TECHNICAL MEMORANDUM

Date: January 31, 2011

To: Janet Gillaspie
Association of Clean Water Agencies

From: David Stone, Ph.D.

Re: Estimates and sources of polychlorinated biphenyls and DDT/DDD/DDE to wastewater treatment influent from human excretion and food waste

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Summary:

Polychlorinated biphenyls (PCBs) and the DDT complex (DDT, DDE and DDD) are highly persistent and bioaccumulative compounds. Despite a decades-old ban, these chemicals continue to be detected in human samples, the environment and foods. The general population is primarily exposed through dietary intake. Some of the ingested PCBs are not absorbed across the intestinal wall and are excreted in their parent, or unchanged, form. A limited number of recent studies were found in which parent PCBs were measured directly in human fecal matter. **Based on data from a mid-sized Oregon community, a daily average of 0.16 ng of PCBs/L is estimated to reach wastewater influent as a result of human excretion.** Unlike PCBs, DDT is extensively metabolized, and excreted in the urine as a water-soluble acid. Negligible amounts of the DDT complex would be expected in fecal matter in their parent form. Disposal of food waste contaminated with PCBs and the DDT complex is an additional source of pollution to wastewater influent, particularly for discarded dairy, meat and seafood. Food waste is generated in households, restaurants, grocery stores and food processing facilities, representing multiple sources for these persistent compounds to reach wastewater.

I. Polychlorinated Biphenyls (PCBs)

Background and Excretion:

Through national and international agreements, PCBs have been phased out and heavily regulated due to their persistence, toxicity and bioaccumulative nature. Since the U.S. ban on the sale and manufacture of PCBs in 1977, levels in the environment and biota have steadily declined (ATSDR 2000). Despite this attenuation, measurable body burdens continue to persist in people across the U.S. and worldwide (CDC 2009). The diet is considered to be the most significant source of PCBs to the general public (Nicholson and Landrigan, 1994; Duarte-Davidson and Jones, 1994; CDC 2009).

Inside the body, PCBs distribute into lipid-rich tissues and are characterized by lengthy half-lives prior to elimination. Excretion of PCBs occurs primarily in the bile and feces. While the majority of PCBs are absorbed and subsequently metabolized, a measurable fraction of unabsorbed PCBs are eliminated as unchanged parent compounds.

Once ingested, PCBs are subject to the physiological processes of absorption, distribution, metabolism and excretion (ADME). PCBs consist of a family of 209 different forms, known as congeners, which vary in their degree of chlorination. The extent and rate at which ADME occurs is variable among congeners (ATSDR 2000). These differences influence the amount of parent PCB that is excreted into municipal sewers. For example, some of highly chlorinated PCB congeners, notably PCBs 138, 153, 180, 187 and 194, are poorly absorbed across the gastrointestinal (GI) tract and thus undergo little metabolism. PCBs that cross the GI tract and enter the blood stream are mostly metabolized with little parent compound remaining in bile. Parent, unmetabolized PCBs can be found in the feces via three physiological routes:

i. lack of absorption across the GI tract from dietary intake
ii. intestinal excretion via exfoliation of epithelium (i.e. sloughing)
iii. biliary excretion (this component consists primarily of metabolites).

Estimating PCB Loading to Wastewater Influent:

To estimate the loading of PCBs to wastewater influent, several variables are required. These include the daily amount of PCB excreted in fecal matter per person [ng PCB estimated/(day•person)], the number of people in a community and the daily flow of influent to a wastewater treatment plant [1 day/total flow to influent (L)]. This model is illustrated in Equation 1 below:

Equation 1,

$$\frac{\text{ng PCB estimated}}{\text{day•person}} \times \text{number of people in community} \times \frac{\text{1 day}}{\text{total flow to influent (L)}} = \frac{\text{ng PCB}}{\text{L}}$$

The experimental factor in this model is the concentration of PCBs excreted in fecal matter. A limited number of studies were identified in which PCBs were measured in humans and met the following criteria: 1) the studies were conducted within the last twenty years; 2) the investigators measured parent PCBs directly in human fecal samples; and 3) the study excluded occupational exposures or exposures related to poisoning incidents. Eligible studies were conducted on adult volunteers in Europe using a mass balance approach that measured PCBs in the diet and fecal matter (Juan et al. 2002; Moser and McLachlan, 2001; Schlummer et al. 1998). Recent PCB biomonitoring in the U.S. has focused on blood serum, as opposed to fecal excretion studies. U.S. blood levels can be compared with blood levels from populations in Europe to allow for comparisons in anticipated fecal excretion and dietary exposure.

In one study, five volunteers, aged 24 to 30 years, were monitored for fecal excretion of 20 PCB congeners using a duplicate meals method (Juan et al. 2002). Most of the PCBs showed a net absorption. However, several congeners, notably the highly chlorinated PCBs, had net excretion rates (i.e. fecal excretion > absorption). An input-output balance was calculated based on dietary intake and measurements of parent PCBs in food and fecal matter. The average excretion per person was **150 ng PCBs/day**. It should be noted that this study examined only 20 out of 209 congeners, likely under-estimating the actual sum of total PCB congeners in fecal matter.

In another study, seven volunteers in Germany, aged 24 to 81 years, were studied using a mass balance approach of PCB dietary intake and fecal excretion (Schlummer et al. 1998). The levels of PCBs were reported in feces on a dry weight (dw) basis and ranged from non-detectable for the lower chlorinated congeners to 14,500 ng/kg dw for PCB 153. The results of this study support the findings of Juan et al. (2002), particularly in characterizing relatively high levels of hexachlorinated PCBs in fecal matter.

The study of Juan et al. (2002) was used to model PCB loading to wastewater influent from human excretion. The study design of Juan et al. met the criteria for inclusion, reported findings that were directly applicable to Equation 1 and used a robust methodology to quantify excretion rates and levels of PCBs in fecal matter. A limitation to using this study as a surrogate for estimating PCB load for an Oregon community is that Juan et al. recruited healthy adult volunteers. Healthy adults are expected to have a greater dietary intake and increased excretion mass compared with adolescents, individuals with pre-existing disease and the elderly. However, Juan et al. did not measure PCB loadings due to lactation or excretion from nursing infants, which are known sources for PCBs (Rogan et al. 1986). An additional limitation is that the participants' dietary intake was consumed in the United Kingdom (U.K.). While no fecal output dietary studies were located in the U.S., the levels of PCBs in blood serum collected during the Center for Disease Control (CDC) survey in 2003, were similar to a survey of 154 people across the U.K. in the same year (Thomas et al. 2006; CDC 2009). The similarity of PCB blood levels between U.S. and U.K. surveys suggests that excretion patterns would be analogous as well.

In addition to the results of Juan et al. (2002), additional parameters of community size and average daily flow were utilized for Equation 1. A mid-sized community and treatment plant in the Willamette Valley was modeled using recorded data from 2010. The average annual daily flow was reported as 36.2 million L/day, with an estimated service population of 39,400 individuals (based on the number of service accounts multiplied by 2.3 and 4 for single residential and multi-family units, respectively). This resulted in an annual average estimate of **0.16 ng PCBs/L** (or 160 pg PCBs/L) per day excreted into wastewater as a result of human elimination. This estimate ranges from an average of 0.12 ng PCBs/L per day in the wet season (November – April) to 0.24 ng PCBs/L in the dry season (May-October).

II. DDT Complex

Background and Excretion:

DDT, or dichlorodiphenyl trichloroethane, is a legacy insecticide belonging to the organochlorine class of pesticides. DDT is a mixture of several isomers, with the p,p'-form associated with insecticidal activity and persistence. The main breakdown products of DDT are dichlorodiphenyl dichloroethylene (DDE) and dichlorodiphenyl dichloroethane (DDD). The United States Environmental Protection Agency (EPA) banned most registered uses of DDT in 1972. Peak production of DDT occurred in the 1960s, reaching 188 million pounds produced on an annual basis (ATSDR 2002). While DDT is not allowed for use on crops grown in the U.S., it is common to import crops from countries that continue to use DDT on a variety of crops. Total worldwide production in estimated to remain at millions of pounds of DDT produced annually (Turusov et al. 2002).

Inside the body, DDT is metabolized primarily to DDE and DDD and distributes into lipid-rich tissues, similar to the PCBs. Excretion of the DDT complex occurs almost entirely in the urine as the metabolized, water-soluble acid known as 2,2-bis, 4-chlorphenyl acetic acid (DDA) (Chen et al. 2009). Negligible amounts of parent DDT or DDE and DDD are anticipated to occur in bile or fecal matter.

Estimating DDT Loading to Wastewater Influent:

No suitable studies were located for DDT that met the inclusion criteria outlined in Section I for estimating chemical loading to wastewater influent. This may due to the following two factors: 1) since 1956, the scientific community has known that DDT is extensively metabolized and excreted as a water soluble acid prior to excretion (Hayes et al. 1956), precluding the need to investigate feces; and 2) the majority of human ADME studies were conducted prior to the ban of DDT in 1972. The DDT complex continues to be found in blood serum samples collected from the general population, mainly in the form of DDE (CDC 2009). The presence of these compounds in blood serum can be the result of lengthy half-lives in the body, as well as on-going exposure to DDT residue in the diet (discussed in Section III).

III. Food and feed residues for PCBs and the DDT Complex

PCBs continue to be detected in the U.S. Food and Drug Administration (FDA) Market Basket surveys, conducted as part of their Total Diet Study (FDA 2006). PCBs were found in lipid-rich foods, such as eggs, oils, butter and seafood, at low parts per billion levels. Seafood was associated with the most frequent PCB detections compared with other food products (FDA 2006). In addition to the elimination of parent PCBs from humans,

PCB contamination in foods will reach wastewater through direct disposal of food waste.

DDT, DDE and DDD residues in food continue to be monitored through the United States Department of Agriculture's (USDA) Pesticide Data Program. In 2008, the p,p'- isomers of DDT, DDE and DDD were detected in multiple food items (USDA 2009), including: catfish (detected in 84.6% of samples), spinach (34.7%), collard greens (28.7%), kale greens (28.0%) and celery (13.9%). The DDT complex was detected in rice and corn samples as well, but at much lower levels and frequency compared with meat, fruits and vegetables. p,p'-DDE was the most frequently detected isomer, with levels ranging from 3 to 40 parts per billion. The USDA noted that all of the detections were below the FDA action levels for food. FDA action levels range from 0.05 parts per million (ppm) in grapes, hops, tomatoes and lettuce to 3 ppm in fish and meat fat (FDA 2002). Similar to PCBs, the direct disposal of meats, fruits, vegetables and grains will contribute to the loading of DDT, DDE and DDD to wastewater.

A high amount of food is disposed of as waste in the United States (Gallo 1980). In a survey of 243 Oregon households, over 1500 grams of food were discarded over a 7-day period (Garde and Woodburn, 1987). A USDA study of America's food disposal and losses highlighted that a high percentage of disposed foods were dairy products, which tend to have relatively high PCB levels compared with other food categories (Kantor et al. 1997). Food waste disposal occurs at residential, restaurant, retail and food processing facilities.

While not possible to provide a quantitative estimate of PCB and DDT loading to wastewater as a function of food disposal, it is reasonable to conclude that food waste presents a significant source of contamination to wastewater. This qualitative assessment is based on the following observations: 1) these compounds continue to be detected at part per billion levels in dairy, meat and seafood; 2) diet constitutes the main source of exposure for PCBs and the DDT complex to the general population; and 3) a high percentage of food in the U.S. is disposed as waste.

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