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Bioaccumulation Potential of Persistent Organic Chemicals in Humans

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A model was used to explore the influence of physical-chemical properties on the potential of organic chemicals to bioaccumulate in humans. ACC-HUMAN, a model of organic chemical bioaccumulation through the agricultural and aquatic food chains to humans, was linked to a level I unit world model of chemical fate in the physical environment and parametrized for conditions in southern Sweden. Hypothetical, fully persistent chemicals with varying physical-chemical properties were distributed in the environment, and their bioaccumulation to humans was calculated. The results were evaluated using the environmental bioaccumulation potential (EBAP), defined as the quotient of the chemical quantity in a human divided by the quantity of chemical in the whole environment. Since the latter is closely related to emissions, EBAP is potentially a more useful tool for comparative risk assessment of chemicals than currently used medium-specific measures such as the fish−water bioaccumulation factor. A high environmental bioaccumulation potential, defined as >10% of the maximum EBAP, was found for chemicals with $2 < \log K_{OW} < 11$ and $6 < \log K_{OA} < 12$. While these chemical partitioning properties clearly influenced bioaccumulation at each trophic level, these effects tended to equalize over the food web. The fact that the transfer from the environment as a whole to humans was quite uniform over a large chemical partitioning space suggests that these partitioning properties are relatively unimportant determinants of human exposure compared to other factors such as the substance's persistence in the environment and in the food web.

Introduction

Persistent organic pollutants (POPs) are characterized not only by their persistence but also by their toxicity, their potential for long-range transport, and their potential for bioaccumulation. The combination of these four properties renders a chemical particularly hazardous in the environment. International treaties have been adopted to restrict and eliminate the production, use, and release of POPs to protect human health and the environment (*1*−*3*). Developing measures of persistence, toxicity, long-range transport, and bioaccumulation is therefore crucial for the assessment and management of chemicals. In this paper we explore measures of bioaccumulation.

Currently the criterion for identifying bioaccumulative chemicals is a bioaccumulation factor or a bioconcentration factor in aquatic organisms in excess of 5000 or, if these data are not available, a log octanol−water partition coefficient (K_{OW}) greater than 5 (*3*, *4*). The characterization of bioaccumulation based on K_{OW} is based only on studies of bioaccumulation in aquatic organisms. Recently, bioaccumulation in the terrestrial environment was shown to be governed by other physical-chemical properties (e.g., octanol−air partition coefficient, K_{OA}) which are related to volatility, not hydrophobicity (*5*−*8*). Moreover, Kelly and Gobas (*6*, *7*) and Gobas and co-workers (*8*) showed that in the arctic lichen−caribou−wolf food chain chemicals with log K_{OW} as low as 2 were biomagnified if their log K_{OA} values were high enough (>5) and their rate of transformation or metabolism was low. Note that in this paper biomagnification is defined as an increase in fugacity from prey—including plants—to predator, as this is the most useful definition from a mechanistic perspective.

Although protecting human health is the top priority of chemical management, measures of bioaccumulation to humans have not been studied to our knowledge. Humans are top predators in both aquatic and terrestrial food chains, and the evidence above suggests that the current criterion for bioaccumulation based on aquatic food chains only may not be appropriate for humans. In the work presented here, we used a recently developed bioaccumulation model (ACC-HUMAN) to explore the physical-chemical properties determining bioaccumulation in humans.

Model Description

A level I unit world model of chemical fate in the physical environment (*9*, *10*) was linked to a bioaccumulation model that describes chemical transfer through the aquatic and agricultural food chains to humans (see Figure 1). In numerical experiments, chemicals with varying physical-chemical properties were discharged into the unit world and the degree to which they were transferred to humans was compared. Note that although the algorithms used in the model to describe sorbed phase partitioning were developed for nonpolar organic chemicals, polar chemicals were also included in the experiments since their transfer to humans is generally not affected by sorbed phase partitioning.

Physical Model. Chemical fate in the physical environment was described with a fugacity-based level I unit world model consisting of the four compartments air, water, soil, and sediment (*9*), and employing the parametrization recommended in ref *10*. One-tenth of the unit world was water covered, while the remainder was soil. The depths of the air, water, soil, and sediment compartments were 1000 m, 20 m, 10 cm, and 1 cm, respectively. The organic carbon contents of soil and sediment were set to 2% and 4%. Phase partitioning was modeled according to ref *11*, and a partitioning equilibrium was assumed.

Bioaccumulation Model. Bioaccumulation from the physical environment to humans was calculated using the recently developed model ACC-HUMAN, which is described in detail in ref *12*. Here we will only give a brief description. ACC-HUMAN is a non-steady-state, mechanistic, fugacity-based model. It focuses on the primary dietary sources of background human exposure to POPs, namely, fish, dairy products, and beef. The model is subdivided into an agricultural food chain represented by grass, milk cows, and beef cattle and an aquatic food chain represented by

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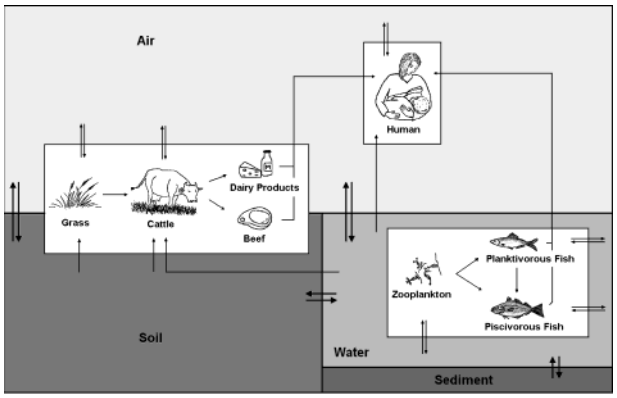

FIGURE 1. Model structure.

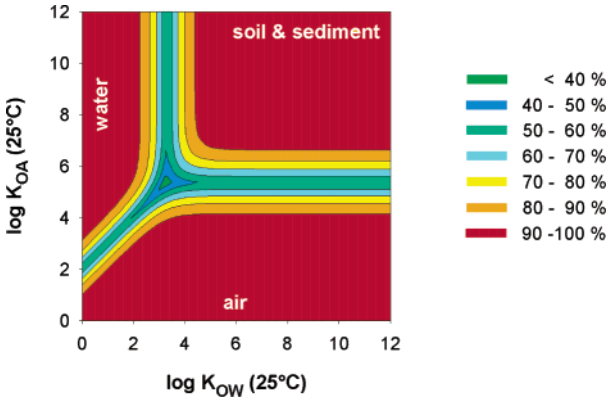

FIGURE 2. Distribution of the chemicals in the unit world as a function of the octanol–water and octanol–air partition coefficients K_{OW} and K_{OA}. The three distinct regions within the chemical partitioning space show the phase into which the main fraction of a chemical with a defined K_{OA}/K_{OW} combination partitions.

zooplankton, planktivorous fish, and piscivorous fish. The human is linked to both systems as the top predator. The contaminants enter the food chains via uptake from air, water, and soil, and are transferred to higher trophic levels via predator–prey interactions. In addition to ingestion, the following uptake mechanisms are considered: gill ventilation (fish), atmospheric deposition and root uptake (grass), ingestion of soil with feed (cattle), and inhalation and drinking (cattle and humans). The elimination pathways treated depend on the organisms and include egestion, urination, exhalation, gill ventilation, lactation, percutaneous excretion, and metabolism. Growth and the temporal variation of physiological and environmental parameters such as the ingestion rate and temperature are taken into account. An exception is the milk cow, which is assumed to be at a steady state. Beef cattle live for 28 months before being slaughtered, while fish and humans are modeled for a lifetime of 10 and 80 yr, respectively. Each year a new generation of the animals is generated, and every 10 yr a new human is born into the unit world.

The model was parametrized according to environmental and agricultural conditions in southern Sweden as described in ref *12* and documented in the supplementary material for that paper archived at the journal's website. The dominant commercial fish species in the Baltic Sea, herring and cod, were chosen as model organisms for planktivorous and piscivorous fish, respectively. In a model validation exercise using this parametrization, good agreement was found between predicted and measured concentrations of polychlorinated biphenyls for cows' milk, beef, herring, cod, and human tissue in southern Sweden (*12*).

The one change made to the model parametrization was to use dietary habits for Germany instead of for Sweden, as the former was thought to be more representative of dietary habits in Europe and North America than the high-fish diet of the Swedes. For a 25 yr old woman the modeled daily ingestion rate was 383 g of dairy products (4.4% lipid), 39 g of beef (20% lipid), and 63.3 g of fish wet weight (2% lipid) (*13*, *14*). The age composition of the diet fish was estimated on the basis of data on average landings of herring and cod in the Baltic watershed (*12*, *15*), where 50% of the total fish were assumed to be herring and 50% to be cod. The dietary composition was assumed to be constant over the humans' lifetime. The ingestion rate on the other hand was dependent on age following the approach in refs *12* and *16*.

Model Experiments. In the physical model, the partitioning properties of soil and sediment are defined using the chemical's solubility in octanol. As a consequence, the partitioning behavior between the four compartments air, water, soil, and sediment is completely defined by any two of the air–water, octanol–air, and octanol–water partition coefficients K_{AW}, K_{OA}, and K_{OW} (*17*). In the bioaccumulation model, K_{OA} and K_{OW} are the only physical-chemical properties apart from degradation rate constants used in the predictive algorithms. Therefore, any two persistent chemicals with the same combination of K_{OA} and K_{OW} will display the same environmental partitioning and bioaccumulation behavior.

To explore the influence of physical-chemical properties on bioaccumulation in humans, hypothetical chemicals with varying combinations of K_{OW} and K_{OA} were defined. The molar mass for each of the chemicals was set to 100 g mol^{-1}. The heats of air–water, octanol–air, and octanol–water phase transfer were set to 60, −80, and −20 kJ mol^{-1}, respectively. To obtain a measure of the maximum possible bioaccumulation, the chemicals were assumed to be completely persistent; i.e., the rate constants for degradation were set to zero. A fixed amount of each of these hypothetical chemicals was distributed in the level I unit world.

To ensure that the bioaccumulation model reached steady state before the simulation of the human started, the model was run for 40 yr before the human was born. The lifetime accumulation of the chemicals in a girl/woman was then simulated. The woman's body burden after completing nursing of her first child at 30 yr of age was used as the basis of comparison for the bioaccumulation behavior.

To investigate the influence of the different food chains on bioaccumulation in humans, three model runs were conducted. In the first, dietary exposure was only due to food of marine origin. In the second, only exposure from the agricultural diet was considered. In a third model run, exposure occurred via both marine and agricultural food.

Results and Discussion

Environmental Partitioning Behavior. Even though the area-based environmental concentrations are equal for each of the hypothetical chemicals, the exposure of the biota differs depending on the chemicals' partitioning behavior. Figure 2 shows how the major environmental phase in which a chemical resides (air, water, or soil/sediment) varies depending on its partition coefficients K_{OA} and K_{OW}. When K_{OA} is low, virtually all of the chemical in the unit world is present in the atmosphere. However, when log K_{OA} exceeds 3.5, the fraction in the atmosphere begins to decrease due to sequestration into soil and sediment. When, in addition, log K_{OW} is low, sequestration into water becomes important, and the fraction in the atmosphere begins to decrease at even lower values of log K_{OA}. Therefore, the exposure of the agricultural food chain, which occurs primarily via the atmosphere, is highest for chemicals with low K_{OA}. On the other hand, the concentrations in water and hence the exposure of the aquatic food chain are highest for chemicals with high K_{OA} and low K_{OW} (i.e., low K_{AW}). Note that a change

of K_{OA} or K_{OW} by 2 orders of magnitude can cause an almost complete redistribution from one phase into another.

Environmental Bioaccumulation Potential of Humans. Bioaccumulation is generally quantified using the ratio of the chemical concentration in the organism of interest to the chemical concentration of the medium in which the organism lives and feeds. For humans, and indeed for some other top predators, this definition presents difficulties. Humans obtain their food from diverse food chains, and the relevant physical medium differs, e.g., water for herring, air for some chemicals in dairy products, or sediment for shellfish. Which of these physical media is most important for human bioaccumulation depends on the properties of the chemical and on dietary habits; it is therefore not possible to define a single medium for the generic assessment of bioaccumulation to humans.

To overcome this difficulty, we chose to assess bioaccumulation in humans on the basis of the concentration in the environment as a whole. This concentration was expressed as the quantity of chemical in the environment of interest divided by the surface area of that environment, yielding units of g m^{-2}. Furthermore, we chose to express the concentrations in humans (in this case a 30 yr old woman) in g $person^{-1}$ for similar reasons. The measure of bioaccumulation was the quotient of these two numbers (C_{human}/C_{env}) with units of m^2 $person^{-1}$. To differentiate from medium-specific bioaccumulation, we call this environmental bioaccumulation, and because we have assumed the chemicals to be completely persistent in this paper, we have dubbed this measure the environmental bioaccumulation potential (EBAP). It expresses the potential of a chemical to bioaccumulate in humans if indeed it were completely persistent. The magnitude of EBAP can be viewed as the quantity (surface area) of the environment that contains the same amount of chemical as the person. A large EBAP implies stronger bioaccumulation, because the chemical from a larger portion of the environment is concentrated in the individual. The EBAP could have been defined using the total quantity of chemical in the environment (instead of the area-normalized concentration), but this would have made its magnitude dependent on the overall size of the environment. As defined here, the EBAP can be compared between different environments and between different chemicals.

In addition to resolving the ambiguity regarding the reference matrix in measuring bioaccumulation of top predators that was addressed above, EBAP has a further advantage. Because the chemical concentration in the entire environment is used and the chemicals are assumed to be completely persistent, the reference (environmental) concentration in EBAP is at a first approximation proportional to emissions. Consequently, EBAP gives information on the relationship between concentration in the organism and the emissions. This is not the case for the bioaccumulation factor, for which the reference concentration is the concentration in a single medium (e.g., water). Thus, EBAP is of direct relevance in comparative risk assessment. Comparing the value of EBAP between chemicals gives a relative measure of the human body burden that can be expected from identical emission of the chemicals (again, assuming they are completely persistent). We suggest that EBAP represents an attractive alternative to the currently used organism/water BAF for identifying potentially bioaccumulative compounds.

In Figure 3 EBAP is plotted against K_{OW} and K_{OA} for the three different model runs: marine diet only, agricultural diet only, and total diet. The EBAP isolines represent the percentage of the maximum EBAP obtained within the chemical partitioning space. The maximum EBAP for the total diet occurs for a chemical with log $K_{OW} \approx 7$ and log $K_{OA} \approx 8.5$ and has a value of 120 m^2 $person^{-1}$. For comparison purposes, EBAP was estimated from a PCB inventory for the United Kingdom at the beginning of the 1990s and an average PCB burden for a British person (*18*). The estimated EBAP value for PCB 180, a persistent chemical with partitioning properties close to the maximum in Figure 3a, was 87 m^2 $person^{-1}$. The good agreement between the model prediction and the observed value is evidence that the model gives a reasonable approximation of chemical transfer to humans.

Taking the 10% isolines as a threshold for potentially bioaccumulative chemicals, Figure 3 indicates that regardless of the diet's origin only chemicals with a log $K_{OA} > 6$ accumulate in humans. Chemicals with lower K_{OA} values are more volatile. They are largely present in the atmosphere (see Figure 2) and have a low tendency to partition into human tissue. Furthermore, they do not biomagnify because they are rapidly eliminated via exhalation. Gobas and coworkers came to a similar conclusion, describing respiration as the dominant elimination route for chemicals with a log $K_{OA} < 5$ in arctic wolves and caribou (*7, 8*).

Less volatile compounds (log $K_{OA} > 6$) have a strong tendency to partition out of the atmosphere into organic matter such as soil (see Figure 2) or human tissue. Inhalation is of minor importance compared to dietary uptake, and exhalation is an inefficient elimination mechanism, making biomagnification possible.

Human Exposure from a Marine Diet. When the dietary exposure is of marine origin only, the potentially bioaccumulative chemicals are found within the chemical partitioning space defined by $3.5 < \log K_{OW} < 11$ and $\log K_{OA} > 6$ (10% isoline, Figure 3b). Above a log K_{OA} of 8 EBAP is almost independent of K_{OA}, reflecting the bioaccumulation behavior in fish (see below). EBAP has a maximum at a log K_{OW} between 7 and 9. The decrease in EBAP at higher K_{OW} is caused by a reduced gut absorption efficiency in both fish and humans. The decrease at lower K_{OW} is a reflection of the lower EBAP in the ingested fish as illustrated in Figure 4a and discussed below.

Accumulation in Fish. The EBAP concept can also be applied to other organisms. However, due to the difficulty in defining the size of an individual organism in some cases, EBAP(organism) was calculated using the chemical concentration in the organism (as opposed to the quantity in the whole organism used for EBAP(human)), yielding units of m^2 g^{-1} of organism. Note again that this definition considers the total physical environment including air, water, soil, and sediment and is not limited to the surrounding medium of the organism (e.g., water when considering fish). Figure 4 shows EBAP(organism) for the individual links of the food chain. For EBAP(fish), the concentration in the fish consumed by humans was used (i.e., a mixture of fish of different trophic levels and age).

In fish, chemicals within the chemical partitioning space defined by $3.5 < \log K_{OW} < 11$ and $\log K_{OA} > 5$ show a high environmental bioaccumulation potential (10% isoline, Figure 4a). Although the fraction of chemical in the water compartment, and hence the water concentration, was highest for chemicals with high K_{OA} and low K_{OW} values (Figure 2; see also above), EBAP(fish) was low in this region of the chemical partitioning space. Due to their rapid elimination via gill ventilation, these chemicals do not biomagnify. They accumulate in the aqueous phase of the fish at concentrations comparable to those in water, but the EBAP remains low due to the diluting effect of the large quantity of water in the environment. However, above a log K_{OW} of 3.5, the chemical is primarily associated with the organic phase both within the fish and in the environment as a whole (see Figure 2). The ratio of the volume of organic matter in the fish to the volume of organic matter in the environment is greater than the corresponding ratio for water, and therefore, EBAP is higher. Furthermore, with increasing log K_{OW} ($>\sim 4-5$) a change in the dominant uptake pathway from ventilation to

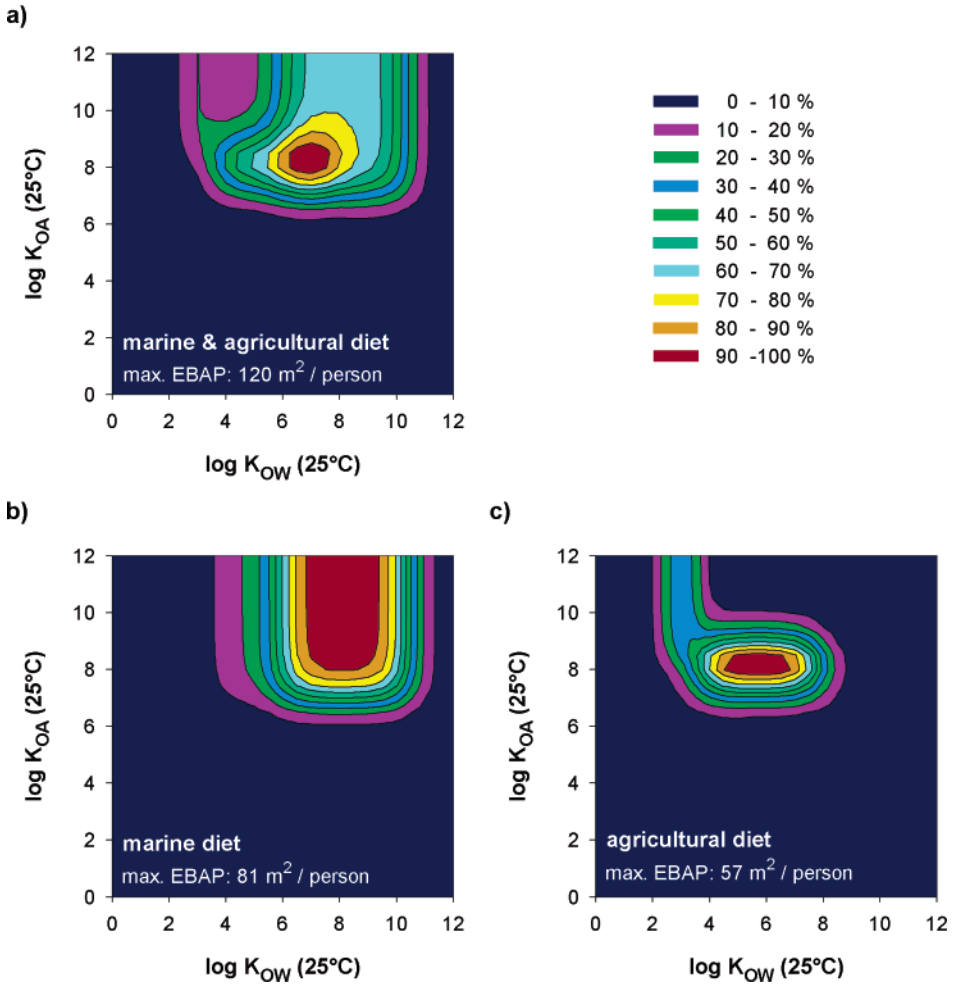

FIGURE 3. EBAP in humans plotted as a function of K_{OW} and K_{OA}. EBAP is the ratio of the quantity of a completely persistent chemical in a human to the quantity of the chemical present in 1 m^2 of the environment. It is shown as a percent of the maximum EBAP within the chemical partitioning space. The results for a 30 yr old mother nursing her first infant are presented whose exposure results from (a) a marine and agricultural diet, (b) a marine diet only, and (c) an agricultural diet only.

ingestion occurs, and biomagnification becomes possible. As a consequence, EBAP(fish) increases further, reaching a maximum at a log K_{OW} of 6.5−9.5. For more hydrophobic compounds, EBAP(fish) decreases again due to a reduced gut absorption efficiency. It should be noted that the predictions for chemicals with log K_{OW} > 7.5 are uncertain due to the paucity of experimental data that can be used for model verification. In particular, the extended plateau at maximum EBAP is particularly sensitive to the assumptions about zooplankton−water partitioning, for which very few empirical data are available. However, the presence of an extended plateau is supported by the work of Arnot and Gobas (*19*), who identified chemicals with a log K_{OW} between ~4 and ~12.2 to be potentially bioaccumulative in an aquatic food chain.

Even though the intrinsic processes involved in the bioaccumulation in fish are dependent on K_{OW} only, EBAP-(fish) shows a pronounced K_{OA} dependency for chemicals with a log K_{OA} < 8 (Figure 4a). This is due to the environmental partitioning behavior of the chemicals, as they are increasingly sequestered into the air phase with decreasing K_{OA} (Figure 2).

At this point it becomes obvious that it is important to consider the total physical environment when bioaccumulation potential is explored. The current criterion for the identification of bioaccumulative substances (namely, log K_{OW} > 5; see above) is based on the bioaccumulation process alone, considering only transfer from water to the organism. For chemical screening it would seem useful to categorize bioaccumulative substances on the basis of the ratio of level in the organism to the quantity emitted to or present in the environment. In this case the chemical's fate in the physical environment must be considered.

Human Exposure from an Agricultural Diet. When the dietary exposure is of agricultural origin only, the potentially bioaccumulative chemicals are located within the chemical partitioning space defined by 2 < log K_{OW} < 9 and 6 < log K_{OA} < 10 (10% isoline, Figure 3c). At high K_{OW} values, EBAP decreases as a result of a reduced absorption efficiency for these chemicals in the digestive tract of humans (*16*) and cattle (*20*) (see below). For an explanation of the other features of this plot it is necessary to examine the bioaccumulation pattern at lower trophic levels in the agricultural food chain.

Accumulation in Grass. EBAP for grass (concentration in grass on a fresh weight basis divided by chemical quantity in 1 m^2 of the environment, units m^2 g^{-1}) is plotted in the chemical partitioning space in Figure 4b. There are two distinct regions of high EBAP that are associated with two different accumulation processes.

The region of elevated EBAP in the right-hand portion of Figure 4b is associated with more hydrophobic chemicals (log K_{OW} > 4) for which uptake occurs mainly via atmospheric deposition. In general, two processes can be distinguished: gaseous and particle-bound deposition, whereby the latter is of relevance only for chemicals with log K_{OA} > 10−11 due to their affinity to airborne particles. Figure 4b shows that

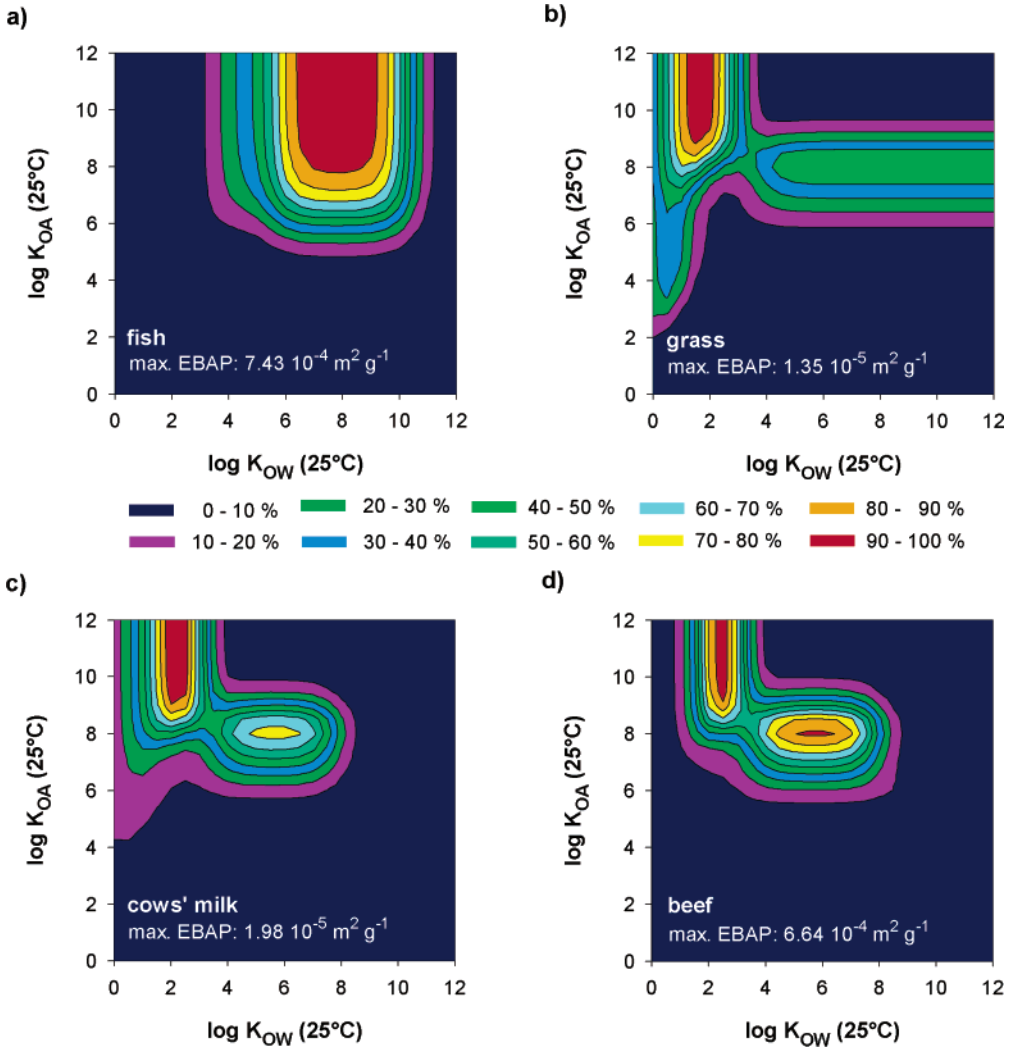

FIGURE 4. EBAP in the lower trophic levels of the food chain plotted as a function of K_{OW} and K_{OA}. EBAP(organism) is defined as the ratio of the concentration in the organism to the quantity present in 1 m^2 of the environment (units m^2 g^{-1} of fresh weight) and is shown as a percentage of the maximum EBAP within the chemical partitioning space. Key: (a) fish, (b) grass, (c) cow's milk, and (d) beef.

EBAP(grass) is independent of K_{OW} in this region, while a distinct K_{OA} dependency is visible, with increasing values for log K_{OA} from 6 to 8 followed by a decrease for log K_{OA} from 8 to 9.5. The initial increase is a reflection of the increasing partitioning capacity of grass compared to soil (the major storage compartment for these chemicals in the unit world) with increasing K_{OA}. The subsequent decrease in EBAP(grass) at higher K_{OA} values is due to the very large grass−air partition coefficients; kinetic limitations prevent the grass from attaining a partitioning equilibrium. This can only be partly compensated by the increasingly important particle-bound deposition (*21*).

The second region of elevated EBAP(grass) is characterized by low log K_{OW} (<4) and high log K_{OA} (>∼8) (Figure 4b, upper left). Chemicals with low K_{OW} are taken up relatively efficiently via the roots and transported in plants with the transpiration stream. In accordance with Briggs, the uptake efficiency is dependent on K_{OW} with a maximum at log $K_{OW} \approx 1.5$ (*22*). This is reflected in the K_{OW} dependency of EBAP(grass) in this region. If the chemical has a low K_{OA} value, then it is rapidly eliminated from the vegetation via volatilization. However, if K_{OA} is high, then volatilization is kinetically limited and the chemical accumulates in the vegetation, leading to high EBAP values. This is comparable to biomagnification in animals; the vegetation can achieve concentrations that are far in excess of its equilibrium concentration with the surrounding environment.

Accumulation in Cattle. For cattle, EBAP was calculated as the concentration in milk and beef (on a fresh weight basis) divided by the quantity of chemical in the environment (see Figure 4c,d). As for EBAP(grass), there are two distinct regions of high EBAP(cattle). The most prominent change compared to EBAP(grass) is a decrease of EBAP(cattle) at high K_{OW} values (>6.5). This is attributable to a decline of the gut absorption efficiency in cattle for chemicals with a log $K_{OW} > 6$ (*20*).

The region of high EBAP(cattle) in the left-hand portion of Figure 4c,d largely mirrors the region of maximum EBAP-(grass). Superimposed on the uptake of the hydrophilic contaminants with drinking water is a high uptake via feed. Urination, which is modeled as an excretion of water in equilibrium with the animal, is a comparatively efficient elimination mechanism for hydrophilic substances. However, its effectiveness is limited by the higher temperature—and hence lower solvent capacity—of urine compared to drinking water and the water in grass. Hence, a temperature-driven biomagnification (*23*) occurs, even if drinking water would be the only source of contaminant. The biomagnification of the hydrophilic substances in grass discussed above amplifies this effect. The peak EBAP is shifted somewhat to the right in cattle compared to grass due to the more rapid elimination of the more hydrophilic compounds. It is also notable that the maximum EBAP was 3 times higher for the left-hand peak than the right-hand peak in grass (Figure 4b), while in

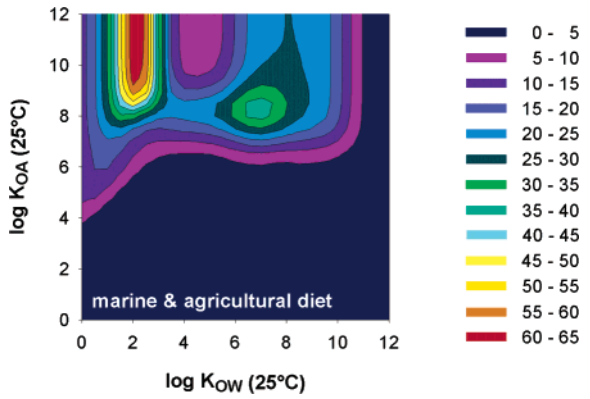

FIGURE 5. Ratio of the fugacity in a 30 yr old woman to the fugacity in the physical environment plotted against the partition coefficients K_{OW} and K_{OA}.

cattle the difference was much less. This can also be attributed to the more rapid elimination of the compounds with lower K_{OW} in cattle.

The trends observed in the bioaccumulation behavior of cattle are reflected in the EBAP in humans (Figure 3c). The same two regions of high EBAP are present in the chemical partitioning space. However, just as between grass and cattle, the left-hand peak has shifted to the right and its relative magnitude has decreased. This can also be explained by the more efficient elimination of the lower K_{OW} compounds in humans.

A Fugacity Perspective on Bioaccumulation in Humans. Returning to Figure 3, it can be seen that the EBAP(human) distribution for the total diet is a combination of the distributions from the marine diet and the agricultural diet, each of which have been explained above. Further insight into bioaccumulation of organic chemicals in humans can be gained by examining the ratio of the fugacity in humans to the fugacity in the physical environment (fugacity can be viewed as the partial pressure of a chemical in the environmental phase of interest; see ref *24*). In Figure 5 this ratio is plotted in the chemical partitioning space in the same manner as for EBAP (note that the fugacity in all compartments of the physical environment was the same as equilibrium partitioning is implicit in the level I unit world model).

If the contaminants' accumulation is only due to diffusive partitioning processes between the environment and the organism, such as gaseous deposition (grass), gill ventilation (fish), or respiration (mammals), the maximum concentration of the chemical in the organism is the equilibrium concentration. As a consequence, the fugacity in the organism does not exceed the fugacity in the environment. Biomagnification on the other hand leads to fugacities higher than in the environment (*23*−*26*). A fugacity ratio of 1 thus implies equilibrium partitioning; a ratio >1 indicates biomagnification.

Figure 5 shows that chemicals with a log K_{OW} < 11 and a log K_{OA} > 6 have a potential to biomagnify in the human food web. There are two distinct regions of high fugacity ratios. The peak on the right-hand side of the figure is almost identical to the maximum EBAP (Figure 3a). However, the second peak, which has the maximum fugacity ratio, lies largely outside the EBAP 10% isoline (Figure 3a). It is surprising that the chemicals that biomagnify most strongly do not have a large EBAP. The comparatively low EBAP values can be explained by the preferential partitioning of compounds with low K_{OW} and high K_{OA} into the water compartment (Figure 2). The ratio of the volume of organic matter in a human to the volume of organic matter in the environment is greater than the corresponding ratio for water, and therefore, for a given biomagnification factor the EBAP values are lower for compounds which preferentially partition into water.

The key to the strong biomagnification of low-K_{OW} compounds in humans lies in the first step in the food chain, namely, their bioaccumulation in grass. As mentioned above, there is an effective contaminant uptake into grass via the roots with the transpiration stream. The model estimates that within one growing season a water volume of ~0.27 m^3 is transpired from a 1 m^2 plot of pasture. This is about 70 times more water than the vegetation contains (3.9×10^{-3} m^3). As volatilization is limited for the chemicals with high K_{OA} values, up to 70 times more chemical can accumulate in the plant than would be present if the plant were in equilibrium with the soil and the air. The fugacity ratio between vegetation and the environment is elevated analogously (results not shown). This effect has been demonstrated experimentally in the past; e.g., the fugacity of Bromacil was estimated to be 6 times higher in soybean leaves than in soil following just 3 days of exposure (*27*). Further biomagnification occurs going from grass to the cow as a result of the temperature increase when the grass is ingested (see above). Although there is no biomagnification in the last step in the food chain from cattle to humans, the biomagnification factor has already reached levels far in excess of those achieved through the more widely recognized digestive tract mechanism for lipophilic substances.

Implications for Chemical Risk Assessment. For chemical risk assessment and management, it is essential to be able to predict the exposure of organisms of higher trophic levels, particularly of humans, on the basis of the emission of a contaminant into the environment. The parameter utilized in this paper, namely, the environmental bioaccumulation potential, the ratio between human body burden and the quantity of chemical in the environment, is a measure of bioaccumulation that can be directly related to chemical risk assessment. It is important to remember that EBAP is defined for a fully persistent chemical. The bioaccumulation of many chemicals will be less than the EBAP due to degradation in the environment and/or metabolism in the food chain.

The discussion above, particularly regarding EBAP in fish, clearly demonstrates the importance of including chemical distribution throughout all of the physical environment when bioaccumulation is evaluated. By assuming a partitioning equilibrium, we have taken the simplest approach to describing this. Using more complex models could yield further interesting insights into bioaccumulation behavior. Another important assumption was the choice of the 10% isoline of EBAP for our interpretation. This implies that a chemical with a EBAP that is within one-tenth of the maximum possible EBAP of any chemical is of potential concern from a bioaccumulation perspective. We consider this assumption to be reasonable, but not especially conservative.

The results of this modeling exercise show that the criteria currently used for identifying bioaccumulative substances based only on K_{OW} (*3*, *4*) and EBAP do not always agree. The K_{OW} method identifies hydrophobic substances that are relatively volatile (log K_{OW} > 5; log K_{OA} < 6) as bioaccumulative, while the EBAP approach predicts they will be rapidly eliminated from humans via respiration. The K_{OW} method does not select less hydrophobic, involatile chemicals (2 < log K_{OW} < 5; log K_{OA} > 6), while the model developed here predicts that these chemicals will bioaccumulate strongly via plants following root uptake from soil. Many pesticides such as atrazine fall into this chemical partitioning space (Figure 6). In studying an arctic terrestrial food chain, Gobas et al. (*8*) and Kelly and Gobas (*7*) also reported that chemicals with a log K_{OW} > 2 and a log K_{OA} > 5 biomagnified.

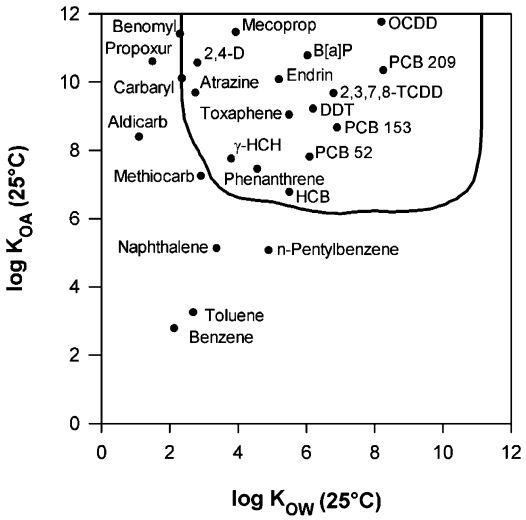

FIGURE 6. Ten percent EBAP(human) isoline showing the location of selected nonpolar organic chemicals within the chemical partitioning space (physical chemical properties from ref *10*).

Many of the substances shown in the low-K_{OW} portion of Figure 6 are polar. Caution must be exercised in applying the model to polar substances because the algorithms describing partitioning to solid phases are applicable only to nonpolar chemicals. However, as discussed above, sorption to solid phases plays little role in the overall fate of low-K_{OW} substances (Figure 2). Furthermore, their bioaccumulation is driven by biomagnification in grass via the transpiration stream and temperature-driven biomagnification in cattle, in which sorption to solid phases is not a factor. Consequently, the conclusions regarding EBAP should be applicable to polar substances with low K_{OW} as long as they do not associate strongly with sorbed phases.

EBAP can be expected to vary depending on human dietary habits, the structure of the key food chains, and the properties of the environment in which the chemical is distributed. The potential influence of human diet is illustrated by the different elements of Figure 3. The contamination of indigenous people of the Arctic with POPs is a good illustration of the influence of food chain structure; the long marine food chain with mammals at the uppermost trophic levels leads to very high bioaccumulation. A comparison of a moist coastal region with an arid continental region illustrates the potential influence of environmental properties. Hydrophilic compounds (low K_{OW} and K_{AW}) will be effectively diluted in the former, leading to much lower EBAP than in the arid continental environment. An additional increase in EBAP would result if transpiration was higher in the arid environment.

Despite the potential for some variability, the most striking feature of the bioaccumulation potential in humans as illustrated in Figure 3a is its uniformity. Over a K_{OW} range of 9 orders of magnitude and a K_{OA} range of 6 orders of magnitude, EBAP varied by just a factor of 10. The chemical partitioning space displaying high EBAP encompasses virtually all nonpolar organic chemicals with the exception of VOCs. While the chemical's partitioning properties clearly influence bioaccumulation at different levels of the food web, these effects tend to equalize over the food web as a whole. As a result, the partitioning properties had surprisingly little influence on the overall extent to which organic chemicals (with the exception of VOCs) are transferred from the environment to humans for the northern European scenario studied here. This implies that a chemical's partitioning properties are relatively unimportant determinants of human exposure compared to other factors such as its persistence in the environment and in the food web.

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