Screening for Persistent Organic Pollutants: Techniques To Provide a Scientific Basis for POPs Criteria in International Negotiations

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The United Nations Environment Programme (UNEP) is currently coordinating negotiations to develop a binding global agreement by late in the year 2000 to prohibit, restrict, or reduce the production, use, or release of certain persistent organic pollutants (POPs). POPs are a small subset of organic chemicals whose characteristics of persistence in the environment, accumulation in biological organisms, and toxicity make them priority pollutants and environmental risks to humans and ecosystems. Under the UNEP negotiation, representatives are developing criteria and procedures for the addition of substances, guided by the initial list of 12 substances or substance groups selected for global action. It is therefore timely to investigate the scientific foundation for POPs screening criteria that have been used in other international, regional, and national programs, focusing on the properties of persistence, bioaccumulation, toxicity, and long-range transport in a policy context. The theoretical, empirical, and multimedia modeling approaches used reveal that guidance for setting POPs screening criteria can be developed using a combination of science and policy input. These approaches suggest that criteria adopted under regional POPs agreements in North America and Europe are reasonable and tend to isolate a limited number of clearly hazardous POPs from the majority of organic chemicals, while not being so stringent that the ability to respond to as yet unidentified risks is seriously compromised.

In 1998, nations throughout the world began negotiations under the United Nations Environment Programme (UNEP) on a binding global agreement to prohibit, restrict, or reduce the production, use, or release of certain persistent organic pollutants (POPs). Risks posed by these chemicals may occur far from the site of initial entry of the POP into the

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environment and include effects in remote, polar, and oceanic regions of the planet (*1–3*). International negotiations have been instituted in response to concerns about the transborder risks posed by POPs and the inability of affected nations to regulate far-off emission sources. The UNEP action is the global counterpart to similar, regional negotiations completed on a POPs protocol under the United Nations Economic Commission for Europe (UNECE) Convention on Long-Range Transboundary Air Pollution (LRTAP) (*4*). In North America, POPs are being addressed under the North American Agreement for Environmental Cooperation (NAAEC) Commission for Environmental Cooperation (CEC) (*5*) and the bilateral United States/Canada agreement to control the discharge or release of POPs in the Great Lakes Basin (*6*).

Clarifying what constitutes a POP warranting international action is central to these negotiations, along with how best to screen and select these substances. The ultimate determinant of whether a substance requires international action under the UNEP agreement is risk to human health or the environment, such that global action is warranted. This determination of risk is complex and is facilitated by the application of screening criteria to prioritize substances for more detailed assessment. Commencing with draft proposals by a criteria expert group (CEG) constituted under the UNEP POPs negotiation (http://www.chem.unep.ch/pops), representatives are developing criteria and procedures for the addition of substances, guided by the initial list of 12 substances or substance groups selected for global action [UNEP/GC.18/32, 1995: DDT, dieldrin, aldrin, endrin, chlordane, heptachlor, mirex, toxaphene, hexachlorobenzene, PCBs, polychlorinated dibenzo-*p*-dioxins, and polychlorinated dibenzofurans]. It is therefore timely to investigate the scientific foundation for POPs screening criteria that have been used or proposed in other international, regional, and national programs (Table 1), focusing on the properties of persistence, bioaccumulation, toxicity, and long-range transport in a policy context.

In this paper, we seek to determine how scientific methods and data can best be used to develop POPs screening criteria, asking the following questions: Are persistence criteria thresholds supported by theoretical analyses of environmental fate properties? How can analysis of the properties of candidate substances contribute to the development of criteria? Is there a scientific basis for determining if exposures following long-range transport pose a health risk? And, can multimedia analysis help determine whether criteria, particularly those developed for the UNECE and CEC, are consistent with estimates of the potential for long-range transport? In asking these questions, we will analyze how science can inform the development of screening criteria in the specific policy context of the UNEP global POPs negotiation.

Theoretical Approaches To Determining POPs Persistence Criteria

An initial approach to setting persistence criteria is a theoretical analysis to determine if thresholds exist beyond which a substance could be considered a candidate POP warranting international action. Figure 1a plots the accumulation over time of two hypothetical chemicals with half-lives of 1 and 12 months, in either soil, water, or sediment. This simple analysis assumes first-order or pseudo-first-order kinetics and that no dispersion pathways are operating. Two modes of release to the environment are shown for each chemical. The first assumes a single release of one hypo-

TABLE 1. National and International Criteria for POPs

	long-range transport[a]			persistence			bioaccumulation		toxicity
	remote measurements	vapor pressure (Pa)	air half-life (days)	water half-life (months)	soil half-life (months)	sediment half-life (months)	BAF/BCF	log K_{ow}	
UNECE−LRTAP 1998 (4)[b]	X	or <1000[c]	& 2	2	or 6	or 6	5000	or 5	risk profile
NAAEC−CEC 1997 (5)	X	or <1000[c]	& 2[c]	6	or 6	or 12	5000	or 5	mutual concern
Canada TSMP 1995 (26)	X		or 2[c]	6	or 6	or 12	5000	or 5	CEPA defined
U.S. EPA 1998 TSCA PBT-ban pending testing				6			5000		develop toxicity data
U.S. EPA 1998 TSCA PBT-release controls (27)				2			1000		develop toxicity data
IJC 1993 immediate action (28)				2			or 5000		or chr. aq. tox. <0.1 µg/L
IJC 1993 initial screen				0.23			or 1000		or chr. aq. tox. <1 µg/L
CMA PTB policy 1996 (29)[d]			5	6	or 12		5000 or calculated, prof judgment		expert judgment; risk assessment

[a] The "&" terminology should be applied before the "or" term, such that a chemical must have an air half-life greater than 2 days plus the vapor pressure requirement, or this combination could be substituted by remote measurements. [b] The UNECE−LRTAP POPs agreement states that alternative criteria may be considered if there is evidence that the substance is otherwise sufficiently persistent or bioaccumulative to make it of concern within the scope of the Convention. [c] The air half-life is sufficient for meeting the persistence requirement. Vapor pressure maxima were incorporated to exclude highly volatile substances. [d] The Chemical Manufacturers Association (CMA) considers long-range transport to be additional to the definition of a PTB chemical.
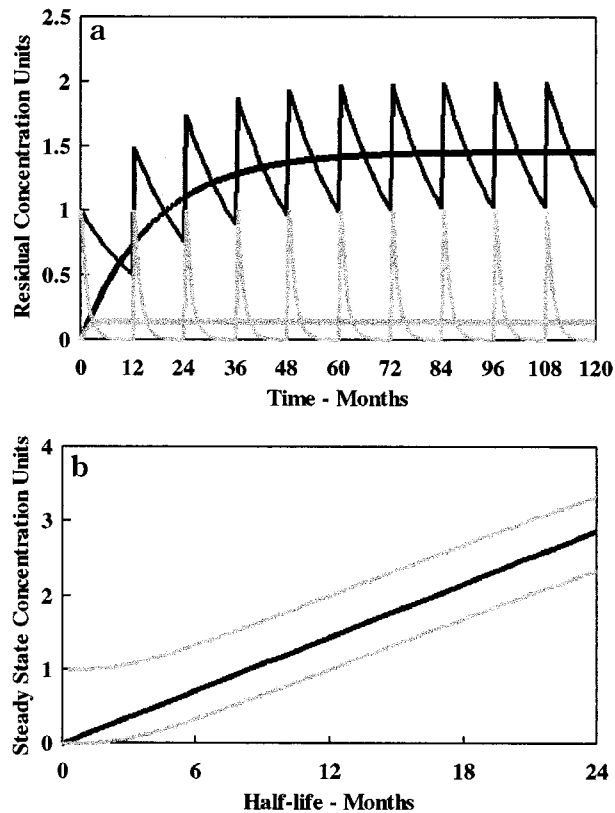

FIGURE 1. Relationship between half-life, accumulation, and steady-state concentration. Panel a graphs the accumulation over time of two theoretical chemicals released at a rate of 1 unit per year, commencing at time zero. The lower graph represents a chemical with a half-life of 1 month; the upper graph represents a half-life of 12 months. Release occurs either at the start of each year (peak/trough appearance) or continuously throughout the year (center line). Panel b graphs the relationship between chemical half-life and the steady-state concentration resulting from a 1-unit annual release rate. The upper and lower lines represent the peak and trough levels at steady state from repeated release at the start of each year. The central line is the steady-state concentration resulting from continuous release over the year.

thetical unit of chemical at the start of each year, and the second assumes a continuous release totaling one unit per year.

The relationship between chemical half-life and concentration at steady state is shown in Figure 1b. For the continuous release scenario, the steady-state concentration (C) passes through the origin and is linearly proportional to the half-life ($T_{1/2}$), following the equation $C = RT_{1/2}/\ln 2$, where R is the release rate. For the annual release scenario (upper and lower lines) in Figure 1b, a similar approximately linear increase is also evident above half-lives of 4−5 months. Below 4−5 months, however, a concave curve is evident, and the residual concentration can approach zero before the next annual release. For substances with half-lives of 6 months or more, the background residue never goes below approximately one-third of the annual release. This is significant for a pesticide in agricultural use, because a persistent residue equivalent to one-third or more of the annual application can be assumed to be potentially hazardous, given that the annual dosage unit is intended to be toxic. On the other hand, for a POP subject to long-range transport, the proportion of the annual release that actually ends up being deposited in remote locations is likely to be far below the toxic level. Whether such a residue constitutes a risk depends on the degree of bioaccumulation and persistence from source to site of deposition being sufficient

to elevate the dose to a toxic level. The more persistent, bioaccumulative, and mobile a chemical, the more likely this is to occur. Hence, no clear threshold criterion for persistence in a long-range context can be deduced using the theoretical approach alone.

Persistence in a Transport Medium

The analysis of persistence in a transport medium, predominantly air for POPs, can differ from the soil/water analysis because it incorporates a finite time limitation, namely, the time necessary for a substance to move from source to site of deposition. The key question is how long a substance needs to remain airborne to constitute a problem warranting international action. This time period is directly related to the geographic scale of interest. For a global negotiation, that scale would presumably be at the trans-oceanic or transcontinental level. Assuming a scale of ca. 4000 km (2500 mi), it can be shown that approximately $7-10$ days would be required for atmospheric transport from source to site of deposition. This is based on average air movement rates across the United States of 7 m/s (7) and computer modeling of air movement on a global scale (8). For a chemical with a 2-day degradation half-life in air, the amount remaining after this approximate 8-day period is $1/16$ (2^{-4}) of the original release. Lower atmospheric half-lives lead to considerably smaller residual amounts after 8 days, due to the shape of the mathematical relationship between the proportion of chemical remaining at time t (m_t/m_0) and the degradation half-life ($\tau_{1/2}$) [$m_t/m_0 = \exp(-\ln(2)t/\tau_{1/2})$]. This suggests that a 2-day half-life may be an appropriate screening criterion for degradation in air.

Empirical Analysis of Degradation Rates and Bioaccumulation

Another approach to setting screening criteria for POPs is to look for trends in measured laboratory and field data for substances already widely acknowledged to be of concern as well as other substances for which data are available. These data can then be compared to proposed criteria guidance values. For this purpose, we initially retrieved degradation and bioaccumulation data on a suite of organic chemicals compiled from U.S. EPA and international lists of priority toxic substances (6, 9–12). From this list, a subset was selected based on the availability of measured environmental degradation and bioaccumulation data. All bioconcentration/bioaccumulation factors (BCF/BAFs) were calculated from measurements in fish. U.S. EPA Great Lakes Water Quality Initiative (13) values were preferred over other lipid-adjusted BCF/BAFs, which in turn were preferred over results from studies that did not measure the lipid content of the fish. All lipid-based BCF/BAFs were normalized to a fish lipid content of 5%. Where more than one equally preferable value existed in a category, the value presented is the arithmetic average of these results. Soil and water degradation data were obtained from laboratory and field studies. Half-lives are based on estimates of probable high and low rates for the most important degradation process within a particular medium, following published methodology (14). Data for structural analogues were used in rare instances where a clear relationship between chemicals existed. Atmospheric half-lives for this analysis were derived from computer models (15).

The findings for soil degradation and bioaccumulation (BCF/BAF) are shown in Figure 2. This graph illustrates the approximate distribution of POPs and other organic chemicals relative to one another and to corresponding screening criteria that were adopted in previous negotiations (Table 1). Degradation half-life values in soil were located on the graph by centering the chemical name on the arithmetic

mean of the upper and lower half-life estimates for the chemical. The full set of soil-degradation half-lives and bioaccumulation values for each chemical are tabulated in the Supporting Information along with the citations for each.

All 12 UNEP POPs (marked in red in Figure 2) or their POP transformation products (aldrin converts to dieldrin, heptachlor to heptachlor epoxide) exceed degradation and bioaccumulation screening criteria adopted under the NAAEC−CEC and UNECE−LRTAP processes, often by large margins. The extent to which these POPs exceed the screening criteria is actually obscured by the truncation of soil half-lives (necessary for ease of presentation) and the logarithmic scaling of the BCF/BAF axis. Figure 2 also demonstrates that, of the substances included in this exercise, few other than the 12 UNEP POPs clearly meet the criteria adopted by the NAAEC and the UNECE, although considerable data gaps exist for a number of chemicals considered in this analysis. Similar findings are evident from graphs of bioaccumulation vs degradation half-life in water (not shown) and from graphs of overall environmental persistence generated by multimedia fate models (see below). This approach serves to identify several additional substances or substance classes that may warrant further consideration under UNEP, based on similar degradation and bioaccumulation properties to the substances under negotiation. These include octachlorostyrene, isodrin, chlordecone (kepone), polybrominated biphenyls, and methyl mercury, the latter cycling through inorganic phases.

Incorporating Toxicity

Screening for toxicity necessarily involves a preliminary assessment of potential dose. For this purpose and given the global nature of the UNEP negotiation, a scoping exercise was performed on the human health risks posed to isolated Arctic Inuit populations by the 12 UNEP POPs, based on available dietary intake studies (16, 17). Where information was lacking, dietary intake was estimated based on measured POP levels in Arctic marine mammal blubber and assuming a daily intake of 60 g of marine mammal fat by Inuit adults. The U.S. EPA cancer slope factors and noncancer reference doses (RfDs) were used to compare dietary intake to levels typically considered acceptable. Additional details on the cancer and noncancer risk calculations are found in the Supporting Information.

Figure 3 suggests that average Inuit doses of many of the 12 UNEP POPs may exceed levels considered safe. The upper-bound cancer risk estimates in this scoping exercise are based on the upper 95th percentile confidence limit on the lifetime cancer risk derived principally from rodent bioassays. The true cancer risk from individual pollutants probably lies below this estimated level and may be zero. Depending on the regulatory authority, estimated cancer risks between 10^{-4} and 10^{-6} have been judged to warrant regulatory consideration and possible intervention, whereas risks above 10^{-4} usually require action. The potential for noncancer health effects was estimated for this exercise based on the ratio of Inuit dose to the corresponding U.S. EPA RfD (no RfD is available for toxaphene). RfDs are most often derived from no-observed-adverse-effect-levels (NOAELs) in animal studies divided by a composite uncertainty factor of $100-1000$. These uncertainty factors are based on animal to human extrapolation, interindividual variability, and database limitations. RfDs are not intended to delineate the onset of hazard or risk, but rather to indicate a dose region below which long-term intake is unlikely to be hazardous. It is worth noting that neither the cancer nor the noncancer risk scoping exercises described here incorporate an explicit assessment of the risks posed in utero or to infants and children, particularly the risks posed to breast-feeding infants ex-
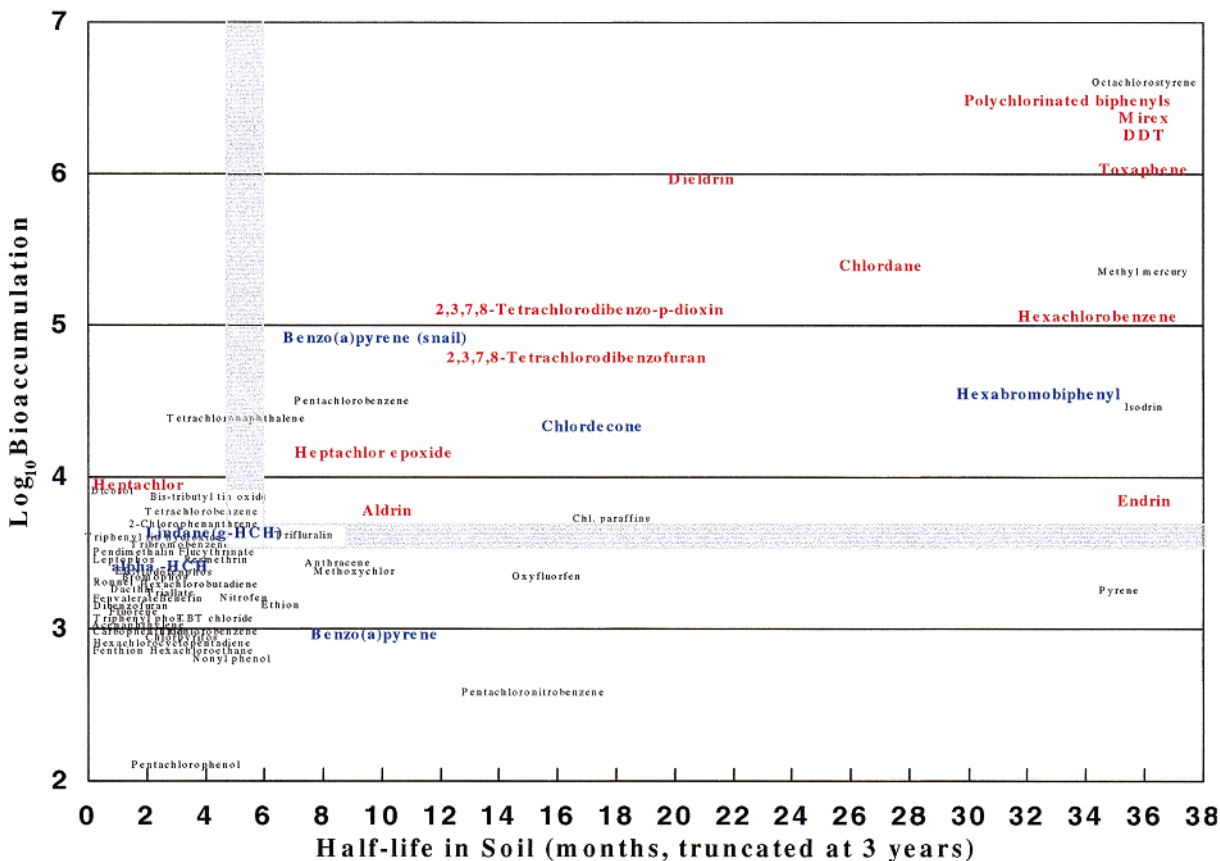
FIGURE 2. Bioaccumulation vs half-life in soil. The logarithm of the bioconcentration/bioaccumulation factors of selected organic chemicals versus their estimated half-lives in soil is graphed. UNEP POPs are displayed in red; additional substances included in the UNECE−LRTAP POPs protocol are displayed in blue. The shaded lines represent the UNECE−LRTAP and NAAEC−CEC guidance criteria.
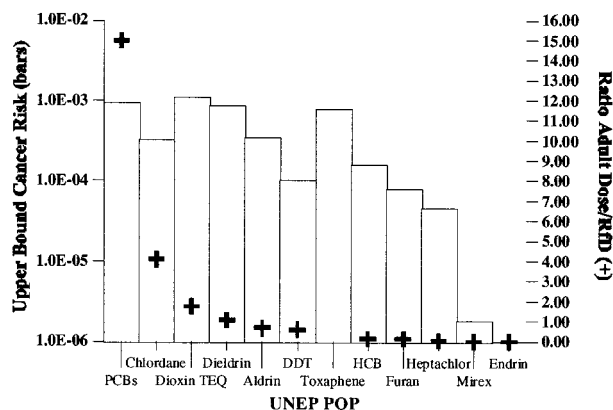

FIGURE 3. Health risks to Inuit adults. The estimated dietary exposure levels for Arctic Inuit adults are compared to U.S. EPA health guidance figures. Upper-bound cancer risk estimates following lifetime exposure are based on the 95th percentile upper confidence limit derived principally from animal bioassays. Noncancer health risk estimates are based on the ratio of the Inuit dose to the corresponding U.S. EPA reference dose (RfD).

periencing high, short-term dose rates of lipophilic POPs at critical developmental times (*16*). Similarly, risks posed to ecosystems by the long-range transport of POPs are not addressed by this exercise.

Multimedia Persistence and Long-Range Transport Models

The demonstration of potential risk from exposure to POPs in locations distant from their sites of release provides compelling evidence that long-range transport is a significant concern. To explore further how a consistent set of criteria for persistence and long-range transport might be established, we used a multimedia fate model to estimate the overall environmental persistence of chemicals coupled with a simple atmospheric transport algorithm to estimate the potential for long-range transport. The use of these models can help account for mode-of-entry and intermedia transport, factors which may strongly influence persistence and long-range transport potential.

For this analysis, the evaluative steady-state model of Mackay (*18*, *19*) was adapted to a spreadsheet format, maintaining the same spatial proportions but with modifications to the advection rate parameters (*20*). The multimedia model was then linked to a simple distance function algorithm. In this combined model, a chemical is released to the environment through a combination of air, water, and soil pathways; reaches steady state in the initial mixing zone through a combination of intermedia partitioning, media-specific degradation, and advection; travels as a result of atmospheric transport; and declines in concentration subject to atmospheric degradation and remote intermedia partitioning. The multimedia model is used to calculate three parameters for the transport model: concentrations in the atmosphere of the initial mixing zone associated with a chemical release rate and release medium, net atmospheric removal rate constant (k), and concentrations in the environment at a remote distance.

The concentration of a chemical in the initial mixing zone is a function of its release pattern to air, water, and soil. For analysis and comparison purposes, we assumed equal releases to air, water, and soil totaling an arbitrary but fixed 3000 kg/h. Typical of multimedia models, the air at the source was assumed to be well mixed with clean incoming air (analogous to a continuously stirred tank). A constant wind
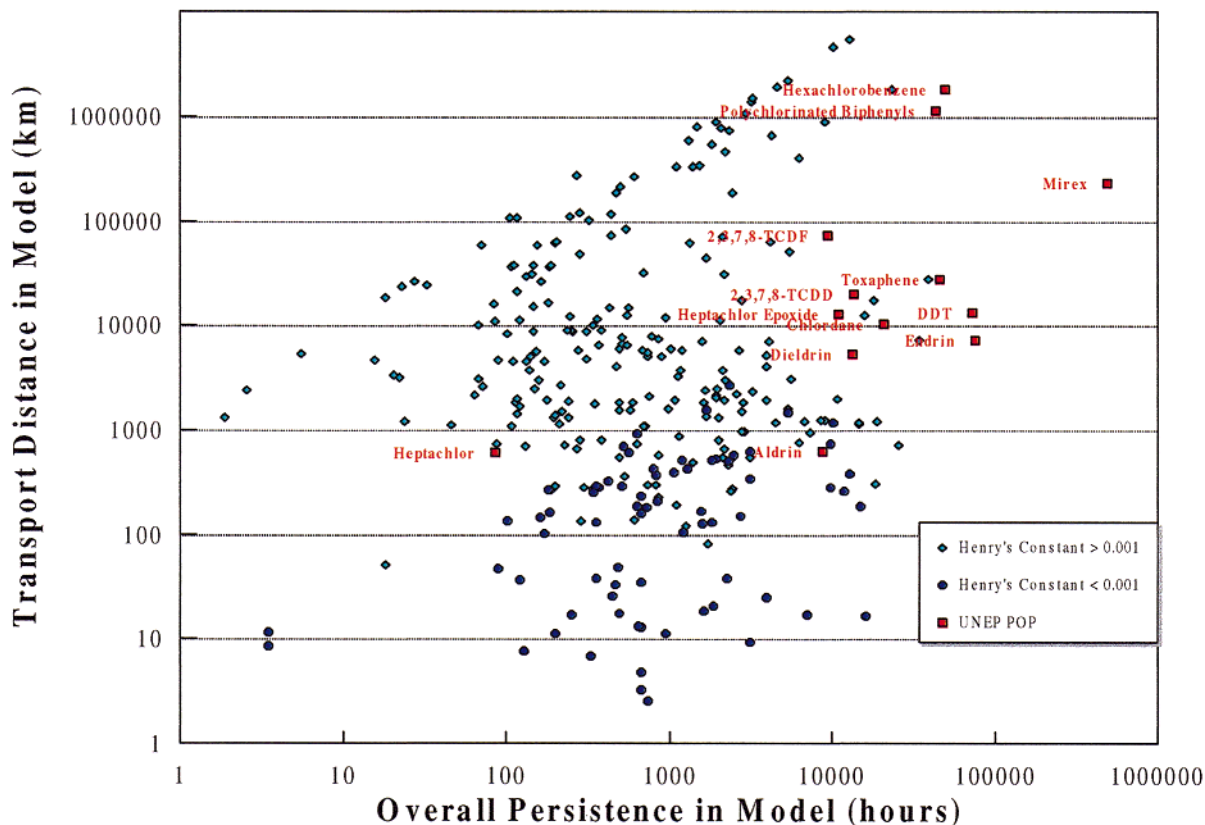
FIGURE 4. Transport distance vs overall environmental persistence. The transport distance predicted using the multimedia−atmospheric transport model versus overall environmental persistence for 316 chemicals is graphed. The 12 POPs under initial consideration in the UNEP negotiation are labeled in red. Both axes are log scaled.

speed of 7 m/s was assumed into and from the initial mixing zone.

On leaving the initial mixing zone, the chemical was assumed to be subject only to atmospheric transport. The net atmospheric removal rate (k) was calculated using the multimedia model to account for both atmospheric degradation and net intermedia transport to soil and water. As wind speed influences the distance traveled but not the removal of the chemical, the net atmospheric removal rate (k) is assumed independent of the advection rate. A closed system model (no advective loss) was therefore used to calculate this rate constant. The mass distributions between environmental compartments after the initial mixing zone were also calculated to reflect input solely via atmospheric transport rather than equally to air, soil, and water.

The decline in concentration with distance from the origin is represented by a first-order atmospheric transport model (analogous to "die-away" models):

$$X = \frac{v}{k} \ln \left(C_x / C_0 \right)$$

where X is relative distance traveled, k is the net atmospheric removal rate, v is the advection velocity, and C is the atmospheric concentration. Subscript 0 denotes source, with C_0 being calculated from the multimedia model. To provide a consistent basis for comparing substances, an average remote concentration of 1×10^{-11} g m^{-3} was arbitrarily specified as the cutoff point. Average environmental concentration is the total chemical mass divided by the volume of the multimedia model and can be related to the atmospheric concentration C_x used in the distance equation. The remote cutoff concentration was chosen to minimize the number of chemicals with a lower concentration in the initial mixing zone, which results in negative distances being

calculated for the set release rate. It should be noted that the time to reach steady state at the source or distance X is not considered in this model, and therefore the distances calculated represent maximums that may ultimately be achieved.

Unlike a number of other available models (*21, 22*), the model presented relates the relative distance traveled to the mass release rate of the chemical rather than to the initial atmospheric concentration. This modification is necessary because different chemicals may have equivalent atmospheric concentrations and net atmospheric removal rates but yet have markedly different average environmental concentrations due to intermedia partitioning and persistence.

We performed two analyses to elaborate the role that multimedia models could play in developing and applying POPs criteria. Both analyses are based on predominantly measured data for a list of 316 chemicals from the U.S. EPA chemical screening initiatives (*20*). In this analysis, upperbound estimates of persistence half-lives rather than mean estimates have been used to maximize data availability and applicability in a screening context.

Figure 4 plots the distance traveled in the model for the 316 chemicals versus their overall environmental persistence (14 chemicals fell below the *Y*-axis cutoff, with transport distances below 1 km). The *Y*-axis, distance traveled, is dependent on arbitrary values in the model, namely, the release rate (3000 kg h^{-1}) and the average concentration at the cutoff point (10^{-11} g m^{-3}), and should only be considered in relative terms. The *X*-axis, overall environmental persistence (*T*), is a measure of a chemical's tendency to remain in the environment unless removed by degradation. It is not a function of advective removal and is calculated as the weighted sum by mass fraction (*f*) of the degradation half-
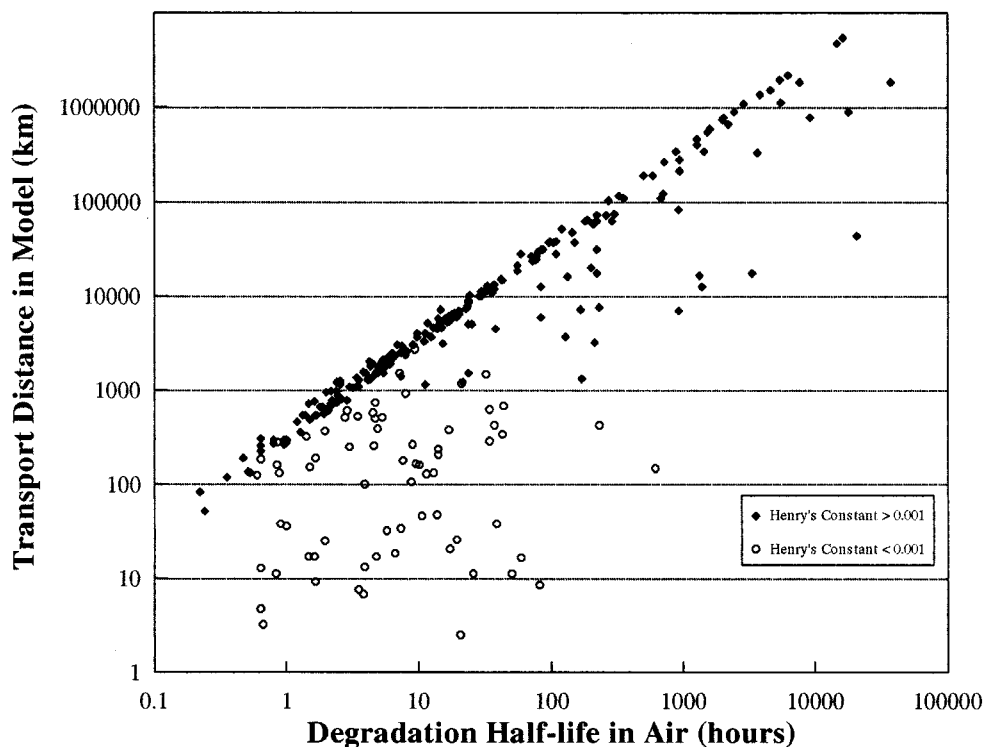
FIGURE 5. Transport distance vs half-life in air. The transport distance predicted using the multimedia−atmospheric transport model versus the degradation half-life in air for 316 chemicals is graphed. *Y*-axis distance estimates result from arbitrary but consistent input assumptions into the model and should be used only for purposes of comparing chemicals. Chemicals with Henry's law constants below 0.001 Pa m^{-3} mol^{-1} are indicated with open circles. Both axes are log scaled.

lives (τ) in each medium:

$$T = \frac{1}{\dfrac{f_a}{\tau_a} + \dfrac{f_w}{\tau_w} + \dfrac{f_s}{\tau_s} + \dfrac{f_{sed}}{\tau_{sed}}}$$

From Figure 4, it is evident that the 12 UNEP POPs, or their transformation products, are again persistent relative to other chemicals in this multimedia analysis and exhibit the potential for long-range transport. Based on a different set of chemicals and data, Scheringer (*23*) obtained analogous results for several UNEP POPs using a similar approach. The effect of the Henry's law constant on the distance traveled is also evident in Figure 4. Chemicals with constants below 0.001 Pa m^{-3} mol^{-1} exhibit reduced potential for transport, an effect that is much less evident at values above 0.01 Pa m^{-3} mol^{-1}.

Figure 5 continues the analysis of multimedia modeling by exploring the relationship between half-life in air and calculated transport distance in the model. For chemicals with Henry's law constants above 0.001 Pa m^{-3} mol^{-1}, the degradation half-life in air is a significant determinant of the transport distance ($R^2 = 0.51$). The impact of a low Henry's law constant in this model is again highlighted by the clustering of these chemicals toward the bottom left. This analysis therefore supports the use of the atmospheric half-life as a simple first screening step for long-range transport potential, followed by a more detailed multimedia analysis incorporating other chemical properties, such as the Henry's law constant.

Data and Model Uncertainties: A Cautionary Note

Caution must be exercised in the above analyses against unjustified reliance on point estimates of degradation half-lives in models. As stressed by Wania and Mackay (*24*), half-lives for chemical transformation cannot be viewed in the same way as half-lives for radioisotopes, which are fundamental, reproducible properties of the radionuclides. Degradation half-lives of organic substances depend not only on chemical properties and structure but also on characteristics of the surrounding environment. There are many environmental factors that can affect a substance's half-life, including, for example, temperature, pH, sunlight intensity, hydroxyl radical concentration, and microbial activity. As a result, there is substantial variability in environmental half-lives in both space and time, and this variability is reflected in available measured data. In addition to real environmental variability, a lack of standardization of test conditions and complexities in extrapolating from laboratory to field introduces further uncertainty. Similar caution is necessary in assessing the bioaccumulation and toxicity data.

Toward Policy Recommendations on POPs Criteria

The purpose of POPs screening criteria in international negotiations is to provide guidance on the type of substance that may pose unreasonable long-range, transboundary risks to humans or ecosystems. Although there is no set of numerical criteria that can distinguish POPs from other substances with certainty, the application of screening criteria with sufficient margins for uncertainty can provide a valuable basis for substance prioritization. The many uncertainties associated with screening criteria indicate that they should neither substitute for nor be given preference over the ultimate determination of unreasonable risk based on the weight of evidence. Moreover, criteria should be applied using expert judgment and flexibility. This means, for example, that a substance exhibiting relatively high values for one or more end points should not automatically be dismissed as a potential POP because it falls just short of another criterion. These considerations argue for a tiered system for the addition of substances to the UNEP treaty beyond the original 12 UNEP POPs. In such a system, POPs screening criteria would be applied in tier 1 followed by a more rigorous, detailed tier

2 evaluation for substances not eliminated at the first level. This approach forms the basis of a framework for substance evaluation that emerged as the consensus of international experts recently convened to discuss scientific issues relating to persistence and long-range transport (*25*).

The analyses described in this paper reveal that guidance for setting POPs screening criteria can be developed using a combination of science and policy input. Methods such as theoretical and empirical analysis of substance properties and multimedia fate modeling are valuable scientific tools that can be used to inform the decision-making process. Our results are intended to be applicable in the context of the UNEP POPs negotiation where the criteria must reflect the international geographic and political scope of the negotiation and be targeted toward those POPs most warranting this level of international attention. They suggest that criteria adopted under the NAAEC−CEC and UNECE−LRTAP agreements are reasonable and tend to isolate a limited number of clearly hazardous POPs from the majority of organic chemicals, while not being so stringent that the ability to respond to as yet unidentified risks is seriously compromised.

Acknowledgments

Supporting Information Available

Tables giving the bioaccumulation values in fish, persistence values, and human cancer risk calculations for Inuit intake of POPs and references (14 pages). This material is available free of charge via the Internet at http://pubs.acs.org.

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