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Edmund G. Brown Jr.
Governor

TO: Richard W. Corey, Chief
Stationary Source Division
California Air Resources Board

FROM: 
Victoria A. Whitney, Deputy Director
DIVISION OF WATER QUALITY

DATE: JUN 20 2011

SUBJECT: **RESPONSE TO COMMENTS SUBMITTED BY THE ALKYLPHENOLS
AND ETHOXYLATES RESEARCH COUNCIL ON AIR RESOURCES
BOARD'S PROPOSALS TO PROHIBIT USE OF ALKYLPHENOL
ETHOXYLATE SURFACTANTS IN CERTAIN CONSUMER PRODUCTS**

As you requested, I am responding to comments submitted by the Alkylphenols and Ethoxylates Research Council (APERC) on the Air Resources Board's (ARB) proposed amendments to the Consumer Products Regulation. Specifically, on November 17, 2010, APERC submitted detailed comments on both the proposed prohibitions on use of alkylphenol ethoxylate (APEO) surfactants in certain consumer product categories, as well as on our memorandum to you dated September 20, 2010. Our response to these comments is attached to this memorandum.

In our September 20, 2010 memorandum, we summarized information received from the Southern California Coastal Water Research Project (SCCWRP) and the San Francisco Estuary Institute (SFEI) that indicated the level of aquatic toxicity posed by APEOs is high enough to cause concern. These surfactants are being discharged into coastal, estuarine, and freshwater by means of wastewater treatment plants (WWTP), stormwater, and other sources. We also indicated that APEOs tend to bioaccumulate in marine vertebrates and invertebrates, and persist in environmental compartments such as sediments. Both SCCWRP and SFEI are well-respected scientific entities whose missions are to provide a scientific basis for developing strategies to protect the California coastal and/or estuary environments. Our memorandum went on to support ARB actions to prohibit use of APEOs. Based on our memorandum, your staff proposed, and your Board approved, prohibitions on use of APEO surfactants in certain consumer products categories.

California Environmental Protection Agency

The comments submitted by APERC do not change the information or recommendation conveyed in our previous memorandum to you on this subject dated September 20, 2010. To reiterate, information submitted by SWAMP, SFEI and SCCWRP indicate that the level of aquatic toxicity posed by APEOs is high enough to cause concern. In fact, levels measured in several California waterways exceed WQC established by some jurisdictions. Given the wide availability of effective, safer alternatives, coupled with U.S. EPA and other jurisdictional actions, we continue to support ARB's prohibition on use of APEOs in non-aerosol General Purpose Cleaners, Glass Cleaners, General Purpose Degreasers, Heavy Duty Hand Cleaner or Soaps, and all forms of Oven Cleaners. If you have any questions, please contact either Ms. Liz Haven, Assistant Deputy Director, at (916) 341-5457 or Ms. Kim Ward at (916) 341-5586.

Attachment

Response to Comments from APERC

The Alkylphenols and Ethoxylates Research Council (APERC) comments were, on the whole, critical of our recommendation and of Southern California Coastal Water Research Project's (SCCWRP) and San Francisco Estuary Institute's (SFEI) interpretations of relevant literature on the toxicity and/or environmental fate of alkylphenol ethoxylates (APEOs) and their breakdown products in aquatic environments. The APERC reviewers made numerous specific comments regarding SCCWRP and SFEI concerns, offered alternative interpretations of relevant studies, and also cited other literature not originally referenced by SCCWRP and SFEI in their previous reviews. APERC's comments can be summarized as follows:

- 1) Concentrations of APEOs in California waterways are low and are not expected to exceed the United States Environmental Protection Agency's (U.S. EPA) water quality criteria (WQC) for nonylphenol (NP), a metabolite of nonylphenol ethoxylate (NPE);
- 2) Concentrations of APEOs in sediment are below predicted no effect concentrations (PNEC) for NP;
- 3) APEOs are highly treatable in Waste Water Treatment Plants (WWTPs) and their degradation intermediates are not persistent or bioaccumulative in the environment;
- 4) APEOs are not a major source of estrogenic activity in wastewater treatment effluent; and
- 5) Rather than prohibiting use of APEO surfactants, concentrations should be monitored to evaluate occurrence of increases.

We would like to take this opportunity to respond to APERC's comments. In responding to APERC we note that our comments are reflective of SFEI's and SCCWRP's original commentary and analysis, supplemented by additional background materials obtained during our review of APERC's November 17, 2010, comment letter. Also note that our response does not address all potentially relevant issues which may pertain to aquatic APEOs, *e.g.*, APEO bioconcentration and endocrine disruption in aquatic/marine mammals and birds.

For clarity I also would like to reiterate that APEOs are used in various applications as surfactants. This family of chemicals includes the most commercially used NPE and its corresponding metabolites NP, as well as octylphenol ethoxylates (OPE) and their degradation products octylphenols (OP). APEOs enter surface waters, for the most part, via discharges of treated effluent from industrial and municipal WWTP. Spontaneous environmental biodegradation and wastewater treatment processes form degradation products, NP, OP, alkylphenoxy carboxylates (APECs), and numerous forms of mono-, di-, and tri-ethoxylates. In reading this memorandum, the terms APEO, NPE, OPE, NP, and OP will be used.

1) APERC contends that concentrations of APEOs in California waterways are low and are not expected to exceed U.S. EPA's WQC for NP

First of all, many of APERCs comments focused on the U.S. EPA's Aquatic Life Ambient Water Quality Criteria – Nonylphenol (U.S. EPA, 2005). We have reviewed these NP acute and chronic WQC established by U.S. EPA for both fresh and salt water. U.S. EPA concluded that these WQC represent concentrations where aquatic life and their uses should not be affected under specified durations and frequencies. Division of Water Quality (DWQ) staff generally agrees that California specific data indicate that concentrations of APEOs in several California waterways are below the NP acute and chronic fresh and salt WQC established by U.S. EPA. However, whether NP is an appropriate benchmark for evaluating relative toxicity of this entire class of diverse organic compounds is unclear. We are not in a position to make far-reaching generalizations about the environmental fate and comparative toxicity of this large class of substances from the relatively narrow dataset for NP, OP, and their ethoxylates.

APERC also appended to their comments summaries of acute, subchronic, and chronic studies conducted on numerous aquatic species. These studies suggest concentrations of APEOs in California waterways are below U.S. EPA's established NP WQC. We appreciate the opportunity to review these studies. On the whole, these studies tend to support that, at present, aquatic species are not being exposed to toxic concentrations of NP. However, SFEI noted that some studies suggest that the effects of APEOs and their degradation products may be additive. As stated earlier, it is unclear whether NP WQC are suitable surrogates for evaluating all potential impacts of this entire class of chemicals.

Environment Canada has also established water quality guidelines for NP and its ethoxylates for the protection of aquatic life. The interim values are 1.0 microgram per liter ($\mu\text{g/L}$) for freshwater and 0.7 $\mu\text{g/L}$ for marine waters (Canada, 2002). The European Union (EU) has also established an Environmental Quality Standard for NP. Under their Water Quality Directive the annual average concentration is not to exceed 0.3 $\mu\text{g/L}$ (OSPAR, 2009) with the maximum allowable concentration in inland and other surface water of 2.0 $\mu\text{g/L}$. We note that several reported concentrations in California waterways, particularly those from the Surface Water Ambient Monitoring Program (SWAMP), are near or exceed these levels (State Water Resources Control Board, 2010b). These criteria values support our contention that existing concentrations of APEOs are of concern.

In summary, we concur with APERCs conclusions that concentrations of APEOs are generally below U.S. EPA's WQC for NP. As discussed above, however, this does not necessarily mean that the current concentrations are safe for aquatic species. APERC does not dispute that these compounds are toxic to aquatic species. We would suggest that, with the wealth of effective alternative, safer surfactants available, there is no need for exposing aquatic species to even low concentrations of these chemicals which are known to be estrogenic and whose effects may be additive. Therefore, our opinion as

to restricting use of APEOs in consumer products has not changed. We continue to support the California Air Resources Board's (ARB) actions to prohibit use of APEOs in the nonaerosol forms of General Purpose Cleaners, Glass Cleaners, General Purpose Degreasers, Heavy Duty Hand Cleaner or Soap, and all forms of Oven Cleaner.

2) APERC contends that concentrations of APEOs in sediment are below PNECs for NP

APERC also disputed statements from SCCWRP that indicated sediment threshold levels protective of aquatic marine or freshwater life are not available. APERC provided NP PNEC sediment values calculated by the Canadian government (2002), as well as NP PNECs for benthic organisms (APERC, 2010). These NP PNECs tend to indicate that, in general, sediment concentrations of APEOs measured in several California locations are below levels of concern. However, SCCWRP cited 2005 data from Schlenk *et al.* which measured some sediment concentrations in excess of the PNECs calculated for marine environments. SCCWRP further indicated that more recent data showed concentrations to be an order of magnitude lower than those found in 2005 (SCCWRP, 2010). Whether this same trend would be true for other waters is not known. As further noted by SCCWRP, very little APEO concentration data exist for other permitted discharges or in water bodies receiving these discharges.

Nevertheless, we believe concentrations of APEOs detected in sediments remains a serious concern. For example, results of model calculations (Huang *et al.*, 2007) showed that over 86 percent of all NP input for an aquatic microcosm consisting of four compartments (surface microlayer, water phase, water sediment, with zebra fish as biota) was removed by advective outflow, while of the remaining NP over 60 percent was distributed to the sediment phase. This finding demonstrates that sediment plays a key role in the fate of NP and acts as a sink in the aquatic environment. The data also support that measures to reduce these substances are important for water quality.

3) APERC contends that APEOs are highly treatable in WWTP and their degradation intermediates are not persistent or bioaccumulative in the environment

APERC's comments include the following statement (p. 6): "While APE[O]s and their degradation intermediates are not 'readily biodegradable'...they are highly treatable and removed from the effluent stream in wastewater treatment plants and they are inherently biodegradable."

SFEI's July 2010 review reached a different conclusion regarding APEOs: "Wastewater removal efficiencies are extremely variable (9 to 94%) and sorption to sludge is the principal pathway for removal...recent studies suggest that urban runoff may also be a pathway for entry of APEOs to aquatic environments."

Related to whether APEOs are persistent or bioaccumulate in the environment, APERC's comment letter appears to question the validity of various researchers' observations that generally support the view that APEOs exhibit a tendency toward biomagnification, bioaccumulation, and/or bioconcentration in marine and freshwater settings. However, we note that recent studies not cited by APERC, SFEI, or SCCWRP also indicate that these phenomena do occur. Among those, for example, Huang *et al.* (2007) found that a comparatively high bioconcentration factor for NP in zebrafish was indicative of substantial potential for bioaccumulation in a food chain, and that depuration was slow and ultimately incomplete within this study's time frame. These observations are consistent with Sumi *et al.*'s findings (2007) on the propensity of common carp to bioaccumulate NP, in addition to exhibiting various signs of endocrine disruption when exposed to ambient concentrations found in several rivers and a lake in Japan. We also note that in its "Nonylphenol (NP) and Nonylphenol Ethoxylates (NPEs) Action Plan" (Action Plan) U.S. EPA indicates that NPEs are "moderately bioaccumulative in mollusks, are persistent in the aquatic environment, and accumulate in soils and sediments" (U.S. EPA, 2010).

Moreover, the EU determined NPEs containing more ethoxylate groups may not be broken down entirely during wastewater treatment. The NP group is particularly stable and can remain intact throughout normal wastewater processing. Data also indicate that NP and NPEs continue to be discharged from wastewater treatment works (Environment Agency, 2010). Further, the EU determined that these chemicals have a tendency to accumulate in the tissues of plants and animals (Environment Agency, 2010).

To be inclusive, we note that Canada found that APEOs, while mildly bioaccumulative in aquatic organisms, did not meet criteria to be considered bioaccumulative under their Persistence and Bioaccumulation Regulations (Canada, 2002).

4) APERC contends that APEOs are not a major source of estrogenic activity in wastewater treatment effluent

APERC's comments express some skepticism about the capacity of APEOs to act as endocrine disruptors. However, recent research not referenced by APERC indicates that APEOs such as NP have some capacity to act as endocrine disruptors. For example, Baker *et al.* found (2009) that NP induced up-regulation of vitellogenin in some fish species; this metabolic response is typically associated with exposure to estrogenic substances, and is thus frequently employed in assays of a prospective endocrine disruptor's biological activity. Vajda *et al.* (2008) assessed the impact of an estrogenic WWTP effluent on white suckers' (*Catostomus commersoni*) reproduction. Gonadal intersex, altered sex ratios, and other changes associated with exposure to estrogenic wastewater contaminants were identified in fish downstream from the WWTP outfall but not at the upstream site. Chemical analyses determined that the WWTP effluent contained a complex mixture of endocrine-active chemicals including alkylphenols. The evidence of endocrine disruption has also been observed in male

fathead minnows (*Pimephales promelas*) exposed to WWTP effluent (Barber L. *et al.*, 2007). Results indicated that the reproductive potential of native fishes may be compromised in wastewater-dominated streams. Of particular concern would be effluent concentrations for discharges to saltwater because the WQC for salt water are much lower than those for fresh water.

In addition, Xie *et al.* (2005) found that certain herbicide formulations containing APEOs as surfactants were “estrogenic to trout at environmentally relevant concentrations” and noted that these effects seemed to be additive at some concentrations. The EU also determined that NP has the potential to mimic hormones, namely estrogen (Environmental Agency, 2010). Yet another study concluded that the increasing occurrence, distribution, and concentration of environmental contaminants, including environmental estrogens (EE), in aquatic habits may also compromise the hypo-osmoregulatory ability of fish (Hanson *et al.*, 2010). Reported results indicated that EE reduced salinity adaptation of rainbow trout (*Oncorhynchus mykiss*) used in this study.

APEOs such as 4-nonylphenol may also exert nonendocrine related deleterious effects on the shoaling behaviors of various fish species, as reported by Ward *et al.* in 2008. These researchers found that 4-nonylphenol, a common anthropogenic contaminant of coastal waters, could disrupt shoaling behavior at environmentally relevant concentrations. Shoaling is a group behavior common to many fish species that is important in reducing mortality due to predation, enhancing foraging success, improving hydrodynamic efficiency, and for providing enhanced opportunities for locating suitable mates. Though not emphasized by SCCWRP or SFEI reviewers, we consider that this line of research warrants further consideration in evaluating the potential aquatic toxicity of APEOs.

5) APERC contends that rather than prohibiting use of APEO surfactants, concentrations should be monitored to evaluate occurrence of increases

We agree that APEO concentrations should be monitored. Through U.S. EPA’s Action Plan additional data may be generated if NP and NPE are added to chemicals to be reported under the Toxic Release Inventory (TRI). However, we do not believe that more data are necessary to determine that APEOs are at levels of concern in California waterways. Moreover, we do not believe it is prudent policy to take action only if data indicate APEO levels are increasing over existing concentrations.

General Comments

In addition to responding to APERC comments, I believe it is worth acknowledging other national and global actions related to restricting/prohibiting use, or conducting further testing of APEOs. First, we note several U.S. EPA actions related to APEOs. In an Advance Notice of Proposed Rulemaking (ANPR) dated June 17, 2009, U.S. EPA announced that it is initiating a rulemaking not only because of aquatic toxicity concerns, but also due to human health concerns (U.S. EPA, 2009a). For example,

U.S. EPA states in the ANPR that they are trying to determine whether a study of industrial laundry workers' exposure to NPEs is warranted. The rulemaking is also being pursued to determine whether additional aquatic toxicity testing is necessary. Earlier, as part of U.S. EPA's Design for Environment Program, the Safer Detergents Stewardship Initiative (SDSI) was put in place. This program recognizes manufacturers that voluntarily switch to safer surfactants to 'help protect aquatic life in both fresh and salt water' (U.S. EPA, 2009b).

Most recently, on August 18, 2010, U.S. EPA released their Action Plan. This Action Plan is designed to "address [U.S. EPA's] concerns about potential ecological effects due to the manufacturing, processing, distribution in commerce, and use of NP and NPEs. Additionally, EPA continues to have some concern about potential risk to human health. EPA anticipates that the action put forward in the Action Plan will further reduce human exposure thereby reducing concerns associated with NP and NPEs." ".....EPA also intends to evaluate the potential for disproportionate impacts on children and other sub-populations." Other potential actions include developing a Significant New Use Rule (SNUR) and continuing to encourage manufacturers of all NPE-containing direct-release products to move to NPE-free formulations. The Action Plan further states that household detergent manufacturers have cooperated to greatly reduce use of NPEs through the SDSI, and that the majority of industrial launderers have agreed to expedite a phase out of the use of NPEs by 2014 (U.S. EPA, 2010).

NP and NPEs are on the List of Chemicals for Priority Action under OSPAR [Oslo and Paris Conventions for the Protection of the Marine Environment of the North East Atlantic] (OSPAR, 2009). As a result, OSPAR countries have been required to implement best available techniques and best environmental practices, and to achieve specified limit values for emissions and discharges of various waste streams, as well as the phaseout of specific substances. Specifically, NP and NPEs used as cleaning agents were recommended to be phased out in 1995 for domestic use, and in 2000 for industrial use (OSPAR, 2009). Because of the risks these chemicals pose to the aquatic environment, in the EU use of NP and NPE has been restricted in various categories including industrial, institutional and domestic cleaning; cosmetics and personal care products; and pesticides and biocides. These restrictions apply to both concentrated and ready-to-use products (Environment Agency, 2010). We also note that under the Helsinki Convention (Baltic Marine Environment Protection Commission; HELCOM), NP/NPEs and OP/OPEs are on the List of Priority Hazardous Substances (Backer *et al.*, 2010, HELCOM, 2010).

Since putting these directives in place, periodic monitoring by OSPAR has found lower concentrations of NP/NPE in marine waters protected by OSPAR/EU countries (OSPAR, 2009). However, data also show that NP and NPEs continue to be discharged from wastewater treatment works in England and Wales (Environment Agency, 2010). This is an indicator that APEOs are not completely removed from WWTP.

Other states, including Minnesota and Maine have taken steps to address exposure to APEOs. Recently, the Minnesota Pollution Control Agency developed draft numeric acute and chronic WQC for total NPE and developed a separate lower value of 2.4 µg/L (considerably lower than that of U.S. EPA's) for the protection of cold-water fish (Minnesota, 2010). We note that NP and NPE concentrations measured in several California waterways would exceed this cold-water fish criterion (SWRCB, 2010b). While not water quality related, the Maine Board of Environmental Policy recently approved a regulation to designate the chemical class of NP and NPE as Priority Chemicals under the Maine Regulation of Chemical Use in Children's Products (Maine, 2010).

In combination, these U.S. EPA and international actions have likely contributed to lowering concentrations of APEOs in California waterways as manufacturers have shifted from use of APEOs to comply with these actions. Clearly, ARB is not acting alone to reduce exposures to APEOs.

In preparing our response to APERC we also reviewed other comments on ARB's proposals. We were pleased to read that a large industry association, representing manufacturers of the products that would be affected by ARB's proposed amendments, is not opposed to the proposed prohibition on use of APEOs (CSPA, 2010).

Summary

In conclusion, the comments submitted by APERC do not change the information or recommendation conveyed in our memorandum to you on this subject dated September 20, 2010. To reiterate, information submitted by SWAMP, SFEI and SCCWRP indicate that the level of aquatic toxicity posed by APEOs is high enough to cause concern. In fact, levels measured in several California waterways exceed WQC established by some jurisdictions. Given the wide availability of effective, safer alternatives, coupled with U.S. EPA and other jurisdictional actions, we continue to support ARB's prohibition on use of APEOs in non-aerosol General Purpose Cleaners, Glass Cleaners, General Purpose Degreasers, Heavy Duty Hand Cleaner or Soaps, and all forms of Oven Cleaners.

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