also used studies on the long-term effects of NP in deriving the chronic NP WQC. These types of studies look at effects at the level of the fish or organism, such as their ability to reproduce and developmental effects in offspring. These types of effects reflect the culmination of changes that can occur at the molecular, biochemical and tissue level. Therefore, the NP WQC address all mechanisms of toxicity– including any that might be due to the compound's weak estrogenic activity.

EPA finalized the Water Quality Criteria for NP in 2006 and since that time additional research has been published on this compound. So, APERC sponsored a project to assess the recent data to see if the criteria are still valid. That assessment, which is summarized in the attachment to these comments, found that the most recent data still support EPA's WQC for NP.

NP and NPEs are treatable in wastewater treatment plants and they are neither persistent nor bioaccumulative; therefore they can be effectively managed using WQS and, as necessary, National Pollutant Discharge Elimination System (NPDES) permits to monitor and control effluent concentrations.[5,6,7,8] Hence, incorporating the federal WQC for NP into CT WQS is not likely to be a technical or economic burden on either the DEP or the local business community in Connecticut.

Adopting the federal WQC for NP will provide a clear definition of ambient aquatic concentrations of NP that are protective of the aquatic environment; thereby assuring that surface and ground waters in Connecticut are protected from degradation as required under the Clean Water Act.

Sincerely,

Barbara S. Losey

Barbara S. Losey
Deputy Director

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[5] Melcer, H., Klečka, G., Monteith, H., Staples, C. (2007) Wastewater Treatment of Alkylphenols and Their Ethoxylates : A State of the Science Review. Published by Water Environment Federation, Alexandria, VA.

[6] Environment Canada (EC). (2007). Ecological categorization of substances on the Domestic Substance List; Categorization decisions. (Completed in September 2006). http://www.ec.gc.ca/substances/ese/eng/dsl/cat_index.cfm

[7] Staples, C.A., Klecka, G.M., Naylor, C.G., & Losey, B.S. (2008). C8- and C9-alkylphenols and ethoxylates: I. Identity, physical characterization, and biodegradation pathways analysis. Human and Ecological Risk Assessment, 14 (5), 1007–1024.

[8] Klecka, G.M., Staples, C.A., Naylor, C.G., Woodburn, K.B., & Losey, B.S. (2008). C8- and C9-alkylphenols and ethoxylates: II. Assessment of environmental persistence and bioaccumulation potential. Human and Ecological Risk Assessment, 14 (5), 1025–1055.

**ATTACHMENT
ALKYLPHENOLS & ETHOXYLATES RESEARCH COUNCIL
COMMENTS ON CRITERIA FOR NONYLPHENOL
IN PROPOSED REVISIONS TO
CONNECTICUT WATER QUALITY STANDARDS
(DECEMBER 22, 2009)**

The Connecticut Department of Environmental Protection (DEQ) proposes to adopt the following numeric aquatic life criteria for nonylphenol (NP) in their revised Water Quality Standards (WQS). [9]

**Numeric Aquatic Life Criteria for NP
in Proposed CT WQS
(December 22, 2009)**

	Acute (µg/L)	Chronic (µg/L)
Fresh Water	28	6.6
Salt Water	7	1.7

These values are consistent with those finalized by the US EPA Office of Water as the federal aquatic life ambient Water Quality Criteria (WQC) for NP.[10] In developing these WQC, EPA conducted a significant review of the available data for NP and utilized a statistical extrapolation procedure that draws upon both acute and chronic toxicity data from a wide range of taxa and species. EPA's WQC represent "an estimate of the highest concentration to which an aquatic community can be exposed indefinitely without unacceptable effect." [11]

In the case of NP, EPA used results from acute studies to statistically calculate a Final Acute Value (FAV) along with results for apical endpoints (e.g., reproduction and growth) from chronic tests to calculate acute-to-chronic ratios. Since the chronic endpoints used to derive the chronic NP WQC reflect the culmination of molecular, biochemical and tissue-level effects at the whole organism level, the NP WQC in turn addresses all mechanisms of action - including estrogenic effects - that result in

[9] Connecticut Department of Environmental Protection (CTDEP). (2009, December 22). Proposed Revisions to Connecticut Water Quality Standards.
http://www.ct.gov/dep/lib/dep/water/water_quality_standards/water_quality_standards_proposed_12_22_09.pdf

[10] US Environmental Protection Agency (US EPA). (2006, February 23). Notice of availability of final aquatic life ambient water quality criteria for nonylphenol. Federal Register, 71 (36), 9337-9339. http://www.epa.gov/EPA-WATER/2006/February/Day-23/w2558.htm.

[11] US Environmental Protection Agency (US EPA). (2005). Aquatic life ambient water quality criteria - nonylphenol. Report 822-R-05-005. US Environmental Protection Agency, Washington, DC, USA.
http://www.epa.gov/waterscience/criteria/nonylphenol/final-doc.pdf

measurable alterations in these apical endpoints. Although NP has been shown to have weak estrogenic activity, EPA noted in the NP WQC document that "the ability of nonylphenol to induce estrogenic effects has seldom been reported at concentrations below the freshwater Final Chronic Value of 6.5965 μg/L."[12]

The EPA WQC were developed using data available for NP as of 2005 and the Agency's conclusions were consistent with a species sensitivity distribution analysis based on essentially the same chronic data set conducted by Staples *et al.* (2004), which calculated a similar freshwater chronic value based on 90 chronic toxicity values for NP reported for 16 species of freshwater aquatic invertebrates and vertebrates.[13]

Since the finalization of the NP WQC additional ecotoxicity data have been reported; therefore, the Alkylphenols & Ethoxylates Research Council (APERC) recently undertook a comprehensive literature search to identify any studies published on NP since EPA finalized the NP WQC as well as any that may have been missed in EPA's review. The practices and criteria employed by EPA to determine whether studies were valid and relevant for use in development of WQC were also used in the APERC literature review. Studies were deemed valid if they contained a thorough description of the experimental design, had a clear linkage between reported findings and the experimental design, contained an ecologically relevant apical endpoint, such as growth, survival or reproduction, and exhibited adequate performance of controls.

APERC also examined studies investigating the effects of NP on secondary endpoints, such as behavioral effects, induction of biochemical markers, or alterations in cells within tissue. From these studies, the types of endpoints being measured, the range of effect concentrations associated with NP, and the possible mechanisms of action in various aquatic species were examined. In summary, there were a total of 30 recent studies (17 with freshwater and 13 with marine species) that examined apical endpoints (survival, growth and reproduction) relevant for risk assessment of NP in a broad range of species (i.e., fish, frogs, echinoderms, crustaceans, mollusks, and diatoms). All but a few studies, which are discussed below, reported no observable effect concentration (NOEC) results that were greater than the chronic NP WQC; providing a weight-of-evidence that supports EPA's current NP WQC.

Freshwater Studies

From all the fresh water studies on NP identified subsequent to development of the EPA WQC in 2005, only one study by Seki *et al.* (2003) reported an ecologically relevant

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[12] US EPA. (2005).

[13] Staples, C., Mihaich, E., Carbone, J., Woodburn, K., & Klečka, G. (2004). A weight of evidence analysis of the chronic ecotoxicity of nonylphenol ethoxylates, nonylphenol ether carboxylates, and nonylphenol. Human and Ecological Risk Assessment, 10 (6), 999-1017.

NOEC for NP that was below the current chronic fresh water WQC of 6.6 μg/L.[14] In this study, medaka fish (*Oryzias latipes*) were exposed to NP continuously from fertilized eggs to 60 days post-hatch. Sex ratio was altered and the biomarker plasma VTG was induced at 11.6, but not 6.08 μg/L. A chronic value (ChV), calculated as the geometric mean of the NOEC and the lowest observed effect concentration (LOEC), was calculated for this study based on altered sex ratio. The use of ChVs is in accordance with the approach used by EPA to calculate WQC. The ChV for the Seki *et al.* (2003) study is 8.4 μg/L, which is greater than - and therefore supports - EPA's freshwater chronic WQC for NP.

Saltwater Studies

With regards to recent studies with saltwater organisms, five recent toxicity studies with Pacific oysters, sea urchins, and mysid shrimp reported LOECs below the current saltwater chronic WQC of 1.7 μg/L for NP.[15,16,17,18,19]

Closer inspection of the oyster studies by Nice *et al.* studies (2001, 2003) found experimental uncertainties including: low sample sizes, inconsistent results for some endpoints, possible solvent effects, and a reported 91% loss of test substance after 6 hours. Therefore these studies by Nice *et al.* are not suitable for WQC hazard assessment as described by EPA guidance.[20] A previous oyster study by Nice *et al.* (2000)[21] that reported developmental effects at 100 μg/L NP on the Pacific oyster (*Crassostrea gigas*) exposed for 72 hours was considered by EPA in development of the NP WQC.

As reported by Arslan *et al.* (2007), spermiotoxicity effects on larval development of the sea urchin *Paracentrotus lividus* were observed at all concentrations of NP down to 0.937

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[14] Seki, M., Yokota, H., Maeda, M., Tadokoro, H., & Kobayashi, K. (2003). Effects of 4-nonylphenol and 4-tert-octylphenol on sex differentiation and vitellogenin induction in medaka (Oryzias latipes). Environmental Toxicology and Chemistry, 22, 1507-1516.

[15] Nice, H.E., Crane, M., Morritt, D., & Thorndyke, M. (2001). Endocrine disrupters, critical windows and developmental success in oyster larvae. In: Atkinson, D., & Thorndyke, M. (Eds). Environment and Animal Development. BIOS Scientific Publishers, Oxford, UK. 203-217.

[16] Nice, H.E., Morritt, D., Crane, M., & Thorndyke, M. (2003). Long-term and transgenerational effects of nonylphenol exposure at a key stage in the development of Crassostrea gigas. Possible endocrine disruption? Marine Ecology Progress Series, 256, 293-300.

[17] Arslan, O.C., Parlak, H., Oral, R., & Katalay, S. (2007). The effects of nonylphenol and octylphenol on embryonic development of sea urchin (Paracentrotus lividus). Archives of Environmental Contamination and Toxicology, 53, 214-219.

[18] Arslan, O.C., & Parlak, H. (2007). Embryotoxic effects of nonylphenol and octylphenol in sea urchin Arbacia lixula. Ecotoxicology, 16, 439-444.

[19] Hirano, M., Ishibashi, H., Kim, J., Matsumura, N., & Arizono, K. (2009). Effects of environmentally relevant concentrations of nonylphenol on growth and 20-hydroxyecdysone levels in mysid crustacean, Americamysis bahia. Comparative Biochemistry and Physiology, Part C, 149, 368-373.

[20] Stephan, C.E., Mount, D.I., Hansen, D.J., Gentile, J.H., Chapman, G.A., & Brungs, W.A. (1985). Guidelines for deriving numerical national water quality criteria for the protection of aquatic organisms and their uses. PB85-227049. US Environmental Protection Agency. http://www.epa.gov/waterscience/criteria/library/85guidelines.pdf

[21] Nice, H.E., Thorndyke, M.C., Morrittt, D., Steele, S., & Crane, M. (2000). Development of the Crassostrea gigas larvae is affected by 4-nonylphenol. Marine Pollution Bulletin, 40, 491-496.

μg/L..[22] However, these results are in contrast to other work reporting effects on the sea urchin, *P. lividus,* in which sea urchin sperm was exposed to NP for 72 hours after which an EC50 of 270 μg/L based on sperm toxicity was recorded.[23] In addition, embryotoxic effects were observed by the same authors in another species of sea urchin, *Arbacia lixula,* at all concentrations of NP down to 0.937 μg/L.[24,25] However, the results from the studies were confounded by apparent solvent effects and a lack of test chemical measurements in the static tests.

Finally, a 14-day study on effects of NP in the mysid crustacean, *Americamysis bahia* conducted by Hirano *et al.* (2009) examined growth, sex ratio, number of molts and hormone levels. The LOEC for the study, based on significantly reduced body length compared to controls, was reported as 1 μg/L NP, and the NOEC was reported as 0.3 μg/L NP. The fact that there was a lack of test chemical measurements in the test vessels indicates that the results should be interpreted with caution. This approach is particularly warranted considering that the results of another, less equivocal, chronic study in which mysid shrimp were exposed to measured concentrations of NP ranging from 3.9-9.1 μg/L NP for 28-days reported reduced growth at 6.7 μg/L NP, but not 3.9 μg/L NP.[26]

In summary, the studies conducted with Pacific oysters (Nice *et al*., 2001; Nice *et al*., 2003) and sea urchins (Arslan *et al*., 2007; Arslan and Parlak 2007) and mysid shrimp (Hirano *et al*., 2009) exhibited methodological uncertainties in their study design and pre-existing toxicity studies on the same or similar organisms did not corroborate the reported toxicity of NP at concentrations less than the current saltwater chronic value of 1.7 μg/L. Thus, there are no definitive data in the recent literature to contraindicate that the current saltwater chronic value of 1.7 μg/L for NP is sufficiently protective of saltwater communities.

Summary

An abundant data set of apical and secondary endpoints exists for NP and the results of valid studies published since finalization of EPA's WQC for NP in 2006 continue to support the current NP WQC as being sufficiently protective of fresh and saltwater aquatic species.

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[22] Arslan et al. (2007).
[23] Ghirardini, A.V., Novelli, A.A., Likar, B., Pojana, G., Ghetti, P.F., & Marcomini, A. (2001). Sperm cell toxicity test using sea urchin Paracentrotus lividus Lamarck (Echinodermata: Echinoidea): Sensitivity and discriminatory ability toward anionic and nonionic surfactants. Environmental Toxicology and Chemistry, 20, 644-651.
[24] Arslan et al. (2007).
[25] Arslan & Parlak . (2007).
[26] Ward, T.J., & Boeri, R.L. (1991, May 16). Early life stage toxicity of nonylphenol to the fathead minnow, Pimephales promelas. Envirosystems Division, Resource Analyst, Incorporated, Hampton, New Hampshire. Final Report #8979 to the Chemical Manufacturers Association, Washington, DC, USA.