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**Environmental Energy Technologies Division**

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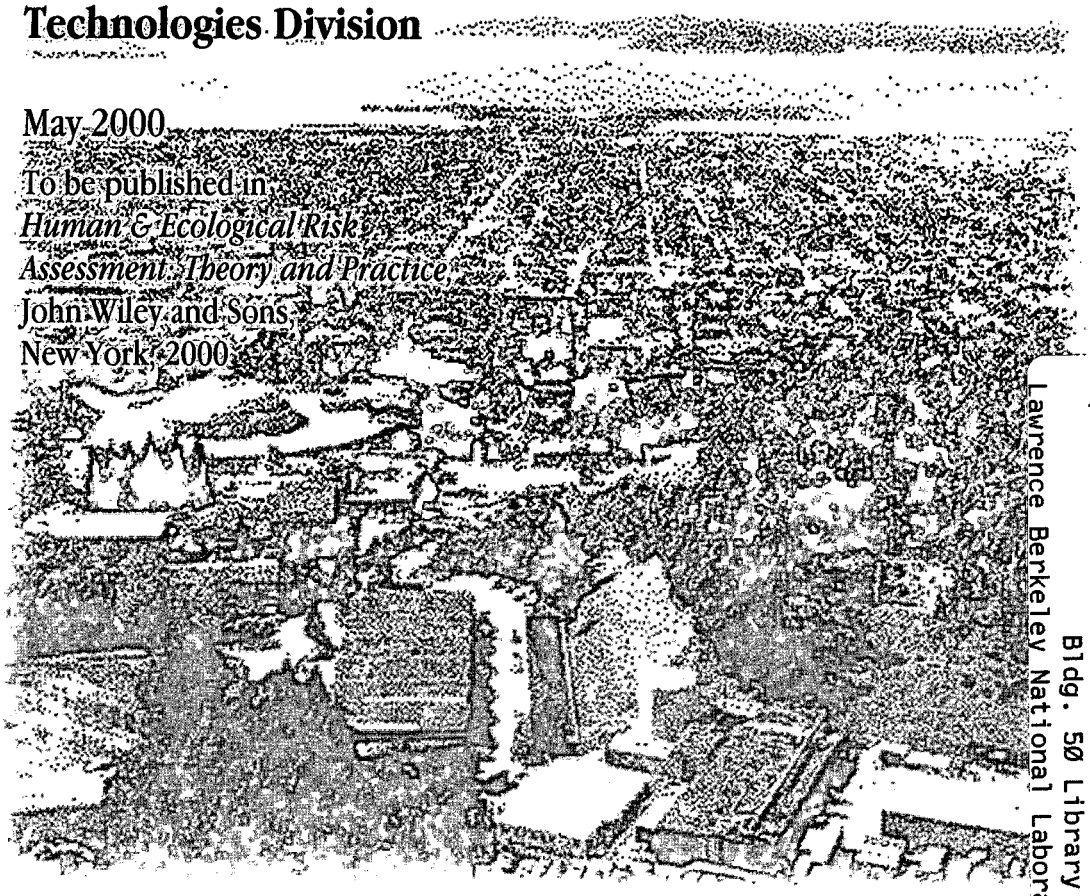

## DISCLAIMER

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## 1 - Introduction

The environmental decision making community is now confronting the potential adverse health and ecological impacts of persistent chemicals such as metals and organic pollutants. The significance of these chemicals is attributed to their potential for health and ecological impacts at low concentrations, coupled with their ability to accumulate and persist in multiple environmental media. Their persistence allows adequate time for environmental interactions and long range transport to populations far from the source. Persistent organic pollutants (POPs) originate from a broad range of human activities, including combustion for energy production and transportation, industrial processes, and agricultural uses of pesticides.

Environmental impacts have often been classified by the medium to which the pollutants are released, such as air, water, or soil. However, many POPs partition into multiple environmental media. Air emissions can result in contaminated soil, and contaminated soil can result in air pollution through the mass exchange between the air and soil.

Another limitation for addressing POPs is that pollution is often regulated as a local problem. However, such an approach is not adequate for many POPs because they can be transported long distances in the environment. When a chemical travels long distances, it can cross local regulatory boundaries. In this sense, the scientific or regulatory community has not

adequately addressed human exposure to POPs through multi-regional, multimedia exposure scenarios.

The objective of this chapter is to present a methodology aimed at answering the following questions:

- What measures can we use to determine whether or not a chemical is a POP with the potential for long range transport?
- How do we properly quantify the population based potential dose resulting from a pollutant with the potential for long range transport?

To answer the first question, we develop a framework to quantify the characteristic time ($\tau$) and characteristic travel distance (*CTD*) for semi-volatile POPs in a multimedia environment. Characteristic time is a measure of temporal persistence; i.e. how long a chemical is likely to remain in a multimedia environment after being released to any compartment of that environment. The *CTD,* on the other hand, is a measure of how far a chemical is likely to travel in the multimedia environment and defines whether a chemical will have a local, regional, or global scale impact.

A multimedia model is used here to incorporate chemical exchange among air, soil, water, and plants as well as chemical degradation in each compartment. This information is used to quantify the steady state spread of pollutants between media.

To answer the second question, we present a conceptual model to characterize the population based potential dose by taking into consideration the *CTD* and the spatially varying population density. If a chemical travels long distances in the environment, more people are exposed to the chemical, although at lower concentrations. If the dose-respose model is assumed linear, then when more people are exposed to the chemical, there is a higher chance of someone experiencing an adverse effect from exposure to the chemical than if only a small number of people are exposed to a chemical. We compare the population based potential dose calculated with this conceptual model to the potential dose calculated using only the locally exposed

population.

The methodologies presented here are appropriate for continuous, large non-point atmospheric emissions of organic chemicals, such as the collective emissions from a large urban area including industrial facilities, and combustion emissions for transportation, heating, and electrical generation. The methodologies are appropriate for ubiquitous chemicals with long atmospheric half- lives (several hours or days); a relatively high value for $K_{ow}$, such that partitioning into vegetation and soil is significant ($K_{ow} > 1 \times 10^6$); and a relatively high vapor pressure (VP) such that there is some partitioning from particles to the gas phase of the atmosphere (VP > $1 \times 10^{-10}$ Pa).

Defining $\tau$ and *CTD* of a chemical will give insight regarding the appropriate scale for regulation. The measure of population based potential dose can be used by decision makers for a variety of analyses to decide what chemical to use for a process or by policy makers to determine appropriate regulations when evaluating a new chemical as it is being introduced. Potential applications for these measures include risk assessment, pollution prevention assessment, health effects studies, pollutant mass balance studies, life cycle analyses, sustainability evaluation, and regulatory impact studies. Regulators can then focus on the chemicals with the highest population based potential dose and corresponding health risk, thereby providing more effective ways to regulate these chemicals.

Two case studies are presented, one for a persistent chemical, 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD), and one for a non-persistent chemical, benzo[a]pyrene. We will calculate $\tau$, *CTD* and population based potential dose and discuss how the results differ when evaluating a persistent and non-persistent chemical.

## 1.1 Review of Environmental Modeling

Historically, environmental contamination has been viewed primarily as a local problem, generally affecting one environmental medium, and regulated accordingly (1). This approach is changing as an increasing number of examples challenge this view. Traces of several POPs have been found in the Arctic although there are no sources of organic pollution there (2, 3). This indicates global scale pollution. Pesticides used in California's central valley have resulted in contamination in the Sierra Nevada Mountain Range (4, 5), an example of both a multimedia problem and long range transport. The chemicals DDT and various PCB's, whose use has been either banned or severely restricted in the US for over 20 years, are currently found in the sediments of the Great Lakes, illustrating persistence (6, 7). The foregoing are all examples of multi-dimensional problems that are difficult to understand using current models and stimulated the research presented in this chapter.

The EPA Science Advisory Board has recommended that reliable, multimedia models would overcome a significant barrier to risk assessment for evaluating chemicals that partition into multiple environmental media (8). At present, two types of multimedia transport models are often used: single region, multimedia models and simplified global multimedia models. Fugacity based models that include different compartments, such as air, water, soil (one or more layers), vegetation, and sediment have been developed by Mackay (9) and McKone (10, 11). These models are comprehensive with respect to the environmental compartments within the model, yet lack spatial resolution.

Multimedia models that include spatial resolution are also available. For example, the SimpleBox model (12) has a nested set of small-, medium- and large-scale "unit worlds". Global scale models have also been developed to determine the distance chemicals are likely to travel in

the environment and to look at temperature dependent trends (13, 14). These models do not include human exposure and thus cannot be used for exposure and risk assessment.

### 1.1 Review of Human Exposure and Dose

Quantifying the potential dose per person has been an essential element in the field of risk assessment since its inception. CalTOX, a fugacity based multimedia model, calculates the dose from multiple environmental media using twenty-three exposure pathways (10). All of these pathways are directly incorporated into the exposure model used in this chapter. Most often, these calculations are carried out for an individual. Sometimes, risk to a population is calculated by multiplying the risk to a representative individual in a given group by the number of individuals in that group. Example calculations of population based dose can be found in case studies by both Thompson and Evans (15) and Webster and Connett (16). A study of global chemicals by Travis and Hester (17) calculated the background cancer risk from eleven global pollutants, based on measured background concentrations, demonstrating a need for research quantifying risk from chemicals with a potential for long range transport.

Starting in 1988, the EPA began requiring companies to report the amount of toxic chemicals released from their facilities through the Toxic Release Inventories program. The simplest method to compare these releases considers the quantity released and the toxicity of the chemical (18). More advanced methods also include critical factors such as persistence, pollutant fate, or exposure factors (19, 20). It was demonstrated that more advanced methods yield significantly different results than simpler methods. The main drawbacks cited for the advanced methods are that they require more data which are often unavailable (19) and that increasing the complexity also increases the uncertainty (20).

## 2. Methods

We first define the multimedia model used to determine the concentrations in the relevant environmental media. The characteristic time is defined and then analytically derived using a multimedia model. We then present a methodology for determining the *CTD* for airborne semi-volatile organic pollutants. A conceptual model for calculating the population based potential dose is also presented. This requires that we determine the exposure from multiple pathways to the multimedia environment.

### 2.1 Defining the Multimedia Model

We use a model with air, surface water, vegetation, and two soil compartments. A schematic of the model used as shown in Figure 1. All phases, (i.e. air, water, solids) in an environmental compartment are assumed to be in chemical equilibrium. From the phase composition of each compartment, the fugacity capacity (e.g. the chemical concentration per unit chemical fugacity) can be defined. The fugacity capacity of each compartment can be found in Reference 21.

The steady state concentration in each environmental compartment is determined from the interactions among the environmental compartments and the decay rate in each compartment. Many of the model compartments and processes needed to define the interactions between compartments have been taken from the CalTOX model (11, 22, 23). We use a fugacity-based model, a common approach for describing partitioning in multimedia systems (9, 24).

The chemical exchange between compartments is determined by the transfer coefficients, or "T" values. These values define the rate of mass transfer per unit mass inventory of the chemical in the compartment from which the chemical is transferred. Table 1 lists all of the transport processes used in the model. The equations for the transfer rates between compartments

can be found in Reference 21. To calculate the flux from one compartment to another, the mass transfer coefficient is multiplied by the mass in the compartment from which it originates. In the case of diffusion, the net flux is calculated from the gross flux in each direction.

Mass transformation is modeled in each compartment based on pseudo-first-order rate constants taken from experimental or field data. This data is often scarce or highly uncertain and reported values often range over orders of magnitude, especially for vegetation (25-27). Transformation in air can include reactions with OH radicals and photodegradation. Transformation rates in soil differ among soil types, and include photodegradation on surface soils and degradation by microbial action in deeper soils. Transformation in vegetation can be rapid and includes photodegradation on the leaf surface (28, 29).

Estimates of parameter values can rarely be characterized accurately by a single value, due to uncertainty in determining a parameter value, spatial variability, or both. A log-normal probability distribution is assigned to each parameter such that the range conforms to the environmental limits of the selected parameters. The parameter values and associated coefficient of variation (the standard deviation divided by the mean value) used in the case studies are found in Table 2.

### 2.2 General Formulation of Characteristic Time

Characteristic time is a measure of temporal persistence; i.e. how likely a chemical pollutant is to remain in a multimedia environment after being released to any compartment of that environment. The characteristic time can be determined by finding the overall decay rate of the chemical in a closed, defined landscape system. Because the decay rates in each environmental

media can differ significantly for a given chemical, determining the $\tau$ in the environment requires knowing both the mass distribution among environmental media and the media-specific half-lives.

The instantaneous mean life or average life expectancy of a molecule in an environmental compartment, $\tau$, is defined as the inverse of the decay rate in the compartment (30). We will refer to this as $\tau$ for that compartment:

$$\tau = \frac{1}{k} \qquad (1)$$

where $k$ is the decay rate, representing radioactive decay or chemical reactions that irreversibly remove the chemical from the system. In a two compartment system, such as in Figure 2, the effective decay rate is mass averaged between the two compartments, leading to the following instantaneous overall decay rate (31):

$$k_{overall} = \frac{M_1 k_1 + M_2 k_2}{M_1 + M_2} \qquad (2)$$

where $M_i$ is the mass in compartment $i$ (kg).

We prefer steady state calculation methods to inform decisions regarding chemical impacts in the environment (31). The steady state distribution accounts for location of the source and for advective phase-transfer processes while retaining sufficient simplicity to complete calculations in a tractable form, such as a spreadsheet, useful if the output of the analysis is to be utilized as a factor in decision making or subjected to an uncertainty analysis. Methods have been developed to classification trees to determine if a chemical is persistent or non-persistent using this measure for characteristic time based on the input properties of the chemical (32). A similar formulation has been developed by Webster et al. (33).

If more than one environmental compartment influences the overall decay rate in the environment, the effective decay rate equation can be expanded to include any number of

environmental compartments. For the multimedia system used in this paper, we use the following equation:

$$k_{overall} = \frac{M_a k_a + M_p k_p + M_g k_g + M_s k_s + M_w k_w}{M_a + M_p + M_g + M_s + M_w} \quad (4)$$

where the indices *a, p, g, s,* and *w* are for the air, plant, ground surface soil, root zone soil, and surface water compartments, respectively.

### 2.3 General Formulation of Characteristic Travel Distance

The *CTD* is a measure of how far a chemical is likely to travel in the multimedia environment and is derived analytically by following the movement of a particular mass of pollutant in a moving Lagrangian air cell as it interacts with the non-moving compartments of the environment (i.e. vegetation and soil). We begin with a two compartment system, a moving (i.e. air) compartment and a non-moving compartment (i.e. vegetation and soil). This system, illustrated in Figure 3, can be thought of as a simple representation of the more complex environmental model shown in Figure 1.

We calculate the change in chemical mass in a moving Lagrangian air cell as the cell travels in a one dimensional band away from the area of release (the source region). The concentration in air is reduced with distance based on degradation in air, transfer to and subsequent degradation in other media. We chose the *CTD* as the distance from the source at which the concentration is reduced by 63% (i.e. reduced to 1/e of the original concentration). By using a continuous differential Lagrangian structure, the results are obtained in closed analytic form as opposed to a more complex numerical form.

We make the following assumptions: the source term is continuous, the system has reached steady state; there is no lateral air dispersion; the long-term average wind pattern can be

represented by an equivalent steady wind rate in one direction; the landscape properties do not vary spatially (or can be spatially averaged); and the atmospheric mixing layer height is constant. We believe these assumptions are justified when considering a large continuous area source, such as the collective sources from a large urban or agricultural region.

The Lagrangian cell represents a small portion of the continuous stream of a pollutant flowing from the source at an average windspeed of u (m/d). At steady state, neither the moving nor the non-moving phases are accumulating mass with time at a particular location. The mass transferred to the non-moving phase is equal to the mass decayed in that phase. Additionally, the ratio of the mass in the ground to the mass in the air is spatially independent.

In the Lagrangian model for the airborne pollutant, we balance the time rate of change of mass with both the decay in the moving phase and the net transfer to the non-moving phase. We define the effective decay rate, $k_{effective}$ (1/s) as:

$$k_{effective} = \frac{M_1 k_1 + M_2 k_2}{M_1} \qquad (5)$$

The effective decay rate is essentially the mass in each compartment multiplied by the decay rate in the corresponding compartment divided by the mass in the moving phase. Solving for the concentration profile as one moves away from the source yields the following result:

$$C_1(x,u) = C_1(0) e^{\frac{-k_{effective}}{u} x} = C_1(0) e^{-x/CTD} \qquad (6)$$

We define the *CTD* to normalize the distance from the source as $CTD = u/k_{effective}$. The characteristic travel distance is the same for the moving and non-moving comparments since the ratio between the concentrations is spatially independent. This implies that the concentration decreases at the same rate with distance from the source in both compartments. The complete derivation can be found in Reference 21.

Again, if multiple environmental compartments influence the *CTD*, it is necessary to include these compartments in the calculation. We expand the definition of *CTD* to the multi-compartment system shown in Figure 1:

$$k_{effective} = \frac{M_a k_a + M_p k_p + M_g k_g + M_s k_s + M_w k_w}{M_a} \qquad (7)$$

The steady state mass in all of the compartments is derived in Reference 21.

This method for calculating the *CTD* has also been applied to other multimedia models (34). It is interesting to note that the *CTD* is not necessarily well correlated to $\tau$. A chemical may be persistent but not have a long travel distance if it primarily partitions into the non-moving phases of the environment (35).

## 2.4 Population Based Potential Dose

We develop a conceptual model for calculating the population based potential dose that incorporates the *CTD* of a particular chemical and the spatially dependent population density. Humans are exposed to chemicals in the environment through multiple pathways. Exposure is characterized by route of entry as inhalation, ingestion, or dermal uptake. Inhalation exposure includes contact with both indoor and outdoor air. The ingestion pathways include tap water consumption; incidental soil ingestion; and intake of fruits, vegetables, grains, and animal products, such as meat, poultry, eggs, and dairy. The dermal route includes exposure through contaminated water from bathing and recreation, as well as from soil on the skin. The pathways are summarized in Table 3. Potential dose is calculated from the contact rate with the exposure media and the

chemical concentrations in these exposure media (i.e. tap water, indoor air, etc.)[1].

For each exposure pathway shown in Table 3, the potential dose is calculated from the concentration in the corresponding environmental medium, the relationship between the exposure medium concentration and environmental medium concentration, intake rate, body weight, activity patterns, and exposure duration as (36):

$$ADD = C_{env} \times R \times \frac{CR}{BW} \times \frac{ED \times EF}{AT} \qquad (8)$$

where *ADD is the a*verage daily dose of chemical via exposure route (mg/kg/day), $C_{env}$ is the chemical concentration in the environmental medium (mg/kg), $R$ is the ratio of the environmental concentration and the exposure concentration, $CR$ is the contact rate (kg/day), $BW$ is the body weight (kg), $ED$ is the exposure duration (years), $EF$ is the exposure frequency (days/year), and $AT$ is the averaging time (days).

The input parameters (e.g. breathing rate, water intake rate, etc.) vary between pathways, thus Equation 8 is written differently for each exposure pathway. The risk to an individual due to exposure to a carcinogen is calculated by multiplying the ADD by a cancer potency factor (CPF). In a risk assessment, this equation is most often applied to a site specific case to determine the risk to an individual. For an air emission, a plume model is used to calculate realistic exposure concentrations for an individual living close to the site. In our case, Equation 8 will be applied to all individuals exposed to the chemical.

***Exposure Concentrations*** - Exposure media concentrations may differ from the ambient

[1] Here we define the potential dose as the amount of chemical that passes into an individual while the actual dose quantifies the amount of chemical that is absorbed into an individual (e.g. the amount of chemical in the air an individual breathes is the potential dose while the actual dose is the portion of that air that passes into the lung tissue). Ideally, risk should be based on the actual dose, but often the potential dose is assumed to equal the dose, an assumption also made in this chapter.

environmental media concentrations and can be calculated from the ambient air, soil, vegetation, and surface water concentrations. For example, the concentration of a chemical in the indoor air differs from the concentration in the outdoor air. The indoor air concentration is influenced by the concentration in the outdoor air, the concentration in the soil gas below the house, the concentration in tap water, and the concentration in resuspended particles in the home attributable to soil tracking, due to shoes, clothing, and the fur of pets or particles in the outdoor air that enter the home and are subsequently deposited. Another example is the concentration in meat, which depends on the animal's ingestion of contaminated soil, pasture and grains, and inhalation of air. Equations relating exposure media concentrations to environmental concentrations were taken directly from the CalTOX model (37).

***Human Activity and Contact*** - The remainder of the terms needed to calculate the ADD in Equation 8 for each exposure pathway relate to various aspects of human activity and contact. Again, the processes used are taken from CalTOX (37). Exposure duration, the length of time a person is likely to be exposed to a contaminant, is needed to calculate the ADD. However, for population dose, we assume a constant total population and thus calculate the dose as a long-term annual average.

***Model of Population Exposure*** - We use the following equation for the population dose:

$$Population\ \ Based\ Potential\ Dose = \iint P(x,y) \times ADD(x,y)\,dx\,dy \qquad (9)$$

where $P$ is the population Density (Persons/m$^2$) and AD$D$ is the dose per Person (mg/kg-d). In this equation, both the dose per person and the population density can vary spatially. We must, however, determine the appropriate scale to use, the system boundaries, and the population

density.

When calculating a population dose, we are not concerned with the variability between members of the population. A Monte Carlo simulation that varies uncertainty and variability simultaneously can be used to predict the mean value of the population based potential dose. If a population risk is determined from the population based potential dose calculated by Equation 9, a linear dose response curve must be used for the risk measure to be meaningful.

We consider an idealized environmental model with the source term located in the urban region where the population density is highest. We assume a steady wind blowing from the urban region toward the suburban and rural regions, which have lower population densities than the urban region.

Figure 4a presents a geometry that accounts for the coupling of the higher population density and higher dose per person in the urban region. We call this the spatial model. The concentration is constant in the urban region and decreases exponentially with distance due to decay in the environment as the distance from the source region increases. Population densities are assumed constant in time in the urban, suburban and rural regions. This geometry uses a wind velocity that always travels in a constant direction with no lateral dispersion. We believe these simplifications are appropriate because the spatial model is designed to compare chemicals, not determine actual risk levels. The equation for the population based potential dose for the spatial model is:

$$Population\ Dose = P_U \times ADD \times w_y w_x + \int_{w_x}^{w_x'} P_{SU} \times ADD \times e^{-(x-w_x)/CTD} \times w_y\, dx + \int_{w_x'}^{\infty} P_R \times ADD \times e^{-(x-w_x)/CTD} \times w_y\, dx \quad (10)$$

the subscripts *U, SU, and R* refer to urban, suburban, and rural, respectivly; $w_x$ is the width of the

urban region in the x direction (m), and $w_y$ is the width of the urban region in the y direction (m) and $w_x'$ is the distance at which the population density changes from suburban to rural, $(A_u+A_{su})/w_y$.

The ADD is calculated from the multimedia, multipathway exposure model. The model region is the size of a representative urban region and we assume an open region (i.e. wind flows out of the region). The size and population densities of the urban, suburban, and rural regions are representative, with the values listed in Table 4. The width and length of the urban region, $w_x$ and $w_y$, are both taken as equal to the square root of the urban area. The *CTD* is calculated using the methods presented in the previous section.

The spatial model is compared to the urban model shown in Figure 4b. In this model, only the urban population is exposed and we use the following equation to calculate the population based potential dose:

$$Population\ Dose = P_u \times ADD \times w_x w_y \qquad (11)$$

## 3. Case Study

To demonstrate the calculation and evaluation of $\tau$, CTD, and population based potential dose, we carried out a case study using two chemicals, one that is considered persistent, 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD), and one that is not considered persistent, benzo[a]pyrene. TCDD is typically released into the air as a by-product from incineration, combustion of fossil fuels, and industrial processes in urban areas, but often contaminates suburban and rural sites as well (38, 39). Airborne 2,3,7,8-TCDD is found in both the gaseous and particulate phases. TCDD is also released to the land and water through industrial sources and landfills. TCDD has limited degradation in soil (40-42) and decays both in air and in vegetation (29, 43-46). In the vapor phase of the atmosphere, reaction with OH radicals is the dominant degradation pathway. TCDD on particles has negligible degradation. The primary degradation process for TCDD in vegetation is

the reductive dehalogenation by sunlight which requires proton donors. Because the lipids in plants are rich sources of proton donors, we expect higher degradation rates in vegetation relative to air (42, 43).

Benzo[a]pyrene is a polyaromatic hydrocarbon that decays rapidly in air and tends to favor the lipid phases of the environment. Benzo[a]pyrene is also a byproduct of combustion. This chemical is found at much higher concentrations in urban regions, indicating that it does not travel a long way in the environment (47). At present, there is no information on the degradation rate of benzo[a]pyrene in vegetation and we assumed a decay rate equal to that in surface soil. The representative values used for all of the chemical properties in calculating the three measures are listed in Table 5.

***Characteristic Time-*** The characteristic times for steady dioxin emissions to both air and soil are plotted in Figure 5. The characteristic time is over an order of magnitude greater when the pollutant is released directly to the soil compartment. The characteristic time is much less if released to air because a significant portion is decayed in air, consistent with the findings of the dynamic mass balance completed by Eisenberg et al. (48). This demonstrates the importance of determining the location of the source term when calculating $\tau$. Benzo[a]pyrene also has a much longer $\tau$ when released to soil than to air. Webster et al. (33) have also developed a method for calculating $\tau$ and also found it important to characterize the medium of entry into the environment.

***Characteristic Travel Distance*** - The *CTD* for benzo[a]pyrene was calculated to be on the order of 30 km. Benzo[a]pyrene has a relatively short *CTD* due to a relatively rapid degradation rate in air. Consistent with this model prediction, field studies have found benzo[a]pyrene in suburban but

not rural regions (47).

In contrast, the *CTD* for TCDD was calculated to be on the order of 600 km. Many field studies have found fairly uniform TCDD concentrations in the northern hemisphere (49) while others have found TCDD concentrations in urban regions approximately one order of magnitude greater than concentrations found in rural regions (50). The calculated *CTD* is on the same order of magnitude as, or greater than, the distance between urban centers. With a *CTD* of 600 km, we do expect TCDD to show some reduction in concentration between urban and rural regions. However, we expect less spatial variation if the urban centers are located less than 600 km apart, explaining the fairly uniform concentrations reported in the literature.

To illustrate the possible effect of one urban center on another, we use two large urban centers located 500 km apart as shown in Figure 6. We assume an average steady wind from Center A to Center B. The size of the urban centers and their corresponding sources are assumed to be equal. Therefore, each center would have the same concentration if considered independently. Since all of the processes are linear, we can use the principle of superposition and sum the concentrations. The fraction of the initial concentration at each urban center and the cumulative total are plotted with distance in Figure 6. Since the centers are slightly less than one *CTD* apart, approximately one-third of the concentration at Center B results from Center A . If we consider the effect of managing the local sources in Center B relative to input from Center A, we determine that although reducing emissions at Center B will impact concentrations, a more regional approach is necessary for additional reductions. Thus, chemicals with long spatial ranges need to be managed regionally. This knowledge can be used to reduce human exposures through regional strategies that address multiple sources in place of single source management.

We completed a Monte Carlo uncertainty analysis with 5000 simulations to generate a

distribution of plausible *CTD* values for TCDD with the Crystal Ball software package (51). The range of values for the *CTD* of TCDD was from 100 to 1000 km. This range results from both uncertainty in chemical properties and variability in landscape properties.

To understand which inputs contribute significantly to the uncertainty in the output, a sensitivity analysis was performed to calculate the rank correlation coefficients between *CTD* and its defining parameters. The rank correlation coefficients are a measure of the strength of the linear relationship between each input and the *CTD*. This technique considers both the range of uncertainty in the input parameter and the influence of the parameter on the *CTD*. The rank correlation coefficients are squared and the values normalized to 100% to determine the approximate contribution to variance. The most influential parameters are displayed in Figure 7. Decreasing the uncertainty and variability in these parameters would have the largest impact for reducing the estimated uncertainty range of *CTD*. The wind speed, rainfall rate, temperature, and plant biomass are all highly variable and are dependent on site specific data. The half-life in air, half-life in vegetation, and vegetation-air partition coefficient are all poorly characterized in the literature and thus the uncertainty of *CTD*s could be reduced as these input values are better defined.

***Evaluation of multimedia mass distribution*** - To gain confidence in the four compartment model, we used a stationary system with an area of 250,000 $km^2$, a continuous emission of 1.1 g/d, and a windspeed of 4 m/s through the system (52, 53). These values were selected to represent the conditions in Germany, where background concentrations have been measured (54). The calculated TCDD concentrations appear to be in agreement with background measurements from Germany, as shown in Table 6 (54). Additionally, calculated transfer rates from air to soil are in the range of experimental data (55). We also compared the model to a vegetation scavenging

ratio determined for TCDD, measuring the vegetation concentration resulting from all processes to the air concentration and found it to be the same order of magnitude (56).

***Population* Based Potential *Dose* -** The uncertainty distributions for the population based potential dose for both benzo[a]pyrene and TCDD using the spatial and urban models are shown in Figure 8. For benzo[a]pyrene, the potential dose is nearly the same using the two calculation methods because very little chemical is carried out of the system by advection. In contrast, for TCDD, the urban model predicts approximately one half the population dose as does the spatial model because much of the chemical is advected out of the model system by wind. This difference is comparable to other sources of uncertainty in the calculation. Thus, for TCDD (and any other chemical with a long *CTD* such as DDT or hexochlorobenzene), it is important to consider using the spatial model to calculate population exposure as opposed to the urban model which only considers the locally exposed population.

## 4.0 Conclusions and Recommendations

In any effort to assess the potential adverse effects of a chemical, $\tau$, *CTD*, and the population based potential dose are important components of the analysis. From a policy perspective, these methods need to be transparent and representative of the complex dynamic environment. In this chapter, we demonstrated simple methods for estimating all three measures.

The characteristic time can be used to evaluate whether a chemical is a POP while the *CTD* determines if a chemical has the potential for long range transport. The duration over which impacts are assessed should be on the same order of magnitude as $\tau$ for that chemical. We also showed the importance of using the correct medium receiving releases of a chemical such as

TCDD.

The *CTD* is essential in any effort to assess the potential adverse effects of a chemical. Often established by political or geographical boundaries (air pollution districts, state boundaries, etc.) or by tradition (i.e. a specified distance from a source), the "regulatory scale" is arbitrary and often not defensible by scientific analysis. In this chapter, we have demonstrated simple methods for making preliminary estimates of the *CTD* that defines the effective range of impact for a chemical contaminant. The *CTD* is needed before deciding what scale to use when measuring or modeling the dispersion of an environmental contaminant. This method was evaluated through a case study of TCDD, for which we found a *CTD* on the order of 600 km. This value is realistic when considered in conjunction with available monitoring data. If a chemical has a *CTD* on the order of magnitude of, or greater than, the typical distance between urban centers, it is important to regulate the chemical regionally, as opposed to locally.

When making choices among alternative chemicals for use in a certain process, evaluating a new chemical upon its introduction to commerce, or deciding if one should further regulate a chemical presently in use, we may want to determine the population based potential dose to that chemical per unit release. If a chemical has a long *CTD* in the environment, calculating the risk only to individuals near the site may be insufficient, as we must consider the exposure to the population far from the site. We show that as the *CTD* increases, the differences between calculation methods can be on the same order of magnitude as contributions from other sources of uncertainty in the calculation (this was the case here for TCDD). It is important to note that the comparison between sources of uncertainty is chemical specific. We recommend that as the *CTD* of the chemical pollutant increases, the population based potential dose should be calculated and the model used for the calculation carefully selected.

In all of the calculations, several assumptions were made, including: spatially independent exposure parameters, a linear cancer slope factor at low doses, no dispersion of airborne chemicals, all sources located in urban regions, a uniform population density in each of the three categories, each assumed to be in a uniform geographical pattern. We have not evaluated the effects of these assumptions on the reliability of the results because this model is intended for screening level purposes in order to compare chemicals, not to determine the level of risk.

These measures are useful for determining the potential for exposure prior to introducing a new chemical to the market. The list of possible uses for such measures of persistence include risk assessments, life cycle impact analyses, development of pollution prevention strategies, evaluation of pollutant mass balances, comparisons between toxic release inventories, and regulatory impact studies.

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Table 1: List of Transfer Pathways in Multimedia Model - Calculation methods for each pathway can be found in Reference 21.

| | Air to | Surface Soil to | Root-zone Soil to | Vegetation to | Surface Water to |
|---|---|---|---|---|---|
| Air | | particle resuspension, and diffusion | | particle resuspension, diffusion | diffusion |
| Surface Soil | wet and dry particle deposition, wet gaseous deposition, and diffusion | | | washoff from leaves | |
| Root-zone Soil | | diffusion, advection through rain | | | |
| Vegetation | wet and dry particle deposition and diffusion | rainsplash of particles | root uptake | | |
| Surface Water | wet and dry particle deposition, wet gaseous deposition, and diffusion | runoff and erosion | | | |
| Flows out of model region | advection by wind | | | | advection of surface water |

Table 2: Representative Spatially Averaged Landscape Properties

| Landscape Property | Notation | Mean Value | Coefficient of Variation | Reference |
|---|---|---|---|---|
| universal gas constant (Pa-m$^3$/mol-K) | $R$ | 8.31 | 0 | |
| ambient environmental temperature (K) | $T$ | 288 | 0.02 | (57) |
| yearly average wind speed (m/d) | $u$ | 3.46E+05 | 0.2 | |
| relative humidity | $rh$ | 0.8 | 0.1 | |
| surface area of particles(m$^2$/m$^3$) | $SA$ | 1.50E-04 | 0.1 | (58) |
| washout ratio | $Wr$ | 5.00E+04 | 2 | (58) |
| atmospheric dust load (kg/m$^3$) | $\rho_{ba}$ | 5.00E-08 | 0.2 | (58) |
| dry deposition velocity of air particles (m/d) | $v_d$ | 43.2 | 0.3 | (58) |
| boundary layer thickness in air above vegetation (m) | $\delta_{ap}$ | 0.005 | 0.2 | (59) |
| boundary layer thickness in air above soil (m) | $\delta_{ag}$ | 0.005 | 0.2 | (59) |
| annual average precipitation (m/d) | $rain$ | 2.0E-03 | 1 | (57) |
| soil runoff rate (kg/m$^2$d) | $erosion$ | 3.0E-3 | 0.2 | (60) |
| ground water recharge (m/d) | $recharge$ | 1.2E-04 | 1 | (60) |
| plant-air partition factor, particles (m$^3$/kg[FM]) | $K_{pa}^{part}$ | 3300 | 0.1 | (61) |
| plant dry-mass fraction | $bio_{dm}$ | 0.20 | 0.2 | (60) |
| plant fresh-mass density (kg/m$^3$) | $\rho_p$ | 1.00 E+03 | 0.2 | (60) |
| soil particle density (kg/m$^3$) | $\rho_s$ | 2.60 E+03 | 0.05 | (60) |
| water content in surface soil (%) | $\beta_g$ | 0.17 | 0.2 | (60) |
| air content in the surface soil (%) | $\alpha_g$ | 0.40 | 0.2 | (60) |
| water content of root-zone soil (%) | $\beta_s$ | 0.28 | 0.2 | (60) |
| air content of root-zone soil (%) | $\alpha_s$ | 0.17 | 0.2 | (60) |
| height of the air compartment (m) | $d_a$ | 1000 | 0.1 | |
| Fraction of area that is surface water | fw | 8.15E-03 | 0.2 | |
| Average depth of surface water (m) | $d_w$ | 5 | 1 | |
| thickness of the ground soil layer (m) | $d_g$ | 2.50 E-03 | 1 | |
| plant dry mass inventory (kg[DM]/m$^2$) | $bio_{inv}$ | 0.40 | 0.2 | |
| Suspended sediment in surface water (kg/m$^3$) | $\rho_{bw}$ | 0.8 | 1 | |
| water runoff rate (m/d) | runoff | 2.8E-4 | 1 | |
| organic carbon fraction | $f_{oc}$ | 0.03 | 1 | (60) |
| organic carbon fraction in sediments | $f_{oc}^{sed}$ | 0.02 | 1 | (60) |
| evaporation rate of surface water (m/d) | $evaporate$ | 4.38E-6 | 1 | (60) |
| sediment particle density (kg/m$^3$) | $\rho_{sd}$ | 2600 | 0.05 | (60) |

Table 3: List of Exposure Pathways - Calculation methods for each pathway can be found in the CalTOX manual (37).

| Ingestion Pathways |
|---|
| * All intake values were correlated per unit body weight, preventing data points based on high intake with low body weight |
| Exposed Produce – Including Grains |
| Unexposed Produce – The concentration in the two types of produce and grains are calculated separately. They include exposure to air, soil, and water used for irrigation. |
| Fish – Based on surface water concentrations |
| Meat, Milk, and eggs - livestock products are exposed through inhalation, direct ingestion of water and soil, and indirect exposure through food contaminated by exposure to air and soil. |
| Soil - both adults and children ingest small amounts of soil through inadvertent hand-to-mouth activities |
| Water - while swimming in surface water |
| Tap water – concentration linked to concentration in both surface water and ground water, assumed clean in this model |
| **Inhalation Pathways** |
| * Breathing rates vary by activity level as do location of activity levels. Breathing rate and location need to be linked. |
| Active Outdoors |
| Resting Indoors – The concentration of indoor air includes soil vapors transferred from under the house and soil particles transferred to indoor air |
| Active Indoors |
| While showering or in the bath – Contaminants transferred from tap water |
| **Inhalation Pathways** |
| Showering – From tap water |
| Swimming – From surface water |
| Soil - dermal exposure to contaminants in soil can occur during a variety of activities, such as construction work, gardening, and recreation outdoors. Children playing outdoors also can have rather large soil loading on their skin. |

Table 4: Representative Population Densities and Areas

| | Population Density Persons/$m^2$ | Average Area $m^2$ |
|---|---|---|
| Urban Region | $PD_U$=3.5E-03 | $A_U$=2E+08 |
| Surrounding Region | $PD_{SU}$=6E-04 | $A_{SU}$=5E+09 |
| Background | $PD_B$=8E-05 | |

Table 5: Representative Chemical Properties Used in the Case Study (26, 41, 42, 44, 46, 60)

| Chemical or Landscape Property | Notation | TCDD Mean Value | TCDD Coefficient of Variation | B[a]P Mean Value | B[a]P Coefficient of Variation |
|---|---|---|---|---|---|
| molecular weight (g/mol) | *MW* | 322 | 0.01 | 252 | 0.01 |
| octanol-water partition coefficient | $K_{ow}$ | 5.70 E+06 | 1 | 2.20 E+06 | .72 |
| melting point (K) | *Tm* | 578 | 0.01 | 451 | 0.028 |
| vapor pressure in (Pa) | *VP* | 1.00 E-07 | 2 | 7.13 E-07 | .07 |
| Henry's law constant (Pa-$m^3$/mol) | *H* | 3.75 | 1.5 | 0.092 | 1 |
| diffusion coefficient in pure air ($m^2$/s) | $D_{air}$ | 4.86E-06 | 0.1 | 5.09E-06 | 0.08 |
| diffusion coefficient; pure water ($m^2$/s) | $D_{water}$ | 5.90E-10 | 0.1 | 6.13E-10 | 0.25 |
| organic carbon partition coefficient | $K_{oc}$ | 5.40 E+06 | 0.1 | 2.49 E+06 | 0.9 |
| biotransfer factor, plant/air ($m^3$[a]/kg[pFM]) | $K_{pa}$ | 25000 | 0.85 | 5.92 E+05 | 14 |
| decay rate in air (1/s) | $k_a$ | 8.0E-07 | 1.5 | 1.27E-04 | 1 |
| decay rate is surface water (1/s) | | | | 3.47E-06 | 1.2 |
| decay rate in surface soil (1/s) | $k_g$ | 2.2E-08 | 1.2 | 3.47E-08 | 1.1 |
| decay rate in root-zone soil (1/s) | $k_s$ | 2.1E-10 | 1.7 | 3.47E-08 | 1.2 |
| decay rate in vegetation (1/s) | $k_p$ | 1.3E-06 | 3.0 | | |

Table 6: Fugacity, Inventory, Concentration, and Mass Transformed for Each Compartment for a Fixed Region Using TCDD

| Compartment Name | Fugacity (Pa) | Calculated Concentration ($gm/m^3$) | Measured Concentration (54) ($gm/m^3$) | Mass Transformed (gm/d) |
|---|---|---|---|---|
| Air | 2.02E-14 | 3.50E-15 | 3.60E-15 | 6.04E-02 |
| Vegetation | 3.85E-15 | 1.33E-08 | 5.43E-09 | 3.73E-01 |
| Ground Surface Soil | 3.46E-15 | 5.41E-08 | 5.58E-08 | 6.37E-02 |
| Root Zone Soil | 3.20E-15 | 6.37E-08 | 7.09E-08 | 5.08E-04 |

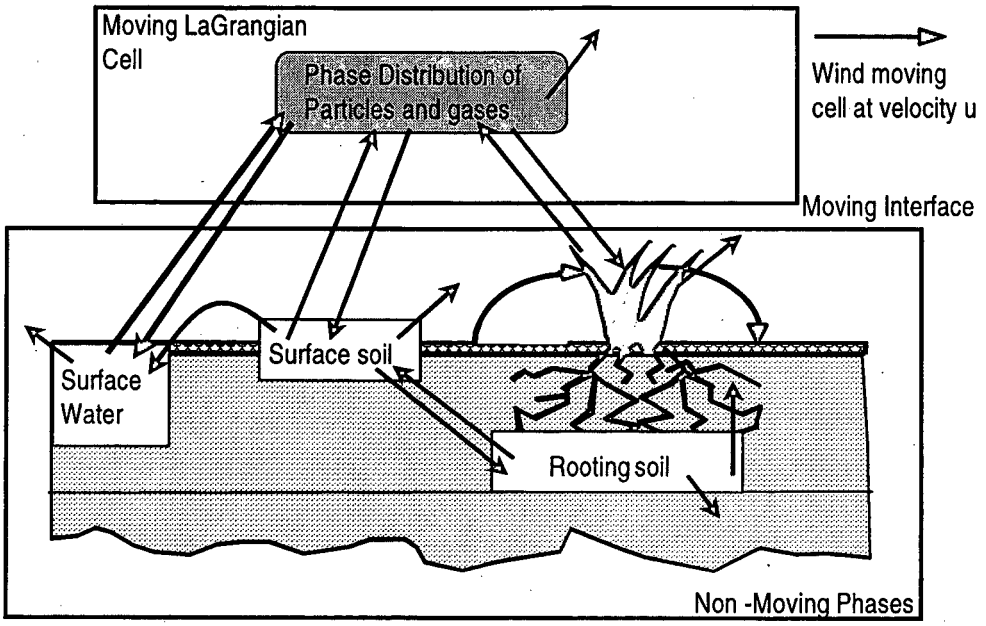


Figure 1: An illustration of a multi compartment model system.

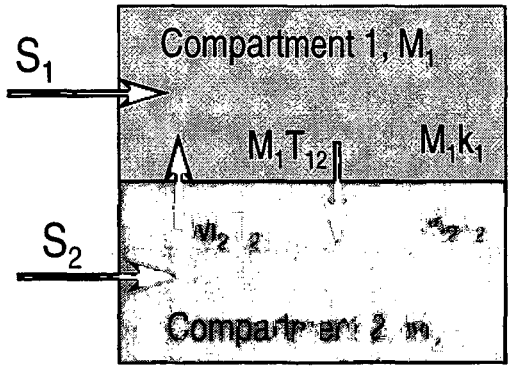

Figure 2: Diagram of the two compartment system used to calculate $\tau$.

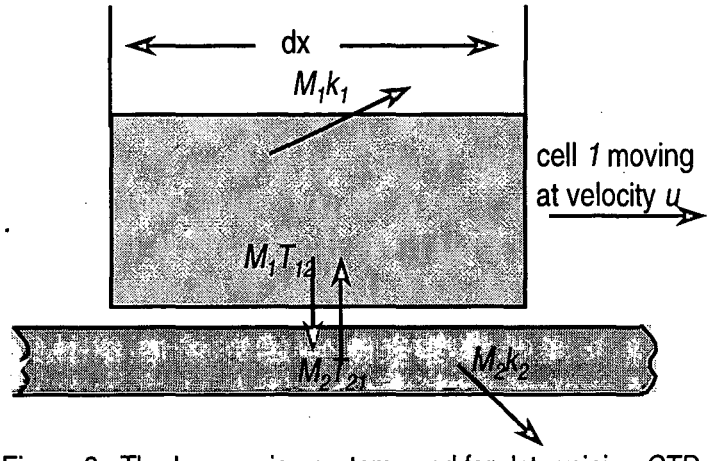

Figure 3: The Lagrangian system used for determining *CTD*

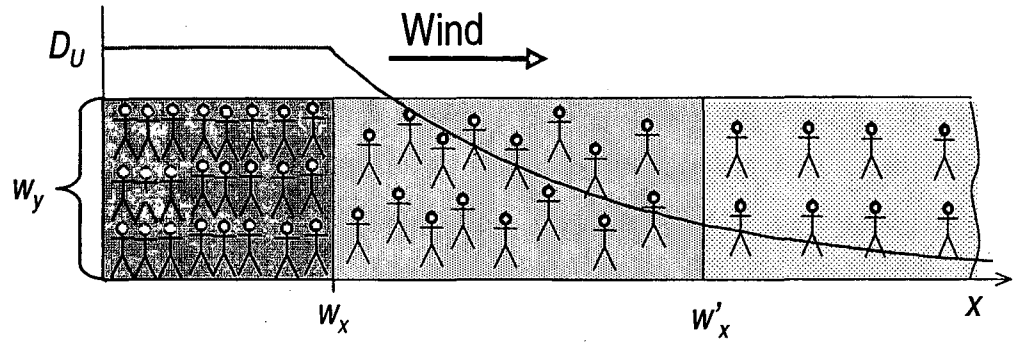


Figure 4a: Spatial System for calculating population based potential dose

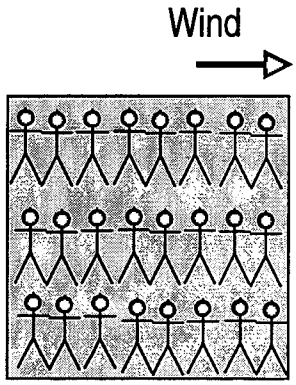


Figure 4b: Open Urban System for calculating population based potential dose

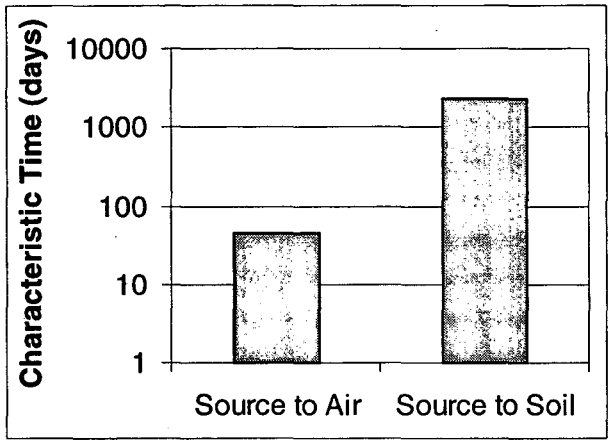


Figure 5: Comparison of $\tau$ for TCDD with a steady source to air and a steady source to soil

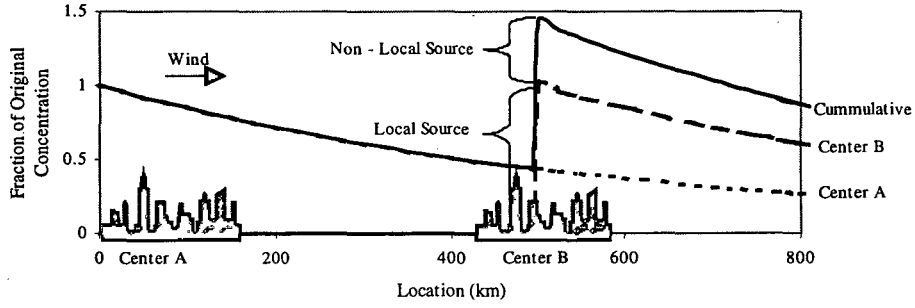


Figure 6: For a case study using TCDD, we plot the fraction of the initial concentration with distance from an urban center for Center A, Center B, and the cumulative total.

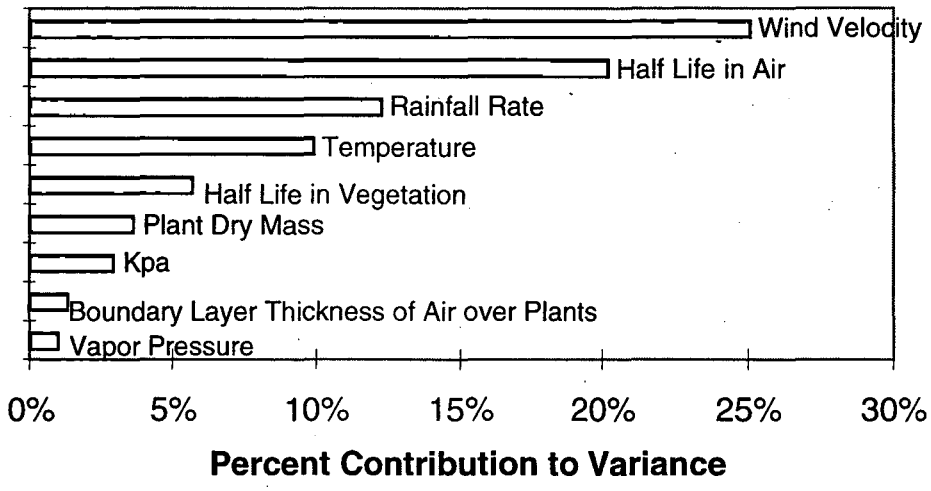


Figure 7: Results of sensitivity analysis for the four compartment steady state system for the TCDD case study.

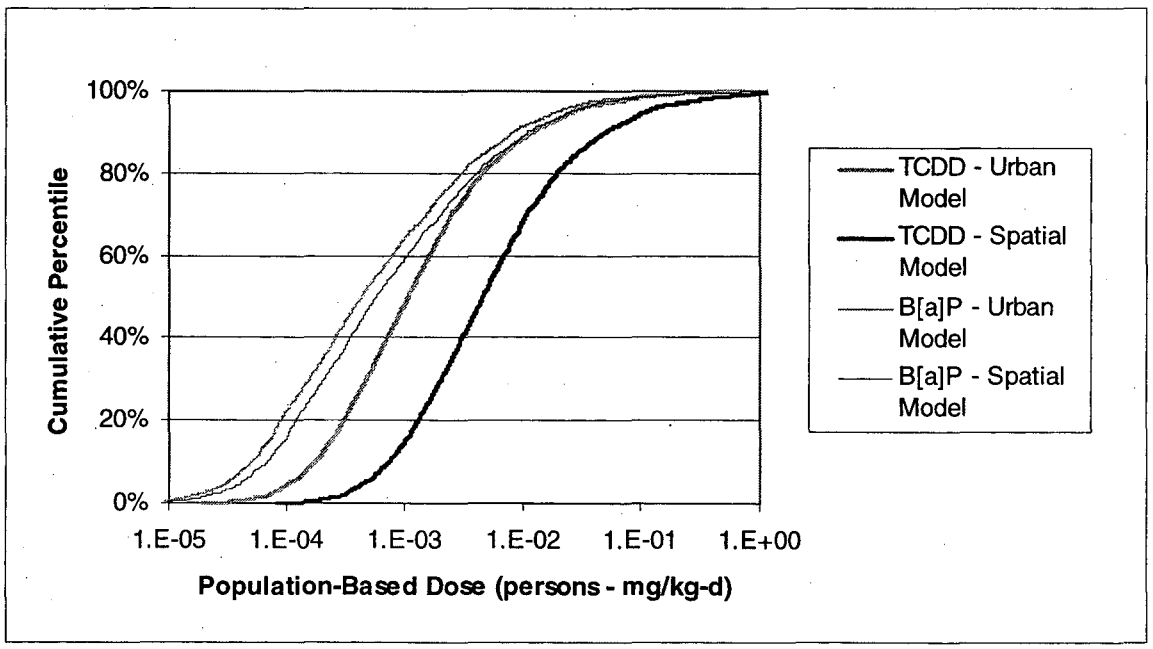


Figure 8: Cumulative percentile distribution of population based potential dose for each calculation method for TCDD (long *CTD*).

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