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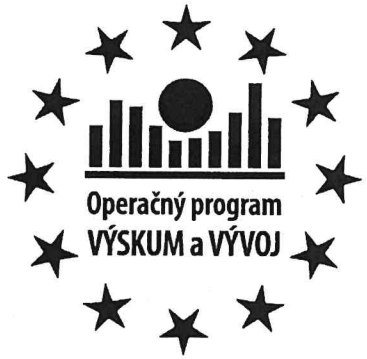

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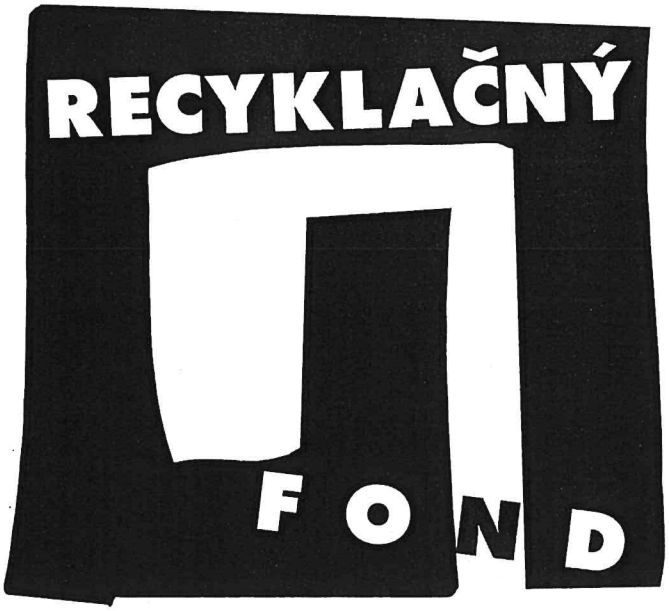

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PREDICTING ECOTOXICITY OF SELECTED BFRS USING ECOSAR

Šenk Nevena [1], Okuka Marija [1], Sekulić Maja Turk [1], Radonić Jelena [1], Miloradov Mirjana Vojinović -[1], Mihajlović Ivana [1], Stošić Milena [1]

Brominated flame retardants (BFRs) are organobromide compounds that have an inhibitory effect on the ignition of combustible organic materials, and are most widely used of all the commercialized chemical flame retardants. BFRs are very effective in plastics and textile applications, e.g. electronics, clothes and furniture. BFRs can be released to the environment during their production and while manufacturing other products, but also during disposal of products containing these chemicals, and, in addition, BFRs may continue to leak out of treated material. Due to their persistence, potential for long-range atmospheric transport, bioaccumulation and biomagnification, BFRs enter all environmental mediums, including aquatic ecosystems. Ecological Structure Activity Relationships (ECOSAR) is a computer software program, which is connecting chemical properties of pollutants with toxicity to aquatic and terrestrial organisms. ECOSAR is used to estimate toxicity of chemicals used in industry and discharged into water. The program predicts the toxicity of industrial chemicals to aquatic organisms such as fish, invertebrates, and algae by using Structure Activity Relationships (SARs). The program estimates a chemical's acute (short-term) toxicity and, when available, chronic (long-term or delayed) toxicity. In this paper, ECOSAR was used for calculating aquatic toxicity of nine emerging BFRs, from the NORMAN's (Network of reference laboratories for monitoring of emerging environmental pollutants) list of emerging substances.

Keywords: Ecotoxicity, BFRs, Software, ECOSAR

Introduction

Emerging substances are defined as substances that have been detected in the environment, but which are not currently included in rutine monitoring programs at European Union level and whose fate, behavior and (eco)toxicological effects are not well understood. Data for

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emerging substances are often scarce and measurement methods are often at the research and development stage or have not yet been harmonised at the EU level. This makes it difficult to interpret and compare the results and represents a major difficulty for regulatory bodies in their decision-making.

Network of reference laboratories for monitoring of emerging environmental pollutants (NORMAN) has identified a list of the currently most frequently discussed emerging substances, with the latest update in March 2011. Emerging substances have been selected by the NORMAN experts, based on citations in the scientific literature, and taking into account the definition of "emerging substances". The NORMAN's list is by definition an open dynamic list, and experts are in charge of regularly revising the list. Emerging substances in NORMAN's list are divided into 25 categories with examples of individual emerging substances more than 700 [1]. One of the categories are flame retardants, and a part of it are brominated flame retardants (BFRs).

Brominated flame retardants (BFRs) comprise a structurally diverse group of aliphatic, cycloaliphatic, and aromatic compounds, of which the most widely produced and studied are tetrabromobisphenol A (TBBPA), hexabromocyclododecane (HBCD), and polybrominated diphenyl ethers (PBDEs) [2]. BFRs are used as additive or reactive components in a variety of polymers, such as polystyrene foams, high-impact polystyrene, and epoxy resins. These polymers are then used in a medley of consumer products, including computers, electronics and electrical equipment, televisions, textiles, foam furniture, insulating foams, and other building materials.

TBBPA is covalently bound to plastic and is used in electronic circuit boards. One moiety of TBBPA's molecular structure is similar to that of the thyroid hormone thyroxine, except that the iodine atoms have been replaced by bromines [2].

HBCD is produced by bromination of cyclododecane in a batch process. It is used in foams and expanded polystyrene. End products include upholstered furniture, interior textiles, automobile interior textiles, car cushions and insulation blocks in trucks and caravans as well as in building materials such as house walls, cellars, roofs and parking decks, against frost heaving in roads and railway embankments, packaging material, housing and electric equipment [2].

The PBDEs theoretically consist of 209 different congeners, varying in both number and position of bromination, but there appear to be many fewer actual PBDE congeners in the commercial mixtures than the theoretical number possible, largely because many of the congeners lack stability and tend to debrominate. These chemicals are major components of commercial formulations often used as flame retardants in furniture foam (pentaBDE), plastics for TV cabinets, consumer electronics, wire insulation, back coatings for draperies and upholstery (decaBDE), and plastics for personal computers and small appliances (octaBDE)[3]. There is growing evidence that PBDEs persist in the environment and accumulate in living organisms, as well as toxicological testing that indicates these chemicals may cause liver toxicity, thyroid toxicity, and neurodevelopmental toxicity. Environmental monitoring programs in Europe, Asia, North America, and the Arctic have found traces of several PBDEs in human breast milk, fish, aquatic birds, and elsewhere in the environment. Particular congeners, tetra- to hexabrominated diphenyl ethers, are the forms most frequently detected in wildlife and humans. The mechanisms or pathways through which PBDEs get into the environment and humans are not known yet, but could include releases from manufacturing or processing of the chemicals into products like plastics or textiles, aging and wear of the end consumer products, and direct exposure during use (e.g., from furniture). In the United States, the manufacture of penta- and octabrominated diphenyl ethers was voluntarily ended in 2004, and in the European Union, the use of penta- and octabrominated diphenyl ethers and polybrominated biphenyls in electronic equipment has been restricted since 2006. [2]

The presence of BFRs in the environment causes concern, and the occurrence of high concentrations of certain representatives and their isomers may be sufficient to cause adverse effects in some wildlife. There is also concern that BFRs could cause adverse effects in sensitive human populations such as young children, indigenous peoples, and fish consumers. Knowledge about these chemicals, their sources, environmental behavior, and toxicity is limited. Many of these substances are persistent and lipophilic and have been shown to bioaccumulate.

Predictive models in the form of quantitative structure activity relationships (QSARs) provide estimates for physical/chemical properties, reaction rates, and biological activity of chemical substances. They are based on the concept that the activity of a substance is a function of its structure and that the activity can be determined through mathematical relationships developed from architecturally similar compounds. Activity data have been generated by experimental testing of the chemical substances using a number of well-defined protocols. With laboratory costs and animal testing on the rise and the need to evaluate many more chemicals, regulatory agencies and other decision makers have become increasingly interested in new methods for determining the potential fate and effects of chemical substances. This is especially true for the toxicity and environmental fate assessments, where the appropriate experimental studies can be very expensive and span many months. In addition, the use of QSARs to predict the toxicity of chemical compounds has the potential to reduce the use of animal testing [4]. The widely used computer program for QSAR prediction of chemical toxicity towards aquatic organisms is ECOSAR (Ecological Structure Activity Relationship).

The Ecological Structure Activity Relationships (ECOSAR) Class Program is a computerized predictive system that estimates aquatic toxicity. The program estimates a chemical's acute (short-term) toxicity and chronic (long-term or delayed) toxicity to aquatic organisms such as fish, aquatic invertebrates, and aquatic plants by using computerized Structure Activity Relationships (SARs)[5].

Materials and method

ECOSAR uses structure-activity relationships (SARs) to predict the aquatic toxicity of untested chemicals based on their structural similarity to chemicals for which aquatic studies are available. Application of structure activity relationships is a technique routinely used by

the U.S. Environmental Protection Agency (EPA) Office of Pollution Prevention and Toxics (OPPT) under the New Chemicals Program to estimate the toxicity of chemicals being reviewed in response to Pre-Manufacture Notices mandated under Section 5 of the Toxic Substances Control Act (TSCA). The toxicity data used to build the SARs are collected from publicly available experimental studies and confidential submissions provided to the U.S. EPA New Chemicals Program. The SARs in ECOSAR express correlations between a compound's physicochemical properties and its toxicity within specific chemical classes [5].

Expansion of the ECOSAR program was supported by the Office to assist the U.S. EPA/OPPT New Chemicals Program scientific staff in developing a complete standard toxicity profile for each chemical reviewed to characterize the potential aquatic hazard concerns. This standard profile consists of acute and chronic effects. Acute effects include 96 hour LC50 for fish, 48 hour LC50 for daphnid, and 72 or 96 hour EC50 for algae. LC50 is the median lethal concentration, the concentration in water which kills 50% of a test batch of fish (or other organisms) within a continuous period of exposure. Chronic effects include chronic values (ChV) for fish, daphnid and algae. The ChV is defined as the geometric mean of the no observed effect concentration (NOEC) and the lowest observed effect concentration (LOEC). This can be mathematically represented as [5]:

$$ChV = 10^{\frac{\log(LOEC \times NOEC)}{2}}$$

Toxicity to these surrogate species (fish, aquatic invertebrates, and aquatic plants) is used to predict toxicity to a general aquatic community. EPA/OPPT has focused resources on models for aquatic toxicity to fresh water organisms because most releases of industrial chemicals go to fresh water bodies. Although some terrestrial and marine species data were available in some cases and programmed into ECOSAR, terrestrial and marine species are only evaluated on a case by case basis depending on the manufacturing, processing, and use of the chemicals. The current version of ECOSAR strives to provide estimates for all 6 standard freshwater aquatic toxicity endpoints listed above for each class programmed into ECOSAR.

ECOSAR Version 1.1 is programmed to identify 111 chemical classes and allows access to 709 QSARs for numerous endpoints and organisms [5]. Selection of the appropriate QSAR within ECOSAR is based on a variety of information depending on the chemical class. This includes factors like the chemical structure, chemical class, log Kow, molecular weight, physical state, water solubility, number of carbons or ethoxylates (or both), and percent amine nitrogen or number of cationic charges (or both) per 1000 molecular weight. The most important factor for selecting an appropriate QSAR is the chemical class, since the QSARs in ECOSAR are class-specific.

In the paper, ECOSAR was chosen for predicting ecotoxicity of nine emerging BFRs to fish, because research papers show that ECOSAR categorised 87% of the chemicals correctly in chemicals classes[6], and it had over 67% correct estimations [6][7]. Also, ECOSAR was highly rated for predictions in comparison with five other QSAR programs [8].

Nine selected emerging BFRs for ECOSAR prediction of toxicity to fish are 1,2,5,6,9,10-Hexabromocyclododecane (HBCD), 2,2',3,3',4,4',5,5',6,6'-Decabromodiphenyl ether (BDE-209), 2,2',3,4,4',5',6-Heptabromodiphenyl ether (BDE-183), 2,2',4,4',5,5'-Hexabromodiphenyl ether (BDE-153), 2,2',4,4',5,6'-Hexabromodiphenyl ether (BDE-154), 2,2',4,4'-Tetrabromodiphenyl ether (BDE-47), Technical octabromodiphenyl ether, Tetrabromo bisphenol A (TBBPA), Tetrabromo bisphenol A bis(2,3 dibromopropylether) (TBBPA-DBPE).

Results and discussion

Nine emerging BFRs chosen for ECOSAR prediction and their basic physical-chemical properties estimations are shown in table 1.

Table 1. Basic physical-chemical properties used in ECOSAR for calculating aquatic toxicity of nine emerging brominated flame retardants (# designates Physical Properties DataBase experimental value, ~ designates EPISuite WSKowwin v1.43 Estimate)

CAS No.	Name	$\log K_{OW}$ (EPISuite Kowwin v1.68 Estimate)	Water Solubility [mg/l]	Molecular Weight [g/mol]
3194-55-6	HBCD	7.742	0.0086#	641.70
1163-19-5	BDE-209	12.110	0.0001#	959.17
207122-16-5	BDE-183	9.440	2.156E-007~	722.48
68631-49-2 (6-01-7)	BDE-153	8.550	4.148E-006~	643.59
207122-15-4	BDE-154	8.550	4.148E-006~	643.59
5436-43-1	BDE-47	6.770	0.001461~	485.80
32536-52-0	Technical octabromodiphenyl ether	10.330	1.285E-006~	801.38
79-94-7	TBBPA	7.203	0.002146~	543.88
21850-44-2	2,3 dibromopropylether	11.517	1.164E-010~	943.62

In ECOSAR, octanol/water partition coefficient (Kow) was the major physical-chemical attribute correlating a chemical structure to toxic effect for nonreactive neutral organic chemicals. The most frequently used relationship is the logarithm of the Kow value versus the median toxicity (LC50 and EC50) value.

Log Kow was estimated using EPISuite Kowwin v1.68 for nine selected BFRs, and water solubility was estimated using EPISuite WSKowwin v1.43 for seven of nine selected BFRs, other two had experimental value in Physical Properties DataBase of ECOSAR.

After estimating log Kow values for nine emerging BFRs, ECOSAR calculated toxicity to fish, where endpoints were LC50, for duration of 96 hours and for 14 days, and also chronic value, all presented in measures of mg/l (ppm). Table 2. shows the results of aquatic toxicity, calculated with ECOSAR.

Table 2. Aquatic toxicity of selected emerging brominated flame retardants, gained with ECOSAR (designates that chemical may not be soluble enough to measure predicted effect, ! designates that toxicity value was estimated through application of acute-to-chronic ratios per methods outlined in the ECOSAR Methodology Document provided in the ECOSAR Help Menu)*

	CAS No.	Name	Fish 96-hour LC50 [mg/l] (ppm)	Fish 14-day LC50 [mg/l] (ppm)	Fish ChV [mg/l] (ppm)	ECOSAR Class
1.	3194-55-6	HBCD	0.004	0.005	0.000671	Neutral Organics
2.	1163-19-5	BDE-209	6.56e-007	1.1e-006	1.93e-007	Neutral Organics
3.	207122-16-5	BDE-183	0.000124*	0.000183*	2.72e-005*	Neutral Organics
4.	68631-49-2 (6-01-7)	BDE-153	0.000693*	0.000983*	0.000138*	Neutral Organics
5.	207122-15-4	BDE-154	0.000693*	0.000983*	0.000138*	Neutral Organics
6.	5436-43-1	BDE-47	0.021*	0.027*	0.003*	Neutral Organics
7.	32536-52-0	Technical octabromodiphenyl ether	2.18e-005*	3.35e-005*	5.27e-006*	Neutral Organics
8.	79-94-7	TBBPA	0.023* (0.009*)	-	0.006* (0.00163)	Phenols, Poly Neutral Organic SAR (Baseline Toxicity)
9.	21850-44-2	TBBPA-DBPE	1.5e-005* (2.2e-006*)	-	8.78e-007*! (6.06e-007*)	Halo Ethers Neutral Organic SAR (Baseline Toxicity)

Seven of nine selected brominated flame retardants (HBCD, BDE-209, BDE-183, BDE-153, BDE-154, BDE-47, Technical octabromodiphenyl ether) were classified as Neutral Organics, and TBBPA was classified as PolyPhenol, and TBBPA-DBPE was classified as Halo Ether.

BDE-183, BDE-153, BDE-154, BDE-47, Technical octabromodiphenyl ether, TBBPA, TBBPA-DBPE may not be soluble enough to measure this predicted effect, and no effects at saturation (NES) are reported.

Chronic value for TBBPA-BDPE was estimated through application of acute-to-chronic ratios per methods outlined in the ECOSAR Methodology Document provided in the ECOSAR Help Menu.

All toxicity results show that LC50 (96-hour and 14-day) and ChV concentrations in mg/l (ppm) are lower than 0.1, and that nine selected emerging BFRs are highly toxic, based on European Union Commission Guideline 93/67/EEC classification [9].

Literature data show that LC50 values for TBBPA, HBCD and BDE-47 are higher than estimated with ECOSAR [10][11][12][13][14]. Also, literature data show log Kow values for nine BFRs are lower than estimated with EPISuite Kowwin v1.68 [12][15][16].

Conclusions

ECOSAR is a widely used computer program for QSAR prediction of chemical toxicity towards aquatic organisms. The program estimates a chemical's acute (short-term) and chronic (long-term or delayed) toxicity to fish, aquatic invertebrates, and aquatic plants using computerized SARs. Calculations done with ECOSAR for nine emerging brominated flame retardants are based on the structure, classification within ECOSAR program, and log Kow values. As it was shown, log Kow values estimated with ECOSAR are higher than literature data show, and brominated flame retardants are classified as highly toxic, but calculated toxicity of these substances is higher than literature data. Also, in ECOSAR estimations, seven of nine selected BFRs might not be soluble enough to measure the predicted effect.
For future work, recommendation is to try other QSAR programs, such as TOPKAT, or DEREK, compare the results with ECOSAR estimations and validate results gained within each program, and, if possible, with experimentally gained values.

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PROGRAM PREDCHÁDZANIA VZNIKU ODPADU SR

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ABSTRAKT

Directive 2008/98/EC on waste introduces a new obligation - to establish and implement waste prevention programmes. The programmes shall set out the waste prevention objectives, describe the existing prevention measures implemented in the countries and evaluate the usefulness of measures proposed in the Directive. This is a challenge for the Slovak Republic as the waste management of the country has been focused rather on waste recycling and waste recovery than on waste prevention. Waste generation has been relatively stable during last five years. Waste Management Programme of the Slovak Republic for 2011 – 2015 approved by the Government, does not specify any prevention objectives. It was decided to develop a separate strategic document – Waste Prevention Programme of the Slovak Republic. The shift from waste recycling to waste prevention should be the main objective of the national programme. It can be implemented through series of informational, promotional and regulatory strategies that should increase the awareness of the society regarding negative environmental impacts of excess production and consumption.

1. ÚVOD

Smernica Európskeho parlamentu a Rady 2008/98/ES o odpade a o zrušení určitých smerníc (rámcová smernica o odpade) prináša do odpadového hospodárstva ES novú hierarchiu, ktorá kladie dôraz na predchádzanie vzniku odpadu a na opätovné používanie výrobkov s cieľom postupného obmedzovania využívania primárnych zdrojov. Jedným z opatrení podporujúcich praktické uplatňovanie hierarchie v odpadovom hospodárstve je povinnosť členských štátov vypracovať programy predchádzania vzniku odpadu.

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