

*Special Issue Honoring Don Mackay*

## MULTIMEDIA PERSISTENCE AS AN INDICATOR OF POTENTIAL FOR POPULATION-LEVEL INTAKE OF ENVIRONMENTAL CONTAMINANTS

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**Abstract**—Although intuitively it is apparent that population-level exposure to contaminants dispersed in the environment must be related to the persistence of the contaminant, there has been little effort to quantify this link formally. In this paper we investigate the relationship between overall persistence and/or overall residence time in a multimedia exposure environment and the population-level intake of contaminants as expressed by intake fraction (iF), the cumulative fraction of chemical emitted to the environment that is taken up by members of the population. We demonstrate that for any given contaminant and emission scenario the definition of iF implies that it is directly proportional to the overall multimedia persistence ( $P_{OV}$ ), or the overall multimedia residence time ( $T_{OV}$ ). The proportionality constant has dimensions of time and represents the characteristic time for population intake (CTI) of the chemical from the environment. We then apply the CalTOX fate and exposure model to explore how  $T_{OV}$  and CTI combine to determine the magnitude of iF. We find that CTI has a narrow range of possible values relative to  $T_{OV}$  across multiple chemicals and emissions scenarios. We use data from the Canadian Environmental Protection Act Priority Substance List (PSL1) Assessments and multimedia  $P_{OV}$  to show that exposure assessments based on empirical observation are consistent with interpretations from the model. Results indicate that  $P_{OV}$  derived from screening-level assessments of persistence, bioaccumulation potential, and toxicity (PBT) is a useful indicator of the potential for population-level exposure.

**Keywords**—Persistence Intake fraction Characteristic time/intake Exposure model Fugacity

## INTRODUCTION

It is an honor to contribute to this special issue of Environmental Toxicology and Chemistry devoted to the scientific career of Don Mackay. Much of Mackay's research is concerned with developing quantitative descriptions of the pathways followed by chemicals on their journey from release into the environment to their ultimate removal by chemical degradation or irreversible sequestration and their entry into human and ecological receptors along the way. Key concepts in this regard are the overall persistence ( $P_{OV}$ ) and overall residence time ( $T_{OV}$ ) of contaminants in the multimedia environment including atmosphere, hydrosphere, and lithosphere, as articulated by Mackay in 1979 [1]. Overall persistence is the residence time of a contaminant in a defined environment due to its degradation to other chemical species. It is related closely to the overall residence time of the contaminant in the environment ( $T_{OV}$ ), which incorporates removals by degradation and physical translocation (advection) out of the environmental system. Both  $P_{OV}$  and  $T_{OV}$  account for the different volumes of, and concentrations and removal rate constants in, the various environmental media. Assessment of  $P_{OV}$  has been an area of active research in recent years [2–6], in part because of international efforts to regulate persistent organic pollutants and regulatory actions designed to identify chemicals that are persistent, bioaccumulative, and toxic (PBT) [7].

Of particular concern to environmental scientists and policymakers is the potential of chemical contaminants to contact and enter humans or wildlife through various exposure pathways and, subsequently, impart adverse effects. The principles pioneered by Don Mackay have provided the basis for mul-

timedia contaminant fate models that have been extended to calculate multipathway exposures to environmental contaminants [8–10]. In his textbook on modeling techniques using the fugacity concept, Mackay states that the task addressed by the book is best summarized by a conceptual diagram showing the pathways followed by environmental contaminants through the environment and into contact with humans [11]. In this paper we use the CalTOX multimedia fate and multipathway exposure model [12–14] to explore the relationship between two aspects of environmental contaminants that are central themes in Mackay's research: Overall persistence and the potential dose to human populations.

The CalTOX model was developed at the University of California for use by the California Environmental Protection Agency in health risk assessments of multimedia pollutants that are exchanged among air, soil, surface water, sediments, and ground water. It has been used widely for chemical classification and multimedia risk assessments. CalTOX consists of a fugacity-based multimedia contaminant fate model linked to exposure models that provide estimated daily intake for a human population attributable to a pollutant source in air, water, and/or soil layers. The exposure models in CalTOX encompass 23 exposure pathways that relate contaminant concentrations in the multimedia model compartments to concentrations in media with which the human population has contact (i.e., personal air, tap water, foods, household dusts, soils, etc.). Average daily pollutant intake is calculated as the product of the exposure concentrations in these media and an intake factor (for inhalation and ingestion) or an uptake factor (for dermal contact) that relates the concentrations to the potential dose within the population.

The CalTOX model quantifies the source-to-exposure re-

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relationship for a system comprised of a pollutant defined by its chemical properties and release scenario, the environment within which it disperses, and the potentially exposed population. The link between source emissions and population exposure can be expressed as an intake fraction (iF), the fraction of material released from a source that eventually is inhaled or ingested. For environmental contaminants, the iF is the simplest possible expression of the source-to-intake relationship, which often is a complicated function of the attributes of the chemical, the environment, and the population. Evans et al. [15] recently have reviewed the development and use of iF in exposure assessments in a variety of applications, and Bennett et al. [16] have suggested a standard formalism for expressing the concept.

It is apparent intuitively that population-level exposure to a dispersed contaminant is a function of the contaminant's residence time in the exposure environment. If all factors except environmental persistence are identical, the cumulative population-level intake of a pollutant that is more persistent in the environment will be higher than intake of one that is less persistent. However, currently there is no quantitative explanation of the relationship between persistence or residence time in the exposure environment and population-level pollutant intake. The goals of this paper are to explore the nature of the relationships between  $P_{OV}$  and iF and  $T_{OV}$  and iF in a multimedia contaminant fate and exposure framework. We make use of both a multimedia, multipathway exposure model and data from the Canadian Environmental Protection Act Priority Substance List assessments (CEPA PSL1). As we demonstrate below, the definition of iF implies that it is directly proportional to  $P_{OV}$  for a closed environmental system and a defined chemical and release scenario. For an open environmental system, iF is directly proportional to  $T_{OV}$ . We note that the proportionality constant in these relationships has dimensions of time and thus represents the characteristic time for population intake (CTI) of the pollutant from the environment. We discuss the relative influence of  $P_{OV}$  or  $T_{OV}$  and CTI in describing population-level intake fraction, and the significance of  $P_{OV}$  as an indicator of potential for human exposure in screening-level environmental assessments for chemicals.

## THEORY AND METHODS

We derive the general relationship between residence time in the exposure environment and iF first for a simple case of a single-exposure medium and single-exposure pathway in order to make clear the concept and our approach. We then consider the more complicated case of multiple-exposure media and multiple-exposure pathways.

### *Single-exposure media and exposure pathway: Inhalation of airborne chemicals*

Evans et al. [15] derived equations that define iF for a conservative, volatile contaminant continuously emitted to a well-mixed volume of air over an urban region, with base area  $A$  (m<sup>2</sup>) and mixing height  $H$  (m). We refer to this volume of air as an air basin. In their example, air pollutants are removed from the system only by advection, with wind blowing at a constant velocity  $u$  (m/d) perpendicular to one of the sides of the air basin. Intake by the population of  $N$  individuals in the basin is by inhalation, with average breathing rate  $B$  (m<sup>3</sup>/d). For this simple model,

$$iF = (B \cdot N) / (u \cdot H \cdot A^{1/2}) \quad (1)$$

Equation 1 expresses intake fraction as the cumulative breathing rate of the population within the region (the numerator) divided by the ventilation rate of an air mass moving through the region (the denominator). The rate of chemical emissions does not appear in the equation because iF is not a measure of absolute exposure of the population. It represents the fraction of chemical put into the system that enters the lungs of the population.

The form of Equation 1 indicates that intake fraction can be viewed as being determined by the rates of two competing processes, intake of pollutants by the population, and irreversible removal from the environmental system. Put another way, iF describes how fast the population is inhaling air relative to how fast air is being flushed out of the region by the wind. This is readily apparent if the numerator and denominator are divided by the volume of the air shed over the region ( $A \cdot H$ ), thus converting the iF expression into the ratio of two rate constants with units of d<sup>-1</sup>.

$$iF = k_b / k_{adv} \quad (2)$$

where the numerator is the rate constant for population intake of air by breathing,

$$k_b = (B \cdot N) / (A \cdot H) \quad (3)$$

and the denominator is the rate constant for advective removal of chemical from the air shed.

$$k_{adv} = (u \cdot H \cdot A^{1/2}) / (A \cdot H) = (u / A^{1/2}) \quad (4)$$

### *Multiple exposure media and multiple exposure pathways*

For a multimedia environment consisting of  $i$  connected compartments representing various environmental media, the overall rate of clearance of contaminants from the system is a function of chemical partitioning and rates of degradation and advection in each compartment. An overall rate constant for clearance of a chemical from the system ( $k_{OV}$ ) can be defined [6] as

$$k_{OV} = \frac{\sum N_i k_i}{\sum N_i} \quad (5)$$

where  $N_i$  is the mass of contaminant in compartment  $i$  and  $k_i$  is the rate constant for removal from that bulk compartment, possibly the sum of rate constants representing advection and degradation processes in the compartment. The reciprocal of  $k_{OV}$  is the overall residence time in the multimedia environment

$$T_{OV} = 1 / k_{OV} \quad (6)$$

Under steady state conditions,  $T_{OV}$  also can be calculated as the ratio of the total inventory of contaminant in the environmental system,  $\sum N_i$  (g), and the emission rate,  $E$  (g/day) [11],

$$T_{OV} = \sum N_i / E \quad (7)$$

and, by definition,

$$iF = I / E \quad (8)$$

where  $I$  (g/day) is the cumulative, steady state, multipathway pollutant intake rate by the population. Multiplying both the numerator and denominator of the right hand side of Equation 8 by  $\sum N_i$  and rearranging yields,

$$iF = \frac{\sum N_i / E}{\sum N_i / I} = \frac{T_{OV}}{CTI} \quad (9)$$

where  $CTI = \sum N_i/I$ , and is defined as the characteristic time for intake of the contaminant by the population. The CTI is a function of the properties of the chemical of interest, the characteristics of the environment receiving emissions of that chemical, and the size and characteristics of the population living in that environment. As we show below, CTI can be characterized according to the dominant exposure pathway for the contaminant of interest. If the exposure environment is closed, i.e., if advective processes do not contribute to removal of contaminant from the environment, then  $iF$  can be expressed as the ratio  $P_{OV}/CTI$ .

In our earlier example of  $iF$  based on inhalation for a contaminant confined to the air shed over a region (Eqns. 1 and 2), we noted that  $iF$  was the ratio of the population breathing rate to air shed ventilation rate. With the introduction of CTI we can reinterpret  $iF$  in the single media example as the ratio of the residence time of air in the urban area, which is the measure of the effective lifetime of a molecule in the air shed, and CTI, a measure of the rate constant with which the molecule is taken into the lungs of a member of the population. We can extend this to the multimedia multipathway system with  $P_{OV}$  or  $T_{OV}$  representing the effective lifetime of a molecule in the multimedia region and CTI representing the overall rate constant for intake (or dermal penetration) of a molecule by the population through multiple exposure routes.

Equation 9 is an informative way of expressing  $iF$  because it makes clear that, for a given chemical in a defined system of environment and population,  $iF$  is directly proportional to the residence time of the contaminant in the environment. The longer a chemical resides in the environment the more likely it is to be taken up by a member of the population.

#### Model-based comparative exposure assessments

In this section we explore the relationships between  $iF$ ,  $T_{OV}$ , and CTI in a model system using the CalTOX model. An interpretation of empirical exposure assessment data using these concepts is presented below.

The CalTOX model [12] contains databases of physico-chemical properties, partition coefficients and media-specific degradation rates for 309 organic chemicals that are contemporary environmental contaminants, landscape parameters for various states and regions in the United States, and exposure factors for population cohorts based on gender and age. In order to explore the relationship between  $iF$  and overall residence time, we ran the CalTOX model for all 309 chemicals in the database using the archetypal "U.S." landscape parameters [12], representing the 48 conterminous United States with an assumed population of 280,000,000 persons, and the LCIA exposure factors, which are appropriate for assessing contaminant intake in the general population of an industrialized country [12]. In this example of contaminant intake by the population of the conterminous United States, the model system includes irreversible advective removal of contaminants from the exposure environment; therefore overall residence time ( $T_{OV}$ ) is the appropriate measure of the removal rate constant from the environmental system. The exposure pathway for population-level intake due to ingestion of breast milk by infants was neglected in this analysis of lifetime equivalent population exposure.

We ran CalTOX for each of the 309 organic chemicals emitted individually to air, water, and surface soil (927 total model runs). Model results gathered for each run included  $iF$  and  $T_{OV}$  (calculated as  $I/E$  and  $\sum N_i/E$ , respectively), CTI (cal-

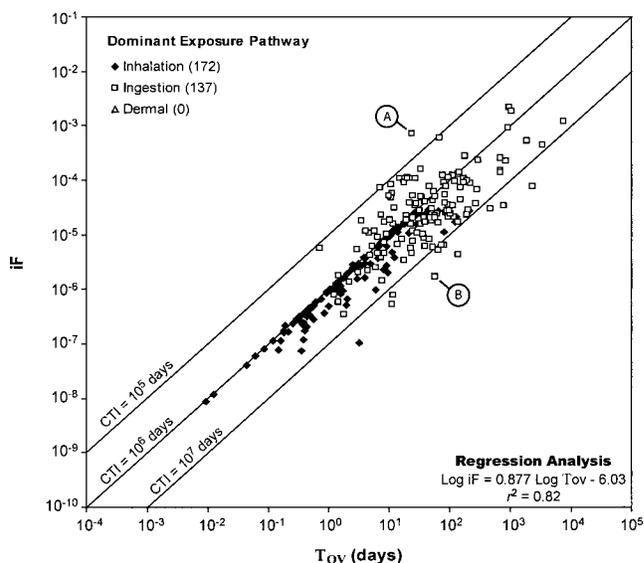


Fig. 1. Relationship between population intake fraction ( $iF$ ) and overall residence time ( $T_{OV}$ ) for 309 organic chemicals emitted to air in the conterminous 48 United States. Lines of constant characteristic time for intake (CTI) are indicated. Data points A (*di-n-octyl phthalate*) and B (*2,6-dimethylphenol*) are discussed in the text.

culated as  $\sum N_i/I$  and verified as numerically equal to  $iF/T_{OV}$ ), and exposure pathways data describing calculated cumulative population exposure by relative contributions from inhalation, ingestion, and dermal routes.

## RESULTS

We first consider results for emissions to air. Figure 1 shows the steady state relationship between  $iF$  and  $T_{OV}$  from CalTOX for the set of 309 chemicals continuously released to the atmosphere. Our regression analysis of these results indicates that 82% of the variability among chemicals in the logarithm of  $iF$  can be explained by the linear relationship with the logarithm of  $T_{OV}$ . The slope of the plot of  $\log(iF)$  versus  $\log(T_{OV})$  is close to unity. If the slope is assumed to be 1, the empirical relationship between  $iF$  and  $T_{OV}$  for this set of chemicals can be expressed as:

$$\log(iF) = \log(T_{OV}) - 6 (\pm 0.96) \quad (10)$$

or, equivalently,

$$iF = \frac{T_{OV}}{10^6} \left( \times / \div 9.2 \right) \quad (11)$$

Equation 11 implies that the median estimate of CTI for the 309 chemicals under this defined scenario is  $10^6$  d. Ninety-five percent of the actual CTI values calculated for the data set fall within a factor of 9.2 of this value. Three lines of constant CTI are plotted in Figure 1 for reference.

Inspection of Figure 1 reveals that  $T_{OV}$  for the 309 chemicals emitted to air spans five orders of magnitude and that the chemicals are distributed fairly evenly across this range. In contrast, CTI spans less than three orders of magnitude, and 296 of the 309 chemicals (96%) lie within an order of magnitude of the central estimate derived from regression analysis,  $10^6$  d, regardless of whether the dominant exposure pathway for the population is inhalation or ingestion of food and water.

In order to further explore this relationship, we consider the two outlying chemicals with similar  $T_{OV}$  values that give rise to short and long CTI relative to the set of 309 chemicals.

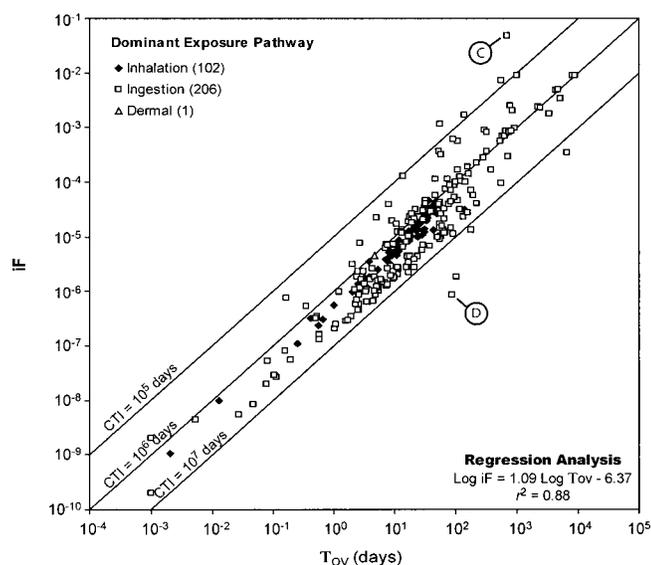


Fig. 2. Relationship between population intake fraction ( $iF$ ) and overall residence time ( $T_{OV}$ ) for 309 organic chemicals emitted to surface water in the conterminous 48 United States. Data points C (Aldrin) and D (benzo[*a*]pyrene) are discussed in the text. CTI = characteristic time for population intake.

These chemicals are, respectively, di-*n*-octyl phthalate (point A in Fig. 1) and 2,6-dimethylphenol (point B in Fig. 1). Di-*n*-octyl phthalate is extremely hydrophobic and has low volatility ( $\log K_{OW} = 9.5$ ,  $\log K_{OA} = 13.8$ ). In the atmosphere it partitions almost entirely to aerosols and is deposited to vegetation, soil, and surface water by wet and dry deposition. In the CalTOX assessment its dominant pathway for human exposure is deposition to vegetation with transfer to agricultural products including meat, milk, and eggs, and subsequent ingestion. The short CTI for di-*n*-octyl phthalate, therefore, is attributable to its high potential for bioaccumulation in the terrestrial food chain. At the opposite extreme is 2,6-dimethylphenol, which is somewhat soluble in water and semivolatile ( $\log K_{OW} = 2.4$ ,  $\log K_{OA} = 6.1$ ). Its exposure pathways for emissions to air are inhalation (35%), transfer to exposed produce followed by ingestion by the population (29%), and ingestion of drinking water (27%). However, greater than 99% of the steady state inventory of 2,6-dimethylphenol is sequestered in soils and groundwater and pathways for transfer from the subsurface environment to exposure media are inefficient. The inventory of contaminant in soil, therefore, controls  $T_{OV}$ , but does not make a dominant contribution to population intake.

Figure 2 shows  $iF$  as a function of  $T_{OV}$  for emissions to surface water. As was the case for emissions to air, a regression analysis yields a slope near unity. Three hundred of the 309 chemicals (97%) have CTI within an order of magnitude of  $10^6$  d, while  $T_{OV}$  spans seven orders of magnitude. The chemical with the shortest CTI for emissions to water is Aldrin (point C in Fig. 2). Aldrin is highly hydrophobic ( $\log K_{OW} = 6.9$ ) and has the highest aquatic bioconcentration factor (BCF) among chemicals in the CalTOX database. The dominant pathway for population exposure to Aldrin emitted to surface water is by ingestion of fish, which accounts for 99.8% of total intake. Benzo[*a*]pyrene (BaP) has the longest CTI in this scenario (point D in Fig. 2). Although BaP has similar hydrophobicity to Aldrin ( $\log K_{OW} = 6.3$ ), its BCF is low because it is assumed to be metabolized by fish. The dominant exposure

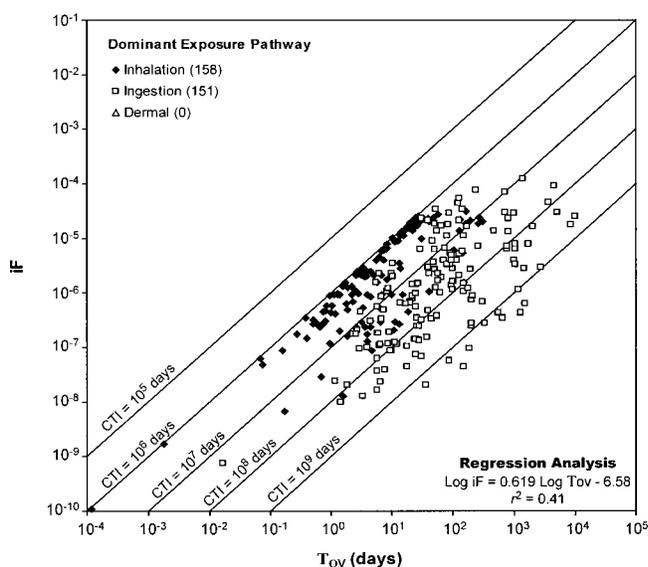


Fig. 3. Relationship between population intake fraction ( $iF$ ) and overall residence time ( $T_{OV}$ ) for 309 organic chemicals emitted to surface soil in the 48 conterminous United States. CTI = characteristic time for population intake.

pathway for BaP emitted to water, therefore, is by ingestion of tap water, which accounts for its long CTI relative to other chemicals in the database.

Figure 3 shows  $iF$  as a function of  $T_{OV}$  for emissions to the surface soil compartment. In this case, we do not see an obvious linear relationship between  $\log(iF)$  and  $\log(T_{OV})$  for the group of 309 chemicals. The absence of the linear relationship is notable especially among chemicals for which ingestion is the dominant pathway for exposure. We believe this is the case because many of the chemicals are not mobilized efficiently from soil into the atmosphere, surface water, or vegetation where they can be consumed in drinking water or food products. Stated another way, the CTI of contaminants that are sequestered in soils is long relative to the same chemicals in air or surface water. Chemicals that efficiently volatilize from soils and have inhalation as the dominant exposure pathway have CTI that is comparable to the other emission scenarios because the rate-limiting process for their uptake is the breathing rate of the population.

In order to determine whether degradation in a specific compartment was controlling the calculated  $iF$ , we compared  $iF$  against the media-specific half-lives used as inputs to the CalTOX model for the 309 chemicals. This comparison is illustrated in Figure 4 using results for emissions to air only. These plots clearly show little or no relationship between  $iF$  and the degradation rates in specific media. Regression of  $\log(iF)$  versus degradation half-lives in water, surface soil, and sediment yielded  $r^2$  less than 0.04 in all cases.

A relationship is evident between  $iF$  and degradation half-life in air, with two distinct regions where the residence time of the contaminants in the atmosphere is dominated by degradation and advective removal. However, for contaminants emitted to air with ingestion as the dominant exposure pathway, degradation half-life in air essentially is uncorrelated with  $iF$  for the population. The results shown in Figure 4 for emission to air are consistent with other emission scenarios. Within limited ranges of environmental behavior and exposure pathway we have found some apparent dependence of  $iF$  on media-specific degradation, but in general degradation half-lives in

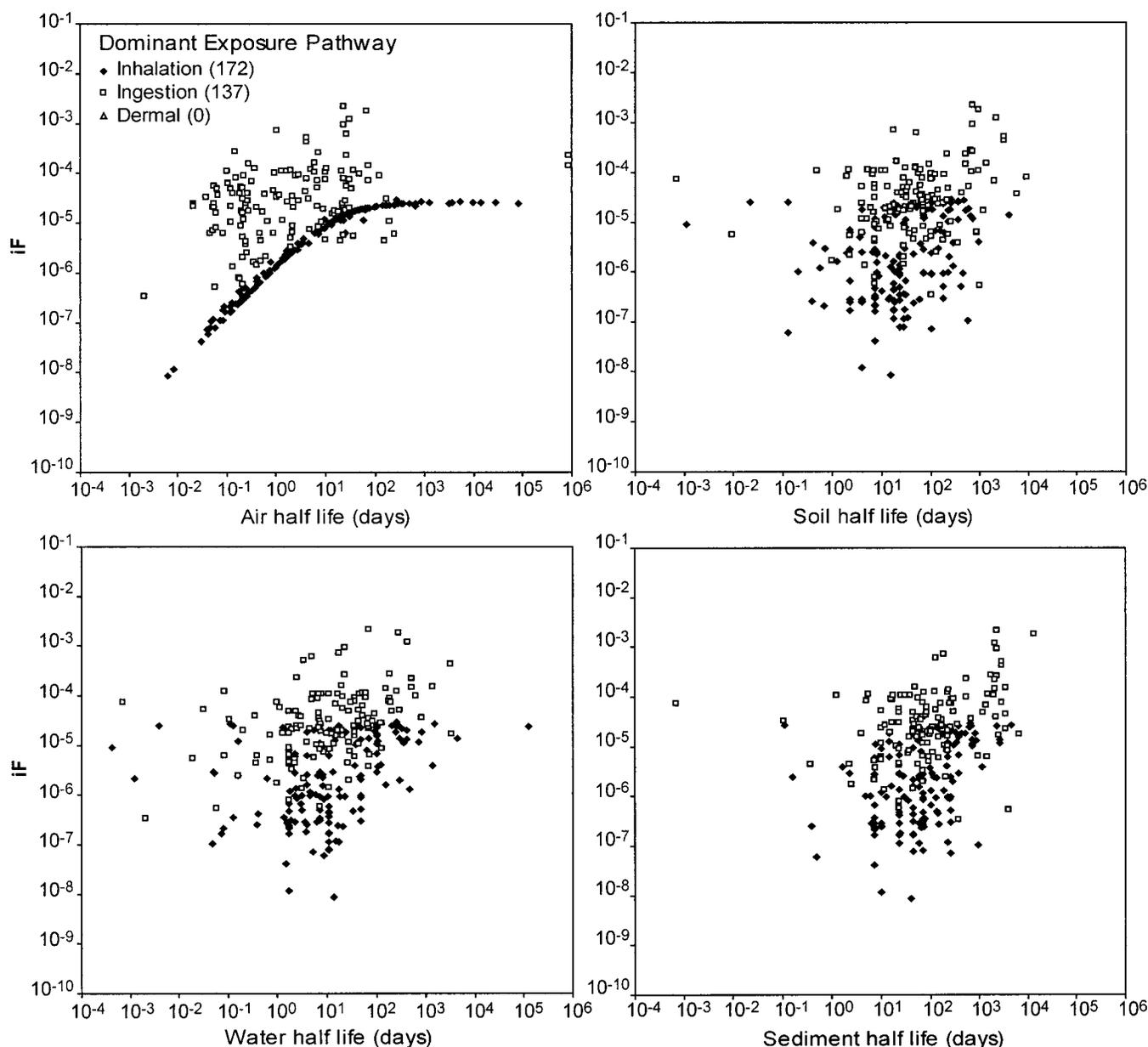


Fig. 4. Relationships between population intake fraction (iF) and degradation half-lives in individual media (air, water, soil, and sediment) for 309 chemicals emitted to air in the 48 conterminous United States.

individual media are not robust indicators of potential for population exposure.

#### Evaluation of empirical exposure assessment data

Because the forgoing analysis is based on model results, it raises the question of whether the relationship between iF and  $T_{OV}$  and/or  $P_{OV}$  can be observed from one or more sets of empirical observations. Few comprehensive assessments of emissions and human exposure have been reported that allow empirical calculation of iF. As a result, a direct confirmation of the relationship is difficult to construct from monitoring data or experiments. However, we are able to carry out an evaluation of the proposed relationship from an independent compilation of emissions and population exposure estimates carried out under the Canadian Environmental Protection Act (CEPA). As an initial evaluation of the model-based premise, we examined the extent to which the CEPA assessment confirms or refutes the relationship between iF and  $P_{OV}$ .

#### CEPA PSLI assessments

Under the CEPA, detailed assessments of emissions, environmental fate, and human exposure have been carried out for a set of priority substances [17]. The goal of these assessments is to determine whether the identified substances enter the Canadian environment in quantities sufficient to constitute a danger to human life or health. Environment Canada and Health Canada have collected and reviewed data relevant to rates of emission and human exposure to substances on the CEPA priority substances list (PSL) to support these assessments. A review of the CEPA PSL reports provides sufficient information to estimate iF for the Canadian population for the 14 chemicals listed in Table 1, using the following equation:

$$iF = \frac{I}{E} \cdot \frac{70 \times 30,000,000 \cdot 365}{10^9} \quad (12)$$

where  $I$  (ug/kg/day) is the estimated daily intake of chemical

Table 1. Canadian Environmental Protection Act Priority Substance List substances with sufficient data in the assessment to calculate intake fraction (iF) for the Canadian population. ( $K_{OW}$  = octanol–water partition coefficient,  $K_{OA}$  = octanol–air partition coefficient,  $K_{AW}$  = air–water partition coefficient,  $P_{OV}$  = overall persistence)

Chemical	Log $K_{OW}$	Log $K_{OA}$	Log $K_{AW}$	Degradation half-lives (h)			
				Air	Water	Octanol	$P_{OV}$ (h)
Benzene	2.18	2.81	-0.63	142	267.6	4,565	142
Chlorobenzene	2.81	3.76	-0.95	396	2,620	1,790	404
1,2-Dichlorobenzene	3.45	4.54	-1.09	840	2,500	5,320	890
1,4-Dichlorobenzene	3.49	4.46	-0.97	924	2,800	12,500	973
Tetrachlorobenzenes	4.50	5.92	-1.42	4,580	1,700	1,700	2,330
Pentachlorobenzene	5.00	6.55	-1.55	6,530	6,210	6,210	6,260
Hexachlorobenzene	5.54	6.91	-1.37	20,300	36,800	11,200	11,600
3,3'-Dichlorobenzidine	3.58	9.57	-5.99	0.05	0.05	2,500	0.200
1,2-Dichloroethane	1.45	2.76	-1.31	1,080	19,600	2,610	1,120
Dichloromethane	0.90	1.38	-0.48	8,090	420	420	7,290
Trichloroethylene	2.50	2.96	-0.46	83	2,920	22,300	83.6
Methyl methacrylate	1.38	3.24	-1.86	1.98	269	420	2.27
Toluene	2.68	3.26	-0.58	57	312	682	57.5
Xylenes	3.11	3.67	-0.56	23.3	420	363	23.6

by adult Canadians and  $E$  (kg/y) is the estimated release rate to the Canadian environment. Both  $I$  and  $E$  were reported for the 14 chemicals in the CEPA PSL1 reports. The constant in this expression converts iF to a dimensionless value by assuming a population of 30 million people in Canada and an average bodyweight of 70 kg. To ensure the intake rate used in the calculation was representative of intake due to environmental contamination, estimates of intake by inhalation of indoor air or during specific activities such as smoking or refueling a car were not included in the calculation of iF.

Overall persistence was calculated for the 14 chemicals in Table 1 using the equilibrium partitioning approach and generic environmental parameters described by Gouin et al. [18]. Partition coefficients and estimated degradation rate constants in air, water, and soil required for the  $P_{OV}$  calculations were taken from the CalTOX database [12].

#### CEPA PSL1

Figure 5 shows iF plotted against  $P_{OV}$  for the 14 chemicals from CEPA PSL1. Error bars represent the upper and lower boundaries of iF in cases where ranges of emissions and/or intakes were reported in the CEPA reports. These results, based on real-world estimates of emissions and contaminant intake, are consistent with the model results presented earlier. Linear regression of the CEPA-derived relationship between iF and  $P_{OV}$  yields a slope that is not statistically different from one and a best estimate of CTI of approximately  $10^6$  d. Although this analysis is not a strict validation of the relationship between iF and  $P_{OV}$ , it represents an independent evaluation from empirical data, accounts for the impact of different assumptions, and provides reasonable confirmation of the approximate relative magnitude of variability in CTI and  $P_{OV}$  between environmental contaminants.

#### DISCUSSION

We present an interpretation of iF as the ratio of rate constants describing two competing processes: Removal of contaminant from the environmental system (characterized by  $T_{OV}$  or  $P_{OV}$ ) and intake by the population (characterized by CTI). The present definition is consistent with other conceptualizations of iF as the ratio of cumulative intake rate to emission rate in exposure assessments [16] and as proportional to the ratio of residence time divided by dilution volume for inha-

lation of contaminated air [15]. Analysis of model results and empirical exposure assessment data indicate that the relationship between log iF and log  $T_{OV}$  or log  $P_{OV}$  in comparative assessments of groups of chemicals is linear with a slope near unity. This implies that interchemical variability in residence time in the exposure environment is larger than interchemical variability in CTI and, therefore,  $T_{OV}$  and/or  $P_{OV}$  can be treated as surrogate measures of potential for population intake in screening-level comparative assessments of chemicals.

The present definition explicitly separates iF into an environmental fate component (represented by  $T_{OV}$  or  $P_{OV}$ ) and a population-level exposure component (represented by CTI). Both  $P_{OV}$  and/or  $T_{OV}$  are functions of chemical partitioning

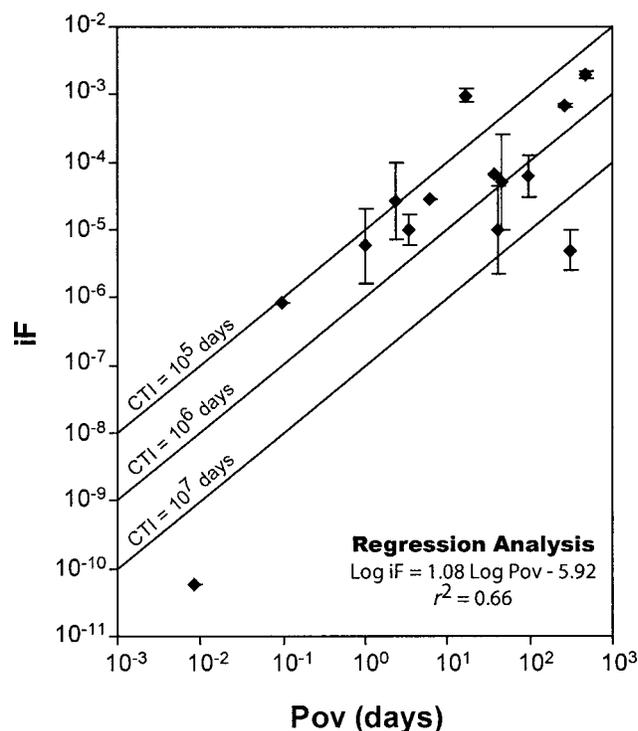


Fig. 5. Relationship between population intake fraction (iF) and overall persistence ( $P_{OV}$ ) for 14 chemicals from the Canadian Environmental Protection Act Priority Substance List Assessments. CTI = characteristic time for population intake.

properties, degradability and properties of the environmental system. The CTI is a function of chemical properties, population characteristics and behavior patterns, and the efficiency of transfer of contaminants from the environmental system into media with which the population has direct contact. Viewed in these terms, it is apparent that chemicals can have a relatively high potential for population exposure for two reasons: They are highly persistent in the exposure environment, or they are transferred efficiently into a pathway for population exposure and intake. The present definition of iF generally is applicable, as we have demonstrated using datasets derived from a fate and exposure model and a real-world population level exposure assessment. These techniques are particularly meaningful when interpreting the results of multimedia fate and multipathway exposure assessments in which there is simultaneous transfer and accumulation of contaminant in air, water, soils, and sediments, and exposure by a combination of inhalation, ingestion, and dermal contact.

In the model-based analysis of a representative group of chemicals emitted to air or surface water in the United States,  $T_{OV}$  varies over seven orders of magnitude and CTI, with only a few exceptions, is restricted to a two order of magnitude range. Differences in CTI are evident for different exposure pathways and for different emission scenarios, but these differences are not as dramatic as the range in  $T_{OV}$  due to inter-chemical differences in degradability and partitioning. These results indicate that, to a first approximation,  $T_{OV}$  in the exposure environment can be viewed as a surrogate measure of potential for population exposure in comparative screening assessments of chemicals. Because iF is the ratio of  $T_{OV}$  and CTI, the range of variability in the data imply that one can predict iF from  $T_{OV}$  with an expected uncertainty of approximately two orders of magnitude. However, from the case studies in this paper, we show in practice that the residual error in the  $T_{OV}$  to iF relationship is one order of magnitude or less. In contrast, relationships between population-level iF and media-specific degradation half-lives are not robust, and therefore assessment of  $P_{OV}$  or  $T_{OV}$  in screening-level persistence, bioaccumulation potential, and toxicity assessments provides additional information about potential for population exposure that is not available by screening against defined half-life criteria only.

For emissions to soil in the United States, the direct relationship between  $T_{OV}$  and iF breaks down, especially for chemicals that remain entrained in soil. Under this scenario, CTI plays a more important role in determining population iF than in the case of emissions to air or water. To some extent this result is a consequence of the environmental boundaries defined in the assessment. The model environment used in the assessment includes soils at depths where contaminants are considered to be in active circulation in the environment, but are unlikely to contact a member of the population, as evidenced by long CTI.

Typical characteristic times for intake calculated for the United States and Canadian populations in these sample assessments are  $10^6$  d, or approximately 2,700 years. The magnitude of the median CTI determined in comparative assessments will be sensitive to landscape properties, population density, and exposure factors used in the assessment. The extremely long CTI reported here should not be misinterpreted as indicating that cumulative contaminant intake will be spread out over several generations. The CTI is a measure of the effective rate constant with which the population takes in en-

vironmentally dispersed chemicals under the defined environmental and population scenario. The correct interpretation of the long CTI is that it would take several hundred years for the population to clear the contaminant burden from the environment by taking it into their bodies through all exposure pathways. From this it is clear that human intake is not a primary mechanism for removal of chemicals from the environment. If it were, iF values would be much larger.

We have focused in this paper on assessing iF for human populations on a national scale. The relationship between  $T_{OV}$  and/or  $P_{OV}$  and iF that is suggested in this study should be further explored and evaluated using other data sets and models. Similar analysis for chemical intake by other species, such as predatory birds or sport fish, is possible, and could provide interesting insights into the relative contribution of contaminant fate and efficiency of exposure pathways in determining the intake of contaminants by wildlife. Similar concepts to those presented here have even been applied to the intake of contaminants by entire ecological systems, as demonstrated by calculations of the efficiency of transfer of contaminants to the Laurentian Great Lakes from different regions of North America [19].

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