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Organic contaminants in municipal biosolids: risk assessment, quantitative pathways analysis, and current research priorities

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Abstract

Basic research and monitoring of the fate and potential effects of PCBs and other xenobiotic organics in biosolids (municipal sewage sludge) used on cropland have identified specific Pathways by which the xenobiotic organics in biosolids can reach and cause exposure to humans, livestock, plants, soil biota, wildlife, etc. In order to provide the scientific basis for the Clean Water Act Regulations (40 CFR 503) on land application of biosolids in the U.S., a Pathway Approach to risk assessment was undertaken. Pathways included general food production; garden food production; soil ingestion by humans, livestock, and wildlife; human exposure through livestock tissues where the livestock were exposed through crop contamination or biosolids/soil ingestion; wildlife exposure through soil organisms; release to surface and groundwater; volatilization into inhaled air, or dust generated by tillage. Two Pathways were found to comprise the greatest risk from persistent lipophilic organic compounds such as PCBs: (1) adherence of biosolids to forage/pasture crops from surface application of fluid biosolids, followed by grazing and ingestion of biosolids by livestock used as human food; and (2) direct ingestion of biosolids by children. Each Pathway considers risk to Highly Exposed Individuals (HEIs) rather than to the general population who seldom have appreciable exposure to biosolids or foods grown on biosolid-amended soils. Because present (1995) biosolids contain very low levels of PCBs in countries which have prohibited manufacture and use of these compounds, the estimated increase in lifetime cancer risk to HEIs from biosolids-borne PCBs applied to cropland or gardens was much less than 10^{-4}. Low biosolids PCBs and low probability of simultaneously meeting all the constraints of the HEI indicate that HEIs have less than 10^{-7} increase in lifetime cancer risk from biosolids-borne PCBs; this provides even higher protection to the general population. We conclude that quantitative risk assessment for potentially toxic constituents in biosolids can be meaningfully conducted because research has provided transfer coefficients from biosolids and biosolid-amended soils to plants and animals needed to assess risk for many organic compounds.

Keywords: Risk assessment; Organic contaminants; Pathways analysis; Biosolids; Sewage sludge

1. Introduction

U.S. approaches to development of regulations for land application of biosolids (municipal sewage sludge) have changed over the years as scientific information about the fate and effects of potentially toxic constituents in biosolids has become more complete. Until the 1970s biosolids were regulated only as a source of infection or as a public nuisance due to odours. In the mid-1970s,

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guidance was provided by the Agricultural Community, accepted by most States, and used by U.S.-EPA in their advice to Publicly Owned Treatment Works (POTWs). By 1979, increasing scientific and public concerns about the contaminants in biosolids lead the U.S.-Environmental Protection Agency (U.S.-EPA) to propose a regulation to control land application. The 1979 regulation was developed as part of a comprehensive rule about landfills, which included a specific section on land application of biosolids.

The 1970s were a period of intense searching for environmental risks, and for research on land application of biosolids. One of the most important areas of risk from biosolids use or disposal which has been identified was the potential for biosolids sold or given to individual citizens (Distributed or Marketed, D&M) without controls. In 1970, many Publicly Owned Treatment Works (POTWs) dried anaerobically digested biosolids on sand beds during part of the year, and these dried biosolids were often given to anyone who would carry them away. As in Europe, research found examples of highly PCB-contaminated biosolids (and Cd, Pb, Hg, etc.) being given away for use on lawns, gardens, and farms (Bergh and Peoples, 1977; Landrigan et al., 1978). This information dramatized the need for regulation of biosolids use in situations other than just use on farmland. Uncontrolled rates of application and uncontrolled exposures seemed to allow human or environmental risk, at least is some cases where the biosolids had high levels of contaminants from uncontrolled industrial discharge to the sewers. The U.S. Government Accounting Office concluded that the practice should either be controlled or prohibited (Comptroller General, 1978). Although the 1979 regulation was the basis for the first U.S. nationwide regulations on biosolids application on land, the rule remained un-finalized despite the GAO warning.

When the U.S. Congress reauthorized the Clean Water Act in the mid-1980s, they decided that further biosolids regulations were needed, partly because the first rule dealt with only a few contaminants known to be in biosolids, and partly because biosolids D&M continued in many cities with no regulation (except for State regulations or local ordinances). No one debated the information that biosolids could contain elements and organic compounds which could cause environmental or human harm in the absence of regulations. At the same time, other Congressional and U.S.-EPA actions were improving industrial pretreatment regulations for many industries which discharged unwanted organic compounds to the sewers. PCBs' use in industry was banned because of the problems found with PCBs' dispersal in the environment, bioaccumulation in aquatic food chains, and harm to some threatened species. This ban, coupled with regulations on acceptable methods for disposal of PCBs already produced, caused PCB concentrations in biosolids to fall remarkably. As part of the pretreatment guidelines, protection of the quality of biosolids was listed as a valid reason to require pretreatment. Therefore, if public acceptance of biosolids application, or if meeting state regulations on biosolids use required better industrial pretreatment, states and cities now had the authority to require lower contaminant levels in biosolids. Results from the 1991 U.S. National Sewage Sludge Survey (U.S.-EPA, 1990), confirmed that concentrations of contaminants in most U.S. biosolids were much lower than found in previous U.S. surveys. This was especially true for compounds such as the PCBs which had been prohibited during this period. However, the U.S.-EPA survey found mostly 'not detected' PCB results, partly because PCBs in biosolids had fallen with the prohibition of PCB manufacture and use, but also because solids content in biosolids samples was not considered in taking wet samples for analysis. For those compounds with many 'non-detected' samples, the Maximum Likelihood Estimation procedure was used to provide a reasonable estimation of the geometric distribution of a constituent in dry biosolids. Using the Maximum Likelihood Estimation Procedure, U.S.-EPA reported that the 98th percentile of biosolid PCB concentration was 0.21 mg/kg dry wt. These findings by U.S.-EPA are supported by findings of low PCBs in samples analyzed in the U.S. (Gutenmann et al., 1994) and the UK (Alcock and Jones, 1993).

During the 1980s, it became clear that addition of equal amounts of an inorganic or organic contaminant in biosolids versus addition as a pure chemical caused lower solubility or phytoavailability for biosolid-applied compounds than found

with the pure chemicals. Biosolids were found to add specific adsorption capacity for inorganics and organics to the biosolids-soil mixture, causing a plateau response between plant metals and biosolid-applied metals (Corey et al., 1987). A similar plateau response of plant concentrations to increasing application rates of a biosolid was observed for biosolids-borne PCBs and PAHs by O'Connor et al. (1990) and by Wild and Jones (1992).

The implication of this relationship (classic adsorption of PCBs to biosolids organic matter) means that biosolids with higher concentrations of a contaminant, the contaminant is bound less strongly, thereby increasing the potential for risk when equal applications (kg/ha·year) of the contaminant are applied from higher contaminant level biosolids. Previously it had been believed that all biosolids sources produced equal effect/risk at equal cumulative applied amounts of a contaminant, and that limiting the cumulative amount of metals or organics was the appropriate form of regulation. With this new information that the concentration of a constituent in a biosolid could strongly affect the potential transfer to plants or animals, there was a scientific basis for regulating biosolids contaminant concentrations rather than only regulating annual or cumulative applications of the contaminant.

The final effect of these improved understandings about the fate and potential effects of contaminants in biosolids was the development of the 'biosolid quality' based regulations and advisories for biosolids use [NOAEL] (No Observed Adverse Effect Level), 'Alternative Pollutant Level' (APL), or 'Exceptional Quality (EQ) Biosolids' in which biosolids constituents are limited to concentrations which comprise low risk to Highly Exposed Individuals (HEIs) (near worst-case scenarios) (U.S.-EPA, 1989a; O'Connor et al., 1989; Jacobs et al., 1989; Chaney, 1990; U.S.-EPA, 1993). These approaches rely on research findings and calculations which showed that application of 1000 dry t of a NOAEL/APL/EQ biosolid/ha comprised no risk to agriculture, livestock, humans, or wildlife/ecosystems. The U.S. Pathway Approach contains the same considerations which other nations and scientists have judged appropriate for the evaluation (Wild and Jones, 1992).

We believe that the comprehensive risk analysis effort which allowed development of the NOAEL approach may be helpful to other nations which are considering biosolids regulations. Many of the field data needed to estimate limits for biosolids applications are difficult to find in the literature, but we are convinced that reliable data are available upon which a valid regulation can be based. Further, the detailed analyses identified specific areas of needed research; research not yet conducted in the agricultural or environmental research communities. Thus, this paper summarizes the new U.S. approach to regulation of PCBs in biosolids, and the general conclusions reached. We conclude that if biosolids can be beneficially used in sustainable agriculture with so low risk to agriculture or environment, that use on farmland should be the preferred method of 'Ultimate Disposal'. Pretreatment of industrial and non-industrial sources of some contaminants may be required to achieve the NOAEL biosolids quality. Technology is presently available to achieve the needed pretreatment.

Some details of the methods and data used for the 503 Rule risk analyses have been described in our papers (Chaney et al., 1991; O'Connor et al., 1991; Ryan and Chaney, 1995) and in official documents about the Rule (U.S.-EPA, 1989a, 1991, 1992, 1993a). Our review of plant uptake of toxic organics discussed at length the potential errors in research methods and methods of analysis which have often caused overestimation of the potential risk of toxic organics in biosolids (O'Connor et al., 1991). After working on this reconsideration of the scientific basis for biosolids regulations for the U.S., we believe that a detailed example of risk analysis for biosolids PCB in the environment illustrates the Pathway Method of quantitative risk assessment which is the basis of the 40 CFR 503 regulation in the United States. Based on these evaluations, we conclude that PCB concentrations will limit use of biosolid from only a few of the 14 000 POTWs in the U.S.

2. General principles of the fate of organic compounds during treatment of domestic wastewater and production of biosolids

Organic chemicals enter wastewater from

human wastes, consumer products, street runoff, and industrial wastes. Some organic chemicals are produced and used and disposed in huge quantities. Although there may be over 100 000 different organic chemicals used in commerce, and some quantity may be found in wastewater of the industries which produce or use the chemicals, the present review is focused on biosolids. Most organic chemicals are present in biosolids in low concentrations (U.S.-EPA, 1990) because (1) many compounds are biodegraded during aerobic wastewater treatment; (2) some compounds are volatilized during activated sludge treatment of wastewater; and (3) some compounds distribute to the effluent during final settling of treated wastewater. The organic compounds which enter the biosolids, and persist during biosolids processing, are mostly the persistent lipophilic compounds bound to the biosolids. Some compounds (e.g. phthalates, nonylphenol detergents) enter the biosolids and are not degraded by anaerobic processing, but are rapidly biodegraded when processed aerobically or mixed with aerobic soils. Other compounds (e.g. polyhalogenated hydrocarbons) are lipophilic and slowly biodegraded even in aerobic soils.

Many known toxic organic compounds have been found to cause adverse effects under the conditions of their commercial use; production or use of these chemicals has been prohibited (e.g. PCBs, dieldrin, DDT, heptachlor, etc.). Further, industrial pretreatment has been very effective in reducing discharges. Because of these regulatory controls, the concentrations of persistent xenobiotics found in modern biosolids are quite low. In the National Sewage Sludge Survey (NSSS) (U.S.-EPA, 1990), nearly all samples had non-detectable levels of PCBs, DDT, etc. In the development of the final 503 Rule (U.S.-EPA, 1993), all of the organic compounds originally considered for regulation were deleted based on a number of criteria: (1) The contaminant has been banned for use in the United States; has restricted use in the United States: or is not manufactured for use in the United States; (2) Based on the results of the National Sewage Sludge Survey, the compound has a low percent detection in sewage sludge; or (3) Based on data from the NSSS, the limit for an organic contaminant in the Part 503 exposure assessment by use or disposal practice is not expected to be exceeded in sewage sludge that is used or disposed. Because these compounds were in nearly every case present at far lower concentrations than would have been permitted under the Pathway Risk Assessment, this action seems responsible. Surrogate compounds were used to represent large groups of organic compounds for the purpose of risk assessment. This was necessary because full details of the fate and potential effects of each individual PAH, PCB, etc., are not available, but adequate information was available to estimate risk for highly toxic representatives of a group of compounds. In the end, not only the PCBs and prohibited pesticides were found to comprise no practical risk when biosolids are used on land, but also compounds such as nitrosamines, and the polycyclic aromatic hydrocarbons such as benzo[*a*]pyrene. Some states have decided to continue to require evaluation of the presence of specific toxic organic compounds in biosolids (using composite sampling to reduce analytical costs), to assure that the concentrations in biosolids products remain very low. We conclude that annual analysis (of samples composited over time) for persistent lipophilic xenobiotics in biosolids destined for land application is a prudent measure. One state has reported to us that of over 400 treatment works in the state, three still had higher than 1.0 mg PCBs/kg, with one of these over 10 mg/kg in 1994. Most of the remaining POTWs had <0.1 mg/kg dry solids, the usual pattern in nations which prohibited these industrial chemicals long ago. In a recent survey of PCBs in the biosolids generated by 16 large U.S. cities by Gutenmann et al. (1994), 15 had lower than 0.25 mg/kg dry wt. (their detection limit), and one had 4.6 mg/kg. In a survey of PCBs in 12 UK biosolids, Alcock et al. (1993) used more sensitive methods and analyzed individual congeners; they found the sum of PCB congeners was 0.106–0.712 mg (ΣPCBs)/kg dry wt. (mean 0.292 mg/kg).

Because risk assessment starts with a consideration of the concentrations of a compound in biosolids, knowing that only very low concentrations of most potentially toxic organic compounds occur in 1995 biosolids is important. Although higher concentrations may be found in biosolids

from a small town which has a factory which produces toxic organic compounds, POTWs and state regulatory authorities have the freedom to consider need for site specific limits or pretreatment requirements

3. Pathway approach for analysis of the risks to humans and the environment from toxic organics in land-applied municipal biosolids

During the 1980s, the U.S.-EPA worked with the research community to develop a 'Methodology' to assess the potential risk of inorganic and toxic organic compound transfer to humans or the environment from all identified pathways for this transfer (U.S.-EPA, 1989a). The EPA Pathways (Table 1) were selected after consideration of many research studies completed by this time (Hansen and Lambert, 1987; Jacobs et al., 1987; O'Connor et al., 1989). Under the Pathway Risk Assessment Approach, risk assessment is conducted in such a way as to protect Highly Exposed Individuals (HEIs). Initially, the Methodology called for protection of 'Most Exposed Individuals', with highly conservative assumptions for each step of the calculation. However, the detailed review of the proposed 503 Rule (Page et al., 1989) indicated that these assumptions defined a population group which did not exist. During consideration of the appropriate response to the review comments on the proposed 503 Rule, EPA revised its definition of the highly exposed and sensitive population which is to be protected as the HEIs. First, the rule ignores the bulk of the population who are not ac-

Table 1
Pathways for risk assessment of potential transfer of biosolids-applied trace contaminants to humans, livestock, or the environment, and the Highly Exposed Individual to be protected by regulation (U.S.-EPA, 1989a)

	Pathway	Highly exposed individual
1	Biosolids → Soil → Plant → Human	General food chain; 2.5% of all food for lifetime
2	Biosolids → Soil → Plant → Human	Home garden 5 years after last biosolids incorporation; 50% of garden foods for lifetime
2 D&M	biosolids → Soil → Plant → Human	Home garden with annual biosolids application; 50% of garden foods for lifetime
3	Biosolids → Soil → Human child	Residential soil, 5 year after last biosolids incorporation; 200 mg soil/day
3 D&M	biosolids → Human child	Biosolids product on soil surface; 200 mg biosolids/day
4	Biosolids → Soil → Plant → Animal → Human	Rural farm families; 40% of meat from livestock produced using crops produced on soils with incorporated biosolids, for lifetime
5 Surface	biosolids → Animal → Human	Rural farm families; 40% of meat from livestock produced on pastures annually sprayed with biosolids, for lifetime
5 Mixed	biosolids → Soil → Animal → Human	Rural farm families; 40% of meat from livestock produced on pastures with incorporated biosolids, for lifetime
6	Biosolids → Soil → Plant → Animal	Livestock fed feed forages and grains, 100% of which are grown on biosolids-amended land
7 Surface	biosolids → Animal	Grazing livestock on biosolids sprayed pastures; 1.5% biosolids in diet
7 Mixed	biosolids → Soil → Animal	Grazing livestock; 2.5% biosolids-soil mixture in diet
8	Biosolids → Soil → Plant	'Crops'; vegetables in strongly acidic biosolids-amended soil
9	Biosolids → Soil → Soil biota	Earthworms, slugs, bacteria, fungi in biosolids-amended soil
10	Biosolids → Soil → Soil biota → Predator	Birds; 33% of bird diet earthworms affected by biosolids
11	Biosolids → Soil → Airborne dust → Human	Tractor operator
12	Biosolids → Soil → Surface water → Human	Water Quality Criteria; fish bioaccumulation, lifetime
13	Biosolids → Soil → Air → Human	Farm households
14	Biosolids → Soil → Groundwater → Human	Farm wells supply 100% of water used for lifetime

For Distributed or Marketed products (D&M), it is assumed that undiluted products may remain on the soil surface.

tually exposed to biosolids or foods grown on biosolids-amended soils. Within the subset of the whole population which are actually exposed to biosolids or foods grown on fields amended with biosolids, there exists a population which are highly exposed to the risks from land application of biosolids. The HEIs are those who use biosolids products on their lawns, gardens, or farms, and who ingest foods produced on biosolids-amended soils as a high portion of their diet for a 70-year lifetime. The HEI can be a child who ingests biosolids or the biosolids-soil mixture, a human adult consuming foods grown on biosolids-amended land for 70 years, livestock which graze biosolids-amended pastures, earthworms, soil bacteria and fungi living in biosolid-amended soils, small mammals or birds which consume large amounts of earthworms or other soil organisms, etc. (Table 1) (see also U.S.-EPA, 1989a, 1992). Because the regulation must protect humans and the environment for the long term, the 503 Rule is based on the assumption that the amended soil has received 1000 dry t/ha, a massive cumulative application. EPA described this as 20 years at 50 t/ha·year in some parts of the rule, and as 100 years at 10 t/ha·year in others. In reality, depending on the nature of the influents, treatment processes, and sludge management methods, a wide range of application rates might be found. In the case where nitrogen in the biosolids is preserved to maximize the beneficial use in the field as N fertilizer (containing 5% total N), after several annual applications only 3 t/ha·year would provide 150 kg N/ha of plant available N; thus application of the presumed loading for the Rule would take 333 consecutive years to avoid exceeding the N fertilizer limit. It is extremely unlikely that the same fields would receive biosolids each year for hundreds of years in a row, so the actual time required to reach the cumulative application assumed for the Rule would be much longer (see Ryan and Chaney (1992, 1996) for discussion of time to reach the regulated loading).

Other reviewers have considered the possible transfer of toxic organics from biosolids to humans (Fries, 1982, 1991; Fries and Marrow, 1982; Harms and Sauerbeck, 1983; Davis et al., 1984; Chaney et al., 1987; Page et al., 1989; Stark and Hall, 1992; Wild and Jones, 1992; Harrad et al., 1994). These reviews have focused on lifetime consumption by a farm family of a large fraction of ingested meats from livestock grazing pastures to which biosolid products were surface applied (Pathway 5-Surface) (the 'home-grown meat scenario'), or the direct ingestion of Distributed or Marketed (Pathway 3-D&M) biosolids products by children. It has become generally accepted that all other Pathways transfer biosolids-applied organics to humans at much lower levels than do Pathways 3-D&M and 5-Surface. Quantitative estimation of the allowed applications of biosolids PCBs under all the pathways clearly demonstrates why these two Pathways are so important. If these are the critical pathways for PCBs, as well as all persistent potentially toxic organic compounds, then research on other organics should focus on the critical transfer coefficients involved in these Pathways.

The Pathways will be reviewed in the sequence of higher human exposures to lower exposures, rather than in the 1–14 order. At the end of this paper, the limits estimated for all pathways will be summarized, but detailed calculations are provided only for the most important pathways which have caused concern to the general public regarding use of biosolids on land.

3.1. Assessing risk from soil/biosolids ingestion (Pathway 3-D&M; biosolids → human)

Direct biosolids ingestion by humans is the simplest Pathway for biosolids-PCBs to reach humans. The importance of this pathway has become increasingly recognized for contaminant transfer after research on Pb exposures of children identified soil and dust as significant possible exposure routes. Perhaps the most convincing demonstration of Pb transfer by housedust ingestion comes from studies to characterize blood Pb in children of workers in the Pb industry who brought their work clothing to their homes and thereby contaminated the housedust to which their children were exposed (Baker et al., 1977). Continuing research on this pathway of Pb exposure has shown the importance of hand-to-mouth play and of pica (intentional ingestion of non-food items) on transfer of soil-Pb and other strongly soil-adsorbed

metals (As) and organics to children (Calabrese et al., 1989; Chaney et al., 1989). Clearly, soil Pb comprises a much greater risk to children with pica than normal children. Because of the concern about contaminant risk from soil ingestion by children, information needed to assess Pathway 3-D&M risks has been assembled for development of regulations.

In the EPA Methodology, the algorithm for calculating Pathway 3 starts by calculating the allowed daily ingestion of PCBs (RIA) so as to protect against development of cancer in the exposed population during lifetime exposure. According to EPA, cancer from excessive ingested PCBs is the most sensitive risk endpoint for humans from chronic ingestion of PCBs. The q_1* is the cancer potency slope from the Cancer Assessment Group of EPA. This slope relates increased cancer incidence vs. dose of a chemical during lifetime feeding experiments with rats and other animals. However, the test substances are fed at the maximum tolerated dose (MTD) to allow more cost-effective testing. The MTD method is generally agreed to overestimate potential cancer risks. Significant artifacts are created due to hyperplasia induced by the toxicity of the high MTD dose to cells (Ames and Gold, 1990). Using the results from these tests, EPA then calculates the upper 95% confidence limit of the actual slope for use as the regulatory limit, in order to add further protection against underestimating the cancer risk from a compound. The q_1* is used with other assumed parameters (RL, BW, RE) to calculate the Adjusted Reference Intake (RIA) (Eq. 1). The RIA is the increase in average lifetime (70 years) human daily intake of biosolids-applied PCBs that may not be exceeded if the Risk Level is to be increased no more than 1 in 10^4 as required for this Regulation by U.S.-EPA policy.

$$\mathrm{RIA} = \left\{\frac{\mathrm{RL}\cdot\mathrm{BW}}{q_1{}^*\cdot\mathrm{RE}}\right\}\cdot 10^3 = \left\{\frac{10^{-4}\cdot\ 10\ \mathrm{kg}}{7.7\cdot 1}\right\}\cdot 10^3 = \frac{1.00}{q_1} = 0.208\ \mu\mathrm{g/day} \qquad (1)$$

where RIA = Adjusted Reference Intake (μg/day).

q_1* = Human cancer potency slope ([mg/kg body wt./day]$^{-1}$).

RL = Risk Level = 0.0001 for HEIs (EPA policy).

BW = Body weight of 2-year-old child = 16 kg

RE = Relative Effectiveness, or bioavailability of the contaminant in ingested biosolids compared to pure chemical form (used in cancer potency slope assessment) added to diet.

The Reference Soil Concentration (RLC) which cannot be exceeded at the selected risk level is then calculated from the RIA using Eq. 2. In this case the Pathway 3-D&M RLC is the maximum allowed biosolids PCB concentration for a biosolids D&M product. Agricultural Use Pathway 3 (ingestion of the biosolids-soil mixture 5 years after cessation of mixing biosolids with the soil) would provide appreciably less exposure than Pathway 3-D&M because of the dilution of biosolids with soil and biodegradation of PCBs over time.

To calculate the RLC, several variables must be defined. For the body weight of the Highly Exposed Individual, we used the weight of 18- to 24-month-old children (16 kg). Children have maximal hand-to-mouth activity at this age, and peak blood Pb of childhood (Bornschein et al., 1985), although soil ingestion is observed in some children throughout the age range of 12–72 months.

How much soil or dust do children consume? The amount of biosolid product or soil which is ingested by Highly Exposed Individuals (1- to 6-year-old children) is also required. Much research has been conducted during the last 5 years to quantify the amount of soil children actually ingest, so that presumed levels of ingestion would not have to be used for this calculation. For the sake of convenience, the soil ingestion estimates are used in lue of biosolid product ingestion estimates since the latter has never been observed or measured; it is only assumed to provide extra measures of protection in case unsupervised children might have access to bags or biosolid products.

Historically, soil ingestion by livestock has been measured by analysis of the feces for soil-borne

elements which are not appreciably absorbed by animals (e.g. Ti, Si, Cr, etc.) (Healy et al., 1974). This method has been called the 'non-absorbed soil tracer method', where the word 'tracer' is used in the sense of isotopic tracers. Because of the potential importance of soil and housedust ingestion in Pb or xenobiotic exposure of children, a number of studies have been conducted to measure the mean and the statistical distribution of daily soil ingestion by children (Binder et al., 1986; Clausing et al., 1987; Calabrese et al., 1989; Davis et al., 1990; Van Wijnen et al., 1990).

Each of these listed studies has experienced an experimental difficulty, failed to measure some variable, made weak assumptions, or studied small populations, such that improved data were badly needed for risk assessment models (Calabrese et al., 1989). Methods for measurement of elements representing soil to allow calculation of soil ingestion by children have been substantially improved in the last few years, correcting unexpected sources of error and reducing variation in the measurements.

Papers from the Calabrese group (Calabrese et al., 1989, as further evaluated in Stanek and Calabrese, 1991a,b; Calabrese and Stanek, 1991; Stanek and Calabrese, 1994; Calabrese and Stanek, 1994), show how an improved method was developed to use the soil tracer approach. Their work has provided the most reliable published estimates for soil ingestion by children at this time. These researchers collected diets (for 6 days) and feces (for 8 days) during a 2-week period for 64 children at urban day care facilities. They analyzed the diets and feces for a number of trace elements (Al, Si, Ti, Ba, Mn, V, Y, Zr) using very reliable analytical methods with excellent QA/QC. These elements are present in soils; some of the elements are not appreciably absorbed by plants or animals, and they are generally present at very low levels in foods. By analyzing the diets (Stanek et al., 1988) as well as the feces, they found that they could not rely on Ti analyses to estimate soil ingestion by children even when non-Ti toothpaste was supplied to the parents of the children, because of dietary Ti levels and variability among children due to other domestic Ti exposures. Similar errors were found with Mn and V; high variability in food element intakes prevented useful measurement of soil ingestion. Most previous soil ingestion measurements (Binder et al., 1986; Clausing et al., 1987; Davis et al., 1990) partially depended on Ti analyses, although Binder et al. (1986) showed nearly 10 times higher estimated soil ingestion based on Ti than on Si or Al. Using inductively coupled plasma emission spectrometry to measure fecal levels of other soil elements which occur in foods at very low levels, and which are very poorly absorbed by animals (Y, Zr, Si, etc.) Calabrese et al. were able to obtain more reliable estimates of soil ingestion.

In order to provide protection of highly exposed individuals from excessive absorption of PCBs from biosolids products, the soil ingestion used in the EPA calculation is supposed to be at about the 95th percentile of the exposed population. With the significant number of missing data for daily soil ingestion due to 'non-detected' measurement of elements above food element intakes, Calabrese et al. developed a method to estimate soil detection limits for each element for each fecal sample (Stanek and Calabrese, 1991; Calabrese and Stanek, 1991; Stanek and Calabrese, 1994; Calabrese and Stanek, 1994). In the examination of the re-evaluated daily soil ingestion measurements of their population of 64 children, Calabrese et al. (1991) reported that the median soil ingestion had fallen significantly compared to the uncorrected results.

It has proven extremely difficult to obtain valid measurement of soil ingestion by children. Because Calabrese et al. used multiple tracer elements to estimate soil in the diets and feces, and attempted to correct for the tracer elements in the diets of the individual test children, the results of Calabrese et al. (1989, 1991, 1994) are the most complete available. Further, as our understanding of how to calculate soil ingestion from the collected data have improved over the last 5 years, these authors have revised their paper in subsequent publications. Even in 1994, new sources of errors were reported. In the original paper, the Calabrese group used as the soil sample to which the children were presumed to be exposed, a calculated mean soil composition based on the analysis of samples collected at their day care center and other samples

collected in play areas at their homes, and house dust from play areas in their homes (in 1994, they used only the soil composition data since they could not distinguish between these two sources by their analyses). The soils were dried, crushed, and sieved to <2 mm. They then ground the sample to a fine powder to minimize variance in the analysis of the soil, used a total metal dissolution digestion method for soils, and analyzed the samples by inductively coupled plasma spectrometry with appropriate standardization and corrections.

Other research has shown that the particle size of soil and dust on children's hands is <100 μm (Duggan and Inskip, 1985; see Chaney and Ryan, 1994 for summary). Most recent research on risk to children of Pb in soils has used the <250-μm soil fraction for chemical analysis so that the sample would represent potential ingestion by hand-to-mouth transfer (Weitzman et al., 1993); the <250-μm fraction was also selected for practical reasons of the time required to obtain a representative sample from the bulk soil sample. It is well known that soil particles vary in the relative concentrations of different elements; Sheppard and Evenden (1994) have reported the important effects this variation can have on estimated composition of the actual soil particles ingested compared to the bulk soil. For example, Si is higher in sand size than in clay size particles, but Ti, Zr and Pb are much higher in the finer particles than in the sand sized particles under most circumstances. Many of the elements in soils are present as adsorbed ions on the hydrous oxides of Fe and Mn which coat the soil clay particles. The variation in composition of soil particles with different sizes is now recognized as another factor which affects the reliability of tracer elements to estimate soil ingested by children. Use of the composition of the <250-μm soil samples to calculate soil in the feces gave improved agreement between the different tracer elements, and the daily soil ingestion estimates declined somewhat (Calabrese, personal communication).

Another improvement within the Calabrese et al. protocol is their effort to estimate the median soil ingestion for the population when some or many children had 'non-detected' levels of soils in feces after correction for diet intakes for each of the tracer elements by using the Maximum Likelihood Estimation procedure. By deleting the elements which suffered from identified sources of error (were outliers by defined criteria), the reliable estimates were assembled. Then the soil and dust intake for each day was evaluated for each child, and those with non-detected soil ingestion were omitted in some calculations or a default value of 1 mg/day was used in other calculations. Because of the missing values for missed fecal samples or for non-detectable soil content of the samples, the median child was near the detection limit. Use of the maximum likelihood estimation procedure allowed estimation of the statistical distribution of soil intakes by the population, including the median and 95th percentiles. Another point not stressed in the consideration of these children was the important role of two children who had repeated substantial soil ingestion during the study period. One of the children averaged nearly 8 g soil/day for the days when samples were reliable, and the high soil ingestion levels occurred in only one of the 2 weeks of sampling (Calabrese et al., 1991). Other researchers have also noted individual children who ingested high levels of soil.

Recently the intent of the 95–98th percentile estimate has become confused by another paper by Calabrese and Stanek (1994) which purports to estimate the 95th percentile of soil ingestion by children during 1 year, by extrapolation of the data from the population of 62 children with 3–8 days of measurements of fecal soils. It is important that the intent of the U.S.-EPA (1989a) calculation be understood. EPA desired to use an estimate of chronic daily soil ingestion at the 95th percentile of childhood daily soil ingestion rates among a population. Because cancer protections are based on lifetime exposures, the intake on a single day is only important in its contribution to the lifetime exposure. Thus, the required limit is the 95th percentile of average daily soil intakes by a population of children. Actually, since the available data are for children nearer the peak of soil and dust ingestion, the available results are probably an overestimation of the average daily soil intake from age 1 to age 6.

Thus, based on the median tracer element-based soil intake estimates, the median soil ingestion was

13 mg/day, and the 95th percentile (log normal) of measured soil/dust ingestion by 2-year-old children was 0.138 g/day. Using the mean tracer element-based soil intake estimates, the median soil ingestion was 45 mg/day, and the 95th percentile was 208 mg/day. Most observers note that the peak soil ingestion period by children is about 1–2 years long. Use of the 0.5 g biosolids/day for 2 years provides a very conservative soil ingestion estimate to minimize potential risk (O'Connor et al., 1989). In the EPA Methodology (1989a), 200 mg soil per day was used for soil intake based on other policy requirements (the number used in the EPA Superfund programme (U.S.-EPA, 1989b)). As it happens, use of either 0.2 g/day for 5 years, or the 0.5 g/day for 2 years is the same amount of biosolids ingestion exposure during childhood, and which provides the same estimate of the increase in cancer incidence due to increased PCB ingestion. We are convinced that this approach is a highly conservative estimate of average daily soil ingestion by a child during childhood.

The Duration Adjustment (DA) must be made when cancer risk slopes for 70-year lifetime exposure are used to estimate allowed exposures for soil/biosolids ingestion which lasts only a part of the lifetime. Thus, the calculation can use 0.2 g/day for 5 years (DA = 0.0714) or 0.5 g/day for 2 years (DA = 0.0286). Values of RIA, I_S, and DA are used to calculate the RLC.

$$\mathrm{RLC} = \frac{\mathrm{RIA}}{I_S \cdot \mathrm{DA}} = \frac{0.208\ \mu\mathrm{g/day}}{[(0.50 \cdot 0.0286) = 0.0143] \text{ or } [(0.20 \cdot 0.0714) = 0.0143]} = 14.5\ \mu\mathrm{g\ PCB/g\ dry\ biosolids} \quad (2)$$

RLC = Reference Soil/Biosolids Concentration of the contaminant (μg/g).
DA = Duration Adjustment for <70 year: 5/70 = 0.0714; 2/70 = 0.0286.
I_S = Soil Ingestion Rate (g dry wt. per day).

Thus, for PCBs, RLC = 14 μg/g dry wt. biosolids (rounded down according to EPA Policy) for marketed biosolid products which may be ingested directly from the soil surface or from the container. The Pathway 3-D&M risk assessment, in which children are assumed to ingest biosolids directly for 2–5 years, indicates that the required biosolids PCB concentration limit is much higher than present levels of PCBs in biosolids (95th percentile, 0.21 mg/kg). Based on data discussed below for bioconcentration of PCB from biosolids ingested by cattle compared to bioconcentration from pure PCB added to cattle diets, the relative effectiveness of PCBs in the biosolids matrix may be about 0.50; if this measured RE had been used, the RLC would been doubled. One soil feeding study with mammals has been reported for PCBs (Fries et al., 1989). If present definitions of relative bioavailability are used, the results for persistent PCBs in soil mixed into diets are about 50% of corn soil gavage PCBs. In this study, chemical PCBs were added to the soil rather than PCBs pre-equilibrated with a biosolid, and the biosolid mixed with the soil. As discussed below, all forms of 'spiked' xenobiotics are more bioavailable than biosolid-incorporated xenobiotics.

Many conservative factors were embedded in this estimate of the allowable PCB limit for marketed biosolid products [Pathway 3-D&M]: (1) sensitive rodent strains were given maximum tolerated doses of PCBs during lifetime feeding studies to estimate the cancer slope; (2) the upper 95% Confidence Interval was used in place of the measured slope; (3) childhood soil ingestion estimates higher than the 95th percentile of a log normal distribution were used as the soil intake; (4) the Relative Effectiveness (bioavailability) was assumed to be 100%; (5) it is highly likely that written directions on the bag and supervision of parents would greatly reduce the average daily intake of biosolid products by children; and (6) young children cannot realistically be expected to have soil exposure each day due to adverse weather conditions. Compared to the HEI, typical children have far lower average daily soil intakes during ages 1–6, the peak period of hand-to-mouth play of children. Thus, a high level of protection was provided by the Pathway Risk Assessment Methodology.

3.2. Risk from biosolids ingestion by grazing livestock used as food by humans (Pathway 5; biosolids → soil → animal → human and biosolids → animal → human)

For Pathway 5, the source of biosolids PCBs

transferred to humans is lifetime consumption of meat which comes from livestock which graze in biosolids-amended pasture fields. The Highly Exposed Individuals in this pathway are the members of a farm family who produce and consume 'home-grown' meat as a high portion of their lifetime meat consumption. Pathway 5 has two quite different kinds of exposure, which should be considered separately. The first involves direct ingestion of biosolids by livestock, where biosolids have been surface applied to pasture crops. Livestock can ingest biosolids adhering to the crops, or biosolids lying on the soil surface. Each year the grazing livestock are presumed to be exposed to freshly applied biosolids with no time for dissipation of the organic chemicals (*Pathway 5-Surface Application*). Alternatively, biosolids can be injected into the soil or mixed with the plow layer soil, and the soil ingested by the grazing livestock is the soil-biosolids mixture. Injected or incorporated biosolids are not at the soil surface; these greatly reduce exposure compared to surface application. The highest exposure under the *Pathway 4-Mixed* scenario would be livestock ingesting soil while grazing a crop seeded immediately after mixing the applied biosolids into the plow layer soil (see below).

Early studies of sheep exposure to Cu in surface applied swine manure first illustrated the adherence of fluid organic wastes to forage crops (Batey et al., 1972). Other studies had shown that livestock regularly ingest soils, and that soil ingestion was able to cause significant transfer of chlorinated hydrocarbon pesticides from soil to edible tissues of grazing livestock (Harrison et al., 1970; Healy et al., 1974). Based on these reports, Chaney et al. (1977), and Chaney and Lloyd (1979) evaluated adherence of spray applied fluid biosolids to forage crops and observed that biosolids adhered to forages for a prolonged period after application. Jones et al. (1979) extended this finding by showing that spray-applied fluid biosolids could be washed off the plants by rainfall immediately after application, but not if the biosolids were allowed to dry on the plants. Chaney and Lloyd also observed that if biosolids are applied to standing forages, the rate of application and solids content of the biosolids affected biosolids adherence (Chaney et al., 1987). N-Fertilizer rate applications of typical fluid biosolids containing 5% solids to standing forage crops caused biosolids to reach about 15% of the forage dry matter on the day of application; plant growth diluted the biosolid residue over time. This evidence led to an advisory against biosolids application on standing forages (U.S.-EPA, 1979), and imposition of a waiting period to minimize direct ingestion of biosolids by grazing livestock. Thus, the potential for biosolids ingestion by livestock from biosolids-amended pastures was substantially reduced by a policy decision on the waiting period.

Available data on field studies of livestock ingestion of biosolids surface applied on pastures are summarized in Table 2. Chaney et al. (1987) reviewed studies in which cattle grazed pastures to which biosolids were surface applied using good practices, and reported several previously unpublished studies. In the Decker et al. (1980) study, fluid biosolids were sprayed on the pastures after grazing or mowing to a low height, and the crop grew for 21 days before cattle were allowed to graze; the study used four 'rotation paddocks' so that each 7 days the cattle were moved to the next pasture which had grown for 21 days after biosolids application. By analysis of the cattle feces after 7 days grazing on the treated pastures, they found that the animals ingested about 2.5% biosolids in their diets. Metals served as a label for biosolids; concentration of metals such as Fe, Zn, Pb, and Cu were much higher in treated forages and feces than in control forages and feces of control treatment cattle. Similar results were observed by Bertrand et al. (1981). However, when biosolids compost was applied the previous grazing season rather than during each 'rotation', the cattle ingested only about 1.0% biosolids (Decker et al., 1980) (Table 2). Surface-applied dewatered biosolids caused much lower adherence to forages than found for fluid biosolids (Boswell, 1975).

Another possible consideration in this Pathway is the low likelihood that all pastures on a farm will receive surface-applied biosolid fertilization in any one year. The U.S.-EPA (1989a) Methodology considered that for the rural farm family who consumed the greatest fraction of 'home-grown' livestock (the HEI in Pathway 4), all grazing fields (pastures) were treated with biosolids each year for

Table 2
Whole grazing season mean adherence of spray-applied fluid biosolids on tall fescue (Decker et al., 1980) or 'Pensacola' bahaigrass (Bertrand et al., 1981), and biosolids content of feces of cattle which rotationally grazing these pastures

Study	Treatment	Solids content (%)	Application rate (cm)	Biosolids in/on forage (% dry wt.)	Biosolids in feces (% dry wt.)
Decker et al. (1980)[a]					
1976	21-day biosolids	4.4	20 × 0.51	5.39	7.1
1976	1-day biosolids	4.8	20 × 0.51	22.3	18.6
1977	21-day biosolids	2.9	20 × 0.51	2.18	7.7
1977	Compost			(0.74)[b]	6.5
1978	21-day biosolids	3.7	24 × 0.51	2.91	6.1
1978	Compost			(0.50)[b]	2.0
Bertrand et al. (1981)[c]					
1979	7- to 26-day biosolids	2.1	9 × 0.84	2.17	4.6
1979	7- to 13-day biosolids	2.1	18 × 0.84	5.17	5.8

[a]Four paddocks grazed on a rotation system; fluid biosolids were applied to clipped pasture 21 days before grazing (21-day biosolids), or to regrown pastures 1 day before grazing (1-day biosolids). Composted biosolids were applied three times in 1977 and one time in 1978, with at least 21 days before grazing of treated pastures. First number is number of applications during grazing season that year.

[b]Estimates based on individual elements were not in close agreement; no significant composted biosolids content.

[c]Data of Bertrand et al. (1981) recalculated using results for Cu, Fe, Pb, and Zn, elements substantially increased by biosolids application. Two paddocks were grazed in rotation. Rotations were made every 12–14 days; depending on forage growth and weather, the biosolids application occurred 7–13 days before grazing commenced. The two biosolids treatments differed in number of biosolids applications made during the grazing season.

a 70-year lifetime. However, if biosolids were applied intermittently, the fraction of biosolids in the chronic grazing animal diet is lower because adhering biosolids exceeded biosolids ingested from the soil surface in season-long grazing studies with biosolids (Chaney et al., 1987; Table 2). Based upon our experience and discussion with regulatory officials in several states, we estimate that in any one year, the maximum fraction of a farm treated with surface-applied biosolids may be 33% rather than 100%. If one presumes that the cattle are rotated among several pasture fields during the season, the actual fraction of the animal diet which the biosolids comprise will be lower than the 2.5% measured with continuous grazing exposure to recently surface-applied pastures (this lifetime approach is necessary because the cancer endpoint is based on 70 years lifetime exposure). To take this into account, the allowed RFC for biosolids-treated forage should be adjusted for the fraction of livestock diet affected by biosolids application. Using 1% biosolids ingestion during non-application periods (2/3 of time), and 2.5% ingestion during biosolids application periods (1/3 of time), the long-term average would be 1.5% biosolids in diet.

Fries (1982, 1991) has raised several other important issues about this Pathway. In most countries, modern milk production practices prevent dairy cattle from having much exposure to pastures. Without possible ingestion of surface biosolids residues from the pastures, the transfer to these cattle would be much lower. Milk cows are given dietary supplements to attain economic production, and conserved forages are fed for a substantial fraction of the diet. Each of these considerations would lower the estimated PCB transfers in Pathway 5.

To estimate PCB transfer from biosolids to grazing livestock to humans, the lifetime average amounts of fat in meat and dairy products (from grazing livestock) ingested per day are needed. The U.S.-EPA (1989a) 'Methodology' estimated the amounts of fat in the human diet from each class

of livestock, for a range of age and sex groups of the U.S. population. The original data were described by Pennington (1983). One can calculate a lifetime diet composition by assuming the number of years each age range of consumption covers. Table 3 shows the estimated average lifetime daily consumption of fat from meats from different livestock. Other foods considered in this paper are also included in Table 3.

The HEI for Pathway 5-Surface is believed to be members of a rural farm family ingesting a high fraction of their lifetime meat consumption from home grown livestock (which are exposed to biosolids on forages or soil-biosolids mixture) for their 70-year lifetime exposure. The calculation starts with estimation of the maximum allowed daily PCB intake to limit 70-year lifetime cancer risk. The adult body weight is used rather than the weight of a 2-year-old child used above for Pathway 3:

$$RIA = \left\{\frac{RL \cdot BW}{q_1^* \cdot RE}\right\} \cdot 10^3 = \left\{\frac{10^{-4} \cdot 70\ kg}{7.7 \cdot 1}\right\} \cdot 10^3 = 0.909\ \mu g\ PCB/day \quad (3)$$

RIA = adjusted reference intake (μg/day)
q_1^* = human cancer potency ([mg/kg/day]$^{-1}$)
RL = risk level = 0.0001 for U.S.-EPA, 1992
BW = body weight = 70 kg
RE = Relative Effectiveness or bioavailability.

After calculating the RIA, one calculates the maximum allowed feed concentration of PCBs (RFC) for Pathway 5:

$$RFC = \frac{RIA}{\Sigma\ (UA_i \cdot DA_i \cdot FA_i)} \quad (4)$$

Table 3
Human consumption of selected foods (fat from meats derived from several classes of livestock; garden crops) (g dry wt./day) for different age groups, and estimated lifetime average food intakes for 70-kg U.S. adult citizens

Food group	Age grouping (years)						
	Baby (0–1)	Toddler (1–6)	Child (6–14)	Teenagers (14–20)	Adult (20–45)	Older adult (45–70)	Estimated lifetime (0–70)
Grazing livestock							
Beef fat	2.45	6.48	11.34	16.22	20.40	14.07	15.50
Beef liver fat	0.05	0.07	0.08	0.10	0.29	0.33	0.25
Lamb fat	0.14	0.08	0.07	0.06	0.31	0.22	0.21
Dairy fat	38.99	16.48	20.46	24.43	18.97	14.51	18.13
Non-grazing livestock							
Pork fat	2.01	8.19	10.47	12.75	14.48	13.04	12.73
Poultry fat	1.10	0.83	1.12	1.41	1.54	1.31	1.34
Egg fat							0.96
Garden food crops							
Potatoes	5.67	10.03	14.72	19.40	17.28	14.79	15.60
Leafy vegetables	0.84	0.49	0.85	1.22	2.16	2.65	1.97
Legume vegetables	3.81	4.56	6.51	8.45	9.81	9.50	8.75
Root vegetables	3.04	0.67	1.20	1.73	1.77	1.64	1.60
Garden fruit	0.66	1.67	2.57	3.47	4.75	4.86	4.15

The child age group (not reported by Pennington, 1983) was assumed to consume the average of that consumed by toddlers and teen-agers. Meats in mixed foods assigned to source by U.S.-EPA (1989a); for Teen, Adult, and Older Adult categories, food intakes of females and males reported by Pennington (1983) were averaged; for other groups, no separation of data by sex was reported; Child intake was set equal to the average of Toddler and Teen.

where

RFC = reference feed concentration of the contaminant (μg/g dry wt.)
RIA = adjusted reference intake (μg/day)
UA_i = uptake response slope of contaminant in the animal tissue food group *i* for organics, on a fat basis, = 2 (μg PCB/g fat)·(μg PCB/g feed dry wt.)$^{-1}$ for *biosolids-borne* PCBs added to test diets
DA_i = Daily dietary consumption of animal food/fat group *i*, g dry wt.
FA_i = fraction of the food group *i* assumed to be derived from amended soil (in this case, from fat in tissues of or milk from cattle consuming 1.5% biosolids averaged over their lives). Assumes that a high fraction of the diet came from cattle raised on the biosolids-treated pastures (44% for meat fat, and 40% for dairy fat for a 70-year lifetime).

UA_i: The best known value for UA for PCB transfer from feed to fat has been discussed by Fries (1982, 1991). Based on his own and other research, Fries concluded that the UA was 4 for beef fat and 4.8 for dairy products. However, this was for pure PCB added to cattle diets or provided in corn oil. Baxter et al. (1983) tested the bioaccumulation ratio for PCB in dried anaerobically digested biosolids containing 24 mg PCBs/kg dry wt. Because biosolids adsorb PCBs strongly, the UA for biosolid-borne PCB in beef cattle was only 1.9. This value was the average for beef cows and steers fed 10% biosolids in their complete diet for 270 days, a period long enough to reach equilibrium in cattle (Fries, 1991). It should not be unexpected that biosolid PCB adsorption properties and oils in biosolids could reduce the bioavailability of biosolid PCBs. Rozman et al. (1982) found adding mineral oils to feeds increased the excretion of hexachlorobenzene from cattle. Fairbanks and O'Connor (1984) found soil organic matter strongly adsorbed PCBs, and the adsorbed PCBs showed strong hysterisis (limited release). Further characterization of release of PCBs from biosolids was obtained by O'Connor et al. (1990) in studies of crop uptake of biosolid-applied PCBs. It is now generally agreed that most organic compounds attain some level of equilibrium soon after being mixed with soils or biosolids. However, part or much of the organics then dissolve 'deeper' into the organic matter of soil or biosolids. Thus the release of adsorbed organics from biosolids is a much slower process that the initial adsorption.

FA_i: The fraction of biosolids in the diet enters into the calculation so the maximum amount of PCB allowed in biosolids can be estimated for *Pathway 5-Surface Application.*

$$RFC = \frac{RIA}{\Sigma(UA_i \cdot DA_i \cdot FA_i)} \text{ or } RFC = \frac{0.909}{27.13}$$

$$RFC = 0.0335\ \mu g/g \text{ dry wt.} \quad (5)$$

Reference Biosolids Concentration (RSC) can be calculated as specified in the Methodology:
Then, RSC = RFC/FS (6)

RSC = Reference Biosolids Concentration
RFC = Reference Feed Concentration
FS = Fraction of livestock diet comprised of biosolids (chronic lifetime exposure = 0.015 according to discussion above)
RSC = 0.0335/0.015 = 2.23 μg PCB/g dry wt. in applied biosolids.

While preparing this article, we were somewhat surprised to find that the algorithm and dataset used in the Technical Support Document (U.S.-EPA, 1992) example of how PCB limits would have been calculated if this had been kept in the 503 Rule, were quite different from the above calculations. Because PCBs were omitted from the rule, we were not asked to review the draft text for the Technical Support Document and presumed they had used our corrected version as supplied to EPA. They apparently used the geometric mean UA for each meat type, using the uncorrected

Table 4
Calculation of $\Sigma(UA_i \cdot DA_i \cdot FA_i)$ needed to estimate human exposure to PCB in fat of meat from grazing livestock for Pathway 5

Food group	Biosolids PCB in diet of grazing livestock			
	DA g/day	UA g/day	FA g/day	$(UA_i \cdot DA_i \cdot FA_i)$ estimated lifetime
Beef fat	15.50	1.9	0.44	12.96
Beef liver fat	0.25	1.9	0.44	0.21
Lamb fat	0.21	1.9	0.44	0.18
Dairy fat	18.13	1.9	0.40	13.78
ΣFat from grazing livestock	34.09	27.13		

UA value from Baxter et al. (1983), based on feeding cattle 10% biosolids for 270 days at which time equilibrium of PCB in fat with the diet would have been reached according to Fries (1991). DA is the lifetime daily average intake of fat from grazing livestock. FA is the fraction of total lifetime ingestion of these meats by the HEIs (members of a rural farm family.

dataset from the proposed Rule. Further, the FA values were 10% for meat and 3% for dairy products; these numbers came from a later USDA survey of home grown foods, but were for the non-metropolitan population, not rural farm families who produce livestock, who may have higher exposure. Because rural farm family members are known to consume much more than 10% 'home-grown' meat when it is available, we chose to use the earlier estimates for rural farm families of 40% of meat and 44% of dairy products. Using the lower exposure estimates, U.S.-EPA calculated RSC = 4.6 mg/kg compared to the 2.23 mg/kg calculated with the more appropriate values shown above. Another factor not considered by U.S.-EPA is the likelihood that small farm owners/operators who produce enough livestock to consume home grown meat so freely will have enough livestock manure requiring disposal that they will not be able to accept much biosolids. If manure supplies all the N required to produce the feeds, no biosolids may be applied under the 503 rule.

Pathway 5-Mixed With Soil This RSC number from the Surface-Application calculation is used for estimating allowed biosolids PCB applications for soil-incorporated biosolids, only now the limit is 2.23 μg PCB/g dry soil-biosolids mixture which is chronically ingested by the grazing livestock at 1.5% of diet (annual average basis; see also Chaney et al. (1987) and Fries et al. (1991)). In the case of biosolids mixed into soils, however, the regulatory approach is quite different because PCBs can be biodegraded or volatilize over time after application. The 2.23 μg PCB/g dry soil-biosolids mixture which must not be exceeded is considered to be the equilibrium reached when PCB dissipation during 1 year equals the annual PCB application. Persistent higher chlorinated PCBs have about a 6- to 10-year half-life; for these calculations, the $T_{0.5}$ is presumed to be 10 years ($k = 0.693/T_{0.5} = 0.0693$/year).

An equilibrium soil PCB concentration would be reached after about $5.6/k = 81$ years (U.S.-EPA, 1989a), and the formula used to calculate the annual PCB application mixed into the soil (RP_A) needed to reach this equilibrium RSC after 81 continuous annual applications is:

$$RP_A = RLC \cdot (2000 \text{ t soil/ha}) \cdot 10^{-3} \cdot [1e^{0 \cdot 0.0693} + 1e^{-1 \cdot 0.0693} + \ldots + 1e^{-81 \cdot 0.0693}]^{-1} \quad (7)$$
$$= 2.23 \cdot 2 \cdot [14.9]^{-1} = 0.299 \text{ kg PCB/ha/year.}$$

In practical terms, 0.299 kg PCBs/ha·year could be applied by an N-fertilizer application of 10 t/ha of a biosolids which contained 29.9 mg PCBs/kg dry wt.:

$$\frac{0.299 \text{ kg PCB}}{\text{ha} \cdot \text{year}} \cdot \frac{1 \text{ ha} \cdot \text{year}}{10 \text{ t biosolids dry wt.}}$$

$$= \frac{0.0299 \text{ kg PCB}}{\text{t biosolids}} \quad (8)$$

$$= 29.9 \text{ mg PCB/kg dry wt.}$$

Thus, Pathway 5-Surface Application comprises significantly greater potential for PCB transfer to humans than does Pathway 5-Mixed With Soil. These calculations remain a very conservative estimation of allowable toxic organic concentrations in biosolids applied to pastures. The 70-year 'lifetime' exposure scenario, a rural farm family ingesting a high proportion of home grown meat and dairy products from livestock which often graze biosolids-amended pastures, is clearly conservative. Further, in one field grazing study which involved 224 t/ha of biosolids incorporated into the plow layer before seeding the pasture, no detectable residue of PCBs were found in sheep, despite measurable residues in the biosolids and the soil-biosolids mixture (Hogue et al., 1984). In *Pathway 5-Surface Application*, the biosolids are assumed to remain on the surface of pastures continuously for 70 years in contrast with known agronomic practices which require that the surface be intermittently tilled with the plow layer soil to incorporate pH-modifying agents, fertilizers, and organic matter into the surface soil. The HEI is conservatively assumed to consume 44% of meat of grazing livestock and 40% of dairy products from livestock raised on biosolids-using farms, but few farms raise both lamb and beef, and dairy cattle are fed feed supplements (grain) to improve production efficiency (Fries, 1991). Although a later survey indicated that maximal home-grown meat and dairy products might have fallen significantly compared to previous surveys (to 10% of meat and 3% of dairy), the data were not for rural farm families which are the identifiable Highly Exposed Individuals for this Pathway.

Surface application of fluid biosolids on standing forage crops has been prohibited, and other management alternatives for biosolids use on pastures are available to minimize the ingestion of biosolids by grazing livestock. The alternatives to surface application of biosolids on pastures include use of biosolids injection, and mixing biosolids with the soil before forage crops are established. These would lead to much lower estimations of risk than found with Surface Application. *Pathway 5-Mixed With Soil* calculations indicate that humans are at about 15-fold lower risk from PCBs in soil-incorporated biosolids than for surface application of biosolids on pastures. The use of biosolids injection minimizes biosolids ingestion by grazing livestock, conserves biosolids nutrients, and prevents malodors and unsightliness of surface applied biosolids. In any case, the HEI is well protected against chronic health effects of biosolids PCBs by limitations appropriate for the method of application. And present biosolids contain far lower PCB levels that would have been allowed.

3.3. Risk from uptake of soil PCB residues by forage and feed crops for livestock used as food by humans (Pathway 4; biosolids → soil → plant → animal → human)

For Pathway 4, biosolids are presumed to be uniformly mixed with the plow layer depth of soil, and forage and feed crops are grown on the biosolids-treated soil. Livestock are not allowed to graze on the biosolids-amended fields, preventing direct ingestion of the soil, but are fed crops produced on the amended soil as 100% of their lifetime diets. As in Pathway 5, the HEIs are rural farm families assumed to consume 44% of their lifetime meat and 40% of their lifetime dairy products from 'home-grown' livestock which eat only crops grown on biosolids-treated soils. This Pathway is similar in approach to Pathway 5-Mixed, but all meats and dairy products are considered, not just the livestock which are allowed to graze on pastures and possibly ingest soil. Fat from non-grazing livestock classes (poultry, swine) are included in Pathway 4 because feed grains can be harvested from biosolids-amended soils in addition to hay and silage harvested for ruminant livestock.

Thus, several new transfer factors must be obtained to complete the calculations for Pathway 4. In this Pathway, soil PCBs enter the livestock diet by either plant uptake from the soil and translocation to the plant shoots, or by PCBs volatilization from the soil and capture by the lipophilic surface of leaves. Transfer factors are needed both for

forage crops (whole plant shoots are consumed by livestock) and grain crops (required by monogastric animals, and fed to increase milk production by cows to reduce costs of milk production, or to increase the efficiency of producing beef on feedlots).

Transfer of PCBs from biosolids-amended soil to edible parts of crops is quite low for the persistent lipophilic compounds which could require regulation. O'Connor et al. (1991) reviewed the literature on 'plant uptake' of these compounds from soils. The well characterized principles of PCB transfer from soil to crops will be summarized here with a minimal review. One of the most important principles of soil PCB transfer to plants was established by Fries and Marrow (1981), using techniques developed for study of plant uptake of DDT and other pesticides. When a compound can volatilize from the soil surface and be adsorbed on the surface of leaves, this possibility must be tested separately from uptake by the roots from the soil and translocation within the plant to the shoots. A two-chamber container with a barrier to volatile movement of PCBs between the soil and plants was used in these studies. By study of ^{14}C-labelled compounds applied to the surface soil, or to the subsurface soil below the volatilization barrier, previous researchers showed that the more strongly adsorbed lipophilic compounds (e.g. DDT) were transferred essentially only by volatilization, while more water soluble compounds such as dieldrin were transferred both by volatilization and by actual plant uptake and translocation.

Using this methodology, Fries and Marrow (1981) showed that three specific PCB congeners moved to the shoots of soybeans essentially only by volatilization. All reports since their work have confirmed that volatilization is the major mechanism of transfer of PCBs both from soil to aboveground parts of plants, and movement within the soil and to subsurface roots such as carrot. Many experiments have found some transfer of isotopic tracers to plant shoots, but the experimenters failed to prevent volatilization transfer. In studies of this type, in order to prevent cross-contamination transfer from soils in one pot to plants in another, the plants are usually surrounded by a polyethylene sleeve. This protection has the effect of significantly increasing the concentration of volatilized organics in the air surrounding the plant shoots, thereby overestimating volatile transfer and/or collection of volatilized chlorinated hydrocarbons by plants.

If the main path for PCB transfer from soil to crops is volatile transfer, the soil properties which control volatilization will control plant uptake. The dominant site of PCB binding in soils is the humic materials (organic matter), so increased organic matter concentration in a soil reduces PCB volatilization. But gas exchange to the air is also affected by soil texture and moisture contents. The transfer coefficient can be as strongly affected by the nature of the plant tissue as by the soil, since the plant leaf or stem surface must 'dissolve' the PCBs from the air to retain them. Plant leaves differ widely in the lipophilicity.

Transfer of soil PCBs, and especially biosolids-applied PCBs, to feed crops has been found to be very low in a number of studies. When reasonable concentrations of PCBs, dioxins, PBBs, DDT, etc. were added to soils, and transfer to crops in the field (and in many pot studies) were examined, non-detectable residues were found (Chou et al., 1978; Kampe and Leschber, 1985; Witte et al., 1988; O'Connor et al., 1990; Ye et al., 1991; Gan and Berthouex, 1994; McLachlan et al., 1994). [These findings are another reason why Pathway 5 has received such high attention since transfer by Pathway 4 remains non-detected unless highly contaminated biosolids were involved (Bergh and Peoples, 1977).]

In this last regard, carrot has been identified as the highest PCB uptake crop in human diets. The peel from carrots had been found to adsorb or accumulate the lipophilic pesticides (e.g. DDT). One study even found cultivar differences in carrot to dieldrin accumulation in carrot roots or peels (Hermanson et al., 1970). Other root crops do not accumulate compounds such as PCBs nearly as well as carrots. For example, sugarbeet accumulated less than one-tenth the PCB residue of carrot in the same test soil with several PCB congeners (Moza et al., 1979). Even carrots accumulate only very small amounts of lipophilic organics when the compound involved has lower vapour pressure, such as the PBBs which were not

measurable in carrot roots or other crops (Chou et al., 1978). Lower chlorinated congeners of PCBs were much more readily transferred to plant shoots and to carrot peel in different studies (Iwata and Gunther, 1979). Root crops occasionally used as cattle feed (beets, turnips) have much lower uptake slopes for PCBs than found for carrot.

Another critical principle is that the source of the PCBs can strongly affect the potential for transfer from soil to plants. Although this pattern should have been evident in the data, the detailed study by O'Connor et al. (1990) made a specific comparison of the transfer of PCBs from soils to crops when either pure chemicals were added (PCB-spiked), or biosolids with 'natural' PCBs were added to the soil. They also compared plant uptake when biosolids with very low PCB levels were added to the soil and incubated, and then the chemical PCB was added to see if the lower transfer to plants might be the result of the higher organic matter content of the biosolid-amended soil. The results clearly showed that pre-equilibrium of the PCB into the organic matter of the biosolid reduced PCB volatile transfer much more than simply increasing the soil organic matter level. It is now generally accepted that both physical mixing phenomena, and chemical adsorption phenomena contribute to this difference. But the conclusion is the same, that any transfer coefficients for PCB movement from soils to plants should be derived from studies with biosolid-applied PCBs. If the transfer to above-ground shoots were the critical process, field studies would be needed, but for carrots growing within the soil, the requirement for field measurements vs. greenhouse measurements has not been tested.

For Pathway 4 (uptake of PCBs by forage/feed plants, or contamination with PCB independent of biosolids or soil ingestion), the EPA (1989a) Methodology presumed that the PCB uptake slope for forage crops was the appropriate transfer slope for feeds for all livestock. However, in calculating the estimate of toxic organic compound transfer to humans by this Pathway, we conclude that it is important to also discriminate between grain crops fed to some classes of livestock and forage crops fed to ruminant livestock because the volatile transfer of soil PCBs to grain is far lower than to leaves. Further, milk cows are supplied grains and other 'concentrates' to provide nutrients and energy for high production rates, and it is inappropriate to presume 100% forage diet for dairy products in the U.S. (Fries, 1991). A modified approach for calculating this limitation is shown below which calculates more appropriate estimates of potential PCB transfer in the risk assessment Pathway. As noted above, these corrected approaches were not used by U.S.-EPA in the example calculations for PCBs included in the Technical Support Document, and the lack of review after the decision to delete PCBs from the final 503 Rule prevented correction of their error.

Another principle of transfer of soil PCBs to livestock has been identified in studies on dioxin in land applied biosolids (McLachlan et al., 1994). Although this principle is important, we conclude that it should be considered as an alternative Pathway 5, and recognition of this sub-Pathway 5 would not cause any changes in the 503 risk assessment since limits based on biosolids ingestion are covered under Pathway 5-Surface. In the McLachlan et al. (1994) study, dioxins were measured in the soils, feedstuffs, and milk produced by several German small farms which produced milk and meat, and did or did not use biosolids as fertilizers. They found an unexpected result. Increased levels of dioxin were found in milk on one of the farms, but it was not related to biosolids use as much as it was related to use of a forage cutters which picked up the surface thatch layer (on top of the mineral soil surface). Whether the dioxins came from aerosol deposition and plant debris falling to the soil, or from biosolids surface application, the residues remain on the soil surface until mixed into the soil. Some forage/silage making farm equipment picks up not only the cut plants, but also picks up the thatch layer. Thus, the equipment design controlled whether the dioxins applied in biosolids were transferred in significant quantities to the livestock diets. This transfer was lower than that estimated in Pathway 5 (see above), and will not be considered separately as 'plant uptake' transfer in Pathway 4.

For 70-year lifetime exposure, the EPA algorithm to calculate RIA for Pathway 4 is the same

as in Pathway 5 (Eq. 3). After calculating the RIA, one calculates the RFC for the feed as under Pathway 5, but adds the fat from other foods as shown in Table 3:

$$RFC = \frac{RIA}{\Sigma(UA_i \cdot DA_i \cdot FA_i)}$$

RFC = reference feed concentration of the contaminant (μg/g dry wt.)
RIA = adjusted reference intake (μg/day)
UA_i = uptake response slope of contaminant in the animal tissue food group *i* for organics, on a fat basis, = 4 for PCB mixtures added to test diets; does not use the data from Baxter et al. used for Pathway 4 because biosolids are not present to adsorb PCB during transit through the gut.
DA_i = Daily dietary consumption of the animal tissue food group *i*
FA_i = fraction of the food group *i* assumed to be derived from amended soil (in this case, from fat in tissues of or milk from cattle consuming feeds which were all grown on soils which received biosolids annually).

The RLC is calculated from the RFC by dividing by the uptake slope for the compound for the crop used to estimate uptake (UC):

$$RLC = (RFC/UC) + BS \quad (9)$$

where:
RLC = Reference Soil Concentration (μg/g dry wt.)
UC = linear response slope of forage or grain crop [μg/g crop dry wt. (μg/g soil dry wt.)$^{-1}$]
BS = background soil concentration of contaminant (μg/g dry wt.) (assumed to be 0 since the cancer risk to be estimated is the incremental risk, not the absolute risk).

Our method of calculation sums a new product of UC_i and DF_i (Diet Fraction) times the old product (Table 5) to directly calculate RLC from RIA:

$$RLC = \frac{RIA}{\Sigma((UC_i \cdot DF_i \cdot UA_i \cdot DA_i \cdot FA_i)}$$

$$= \frac{0.909}{0.04954} \quad (10)$$

= 18.3 mg/kg soil-biosolids mixture dry wt.

The Methodology calculation then estimates an annual biosolids PCB application rate taking into account the rate of dissipation (volatilization or degradation) of PCBs ($T_{0.5}$ = 10 years) so that the RLC is reached at steady state between applica-

Table 5
New approach to estimate PCB limitation for Pathway 4; $\Sigma(UC_i \cdot UA_i \cdot DA_i \cdot FA_i)$ is calculated to allow discrimination between PCB transfer to livestock which consume grain and those consuming forage

Animal tissue group	Feed type	Diet fraction	Uptake (UC_i) slope	UA	DA	FA	Σ
Beef fat	Forage	1.00	0.001	4.0	15.50	0.44	0.02728
Beef liver fat	Forage	1.00	0.001	4.0	0.25	0.44	0.00044
Lamb fat	Forage	1.00	0.001	4.0	0.21	0.44	0.00036
Pork fat	Grain	1.00	0.0001	4.0	12.73	0.44	0.00224
Poultry fat	Grain	1.00	0.0001	4.0	1.34	0.34	0.00018
Dairy fat	Forage	0.50	0.001	4.8	18.13	0.40	0.01740
Dairy fat	Grain	0.50	0.0001	4.8	18.13	0.40	0.00146
Eggs fat	Grain	1.00	0.0001	4.0	0.96	0.48	0.00018
Total							0.04954

In the Technical Support Document, the U.S.-EPA used the uncorrected datasets and algorithms, and calculated RLC = 30.7 mg/kg, and RP_A = 4.3 kg/ha · year.

tion and dissipation (see above for Pathway 5-Mixed).

$$RP_A = RLC \cdot (2000/1000) \cdot (14.9)^{-1} = 2.46 \text{ kg PCB/ha/year} \quad (11)$$

At an annual application rate of 10 t dry biosolids/ha, the biosolids could contain as much as 246 mg PCBs/kg:

$$\frac{2.46 \text{ kg PCBs}}{\text{ha} \cdot \text{year}} \cdot \frac{1 \text{ ha} \cdot \text{year}}{10 \text{ t biosolids dry wt.}} = \frac{0.246 \text{ kg PCBs}}{\text{t biosolids dry wt.}} = \frac{246 \text{ mg PCBs}}{\text{kg biosolids dry wt.}}$$

The high allowable annual application of biosolids PCBs for this Pathway indicates the higher protection offered by mixing applied biosolids with the plow layer soil and growing livestock feed crops rather than allowing grazing of livestock on biosolid-amended pastures. Further, because of prevention of PCB use in the U.S., and effective pretreatment and monitoring programs, modern biosolids contain low PCB levels, generally below 0.1 mg/kg dry wt. These considerations confirm the summary of relative risk from Pathways 4 and 5 noted in the introduction.

Calculations for Pathway 4 also remain very protective for the presumed HEIs. Sources of protection include: (1) the assumption that the rural farm family annually consumes a large portion of 'home-grown meats' for their 70-year lifetime exposure; and (2) that biosolids are applied annually to 100% of the farmland used to produce all forage and grain crops used as animal feed.

The uptake slopes (UC_i) for forage crops was the limit of detection in several recent well conducted studies (O'Connor et al., 1990; Gan and Berthouex, 1994). The uptake slope for grain crops was assumed to be 0.1 that of forage crops due to the covering of grains by other plant tissues.

The absorption/retention of PCBs by livestock may differ between pure PCB chemicals, PCBs in feedstuffs, and PCBs in ingested biosolids; we are aware of no data which indicate that PCBs in crops will have different bioavailability than PCBs added to livestock diets to test absorption. Thus the UA factors reported by Fries (1991) were used in this calculation.

3.4. Risk from home garden food crops (Pathway 2; biosolids → soil → plant → human)

The production of food crops for human consumption on biosolids-amended soils might transfer some biosolids xenobiotics into diets. The Highly Exposed Individuals for this Pathway are home gardeners who grow a large fraction of their diet on biosolids-amended soil, for 70 years. Biosolids are presumed to be applied and mixed into the soil annually, and the soil PCB concentration reaches an equilibrium with PCB loss after 81 years of applications. The exposure is assumed to be at the maximum allowable equilibrium level for the whole 70-year consumption of garden crops grown on the treated soil. The Pathway 2-D&M limit is estimated by assuming a D&M product is applied annually, so no waiting period is allowed. In the Agricultural Use Pathway 2, 5 years are presumed to pass after the last annual biosolids application before garden crops are grown on the treated soil, after conversion of the site to residential use. Because Pathway 2-D&M estimates the highest possible exposure under Pathway 2, it is used for the example calculations. Exposure from Pathway 2-D&M is much lower than from Pathway 3-D&M or Pathway 5-Surface. Pathway 2-D&M will not practically limit use of biosolids with low PCB concentrations presently available.

The method of calculation of food chain transfer of toxic organics according to the Methodology (U.S. EPA, 1989a) (home garden scenario) uses several steps to complete the calculation. The first step is the calculation of daily allowed PCB intake (RIA) based on the cancer potency value (Eq. 3). One then sums the amount of each food group consumed times the slope of plant uptake for the compound times the fraction of food group grown on biosolids-amended soil. This value is divided into the RIA to produce the RLC, the soil PCB concentration which biosolids application may not be allowed to exceed.

From Eq. 3, we know the RIA is 0.909 μg PCB/day. The next step sums the amount of PCB ingested if FC (fraction of the diet) were grown on biosolids-amended soil, if DC (g dry wt. of foods)

Table 6
Calculation of potential transfer of biosolid-applied PCBs from soil to diets of HEIs through home garden crops

Food group	Food intake DC (g dry wt./day)	Uptake slope UC ($\mu g/g(\mu g/g)^{-1}$)	Fraction home-grown FC	($UC_i \cdot DC_i \cdot FC_i$)	
				μg/day	% Total
				μg/g soil dry wt.	
Potatoes	15.6	0.025 · carrot = 0.00188	0.45	0.0132	24.9
Leafy vegetables	1.97	≤0.0010	0.60	0.00118	2.2
Legume vegetables	8.75	0.001 · carrot = 0.000075	0.29[a]	0.00019	0.4
Root vegetables	1.60	0.53 · carrot = 0.03975	0.60	0.03816	72.1
Garden fruit	4.15	0.001 · carrot = 0.000075	0.60	0.00019	0.4
Total				0.05292	100.0

Uses appropriate UC slopes for biosolids-applied PCBs, food intakes (DC), etc., for the calculations for Pathway 2 home garden analysis for PCBs. The absolute slope used for unpeeled carrot was (0.075 μg/g dry wt. carrot) · [μg PCB/g soil dry wt.]$^{-1}$; from O'Connor et al., 1990), after review of PCB uptake by carrot and lettuce from biosolids-PCB treated soil. Other data are best judgement values because slopes are not generally available from properly conducted field experiments with biosolids-applied PCBs.

[a]0.17 for dried legume vegetables (8.412 g dry wt./day) and 0.60 for fresh legume vegetables (3.340 g dry wt./day) gives 0.29 for weighted total legume vegetables.

were consumed daily on average for 70 years, and if UC were the transfer coefficient from soil to each food group (μg PCB/g dry crop per [μg PCB/g dry soil]) grown in the garden.

We conclude that calculation of the $\Sigma(UC_i \cdot DC_i \cdot FC_i)$ for PCBs should rely on the values in Table 6. The DC for garden vegetables lifetime ingestion estimates were shown in Table 3 (lifetime diet model). Table 6 combines the lifetime average food intakes, appropriate crop PCB uptake slopes, and the Highly Exposed Individual fraction of diet from the final U.S.-EPA 503 Rule (U.S.-EPA, 1992) to calculate the $\Sigma(UC_i \cdot DC_i \cdot FC_i)$ for PCBs. Under conditions appropriate for study of PCB uptake from soils amended with biosolids, carrot had measurable levels of PCBs while other crops had non-detectable levels (O'Connor et al., 1990). U.S.-EPA used the 0.001 default PCB uptake slopes for each crop group and estimated the allowable soil maximum PCB concentration to be 67 mg/kg soil-biosolid mixture. We conclude that the important difference between crop types in transfer of biosolid-applied PCBs to edible crop parts is better reflected by the approach in Table 6. Carrot accounts for 53% of the dry matter of the root vegetable group according to Chaney et al. (1987). Thus, the overall root vegetable group is shown as equal to 53% of the uptake slope of unpeeled carrot; PCB uptake by other root vegetables from soils is much lower than found for unpeeled carrots (O'Connor et al., 1991). Although most people peel carrots before consuming them, there is no assurance that carrots will be peeled. Therefore, the uptake slope for unpeeled carrots was used. O'Connor et al. (1990) found little PCB entry to carrot deeper than the normal peel depth. As noted above, transfer of PCBs from biosolids-amended soils to edible parts of plants is nearly exclusively by volatilization of PCBs from the soil particles and capture by lipophilic surfaces of the crop.

Using the appropriate ($UC_i \cdot DC_i \cdot FC_i$) for pathway 2, the estimated RLC is:

$$RLC = \frac{0.909\ \mu g\ PCB/day}{0.05292\ \mu g\ PCB/day\ (\mu g\ PCB/g\ soil\ dry\ wt.)^{-1}} = 17.2\ \mu g\ PCB/g\ dry\ soil \quad (13)$$

This is the maximum allowed concentration of PCB in soil at any time that garden vegetables are

grown on the soil. This can be converted to an annual biosolids-PCB application (RP_A) as done above in Pathway 5-Mixed With Soil, and Pathway 4. The RP_A would be $17.2 \cdot 2(14.9)^{-1} = 2.31$ kg PCB/ha/year. Biosolids applied at 10 t/ha/year could contain 231 mg PCB/kg dry wt.

It is important to consider the hidden safety factors still present in this Pathway algorithm. We believe that very few and possibly no individuals would come close to the high exposure conditions defined for the Highly Exposed Individuals in this Pathway. The following list shows the aspects of this risk assessment which provide hidden safety factors against risk from biosolids-PCBs through the home garden pathway:

1. Most people peel carrots before consumption which would greatly reduce potential transfer of applied PCBs to diets.
2. Biosolids now have very low PCB concentrations in the U.S.
3. It is rare for an individual to have such a high fraction of 'home-grown' garden foods for 70 continuous years.
4. The model would require 151 annual applications of fertilizer rates of biosolids (81 years to reach equilibrium with disappearance and 70-year exposure period); thus it would be extremely unlikely that individuals would ever attain the model levels of exposure.
5. It is generally believed that PCBs are biodegraded or volatilized more rapidly than the 10-year half life used in these calculations. For example, in the study by Gan and Berthouex (1994), the lower chlorinated congeners had far shorter half lives. Although some more highly chlorinated congeners are usually more persistent, they also have lower volatility and are not as readily transferred even to carrots as the transfer measurements of O'Connor et al. (1990).
6. Most previous estimates used plant uptake from 'spiked' soils to which pure chemical PCBs were applied immediately before the crop was grown. The O'Connor et al. (1990) study shows that method greatly overestimates potential PCB uptake by crops.

3.5. Risks from biosolids PCBs to wildlife: Pathway 10

This Pathway estimates biosolids PCBs limits needed to protect wildlife. The fundamental finding which required regulatory prohibition of PCBs and DDT was population declines in raptor birds due to excessive body burden of lipophilic xenobiotics which caused eggshell thinning. These species are at the top of food webs which bioaccumulate lipophilic organics, especially the bioaccumulation of DDT and PCBs in aquatic food-chains where several trophic levels of bioaccumulation can occur to increase the concentrations of the lipophilic organics in the fish species actually consumed by the raptor birds.

The 503 Rule was to protect all kinds of wildlife which might be exposed to PCBs as a result of land application of biosolids on cropland. Thus, the raptor birds which are exposed through aquatic food webs do not appear to be the appropriate Highly Exposed Individuals for limiting PCBs in biosolids applied to agricultural land because of the poor connection between biosolids application and potential adverse effects of the PCBs on these species. If the fish consumed by raptor species have PCB levels which integrate the erosion or deposition of PCBs into lakes, and the biosolids-amended fraction of total land area which may contribute to a lake is very small, then the exposures would be far lower than the worst case in which all land is contaminated surrounding the lake. Biosolids application areas usually are required to be at least some specific setback distance from surface water channels or water bodies; these setbacks substantially reduce the potential for movement (erosion) of applied PCBs (adsorbed to soil particles) into lakes.

Instead, the Highly Exposed Individuals for Pathway 10 are shrews, moles, or other mammals with a small range, and which actively ingest earthworms as part of their diet during their lives. Beyer and Stafford (1992) and Eisler (1976) have discussed wildlife exposures to PCBs, and sensitivity of different wildlife species to PCBs. Because earthworms and other soil biota whose skins have lipophilic characteristics can bioaccumulate PCBs

to higher concentrations than present in the biosolid-amended soil in which they live, and earthworm-consuming small mammals often live their whole life in an area which could all be amended with biosolids (smaller than an agricultural field), this food-chain is much more direct and sensitive to soil PCBs than other wildlife species such as the raptor birds (these species also have a large range, such that only part of the food they consume would be expected to have exposure to biosolids amended soils). Other small mammals might live their life within a biosolids-amended field, but they consume plant leaves or seeds, or above-ground insects which do not have the extent of PCB bioaccumulation found for earthworms (Davis et al., 1981; Forsyth and Peterle, 1984).

The tolerable concentration of PCBs in the whole diet of small mammals was obtained from the literature. Eisler (1986) and Peakall (1986) reviewed the literature on wildlife toxicity from PCBs, and this was updated by Beyer and Stafford (1992). The lowest 'No Observed Adverse Effect Level [NOAEL]' for PCB toxicity to mammals and birds is 5 mg/kg in chickens (Lillie et al., 1975). Although shrews and moles have not been studied, this NOAEL is considered appropriate for earthworm-consuming wildlife. Chickens were more sensitive to dietary PCBs than were mice or rats. On the other hand, these tests were conducted with dietary PCBs, not with PCBs injected into the eggs at the most sensitive stage of fetal development as was done in studies of other xenobiotics.

As in the above Pathways, we needed to make the pathway calculations as realistic as available data allow. Thus, in contrast with the original EPA pathway for wildlife in which ducks were presumed to consume earthworms as 100% of their diet (U.S.-EPA, 1989c), the exposed wildlife species is no longer presumed to consume only earthworms for their lifetime. Ingestion of earthworms as 100% of diet might be appropriate for consideration of acute exposure of wildlife species, but not for chronic exposure. After consideration of maximum chronic consumption of earthworms by wildlife (see review by MacDonald, 1983), 33% of diet was selected as the maximum chronic fraction of dietary earthworms. It should be recognized that the 45% soil content of earthworms as ingested in the field makes them have a much higher dry matter percentage than other soil biota and insects consumed by species such as shrews; thus, earthworms comprise a higher fraction of the diet wet matter than the diet dry matter upon which these calculations are based. Available information indicates that earthworms comprising 33% of the lifetime diet dry matter of shrews is a high-end estimate. Thus, the maximum allowed diet PCB level (5 μg PCB/g diet dry wt.) is multiplied by 3 to calculate the 15 mg PCB/kg dry wt. allowed in whole earthworms which comprise 33% of the diet of HEI wildlife.

Next, the bioaccumulation of PCB or similar compounds in earthworms, compared to soil in which they live, must be obtained from field studies with soil/biosolids PCBs. This transfer coefficient is available from studies published by Beyer and Stafford (1992), Beyer and Krynitsky (1989), Diercxsens et al. (1985), Kreis et al. (1987), Marquenie et al. (1987) and Tarradellas et al. (1982):

For earthworms with soil in their digestive system (as they would normally be consumed in the field), the bioaccumulation factor is about 3.69 mg PCB/kg earthworm-dry wt. (non-purged) per 1 mg PCB/kg dry soil (geometric mean for 1.8, 3.4, 3.3, 4.9, 5.0, and 5.1 [arithmetic mean = 3.9]) from the above noted studies.

$$\text{Thus } 15 \text{ mg PCB/kg worm fresh wt./3.69} = 4.06 \text{ mg PCB/kg soil dry wt.} = 8.12 \text{ kg PCB/ha} \quad (14)$$

For organics, the biodegradation or other dissipation must be considered. Therefore, the allowed loading must be considered the maximum concentration which may be reached at any time rather than the cumulative application. Equal annual applications are usually estimated based on half-life of the organic compound in soil, although the regulation is met as long as the maximum soil concentration needed to protect wildlife is not exceeded.

For biosolids mixed with soil, the annual application of PCB which will not exceed the maximum allowed 4.06 mg PCB/kg dry wt. soil [when equilibrium is reached between annual application and annual dissipation] is calculated using the result from Eq. 7:

$$= (4.06 \text{ mg PCB/kg soil-biosolids mixture dry wt.}) \cdot 2000/1000 \cdot [14.9]^{-1} \quad (15)$$

$$= 0.545 \text{ kg PCB/ha} \cdot \text{year}.$$

If one assumes 10 t of a biosolids would be applied per year as a N fertilizer, the concentration of PCB in that biosolids could be:

$$\frac{0.545 \text{ kg PCB}}{\text{ha} \cdot \text{year}} \cdot \frac{1 \text{ ha} \cdot \text{year}}{10 \text{ t biosolids dry wt.}}$$

$$= 54.5 \text{ mg PCB/kg biosolids dry wt.} \quad (16)$$

Because the concentration of PCBs in modern biosolids is generally < 0.10 mg/kg dry wt., wildlife are highly protected from PCBs in biosolids applied to land.

4. Summary of Pathway limitations

Table 7 summarizes of the calculated allowed biosolids PCB concentrations, or the annual applications of biosolid-PCBs which would have been allowed under the 503 Rule. These limitations on PCBs in biosolids were calculated using the appropriate algorithms and transfer coefficients discussed in this paper.

Although we have previously stressed the remarkable errors in the Proposed 503 Rule (U.S.-EPA, 1989c) (Page et al., 1989), the Proposed Rule was abandoned and the errors were corrected before the Rule was promulgated. EPA requested that an expert committee led by Dr J.A. Ryan work to identify the correct database for development of limits for biosolids applications on land. That committee included Drs Chaney and O'Connor. The information presented in this paper is more complete than provided in the TSD, and organized to show details of the calculations for these most limiting pathways for biosolids PCBs; the Pathway calculations are those which should have been in the final 503 Rule (U.S.-EPA, 1993) and TSD (U.S.-EPA, 1992), but were not corrected by EPA staff.

During development of the final 503 Rule, all participants sought to assure that Highly Exposed Individuals would not be at risk from PCBs and other contaminants in biosolids applied to cropland, and that farmers who allowed beneficial use of biosolids on their land would not suffer future limits on marketing of their produce. The present more realistic risk assessment methods, coupled with conservative protections provided by the de-

Table 7
Comparison of PCB application limits for each pathway from the 503 Proposed Rule (U.S. EPA, 1989b) with the corrected versions based on EPA (U.S. EPA, 1993a,b)

Pathway	Limit units	Limit value	Concentration (annual) (mg/kg biosolids dry wt.)
1 (U.S.-EPA)	mg/kg soil maximum	290.0	3700
	kg/ha · year	37	
2	mg/kg soil maximum	17.2	
	kg/ha · year	2.3	231
2 (U.S.-EPA)	mg/kg soil maximum	67.1[a]	
	kg/ha · year	8.5	
3	mg/kg biosolids dry wt.	14	14
4	mg/kg soil maximum	18	
	kg/ha · year	2.4	246
4 (U.S.-EPA)	kg/ha · year	4.6	460
5-Surface	mg/kg biosolids dry wt.	2.2	2.23
5-Mixed	mg/kg soil maximum	2.2	
	kg/ha · year	0.299	29.9
5 (U.S.-EPA)	mg/kg biosolids dry wt.	4.6	4.6
10	mg/kg soil maximum	4.06	
	kg/ha · year	0.545	54.5

Units are changed in some corrected versions. Error in actual compound residue in plant per unit soil residue was corrected compared to the proposed 503 Rule (U.S. EPA, 1989). The last column shows the PCBs concentrations which would have been required to meet each Pathway annual application rate for PCBs if biosolids were applied at 10 dry t/ha per year.

[a]EPA used default assumption for their calculation which differed from those we judge to be most valid; carrot carries most PCB into garden foods in human diets, not potatoes as calculated by EPA.

finition of the exposed population, and the selection of highly exposed individuals upon which to base the final limits, provide the protections required by law and by common sense. Pretreatment and cessation of PCB use in industry have allowed PCB concentrations in biosolids to fall to very low levels. Thus, although the limits under 503 were initially designed to protect Highly Exposed Individuals at the level of 1 in 10^4 increase in lifetime cancer risk, the concentrations which would have been used to set limits for 503 (Table 7) are far higher than levels currently found in biosolids (typically <0.10 mg/kg dry wt.). Further, the definition of the Highly Exposed Individuals remains very conservative, in that it is hard to find individuals who consume a substantial fraction of their lifetime ingestion of meat and dairy products produced with exposure to biosolids-amended soils under the conditions of the Pathways. We believe it is clear that the rule provides at least 1 in 10^7 protection levels for the highly exposed individuals, and correspondingly greater protection for average consumers who have little chance to achieve significant exposure to biosolids-applied PCBs.

5. Other considerations

Other pathways were also evaluated by EPA, and this information is reported in the Technical Support Document for the 503 Rule. These include human exposure to biosolids-applied PCBs which have moved into surface or groundwater, volatilization of PCBs into air chronically inhaled by humans, resuspended dust from treated fields, etc. However, each of these caused so much lower human exposure than the two more limiting Pathways (human consumption of home-grown meats from livestock grazing pastures which receive surface-applied biosolids; and children ingesting biosolid products), that a detailed evaluation is not worth the work required to assemble the data.

The possible bioaccumulation of PCBs in fish which live in lakes with significant erosion of biosolids or biosolid-amended soils could provide exposures of importance because if there are no limitations on erosion or runoff of applied biosolids, the amount reaching a water body could be appreciable. EPA made several policy decisions regarding the fraction of a watershed which might receive biosolids applications, and the potential for erosion to carry soil long distances from the treated soils to the surface water channels. Because it would be nearly impossible to estimate the possible human or wildlife exposure with any reliability unless site specific characteristics were taken into consideration, EPA decided to make a policy decision regarding the setback distance between land to which biosolids may be applied, and the entry to surface waters. This 'risk management' decision provided a great reduction in potential transfer of biosolids PCBs into lake sediments. Enough data are available to make estimates of the concentrations of PCBs in lake waters in relation to concentrations in sediments, and estimates for bioaccumulation from water levels to levels in higher trophic level fish. But the maximal lifetime exposure of individuals to fish from a single source had poor reliability. Subsistence fisherpersons might be imagined to consume substantial amounts of fish on some days, but inadequate data are available to estimate their lifetime consumption of these fish.

Because the exposure of fish has caused human exposures in the U.S., and public warnings about fish consumption have been issued at numerous locations where massive PCB contamination of streams and lakes occurred in previous decades, there has been concern about land application of biosolids which contain measurable levels of PCBs. However, recent studies of contaminated lakes have shown that lakes are releasing PCBs to the air over time (Hornbuckle et al., 1993), and the airborne PCBs are either photodegraded or deposited onto soils around the globe. Thus, use of modern biosolids with low concentrations of PCBs and other persistent chlorinated hydrocarbons comprises very low risk to aquatic wildlife or subsistence fisherpersons. The combination of the low concentrations of PCBs in biosolids and the required setback distances and use of erosion control practices, provide high protection against these risks.

During the evaluation of critical factors in Pathways 4 and 5, we noted that the 'home-grown'

fraction of dietary meat and milk products consumed by the Highly Exposed Individuals had declined since the previous estimate. A food consumption survey from the 1980s indicates that the 'home-grown' fraction of meat and dairy products consumed by Highly Exposed Individuals may presently have fallen to about 10% of meat and 3% of milk products. The values in this paper are based on 'home-grown' livestock grazed on biosolids-amended pastures supplying 46% of lifetime meat and 40% of lifetime milk products to the HEI. Thus, a further hidden safety factor of about 5- to 10-fold remains built into the Pathways involving consumption of livestock tissues. Comparison of the concentrations of PCBs which must not be exceeded in the present calculations with the Food and Drug Administration limits on PCBs in foods (U.S.-FDA, 1979) indicates close agreement, so that farm products should be expected to readily meet these marketplace limits even under the extreme exposure cases used in the Pathway Risk Assessment Methodology. It is clear that HEIs and consumers have great protection from PCBs entering the food-chain as a result of use of biosolids on cropland.

6. Research needs to improve regulations for xenobiotics in land-applied biosolids

Our understanding of the fate and effects of the myriad of organic compounds which enter wastewater remains incomplete. What compounds end up as persistent residues in biosolids-amended soils? What are the physico-chemical processes which allow lipophilic organics to reach stronger adsorption sites in soil or biosolids organic matter? The mechanism of toxicity and fuller demonstration of cancer induction by exposure to specific compounds remains a general need for most organics.

Transfers from biosolids to livestock or children which ingest biosolids-amended-soils or biosolids are very important, but the transfer coefficients have been measured for only a few compounds. The bioaccumulation of lipophilic xenobiotics in earthworms and other soil macrofauna is only partially characterized. Both the nature of the wildlife food chains starting with soil organisms, and the bioavailability of xenobiotics bioaccumulated by earthworms, etc., should be experimentally characterized in more detail.

In the aquatic pathways, the potential for xenobiotics and methyl-mercury bioaccumulation in food webs has substantial uncertainty. The erosion processes which allow substantial movement of plow layer soils to streams can be better modelled, as can the localization of the deposition of the suspended sediments from the eroded soils. Because persistent lipophilic xenobiotics are adsorbed strongly on fine particles of soil organic matter, the eroded sediment-bound organics are deposited only after water movement has slowed sufficiently, such as in lakes or reservoirs. The basis for the highly variable biomagnification of xenobiotic residues in different lakes needs to be characterized. Is the dilution of dissolved PCBs by increased biomass the total basis for lower biomagnification estimates in some lakes than others?

For PCBs, perhaps the weakest Pathway estimation is that to protect wildlife. Even though available data strongly support the Pathway as calculated in this paper, other wildlife food webs need to be quantitatively examined to assure that all species are protected. This requires valid consideration of the small fraction of land area required for use of biosolids as fertilizer, which reduces the potential risk to carnivores at the top of the food web because of their wider ranges for obtaining food.

Another area of great opportunity and research need is the use of composting to biodegrade xenobiotic organics. A number of factors appear to allow applied composts to remediate xenobiotics more effectively than other bio-remediation approaches. Composts provide balanced soil fertility and microbial nutrition. The carbon sources in composts provide substrates to support growth and metabolism by the microbes. Composts serve as a diverse microbial inoculum. And the organic matter in composts can sorb the xenobiotics, thereby reducing their volatilization, leaching, or toxicity in contaminated soils yet allowing the microbes in the amended soil to biodegrade the compounds. Although composting at thermophilic temperatures reduces microbial diversity and has not been found to achieve biodegradation of a

number of xenobiotics studied, recent success with TNT biodegradation offers hope that improved management of composting conditions may increase the utility of composting of xenobiotics. Studies have shown that application of mature compost is much more effective in biodegradation of soil xenobiotics than addition of microbial inocula or fertilizers. The combination of factors noted above show the present theory of why composts are so effective in biodegrading xenobiotics in soil, but more research will be required to provide the information needed to make this technology as effective as practicable.

Development of analytical methods for xenobiotics in biosolids with improved detection limits, reduced costs, and simple equipment requirements would provide greater ability to provide regular monitoring of residues in biosolids.

Besides these research needs, nearly all of which would better identify potential risks from xenobiotics in biosolids, research is also needed to characterize the hidden safety factors in models for risk assessment. In nearly every case examined during development of the PCB limitations for the 503 Rule, risk was greatly overestimated by the methods initially used. When appropriate data were obtained from the literature rather than using default assumptions, many sources of overestimation of risk were identified. But other overestimation came from the assumed characteristics of the Highly Exposed Individuals. Research is also needed to identify the number of individuals who may actually achieve the modelled high exposures to biosolids contaminants by these HEIs, and to identify regulatory approaches to reduce unnecessary exposures while causing minimum interference with beneficial use of biosolids in agriculture and land reclamation.

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