A PRELIMINARY ASSESSMENT OF ECOLOGICAL RISKS FROM NONYLPHENOL IN MUNICIPAL SEWAGE SLUDGE FOLLOWING WASTEWATER TREATMENT

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Summary

An assessment was made of potential risks to wildlife and plants that inhabit soil following amendment with sewage sludge from biological wastewater treatment plants (biosolids). The results of this preliminary assessment suggest that the presence of nonylphenol in municipal sewage sludge applied to soils does not pose a risk to plants, soil-dwelling organisms, or small mammals, based on relevant toxicity data and typical levels of nonylphenol in the sludge.

Background

NPE surfactants are biodegraded in biological wastewater treatment plants. Final effluents discharging to surface waters typically contain measurable amounts of nonylphenol mono- and di-ethoxylates (NPE1,2) and nonylphenol mono- and di- ether carboxylates (NPEC1,2), but only trace levels of nonylphenol (NP). Potential aquatic risks following discharge of these biodegradation intermediates into surface waters have been extensively examined. However, most NP (and some NPE1,2) formed during wastewater treatment is associated with sludge that is disposed in landfills, incinerated or used as a soil amendment. A detailed assessment of potential risks from sludge-amended soil containing NP and NPE has not been conducted.

NP comprises an average of 88.5 to 97.4% of the combined NP, NPE1 and NPE2 on a molar basis in sludge obtained from several North American treatment plants equipped with various tertiary treatment systems (*e.g*., anaerobic digestion, composting) (Bennie and Webber, 2000; LaGuardia *et al*., 2001). NPE1,2 are known to be somewhat less toxic than NP (Staples *et al*., 1998) and aerobically biodegrade to various carboxylic acids, ring fragments, and carbon dioxide faster than NP (Staples *et al*., 1999; Staples *et. al*., 2001) so the focus of any assessment of sewage sludge residues should be on NP. A preliminary ecological risk assessment is presented here that examined potential risks from NP in sludge-amended soil.

Risk Assessment of NP as a Component of Sludge-Amended Soil

The basic principals of risk assessment were used to assess whether or not NP contained in sludge amended into soil poses risk to the environment. The risk assessment performed here examined the potential concentrations of NP to which soil organisms may be exposed and examined relevant toxicity data to assess potential effects.

Several relevant pathways of exposure could be addressed including directly exposed soil following sludge application and incorporation into soil, runoff into nearby surface water from rainfall, and percolation downwards towards groundwater driven by rainfall. It was assumed that the maximum concentrations to which wildlife would be exposed following application of sludge to soil, and hence the greatest potential for adverse risks, were directly in the sludge-amended soil. Therefore, this risk assessment focused on ecological receptors directly exposed to sludge-amended soil. Studies have been conducted with NP incorporated into soil directly or through sludge amendment using microbial populations, earthworms, insects, and plants. These organisms are considered representative organisms for the evaluation of soil and presumably sludge-amended soil. Thus, the main pathways addressed in this risk assessment of sludge-amended soil were:

1) direct exposure of soil to earthworms,
2) direct exposure of soil to insects,
3) direct exposure of soil to plants; and,
4) ingestion of soil, invertebrates, and plants by small mammals.

Data Available on NP Concentrations in Municipal Sewage Sludge

Data are presented in Table 1 regarding the concentrations of NP in municipal sewage sludge collected from numerous treatment plants in North America. Data were recently summarized in Bennie (1999) and Bennie and Webber (2000). Also included are recent data by LaGuardia *et al*. (2001). Concentrations of NP range overall from about 5 to several hundred mg/kg dry weight (dw). A close examination of the data suggests that a concentration of 500 mg/kg dw is predictive of the concentration of NP in sludge. This value is the average from 11 STPs in the US, the midpoint of the ranges of sludge concentrations in two surveys of Canadian STPs and exceeds the values observed at Los Angeles, Toronto, and Ontario area STPs. Therefore, a concentration of 500 mg/kg dw NP in sludge was used for assessing potential effects to soil organisms following incorporation of sludge into soil. To obtain a potential soil concentration, the sludge was assumed to be applied to soil at a rate of 7.5 metric tonnes per hectare (t/ha) and incorporated to a depth of 15 cm. Assuming that 1 hectare of soil 15 cm deep equals 2000 kg, a biosolid application factor of 0.00375 was calculated. Starting with a sludge concentration of 500 mg/kg dw and using the biosolid application factor of 0.00375, a predicted soil concentration (PEC_{soil}) of 1.9 mg/kg dw was calculated. Using the highest sludge concentrations observed (887 mg/kg dw), a maximal soil concentration (PEC_{max}) of 3.33 mg/kg dw was also calculated.

Table 1. Concentrations of NP in Sewage Sludge from Tertiary Treatment Plants in North America

Concentration Range (mg/kg dw)	Location	Reference
8.4 to 850	Canadian STPs	Bennie (1999)
370	Los Angeles, CA, USA	Bennie (1999)
137 to 470	Toronto area STPs, Canada	Bennie (1999)
290 to 818	Canadian STPs	Bennie (1999)
450	Ontario area STP, Canada	Bennie and Webber (2000)
5.4 to 887 (n=11) average: 491	STPs from California, Texas, Mid-Atlantic, and New England, USA	Laguardia *et al*. (2001)

Data Available on the Toxicity of NP to Soil Dwelling Organisms

Data are available on the toxicity of NP to soil dwelling organisms and are summarized in Table 2. Studies have been performed on microbial populations, plants, insects, and earthworms. A study examining the effect on the production of carbon dioxide from soil to which sludge had been added found that a concentration of 1000 mg/kg NP reduced carbon dioxide production after 40 days, but 100 mg/kg NP did not. Other microbial processes were stimulated by sludge amendment. Three species of plants were exposed to NP added to soil for 21 days and no growth effects were observed at the highest concentration tested, 100 mg/kg dw (Windeatt and Tapp 1987). The lack of effects is likely due to the inability of the plant to take up NP from soil through their root system or translocate the NP from the root system into the aboveground leaf and stem (Kure *et al*., 1999) or hydroponic (Doucette, 2002) systems. The Collembollan insect (springtail) *Folsomia fimetaria* was exposed to soil to which sludge spiked with NP was added.

This soil preparation process more closely mimics the amendment of soil with sludge than direct application of NP to soil. After 21 days, an EC_{10} of 49 mg/kg dw based on reproduction effects was measured. It should be noted that these data have not been published except as a one-page summary in a conference proceedings (Jensen and Krogh, 1997). Therefore, the data are uncertain as full documentation of the test methods, analytical confirmation of concentrations, and the full data are not available. The earthworm *Eisenia fetida* was exposed to soil in a fashion similar to the Collembolla study. No effects on reproduction were found after 56 days exposure to a maximum concentration of 46 mg/kg dw. These data are in contrast to an earlier study with the earthworm *Apporectodea calignosa* that were exposed to NP that was directly added to soil and reported a 21-d EC_{10} of 3.4 mg/kg dw (Krogh *et al*., 1996). It is clear that the bioavailability of NP in sludge-amended soil is significantly reduced compared to direct addition of NP to soil. The lack of bioavailability was noted by Krogh *et al*. (1999) who applied sludge containing NP to soil plots and found stimulation of Collembolla populations, likely due to the addition of nutrients to the soil.

The lowest effect concentration for any study that showed an adverse effect to an organism directly in contact with soil is the 21-day study with springtail that reported an EC_{10} of 49 mg/kg dw. Despite the absence of full documentation of this study, it will be used in this preliminary risk assessment as the basis of a predicted no effect concentration for sludge-amended soil ($PNEC_{soil}$) protective of non-mammalian wildlife (*e.g*., microbial populations, plants, insects, and earthworms). Using an assessment factor (AF) of 10 with the critical toxicity value of 49 mg/kg dw, a $PNEC_{soil}$ of 4.9 mg/kg dw was determined.

In addition to microbes, plants, and invertebrates, small rodents may be assumed to inhabit the soil where sludge has been recently applied. Contributions from all sources to the intake of NP through diet by a representative small mammal (*e.g*., deer mice) were calculated. Intake was assumed to occur via ingestion of small amounts of plants, invertebrates, and soil, as well as soil pore water. An ingestion rate of NP from these sources was calculated to be 0.12 mg/kg/day. (calculations not shown) The assessment of effects on small mammals was based on an estimated NOAEL for NP of 15 mg/kg/day taken from a multi-generation study with laboratory rats conducted by the National Toxicology Program (NTP). The NOAEL was divided by an assessment factor of 10 to estimate the toxicity to small mammals in the field ($PNEC_{sm.\ mammal}$ = 15 mg/kg/day).

Table 2. Toxicity data for terrestrial organisms exposed to NP in soil

Receptor	Key Study	Critical Toxicity Values***
Microbes*	40-d CO_2 production reduced (other processes stimulated)	LOEC/NOEC 1000/100 μg/g dw
Plants*	21-d growth of sorghum, sunflower, soya	no effect at top concentration, ≥100 μg/g dw
Insects**	21-d reproduction reduced, Collembella *Folsomia fimetaria*	EC_{10} = 49 μg/g dw
Earthworms**	56-d reproduction, *Eisenia fetida*	no effect at top concentration, ≥46 μg/g dw
Small mammals	multi-generation laboratory rat study	NOAEL = 15 mg/kg/day

* Study performed using direct application of NP to soil

** Study performed using incorporation of NP into sewage sludge, then amendment of soil with the sludge

*** LOEC = lowest observed effect concentration, NOEC = no observed effect concentration, EC_{10} = the lower bound 10^{th} percentile effect concentration, NOAEL = no observed adverse effect level.

Risk Characterization

The application of sewage sludge to soil was assumed to expose wildlife to NP contained in the sludge. The most directly exposed organisms are those that inhabit the soil and were the receptors addressed in this assessment. In laboratory studies, microbial populations were not adversely affected except at 1000 mg/kg, well above the calculated PEC_{soil} for NP of 1.9 mg/kg dw and the PEC_{max} of 3.3 mg/kg dw. Plants were not affected at all from exposure to NP in soil with no effects at 100 mg/kg dw, also well above the PEC_{soil} for NP of 1.9 mg/kg dw and the PEC_{max} of 3.3 mg/kg/day. Similarly, earthworm tests with exposure based on NP added to sludge, then incorporated into soil, were not affected at the highest concentration tested (37.4 mg/kg dw). Of the test organisms evaluated, only the collembollan insect was affected by NP incorporated into sewage sludge and amended to soil. Comparison of the PEC_{soil} for NP of 1.9 mg/kg dw to the $PNEC_{soil}$ of 4.9 mg/kg dw gives a ratio of 0.4 or below the regulatory criteria of 1.0. PEC/PNEC ratios of less than 1.0 are indicative of adequate safety margins. Comparison of the PEC_{max} for NP (3.3 mg/kg dw) to the $PNEC_{soil}$ also gives a ratio below the regulatory criteria of 1.0 and is therefore also indicative of an adequate safety margin.

The potential risks to small mammals from exposure to sludge-amended soil were calculated by comparing the calculated intake of 0.12 mg/kg/day to the $PNEC_{sm.\ mammal}$ of 1.5 mg/kg/day. A ratio of 0.08 was obtained and is well below the regulatory criteria of 1.0. Small mammals therefore have a lower potential for adverse effects than do insects.

Discussion and Conclusions

Preliminary results show that the potential for adverse risks to the terrestrial environment from sewage sludge applications containing NP are low. Incorporation of concentrations of NPE1 and NPE2 into the analysis would only slightly change the results. NPE1,2 are less toxic than NP and biodegrade faster (Staples *et al*., 1998; Staples *et al*., 1999; Staples *et al*., 2001). Since NP makes up the vast majority of combined NP (Bennie, 1999; Bennie and Webber, 2000; LaGuardia *et al*., 2001) plus NPE1+2 on a molar basis, the overall characterization of risks would be unchanged. Thus, the application of municipal sewage sludge that contain NP as currently measured in North American treatment plants and are applied to soils as a biosolid does not appear to pose adverse environmental effects.

It is imperative that toxicity studies used to evaluate the potential impacts to the environment from the amendment of soil with sludge be conducted so they reflect the exposure pathways being simulated. Laboratory studies show that the bioavailability of NP is rapidly reduced when applied to soil following incorporation into sludge (Jensen and Krogh, 1999; Springborn, 2001). In addition, NP is easily biodegraded under oxic soil conditions. Thus, concentrations and subsequently the potential for toxic effects to exposed organisms decrease when sludge containing relatively high amounts of NP obtained from a treatment plant is applied to soil. For example, Bennie and Webber (2000) showed that the greatest reduction in NP concentrations in soil occurred in the first few weeks following land application of sludge. Krogh *et al*. (1999) showed that sludge containing NP added to soil plots actually caused the stimulation of populations of collembolan insects.

The development of consistent assessment techniques for organic compounds found in sewage sludge would lead to more definitive analysis of potential risks and eventually defensible numerical standards for sludge applied to soil. As noted by Smith (2000), although a number of European countries have adopted standards for organic contaminants in sewage sludge intended for agricultural use, there is no consistent approach to the selection of contaminants or the numerical limits. In addition, it appears that most European standards have no identified toxicological/environmental basis and in some cases they have severely hindered the beneficial use of the biosolids (Smith, 2000). As the US Environmental Protection Agency continues with its evaluation of potential risks from organic contaminants in sewage sludge, it should keep these issues and findings in mind.

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