Identifying the Persistent Organic Chemicals with the Highest Potential for Accumulation in Arctic Residents

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Introduction

Since the first observation of high concentrations of polychlorinated biphenyls (PCBs) in breast milk samples from the Canadian Arctic in the late 1980s, high exposure of arctic indigenous people to persistent organic pollutants (POPs) has been confirmed in a variety of studies [1-3]. As a consequence, the identification of the properties of organic contaminants with the ability to reach the remote Arctic and to bioaccumulate through food chains up to humans became a high priority in chemical risk assessment and management. In general, these properties are i) persistence in air (or water) to survive long-range transport, ii) semi-volatility allowing the chemical to partition into and be transported with the air phase, but on the other hand also be deposited again in the Arctic, and finally iii) lipophilicity making the chemical bioaccumulative.

For the identification and assessment of the governing properties, two concepts have been introduced recently: the arctic contamination potential ACP [4], and the environmental bioaccumulation potential EBAP [5]. The ACP quantifies a chemical's enrichment in the arctic physical environment. It is the ratio of the contaminant's quantity in the arctic surface compartments (soil, water and sediment) and the quantity present in the total global environment [4]. The EBAP is a measure for the potential of a contaminant to bioaccumulate in humans on the basis of the environmental concentrations [5]. It is the quotient of the human body burden and the chemical's amount in the total environment the human is living in (normalized to the surface area of the environment). The EBAP thus has a unit of m^2 per person, i.e. it describes the area containing the same amount of chemical as has been accumulated in one person. In this modelling exercise, the concepts of ACP and EBAP are combined to identify physical-chemical properties enabling a chemical to accumulate both in the physical environment and the human food chain of the Arctic, thus providing an integrated measure of the potential of human exposure in the Arctic to a chemical used in lower latitudes.

Material and Methods

For the simulations, hypothetical completely persistent chemicals were defined, differing from each other only by their air-water and octanol-air partition coefficients (K_{AW} and K_{OA}, respectively), which can be viewed as a measure of the chemicals' volatility and lipophilicity. Thereby, a broad range of the chemical partitioning space was covered, which comprises the majority of the organic contaminants of environmental relevance. In order to receive clear-cut information on the chemicals' potential for arctic enrichment and bioaccumulation, properties such as the persistence were kept constant in the simulations. Adapting the approach presented by Wania [4], the ACP was calculated using the multimedia fate and transport model Globo-POP and a scenario of 10 years of continuous global chemical emission into air distributed according to the human population's global zonal distribution [6]. For the calculations of the EBAP a modelling scenario was chosen in accordance with the approach suggested by Czub and McLachlan [5]: A four-compartment level I unit world [7] was defined and linked to a human food chain model characteristic for the arctic region. 75% of the arctic environment was assumed to be covered by water, an additional 23% by permanent snow and ice, and the remaining area was assigned to soil. The depth of the four compartments air, water, soil, and sediment were set to 1000 m, 100 m, 0.5 cm and 10 cm [8], respectively. Soil was assumed to have an organic carbon content of 2%, sediment of 1% [9]. The hypothetical chemicals were assumed to attain a partitioning equilibrium in the unit world and subsequently entering an arctic food chain, which was described with a modified version of the bioaccumulation model ACC-HUMAN [10, 11]. This model application focuses on the seal food chain, as seal and in particular seal blubber is one of the main vectors of exposure to PCBs for arctic indigenous people living on a traditional diet [12]. The endpoint of the simulation was a 30 yr old Inuit woman living entirely on a traditional

marine diet and nursing her third child (first and second child at an age of 20 and 25 years, respectively; lactation length six months for each child).

Results and Discussion

The ACP of the hypothetical persistent chemicals is shown in Figure 1a as a function of K_{OA} and K_{AW}. It is presented as the percentage of the maximum ACP within the investigated chemical partitioning space (4.5), which was obtained for the rather hydrophilic compound with a log K_{OA} < 7 and a log K_{AW} of about -2 [4]. The partitioning map for the EBAP of the Inuit woman is presented in Figure 1b as percentage of the maximum EBAP (1.1×10^5 m^2 $person^{-1}$). Maximum values were obtained for chemicals with a log K_{OA} > 8 and a log K_{OW} of about 6. In contrast to the ACP partitioning map, a steep gradient can be observed and a large fraction of the investigated partitioning space shows EBAP values less than 10% of the maximum. Following the approach presented in ref. [5], which suggests that chemicals could be classified as potentially bioaccumulative in humans if they possess an EBAP of at least 10% of the maximum EBAP, the bioaccumulation thresholds for the Arctic are log K_{OA} > 6 and 3.5 < log K_{OW} < 9.

The partitioning property combinations of maximum ACP and EBAP are located in different areas within the chemical partitioning space. However, there is also a large area of overlap. By multiplying the (normalized) ACP and EBAP for each hypothetical chemical, those compounds can be identified which have an elevated potential to reach the arctic region AND to bioaccumulate through the food web up to humans, i.e. chemicals being used in the lower latitudes with a particular high potential to accumulate in humans living in the Arctic (Fig. 1c). Using again the 10% isoline as a threshold, only a narrow "band" covering about three to four orders of magnitude of the K_{OA} ($7 < \log K_{OA} < 10$ to 11) features elevated ACP × EBAP levels. In that "band", the lower threshold (on the left) is determined by the bioaccumulation potential, as more volatile compounds cannot bioaccumulate in the arctic food chain. As a consequence, only a low potential for human exposure in the Arctic is predicted for the rather hydrophilic and semi-volatile chemicals for which the highest ACP was identified (Fig. 1a). The upper threshold (on the right) on the other hand is to a large extent controlled by the potential for long-range transport, because the involatile chemicals are not transported to the Arctic very efficiently.

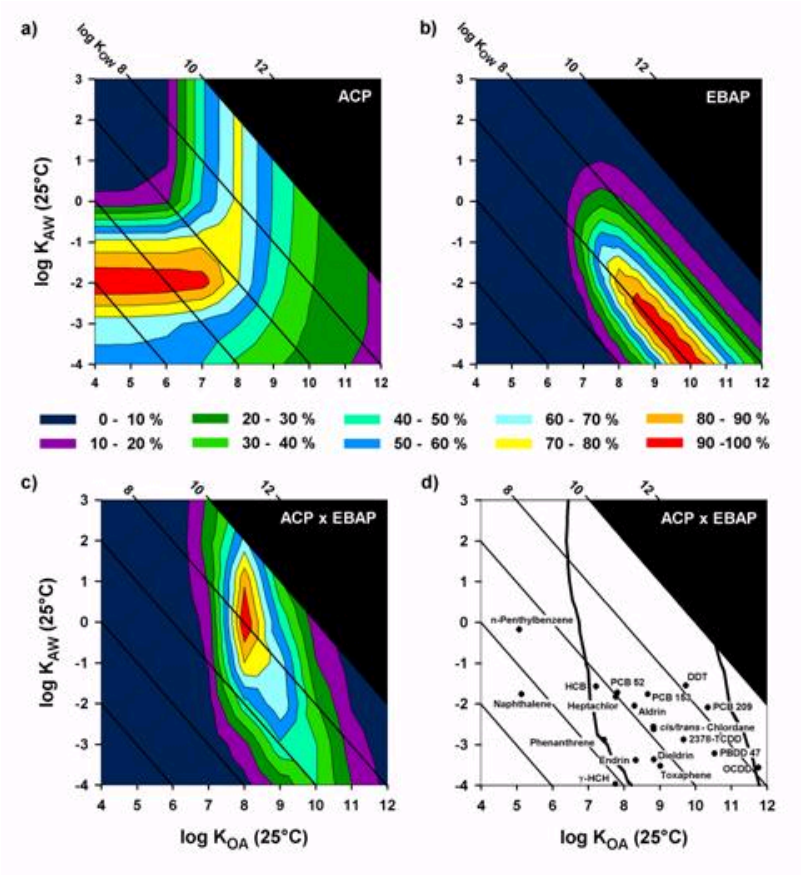

Figure 1. Chemical enhancement and bioaccumulation in the Arctic as a function of K_{OA} and K_{AW}. The results are presented as the percentage of the maximum parameter value within the investigated chemical partitioning space; a) Arctic contamination potential ACP after 10 years of continuous global emission into air; b) Environmental bioaccumulation potential EBAP of a 30 yr old Inuit woman living entirely on a traditional marine diet and nursing her third child; c) Chemical accumulation in the physical environment AND bioaccumulation in the human food chain of the Arctic as described by the product of the ACP and EBAP for each chemical within the investigated partitioning space; d) 10% isoline of the ACP × EBAP overlay in comparison with the location of selected organic chemicals within the chemical partitioning space [13-16].

It is unclear whether there are real chemicals with partitioning properties corresponding to the upper part of the elevated ACP × EBAP region. The maximum value is obtained for chemicals with a log K_{OA} of 8, a log K_{AW} of about

0, and a log K_{OW} of about 8, a rather exceptional combination of partitioning properties, which however might apply to some long-chain perfluorinated alkanes and volatile cyclic organosilicons . Figure 1d shows the location of selected organic substances within the investigated chemical partitioning space [13-16]. It reveals that the 10% isoline indeed encompasses the chemicals of concern in the arctic environment, e.g. PCBs, polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), chlordanes, toxaphene and polybrominated diphenyl ethers (PBDEs). These results suggest that the link of the modeling concepts of ACP and EBAP holds promise as a screening tool, as it illustrates the combined effects of partitioning on long range transport potential and bioaccumulation potential. With further development it could also address chemical persistence, which is also an important determinant of the risk of chemical emissions in the lower latitudes to the indigenous people in the Arctic.

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