Regional differences in the environmental bioaccumulation potential of persistent organic pollutants in humans

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Introduction

The potential to bioaccumulate is one of the properties characterizing persistent organic pollutants (POPs) (1) . Although the protection of human health has top priority in chemical risk assessment and management, the criteria currently used to identify bioaccumulative compounds (defined via the bioaccumulation factor BAF, the bioconcentration factor BCF or the octanol-water partition coefficient $K_{\Omega(W)}$ are only based on bioaccumulation in aquatic organisms $(1, 2)$. A measure recently introduced to quantify the potential of a persistent chemical to biaccumulate in humans is the environmental bioaccumulation potential EBAP⁽³⁾. The major difference compared to the commonly used BAF is the reference matrix used for identifying the magnitude of bioaccumulation: The BAF uses the contaminant concentration in one single environmental phase, whereas the EBAP compares the human body burden with the contaminant's quantity in the total environment the human is living in. Normalizing the environmental concentrations to the surface area yields the unit m^2 per person for the EBAP. It thus stands for the environmental area containing the same amount of chemical as having been accumulated in one person.

Unusually high tissue levels of POPs observed in residents of the remote Arctic (4) indicate regional differences of the general potential of a chemical to bioaccumulate in humans. The aim of this modeling exercise was to identify the major factors causing such regional differences of the EBAP for non-polar organic chemicals. For this purpose, the bioaccumulation up to humans in the Arctic was compared with that in northern Europe, a region which differs not only in the characteristics of the physical environment, but also in the traditional humans' diet and thus the primary exposure pathway of POPs.

Material and Methods

The EBAP for a 30 yr old woman living in northern Europe and nursing her first child was calculated as a function of the chemicals' octanol-air and octanol-water partition coefficients ($\mathcal{K}_{\rm OA}$ and $\mathcal{K}_{\rm OW}$, respectively) in a previous paper $^{(3)}$.

Following the same modelling approach, a level I unit world (5) , consisting of the four compartments air, water, soil and sediment, was defined and parameterized for the Arctic. 75% of the area was assumed to be covered by water with an additional 23% covered by permanent snow and ice. The remaining area was attributed to soil. The organic carbon content was set to 2% for soil and 1% for sediment (7) . The depth of the four compartments was set to 1000 m, 100 m, 0.5 cm and 10 cm $^{(8)}$, respectively. A fixed amount of hypothetical, completely persistent chemicals differing from each other by their octanol-air and octanol-water partition coefficientswas discharged into the unit world, assuming equilibrium partitioning between the four compartments. Subsequently, a human food chain model was linked to the unit world and the human exposure resulting from the contaminants' partitioning behaviour was investigated for each of the hypothetical persistent compounds. While for the EBAP calculations for northern Europe the mechanistic bioaccumulation model ACC-HUMAN was used, which considers human exposure via an aquatic and an agricultural vector $(3, 9)$, the calculations for the Arctic were performed with a modified version of the marine module of ACC-HUMAN⁽¹⁰⁾. As seal, and above all the seal blubber, has been identified to make a major contribution to the exposure to Inuit living on a traditional diet ⁽¹¹⁾, this model focuses on the seal's food chain, consisting of zooplankton, ice amphipods, polar cod and ringed seal, which in turn is consumed by the human. In accordance with ref. (3) a 30 yr old woman who had been living in the contaminated unit world during all her life was chosen as the simulation end point. In accordance with the more traditional lifestyle, the Inuit mother was nursing her third child (the first and second children were born at an age of 20 and 25 yr, respectively; lactation length six months for each child).

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Results and Discussion

The influence of the physical chemical properties on the partitioning behaviour in the physical environment is similar in the two environments. However, as a result of the 2.4 times smaller quantity of organic material in the arctic unit world model (which is mainly due to the about three orders of magnitude smaller soil compartment), some chemicals with intermediate K_{OW} and K_{OA} values that are predicted to be mainly present in the solid phase in northern Europe are primarily sequestered into water or air in the Arctic. Consequently, these chemicals show higher water (or air) concentrations in the Arctic compared to Europe and can thus be expected to be more amenable to bioaccumulation through the food web up to humans.

Figure 1a shows the predicted EBAP for a 30 yr old Inuit women who has lived all her life on a traditional marine diet. The EBAP is plotted as a percentage of the maximum possible EBAP against the partition coefficients K_{OW} and

 K_{OA} . The corresponding partitioning map for the European environment shows a comparable result for a woman

whose dietary exposure to persistent chemicals was solely via the marine diet modeled on a Baltic Sea food chain ⁽³⁾. However, the maximum EBAP in the Arctic (1.1 \times 10⁵ m² person⁻¹) is about three orders of magnitude higher than the maximum for the Baltic Sea marine food chain (80 m² person⁻¹)⁽³⁾, indicating that there is a much greater potential for bioaccumulation in the Arctic. The maximum EBAP occurred for a chemical with log $\kappa_{\rm OA}$ > 8 and log $\kappa_{\rm OW}$ of about 6, which corresponds to the properties of many PCB congeners.

An indication of which factors have an impact on the elevated EBAP in the Arctic can be obtained by comparing the concentrations predicted in the EBAP simulations for each trophic level as well as the water concentration the simulation is based on. The results for a hypothetical chemical with log $K_{OA} = 9$ and log $K_{OW} = 6$ are shown in Figure

1b. For both regions the same quantity of chemical $(10^{-7}$ g m⁻²) was introduced into the environment. The resulting lipid normalized concentration in the Inuit woman was 2200 times higher than in the European woman. This is even greater than the difference in EBAP since EBAP is calculated from body burden, and the lipid mass of the smaller Inuit woman is less than for the European woman.

Figure 1. Environmental bioaccumulation. a) Environmental bioaccumulation potential EBAP in Inuit as a function of K_{OW} and K_{OA} , plotted as a percentage of the maximum EBAP within the chemical partitioning space. The results are for a 30 yr old woman living entirely on a traditional marine diet and nursing her third child. b) Comparison of the predicted concentrations of a hypothetical chemical (log $K_{OW} = 6$, log $K_{OA} = 9$) along the food chains of the Baltic Sea and the Arctic, using the combined region specific unit worlds and bioaccumulation models. Note the two different scales on the y-axes referring to the water concentrations (first pair of bars, axis on the left) and the biota concentrations (axis on the right). The concentrations in the biota reflect the weighted mean concentrations of different age classes in accordance with their contribution to the predator's diet. The 30 yr old European woman was living on

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a typical diet of marine and terrestrial origin and nursing her first child, while the 30 yr old Inuit woman was nursing her third child.

The analysis shows, that only a small portion of the elevated levels in the Inuit woman is attributable to the observed differences in the chemical's partitioning behavior in the physical environment, whereas food chain bioaccumulation was accountable for the major portion, being 850 times greater in the Arctic compared to the Baltic. Each trophic level contributed to the enhanced bioaccumulation, though a strong increase is first observed at the fish predator level. Biomagnification in the ringed seal was 8 times greater than bioaccumulation in Baltic cod, which occupies a comparable trophic level. Mammals are much stronger biomagnifiers of lipophilic organic contaminants due to their more efficient food digestion, slower elimination via respiration, and longer life. However, the greatest contribution to the high levels in Inuit comes from the final link in the food chain. Biomagnification in the Inuit woman was 35 times greater than in the European. This was due to the much greater reliance of Inuit on food from the marine food chain, as reflected in their ~50 times higher dietary intake of marine lipids. The greater number of children of the simulated Inuit woman served to reduce her tissue levels compared to the simulated European woman.

The consequence of the enhanced bioaccumulation potential in the Arctic is that critical levels of human exposure will result from much lower levels of input of persistent chemicals. This makes the Arctic environment particularly sensitive to the impacts of persistent organic contaminants.

Acknowledgements

Financial support from the European Union (FAMIZ grant EVK3-CT-2000-00024) and from the CEFIC Long-Range Initiative is gratefully acknowledged.

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