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Dioxins in Washington State Soils

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Introduction

During 1996 and 1997 citizen concern¹ and media reports² alerted the Washington State Legislature and the Washington State Department of Ecology (Ecology) to the practice of using waste products as components of fertilizers and soil amendments. Concerns focused on potential contamination of these products with heavy metals and dioxins.

In response the State of Washington enacted *The Fertilizer Regulation Act*. This legislation, the first of its kind in the country, established limits on metals in fertilizers based on application rates, and required manufacturers to report metals concentrations in their fertilizers. State agencies were charged with conducting studies of dioxin and metal concentrations in fertilizers, soil amendments, and soils.

One of these studies focused on soil concentrations of polychlorinated-dibenzodioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs), here referred to collectively as *dioxins*. The objective of this testing was to assess typical (or background) dioxin concentrations in Washington State soils³, particularly agricultural soils⁴.

One use for these data was to evaluate potential criteria for dioxin contamination in fertilizers and soil amendments based on a philosophy of non-degradation. A non-degradation standard would ensure that dioxin concentrations in fertilizers were no higher than those in soil. To meet these objectives Ecology analyzed eighty-four soil samples from open, forested, urban and agricultural areas. This paper reports the results of those analyses. The full reports^{3,4} are available at <http://www.ecy.wa.gov/biblio/99309.html> and <http://www.ecy.wa.gov/biblio/99333.html>.

Methods and Materials

Soil samples were collected from open areas (non-urban grasslands or prairies), forest lands, urban areas as defined by the U.S. Census Bureau⁵, and agricultural lands. Agricultural lands were defined as the approximately 5,284,000 harvested acres in Washington^{6,7,8}. Table 1 shows the distribution of samples among these land uses.

Table 1. Number of samples allocated by land use.

Land Use	East of Cascade Mts.	West of Cascade Mts.	Total
Open	4	4	8
Forest	4	4	8
Urban	3	11	14
Agricultural	50	4	54
Total	61	23	84

We randomized site distribution for samples from urban and agricultural land uses (68 of 84 samples). Sites for the land uses with smaller sample sizes (forested and open lands) were equally distributed between eastern and western Washington, and between private and public lands.

Urban sites were assigned to specific urban areas by randomly selecting 14 of 3394 units representing the number of urbanized square kilometers in Washington State. Each of these units was assigned to a specific urban area. Sample sites were selected from available parks and similar grassy areas within each of the urban areas.

We randomized agricultural site distribution by county and crop. The state's agricultural acreage was represented by 5284 units, each equivalent to 1,000 harvested acres. Each unit was associated with a specific county and crop. Fifty-four units were randomly selected. Site selection was, therefore, generally proportional to the distribution of agricultural lands devoted to specific crops in specific counties. For instance, 31 of 54 sites (~57%) were assigned to wheat lands which account for about 52% of Washington's cropland⁶; while 50 of 54 sites (~93%) were assigned to eastern Washington counties which account for about 95% of Washington's agricultural acreage⁶.

Growers were contacted randomly by phone using Farm Service Agency (FSA) lists. Approximately 80% (54 of 68) of those contacted agreed to participate.

Each sample consisted of a composite of 10 subsamples collected from the center and periphery of a circular 1-acre sample unit. Sample units were located away from roads, railroad tracks, buildings, and treated wood poles and fences. The locations of open, forested and urban sites were recorded using GPS. Agricultural site locations were not recorded because of landowner concerns about potential release of this information under public disclosure laws.

Samples were collected from the depth interval of 0 to 5 cm. using a specially cleaned stainless steel scoop. Rocks, vegetation and debris were removed from the composite samples. Samples were shipped in ultraclean sample jars, and stored at 4° C.

Each sample was analyzed for dioxins⁹, total organic carbon (TOC)¹⁰ and grain size¹⁰.

Dioxin results were converted to toxic equivalents¹¹ (TEQs). Results reported here assume that undetected congeners are not present in soils (non-detects = 0). The full reports^{3,4} also include TEQs based on the assumption that congeners are present at detection limits (non-detects = DL) or are present at ½ the detection limit (non-detects = ½ DL).

Results and Discussion

Eighty-four soil samples were analyzed for dioxins, TOC and grain size. These results provide one of the most comprehensive characterizations of background dioxin concentrations in soils available in the literature. Dioxins (one or more 2,3,7,8-substituted congeners) were detected in every sample. Dioxin concentrations ranged from 0.0078 to 19.5 pptr TEQ.

A summary of dioxin results is presented in Table 2. Although this study was not designed to statistically compare results from different land uses, dioxin concentrations appear to be highest in urban and forest soils, followed by soils from open areas. Agricultural soils have the lowest dioxin concentrations.

Dioxin TEQ results appeared to be log-normally distributed for urban, open and agricultural soil samples; TEQ results in forest soils approximated a normal distribution.

Although review of the available literature reveals a relative paucity of “background” data, concentrations of dioxins found in soils from urban, open and forested areas were comparable with results reported in studies from Spain^{12, 13} Germany¹⁴, and Austria¹⁵.

Table 2. Summary of dioxin concentrations in Washington State soils by land use (reported as TEQ, ng/kg = pptr)

Land Use	Range	Mean	Median	Geometric Mean	n
Urban	0.13 – 19	4.1	1.7	1.9	14
Forest	0.033 – 5.2	2.3	2.2	1.2	8
Open	0.040 – 4.6	1.0	0.27	0.24	8
Agricultural	0.0078-1.2	0.14	0.054	0.062	54

n = number of samples

Small sets of comparable data were reported for agricultural soils in Germany¹⁴ and Russia¹⁶. These German and Russian results were at or above the upper range of concentrations found in Washington State agricultural soils.

Forest soils had the highest median TOC content (9.9%), followed by open (7.2%), urban (4.1%), and agricultural (1.1%) soils. The linear correlation ($r=0.66$) between organic carbon content and dioxin concentration was highly significant ($p<0.0005$). We found no correlation between grain size and dioxin concentrations.

Although we do not know why soils from different land uses appear to have different dioxin concentrations, the following factors may play a role:

- Urban soils are nearer to many of the known or suspected sources of dioxin air emissions in Washington State.¹⁷
- Agricultural lands are generally tilled. This may mix surface deposition with deeper soils, thereby lowering the concentration in the top 5 cm.
- The high correlation between organic carbon and dioxin concentrations may imply that dioxin accumulation in soils is associated with adsorption onto leaves and needles that are subsequently cycled into the soil

To estimate a statewide median for typical dioxin concentrations in Washington State soils, we ran a Monte Carlo simulation using the Palisade @Risk add-in for Microsoft Excel. For this exercise the total land area devoted to each of the four uses was estimated from data compiled by USDA¹⁸. Soils represented by forested land were estimated to comprise 57.7%, urban lands 3.1%, open land 18.8% and agricultural lands 20.4%. Probability plots were generated for the TEQ distribution for each land use. Ten thousand random samples were drawn from these probability plots in proportion to the percentage of land in each land use category. Using this technique we estimated a statewide median of 1.4 ppb TEQ. This value could serve as a starting point for developing non-degradation criteria for land-applied products.

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