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General Formulation of Characteristic Travel Distance for Semivolatile Organic Chemicals in a Multimedia Environment

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A relatively simple but informative methodology is introduced to determine the characteristic travel distance (CTD) for airborne semivolatile organic pollutants. The CTD is derived from a moving Lagrangian cell (representing the air) and a nonmoving compartment (representing soil or vegetation). The methodology is expanded to a fugacity based steady-state multimedia environmental framework including air, vegetation, and soil. Chemical transformations in air as well as partitioning to, and transformation in, vegetation and soil are considered. Concentrations are determined by interactions among the compartments and transformation rates. This method is most appropriate for continuous, large nonpoint emissions (such as emissions from an urban airshed). A case study for 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) reveals that the CTD is on the same order of magnitude as the typical distance between urban centers. Vegetation is important for defining the regional transport processes for TCDD.

Introduction

Semivolatile organic pollutants originate from a broad range of human activities, including combustion for energy production and transportation, industrial processes, and agricultural uses of pesticides. Recent studies have reported surprising levels of regional atmospheric dispersion, deposition to and uptake by vegetation of PAH and PCDD/F compounds (*1*−*5*). A methodology to determine the characteristic spatial scale for airborne chemical pollutants is needed to evaluate and classify the potential human health and ecological impacts of a chemical released into the environment and to determine whether the pollutant can have a local, regional, or global scale impact. As the characteristic spatial scale approaches the distance between source regions (i.e., between urban/industrial centers), the chemical pollutant may be considered a regional or global scale pollutant as opposed to a local scale pollutant.

There are a variety of regional-scale models, such as the nonspatial, zero-dimensional multimedia box models, a concept introduced by Mackay (*6*). Models with spatial resolution include the SIMPLEBOX model (*7*) with a nested set of “unit worlds” or Wania and Mackay’s (*8*) model of global transport of persistent organic pollutants (POPs) from temperate to arctic latitudes with a series of connected boxes. Scheringer (*9*) demonstrated that the spatial scale needed to characterize the multimedia dispersion of organic chemicals is dependent on the chemical properties using a series of connected boxes in a one-dimensional band. Van Pul derived a characteristic distance based on degradation and deposition processes in an uncoupled air parcel (*10*).

In this paper, we examine a chemical transported through air by means of a Lagrangian cell that tracks the movements of a contaminated air parcel instead of using a fixed reference box model. Starting with two compartments, a moving (i.e., air) compartment and a nonmoving compartment (i.e., vegetation or soil), we consider chemical transformation in the moving phase as well as partitioning to and subsequent transformation in the nonmoving phase, i.e., a coupled system. From this model, we introduce the concept of a characteristic travel distance (CTD) derived from a multimedia model. CTD is the distance from the source region at which the concentration in the Lagrangian cell is reduced by 63%. We next expand this model to a steady-state multimedia environmental framework including air, vegetation, and two soil layers.

Vegetation has only recently been considered in the interpretation of regional air pollution measurements (*2, 3*) and is not yet well characterized in terms of storage and transformation capacity. The ratio of concentrations in air to vegetation and air to soil is controlled by the following processes: chemical specific vegetation/air and soil/air partitioning; the kinetics of mass transfer rates among air, vegetation, and soil; and the degradation rates in air, plant tissue, and soil. These processes have been studied extensively (*11*−*15*). We assess how these chemical specific processes impact the geographical zone of influence of pollutant releases.

The methodology presented here is appropriate for continuous, large nonpoint atmospheric emissions of organic chemicals, such as combustion emissions from transportation, industrial facilities, heating, and electrical generation. The methodology is appropriate for ubiquitous chemicals with long atmospheric half-lives (several hours or days); a relatively high K_{ow} value, such that partitioning into vegetation and soil is significant (at least 1×10^6); and a high enough vapor pressure such that there is some partitioning from particles to the gas phase of the atmosphere (at least 1×10^{-10} Pa).

Methods

Defining Characteristic Travel Distance. To derive the characteristic travel distance (CTD), we consider the system in Figure 1a with moving and nonmoving phases. This is an idealized representation of the system in Figure 1b. We calculate the changing chemical mass in a moving

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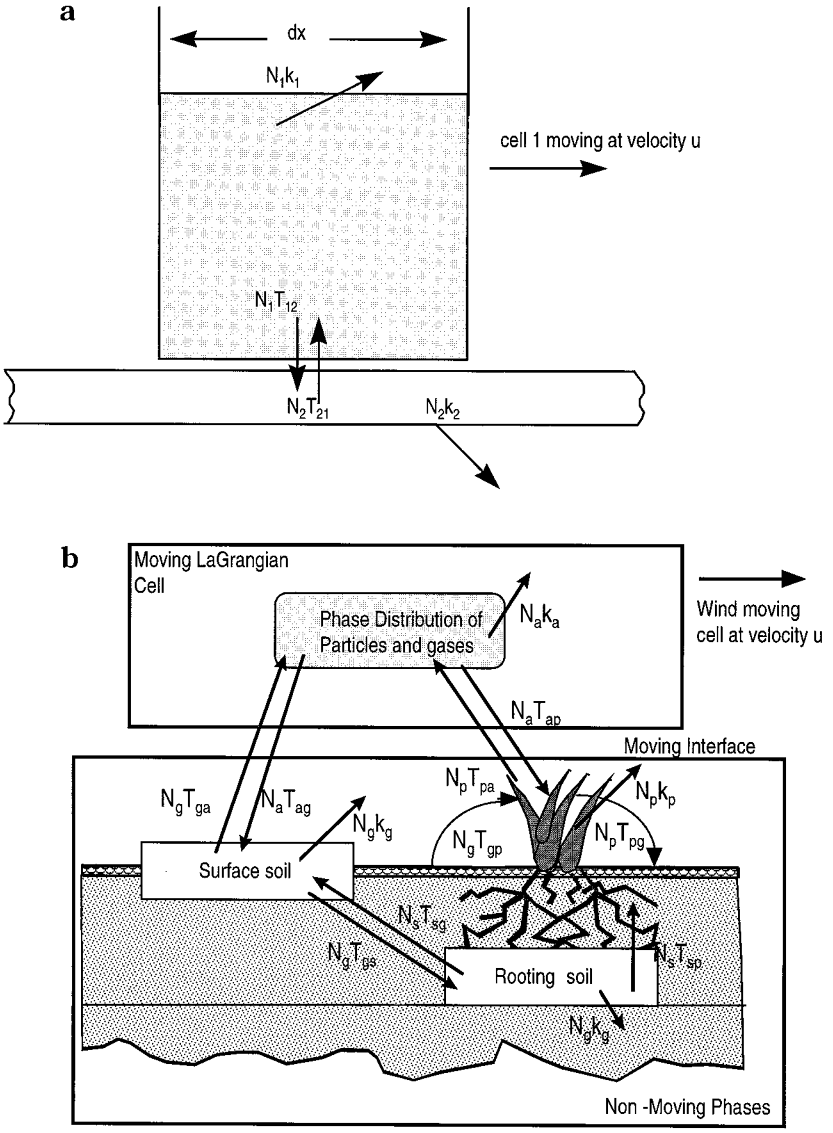

FIGURE 1. (a) The two compartment Lagrangian system used for determining the CTD. (b) An illustration of the multicompartment model system used in the case study.

Lagrangian air cell as the cell travels in a one-dimensional band away from the area of release (the urban source region). Transformation in the moving phase, transport to, and subsequent degradation in the nonmoving phase reduce the concentration in the air cell. The CTD for a given chemical is the distance it takes to reduce the chemical concentration in the Lagrangian cell by 63% (i.e. reduced to 1/e of the original concentration). By using a continuous differential Lagrangian structure, equations defining the CTD are obtained in closed analytic 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 a steady wind rate in one direction; the landscape properties do not vary spatially (or are spatially averaged); and the atmospheric mixing layer height is constant.

The Lagrangian cell represents a small portion of the continuous stream of pollutant flowing from the source region. At steady state, neither the moving nor the non-moving phases are accumulating mass at a particular location. Additionally, the ratio of the mass in the ground to the mass in the air is spatially independent.

The differential equations defining the mass balance in the cells shown in Figure 1a are

$$\frac{dN_1(t)}{dt} = -N_1(t)k_1 - N_1(t)T_{12} + N_2(t)T_{21} \qquad (1)$$

$$N_1T_{12} = N_2 \times (k_2 + T_{21}) \qquad (2)$$

where k_i is the transformation rate (1/s), N_i is the chemical mass in a compartment (kg), T_{ij} is the diffusion/advection transfer rate from compartment i to j (1/s), t is the time traveled from source region (s), and 1 and 2 are the indices for moving phase and nonmoving phase, respectively.

The distance and elapsed travel-time from the source region are related through the relationship $x = tu$ where u is the windspeed (m/s). Because the system is in steady state and both the transfer factors and the decay rates are assumed to be spatially independent, the ratio N_2/N_1 is spatially independent. The net flux at the moving, non-moving interface equals the amount decayed in the non-moving compartment, because at steady state the mass in the soil has increased to a level such that decay compensates the flux. A full derivation can be found in Supporting

TABLE 1: Fugacity Capacity of Each Compartment

compartment	fugacity capacity
air	$Z_a = Z_{\mathrm{air}} + Z_{ap},\ Z_{\mathrm{air}} = 1/RT,\ Z_{ap} = \dfrac{0.17 \times SA}{RT \times VP \times e^{\left(6.81\left(1 - \frac{Tm}{T}\right)\right)}}$
plant	$Z_p = K_{pa} \times \rho_p \times Z_{\mathrm{air}} + K_{pa\_part} \times \rho_p \times Z_{ap}$
surface soil	$Z_g = Z_{\mathrm{air}}\alpha_g + Z_{\mathrm{water}}\beta_g + (1 - \alpha_g - \beta_g)Z_{gp},\ Z_{\mathrm{water}} = 1/H,\ Z_{gp} = \dfrac{H \times Kd \times \rho_s}{1000}$
root zone soil	$Z_s = Z_{\mathrm{air}}\alpha_s + Z_{\mathrm{water}}\beta_s + (1 - \alpha_s - \beta_s)Z_{gp}$

Information, Section 1. Using these simplifications, it can be shown that

$$\frac{\mathrm{d}N_1(t)}{\mathrm{d}t} = \frac{N_1k_1 + N_2k_2}{N_1}N_1(t) = k_{\mathrm{effective}}N_1(t) \qquad (3)$$

Where the effective decay rate, $k_{\mathrm{effective}}$ (1/s), is defined as

$$k_{\mathrm{effective}} = \frac{N_1k_1 + N_2k_2}{N_1} \qquad (4)$$

The effective decay rate is essentially the mass in a compartment times the corresponding decay rate divided by the mass in the moving phase. Solving eq 3 and transforming the time variable into a spatial variable, i.e., the distance from the source, leads to the following result

$$N_1(x,u) = N_1(0)\ \exp\left(\frac{-k_{\mathrm{effective}}x}{u}\right) = N_1(0)\ \exp\left(\frac{-x}{\mathrm{CTD}}\right) \qquad (5)$$

We define the characteristic length to normalize the distance from the source as

$$\mathrm{CTD} = u/k_{\mathrm{effective}} \qquad (6)$$

The characteristic length is the same in both the moving and nonmoving phases because the ratio between the concentrations is spatially independent. This implies that the concentration decreases at the same rate with distance from the source region in both phases.

To more accurately evaluate CTD, we use a more comprehensive environmental representation, illustrated in Figure 1b. In devising this representation, our goal is to balance the amount of detail in order to be useful and representative yet not so complex as to limit the understanding of the system interactions (*16*). The relatively simple system allows for the ability to complete an uncertainty analysis to understand the effects of uncertain and variable parameters. The moving air phase interacts with three nonmoving phases, vegetation and two soil layers, each in a steady-state relationship with one another. The effective transformation rate for this system is

$$k_{\mathrm{effective}} = \frac{N_ak_a + N_pk_p + N_gk_g + N_sk_s}{N_a} \qquad (7)$$

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

Defining the Multimedia Model. The steady-state concentration in each environmental compartment is determined from interactions among the compartments and the transformation rate in the compartment. Many of the processes defining the interactions between compartments have been discussed elsewhere (*12, 16, 17*). The equations for the steady-state concentrations based on a mass balance between compartments can be found in Supporting Information, Section 2.

We use a fugacity-based model where the fugacity capacities (the chemical concentration per unit chemical fugacity) are defined in Table 1. All of the phases (i.e. air, water, solids) in a compartment are assumed to be in chemical equilibrium. The exchange between compartments is based on transfer coefficients, or "*T*" values, found in Table 2. These values are transfer rates applied to unit mass inventory.

Environmental Compartments. The interactions between air and vegetation vary by vegetation type (e.g. deposition rates (*18*)). Chemicals traveling long distances will interact with several vegetation types. Processes and properties depend on vegetation type, but, for simplicity, we assume all properties and processes can be spatially averaged; hence representative properties are used.

The capacity of the root-zone soil to take up and retain a chemical from the atmosphere depends on the defined root-zone soil depth and the water and organic-carbon content. Because our interest is in the role of soil as a reservoir for steady-state air emissions, we use a chemical-dependent "effective soil depth". This equals the steady-state penetration depth (*19*), derived from a modified Damkoehler number, which expresses the ratio of reaction losses to diffusion/advection transport in a given soil depth. When the soil in one penetration depth reaches a steady-state relationship with the air, it contains the same amount of chemical as an infinite column of soil; one with a concentration gradient corresponding to the steady-state solution of the dispersion/advection equation with a fixed concentration at the surface and zero concentration at infinite depth. Details of this derivation are in Supporting Information, Section 3.

Transformation Processes. Mass transformation is modeled in each compartment based on pseudo-first-order rate constants taken from experimental or field data. These data are often scarce or highly uncertain, and reported values often range over orders of magnitude, especially for vegetation (*20*−*23*). Transformation in vegetation can be rapid and include photodegradation, a process that is well-known for pesticides applied to plant foliage (*24, 25*).

Interactions with Air. Diffusive chemical transport between compartments is based on the compartment fugacity and the mass transfer resistance between compartments. The resistance, based on a boundary layer model at the contact point with vegetation or soil, contains a concentration gradient defining the mass transfer rate between two compartments. The air compartment is assumed to be well mixed. For plants, we also assume a mass transfer resistance in the leaf cuticle. The diffusion between vegetation and air is based on two pathways in parallel, the stomata and the cuticle both in series with air (*15*). The conductance of chemicals through the stomata is correlated to the conductance of water vapor through the stomata (*26, 27*). The resistances for the diffusive processes are defined in Table 3.

Advective processes from air to vegetation include wet and dry particle deposition. The particle-bound fraction in air is estimated by the Junge equation (modified and incorporated into the fugacity capacity of the air, in Table

TABLE 2: Fugacity Based Transfer Factors

ground−air interactions, diffusive	**air flow**
$T_{ag} = \frac{\gamma_{ag}}{Z_a d_a}, \; T_{ga} = \frac{\gamma_{ag}}{Z_g d_g}$	$T_{\mathrm{aflow}} = \frac{u}{\mathrm{length}}$
air to ground, advective	**ground to air, advective**
$T_{ag} = \frac{\mathrm{rain} \times Z_{\mathrm{water}} + ((1 - If_d)v_d + (1 - If_w)v_w)Z_{ap}\frac{\rho_{ba}}{\rho_s}}{Z_a d_a}$	$T_{ga} = \frac{v_d Z_{gp}\frac{\rho_{ba}}{\rho_s}}{Z_g d_g}$
air to plant, diffusive	**plant to air, diffusive**
$T_{ap} = \frac{2 \times \gamma_{ap} LAI}{Z_a d_a} + \frac{g_{\mathrm{stomata}} LAI}{Z_a d_a}$	$T_{pa} = \frac{2 \times \gamma_{ap} LAI}{Z_p d_p} + \frac{g_{\mathrm{stomata}} LAI}{Z_p d_p}$
air to plant, advective	
$T_{ap} = \frac{(If_d v_d + If_w v_w)Z_{ap}\frac{\rho_{ba}}{\rho_s} + If_w Z_{\mathrm{water}}\mathrm{rain}}{Z_a d_a}$	
plant to ground surface soil, advective	**surface soil−root soil interactions**
$T_{pg} = \frac{(If_d v_d + If_w v_w)Z_{ap}\frac{\rho_{ba}}{\rho_s}}{Z_p d_p} + \frac{1}{180}$	$T_{gs} = \frac{\gamma_{gs}}{Z_g d_g} + \frac{\mathrm{recharge} \times Z_{\mathrm{water}}}{Z_g d_g}, \; T_{sg} = \frac{\gamma_{gs}}{Z_s d_s}$

1) (*28*). A representative dry-particle deposition velocity determines the dry deposition rate (*28*). The interception factor determines the fraction of particles landing on the vegetation versus landing on the soil and is calculated from plant biomass (*29, 30*).

The wet deposition of particles is calculated from the rainfall rate and washout ratio, a measured chemical dependent parameter relating the rate of particle deposition to the rainfall rate (*28*). The interception factor for wet deposition depends on the rainfall rate and the canopy water storage capacity (*31*). It is assumed there is no wet gaseous deposition to vegetation, because the water remains on the plant surface or is washed off to soil as opposed to penetrating the surface.

Advective interactions between air and ground surface soil include both wet and dry deposition of particles (similar to deposition to vegetation) and wet gaseous deposition (based on the concentration in rainwater determined by the Henry's law constant and the rainfall rate). The equations related to the advective processes are found in Table 3. Transfer rates for the system are in the range of experimental data (*32*). The mass-transfer resistances are based on a boundary layer in air and a soil mass transfer resistance, found in Table 3 (*17, 33*).

Vegetation−Soil Interactions. On an annual basis, the plant growth rate is assumed equal to the litterfall, harvest, senescence, and biomass increase rates. Litterfall and senescence are the processes of plant foliage falling to the soil as a result of shedding leaves and plant death, thus adding the chemical within the plant foliage to the surface soil, referred to collectively as litterfall in this paper (*30, 34*). We assume that litterfall is the dominant loss mechanism of plant biomass and thus litterfall equals the growth rate. By maintaining a constant volume, dilution resulting from growth is modeled as chemicals are removed through litterfall. The litterfall rate is assumed to be equal to one divided by the length of the growing season. Under steady-state conditions, particle deposition processes are balanced by washoff to the soil, thus maintaining a constant mass of particles on the leaves. It is assumed the particles on the foliage are in equilibrium with the plant tissue and are included in the fugacity capacity of the plant (*35*).

Soil Interactions. Soil is inherently heterogeneous and has a contaminant concentration gradient. To account for this, two soil layers are used: the surface soil and the underlying soil. Mackay (*16*) suggested using multiple vertical soil compartments or a boundary layer approach with a nonlinear concentration gradient from the surface to a specified soil depth. McKone (*17*) developed a soil model using a nonlinear "boundary layer" derived by best fitting a two-layer soil model to the solution of the dispersion/advection equation for mass transfer in soil layers. The model consists of two soil layers with transport between them defined by the effective chemical diffusivity in soil. The two compartments are assumed to be well mixed with a defined gradient at the interface.

Case Study. To demonstrate the calculation and evaluation of travel distance, we complete a case study using 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD), a compound that transforms both in air and vegetation (*20, 36−38*). Soil degradation is limited for this compound (*39, 40*). In the vapor phase of the atmosphere, OH radical reactions dominate degradation while particle degradation is negligible. The primary degradation process for TCDD in vegetation is the reductive dehalogenation by sunlight, requiring proton donors. Because plant lipids are rich sources of proton donors, higher degradation rates in vegetation are expected (*20, 37, 40*).

The representative values used for all landscape and chemical properties in calculating the characteristic length are listed in Supporting Information, Table 1. TCDD is

TABLE 3: Diffusion and Advection Parameters

Process	Equation	Reference
Air resistance over soil surface (d/m)	$\frac{\delta_{ag}}{D_{air}}$	(*17*)
Soil resistance -ground surface soil (d/m)	$\frac{\delta_g}{D_g} = \frac{0.108(10^{0.229 Log(D_g)})}{D_g}$	(*17*)
Soil resistance in root zone soil (d/m)	$\frac{\delta_s}{D_s} = \frac{318.4(10^{0.683 Log(D_s)})}{D_s}$	(*17*)
Diffusion coefficient in soil (m^2/d)	$D_{g,s} = \frac{\beta_{g,s}{}^{10/3} D_{water} Z_{water} + \alpha_{g,s}{}^{10/3} D_{air} Z_{air}}{Z_{g,s}(\alpha_{g,s} + \beta_{g,s})^2}$	(*33*)
Air resistance over plant surface (d/m)	$\frac{\delta_{ap}}{D_{air}}$	(*17*)
Conductance plant cuticle (m/d)	$g_{cuticle} = \left(\frac{10^{(0.704\, LogKow - 11.2)}}{Z_{water} / Z_{air}} \right) \times 86400$	(*15*)
Conductance- air and stomata of plant (m/d)	$g_{stomata} = \sqrt{18/MW} \times g_{water}$ $g_{water} = \frac{461 \times T \times Z_{air}}{\left((1 - rh) \times 611 \times 10^{\frac{7.7(T-273)}{(237+(T-273))}} \right)}$	(*26*)
Overall Soil –Air Resistance (mol/Pa-m^2d)	$\gamma_{ag} = \left(\frac{\delta_{ag}}{Z_a D_{air}} + \frac{\delta_g}{Z_g D_g} \right)^{-1}$	
Overall Plant –Air Resistance (mol/Pa-m^2d)	$\gamma_{ap} = \left(\frac{\delta_{ap}}{Z_a D_{air}} + \frac{1}{Z_p g_{cuticle}} \right)^{-1}$	
Overall Soil –Soil Resistance (mol/Pa-m^2d)	$\gamma_{gs} = \left(\frac{\delta_s}{Z_s D_s} + \frac{\delta_g}{Z_g D_g} \right)^{-1}$	
Interception factor –dry deposition (unitless)	$If_d = 1 - \exp(-2.8 \times bio_{inv})$	(*30*)
Interception factor –wet deposition (unitless)	$If_w = S_{bl} \times LAI \times e^{\left(\frac{-\ln 2 \times rain}{3 \times S_{bl}} \right)}$	(*31*)
Wet deposition (m/d)	$v_w = Wp \times rain$	(*28*)

typically released into the air from urban area sources but often contaminates suburban and rural sites as well (*3*, *4*). Airborne TCDD is mostly in the gaseous state and is emitted from incineration, combustion of fossil fuels, and industrial processes.

The characteristic length for TCDD was calculated using the four-compartment Lagrangian system described above and was found to be approximately 600 km. This value is on the same order of magnitude as, or greater than, the distance between urban centers and provides insight on the fairly uniform TCDD concentrations found in the northern hemisphere (*5*).

Figure 2 compares the concentration profiles for the following systems: a four compartment system (air, plant, two soils), an air/vegetation system, an air/soil system, and an air only system. From the comparisons, we find that including vegetation decreases characteristic length by an order of magnitude. This illustrates the importance of vegetation on regional transport for TCDD. In contrast, soil appears to have less of an effect on travel distance.

To understand these results, we determined the percentage of mass in each compartment as well as the percentage of total mass degraded in each compartment, as shown in Figure 3. Although the root-zone soil compartment contains the majority of the chemical mass, little mass is degraded in this compartment. In contrast, the vegetation compartment, with its rapid reaction rate, transforms a significant portion of the chemical, explaining the importance of including

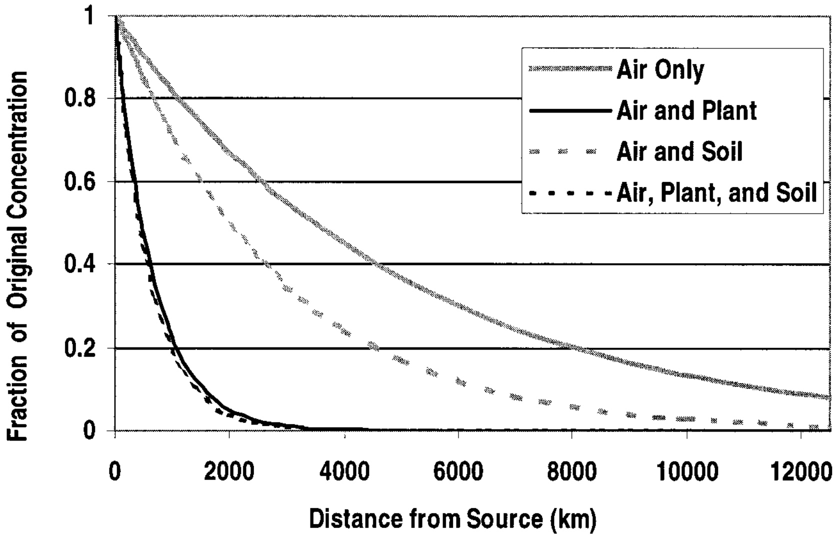

FIGURE 2. Fraction of original concentration plotted with distance for four environmental models, each for a case study using TCDD.

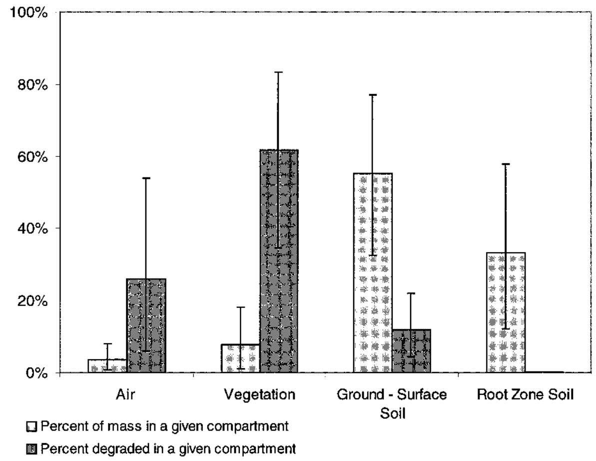

FIGURE 3. Percent of mass in each environmental compartment as well as the percent degraded in each environmental compartment at the mean, 10th, and 90th percentile for the TCDD case study.

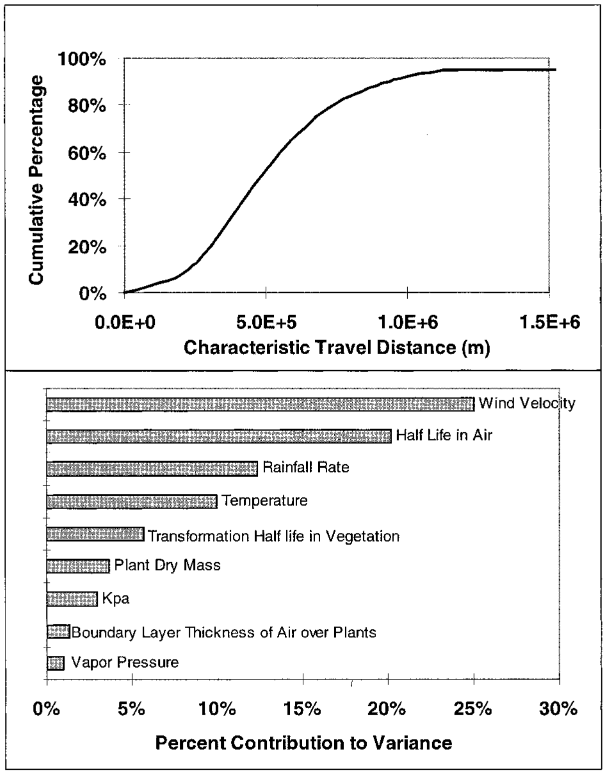

FIGURE 4. Cumulative probability distributions resulting from propagating uncertainty through a steady-state model with the corresponding contribution to variance of parameters that contribute significantly to the range of uncertainty for a case study using TCDD.

vegetation in the calculation for the CTD. There is also an analytical method for evaluating the importance of a compartment, which can be found in Supporting Information, Section 4.

An uncertainty analysis was completed for the case study. Using Monte Carlo techniques, we repeatedly calculated the CTD by randomly selecting values from the probability distribution of values for each parameter. All of the distributions are log-normal, with coefficients of variation defined in Supporting Information, Table 1. We used 5000 simulations to generate a distribution of plausible CTD values. The results are illustrated in Figure 4 and indicate that it is plausible for the CTD of TCDD to be anywhere 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 output uncertainty, the rank correlation coefficients between the CTD and its defining inputs are calculated. The rank correlation coefficients measure the strength of the linear relationship between each input and the CTD, considering both the range of uncertainty of an input and the influence of that input on the CTD. The rank correlation coefficients are squared and normalized to 100% to determine the approximate contribution to variance. The most influential parameters are displayed in Figure 4. Decreasing the uncertainty or variability in these parameters would have the largest impact for reducing the estimated range of the 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 range of characteristic lengths could be reduced as these values are refined.

To make an initial validation of the model, we employed a stationary system with an area of 250 000 km^2, a continuous emission of 1.1 g/d, and a windspeed through the system of

TABLE 4: Fugacity, Inventory, Concentration, and Mass Transformed for Each Compartment for a Fixed Region Using TCDD

compartment name	fugacity (Pa)	calculated concentrations (g/m^3)	measured concentrations (*42*) (g/m^3)	mass transformed (g/d)
air	2.02E−14	3.50E−15	3.60E−15	6.04E−02
vegetation	3.85E−15	1.33E−08	5.42E−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

4 m/s (*41*). These values are representative of the conditions in Germany, where the background concentrations have been measured (*42*). The calculated concentrations of TCDD appear to be in agreement with background measurements from Germany, as seen in Table 4 (*42*). We also compared the model to a 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 (*43*).

Discussion

The characteristic travel distance 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 distance is arbitrary and not defensible by any scientific analysis. In this paper, we have demonstrated simple methods for making preliminary estimates of the CTD which defines the effective range of impact for a chemical contaminant and is needed before deciding what scale to use when measuring or modeling the dispersion of an environmental contaminant. Thus, determining the CTD of a chemical is an important component in a number of analyses, including risk assessment, pollution prevention, health effects, pollutant mass balance, and regulatory impact studies.

The analysis of the travel distance of TCDD leads to two primary conclusions. First, the CTD exceeds the distance between urban centers, a possible explanation for the uniform concentrations of TCDD. This implies that TCDD should be regulated on a regional or global scale, as opposed to a local scale. Second, a significant quantity of TCDD is removed from the atmosphere by transport to, and subsequent degradation in, vegetation. Based on this analysis, vegetation is a potentially important compartment for transformation for some multimedia compounds. To date, this pathway has been either underestimated or excluded from most models, largely because of a lack of data on uptake and degradation rates. To further address these issues, there is a need for more data on the degradation rates as a function of chemical and vegetation types. This need is particularly important for chemicals posing a potential human health or ecological risk and for chemicals with a potentially long half-life in the environment.

As vegetation degradation rates become available, it is likely that properly accounting for both uptake to and degradation in vegetation could become a critical issue for environmental policies and regulatory actions. In particular, the spatial distribution and effective lifetime of persistent organic pollutants cannot be accurately characterized without more information on how these chemicals interact with vegetation. This need applies both to quantitative assessments of the regional and global behavior of a chemical and to evaluations of field data. In either case, conclusions about what drives regional changes in concentration could depend in large part on the assumptions made regarding the role of vegetation.

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Supporting Information Available

Section 1, derivation of characteristic travel distance; Section 2, mass balance equations to solve for mass in each compartment; Section 3, root zone soil depth; Section 4, analytical method for evaluating the importance of a compartment; and Table 1, chemical properties and landscape properties used in the case study (10 pages). Ordering information is given on any current masthead page.

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