

Targeting aquatic microcontaminants for monitoring: exposure categorization and application to the Swiss situation

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Abstract

Background, aim, and scope Aquatic microcontaminants (MCs) comprise diverse chemical classes, such as pesticides, biocides, pharmaceuticals, consumer products, and industrial chemicals. For water pollution control and the evaluation of water protection measures, it is crucial to screen for MCs. However, the selection and prioritization of which MCs to screen for is rather difficult and complex. Existing methods usually are strongly limited because of a lack of screening regulations or unavailability of required data.

Method and models Here, we present a simple exposure-based methodology that provides a systematic overview of a broad range of MCs according to their potential to occur in

the water phase of surface waters. The method requires input of publicly available data only. Missing data are estimated with quantitative structure–property relationships. The presented substance categorization methodology is based on the chemicals' distribution behavior between different environmental media, degradation data, and input dynamics.

Results Seven different exposure categories are distinguished based on different compound properties and input dynamics. Ranking the defined exposure categories based on a chemical's potential to occur in the water phase of surface waters, exposure categories I and II contain chemicals with a very high potential, categories III and IV contain chemicals with a high potential, and categories V and VI contain chemicals with a moderate to low potential. Chemicals in category VII are not evaluated because of a lack of data. We illustrate and evaluate the methodology on the example of MCs in Swiss surface waters. Furthermore, a categorized list containing potentially water-relevant chemicals is provided.

Discussion Chemicals of categories I and III continuously enter surface waters and are thus likely to show relatively steady concentrations. Therefore, they are best suited for water monitoring programs requiring a relatively low sampling effort. Chemicals in categories II and IV have complex input dynamics. They are consequently more difficult to monitor. However, they should be considered if an overall picture is needed that includes contaminants from diffuse sources.

Conclusions The presented methodology supports compound selection for (a) water quality guidance, (b) monitoring programs, and (c) further research on the chemical's ecotoxicology. The results from the developed categorization procedure are supported by data on consumption and observed

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concentrations in Swiss surface waters. The presented methodology is a tool to preselect potential hazardous substances based on exposure-based criteria for policy guidance and monitoring programs and a first important step for a detailed risk assessment for potential microcontaminants.

Keywords Biocides · Micropollutants · Chemical risk assessment · Emerging pollutants · Pesticides · Pharmaceuticals · Priority substances · Prioritization · Surface water

1 Background, aim, and scope

Aquatic microcontaminants (MCs) are important when considering freshwater quality and water pollution control. Although MCs are typically only present at low microgram per liter or even nanogram per liter levels, some of these chemicals raise toxicological concerns, in particular when present in mixtures (Schwarzenbach et al. 2006). MCs comprise diverse chemical classes such as pesticides, biocides, human and veterinary pharmaceuticals, consumer products, and industrial chemicals. Due to the increasing sensitivity and lower detection limits of recent analytical methods, which allow quantification of chemicals at low nanogram per liter levels, an increasing number of xenobiotic chemicals have been detected in surface waters during the last years (Stoob et al. 2005; Ternes 2007; Stamm et al. 2008). Some MCs, particularly polar pesticides that are mainly from agricultural sources, have been found in concentration levels where toxic effects on aquatic organisms have been reported (Chèvre et al. 2006). Beside diffuse sources such as agriculture, atmospheric long-range transport, and runoff from waste deposits, municipal wastewater has been identified as a major point source of organic MCs. Approximately 300 million tons of xenobiotic chemicals annually used in industrial and consumer products are partially entering natural waters through wastewater discharge, sewage overflows, and canalization leakages (Schwarzenbach et al. 2006). Additionally, moderately–highly persistent hydrophilic compounds, such as pharmaceuticals, are not efficiently removed by current state of the art sewage treatment plants (STPs) and may enter natural surface waters in high amounts (Joss et al. 2006; Ort et al. 2009). Recently, novel water protection measures to reduce these point sources, such as additional treatment steps in STPs, have been discussed (Benner et al. 2008; Joss et al. 2008). To evaluate the effect of such measures and to control the surface water quality relevant to MCs, some representative chemicals have to be selected for monitoring. However, because of the large number of potential MCs, it is difficult to identify which compounds should be considered.

For persistent organic pollutants (POPs) and arctic contaminants, which are subject to atmospheric long-range transport, many screening prioritization methods based on physical–chemical compound properties have been developed (Scheringer 1996; Brown and Wania 2008). However, these methods focus on global contaminants and do not contain criteria to identify aquatic MCs, which are often more polar and less persistent than POPs and, thus, primarily of regional impact. Existing prioritization methodologies for aquatic contaminants usually consider one or two selected compound classes or selected input pathways only. For example, some recently published prioritization methods focus on current-use pesticides or pharmaceuticals (Baun et al. 2006; Besse and Garric 2008). Reemtsma et al. (2006) presented a categorization method for substances entering the environment through STPs. By dividing the effluent concentration of a compound by its relative removal in STPs, they calculated a water cycle spreading index for each compound. Besides these methods, several environmental risk assessments (ERAs) that calculate risk quotients of chemicals have been published (Jones et al. 2002; Carlsson et al. 2006). In principle, ERAs are appropriate to rank and prioritize chemicals. However, ERAs require predicted no effect concentrations (PNECs) for an assessment of chemicals, whereas one of the main findings across recent studies is the lack of experimental ecotoxicological data to validate PNECs (Jones et al. 2002; Carlsson et al. 2006). In contrast to physical–chemical properties, the estimation of PNECs with quantitative structure–property relationships (QSPRs) is more complex and uncertain due to the typically unknown toxicological modes and sites of action (Escher and Hermens 2002). This lack of ecotoxicological data limits the outcome of ERAs and leaves a significant amount of chemicals unclassified. Similarly, currently used methodologies to identify emerging pollutants under the Water Framework Directive (WFD) of the European Union (EU) leave approximately half of the candidate chemicals unevaluated because of insufficient information (Wilkinson et al. 2007).

Currently, no methodology exists that categorizes the various MCs among different compound classes according to their properties and that provides a systematic overview of existing aquatic MCs. In practical application, this often leads to a selection of compounds for monitoring based on expert knowledge, which may be driven by temporary trends and individual analytical capabilities.

Here, we present a categorization methodology that is based on physical–chemical properties and information about degradation and input dynamics. To minimize the number of unclassifiable chemicals, ecotoxicological data are not considered. The minimization of the group of unclassifiable chemicals is crucial to avoid that potentially hazardous chemicals are a priori ignored because of a lack of data. With the developed methodology, different exposure categories

which integrate chemicals of similar potential to occur in surface waters and of similar input dynamics are distinguished. The goal of this categorization methodology is to support the selection of compounds for water protection policy guidance and the identification of appropriate monitoring strategies. To derive a chemical list that is suitable for monitoring programs, further criteria such as the availability of a state of the art analytical method have to be considered. In addition, chemicals for further ecotoxicological investigations can be identified and prioritized. The presented exposure-based categorization methodology can also be used as a first step in an ERA. However, to assess the hazard of the regarded chemicals, it is necessary to include ecotoxicological data.

We illustrate the presented method on the example of potential aquatic microcontaminants in Switzerland. We evaluate the categorization methodology using Swiss data on annual consumption and compare them with measured concentrations in Swiss surface waters. This is practical for some representative chemicals only, because consumption data are often not publicly available and may not be accurately estimated with the available information. Finally, we offer some recommendations for monitoring programs in Switzerland, based on the developed categorization methodology and Swiss monitoring data.

2 Methods and models

2.1 Candidate substance list for Switzerland

The candidate substance list of potential MCs was based on three criteria. The compounds (a) were listed in the EU WFD, (b) were listed in the list of relevant substances for the river Rhine, or (c) had been measured in Swiss surface waters (European-Commission 2006; IKS 2006). The candidate list was used as a training set to illustrate the developed methodology and to contrast this categorization with findings from experimental data. Moreover, it represents a case study for Switzerland and the developed list can be used for the selection of chemicals for monitoring in Swiss surface waters. However, the presented methodology can be applied to categorize MCs in general.

Annual Swiss monitoring data were provided by the Swiss Federal Offices (AWEL 2008; FOEN 2008a, b; CIPEL 2008) and additional data were collected from the literature (Freitas et al. 2004; Stoob et al. 2005). Compounds measured in the River Rhine at the permanent monitoring station of the IKS at Weil were of particular interest. The River Rhine has a catchment area in Switzerland of 36,494 km², which is about 80% of the total area of Switzerland. Thus, screening measurements in Rhine water are well suitable to gain a broad overview on persistent and mobile chemicals used in Switzerland.

The candidate list contained 250 substances from various compound classes. Compound classes are presented in Table 1 and the complete compound list is given in Table S1 in the Supporting Information. The candidate list includes biocides, pesticides, human and veterinary pharmaceuticals, estrogens and phytoestrogens, personal care products, mycotoxins, industrial chemicals, and metabolites. Two input pathways to surface waters were distinguished: (a) point sources that are inputs through municipal and industrial STPs and (b) diffuse sources that are not only inputs from agricultural applications and contaminated sites but also include inputs through atmospheric long-range transport and subsequent wet and dry deposition.

2.2 Categorization of candidate substances

In total, seven exposure categories are distinguished: (I) highly persistent chemicals that are continuously released into surface waters, (II) highly persistent chemicals with a complex input dynamic, (III) moderately persistent chemicals with a continuous input, (IV) moderately persistent chemicals with a complex input dynamic, (V) volatile and strongly sorbing chemicals, (VI) rapidly degradable chemicals, and (VII) unclassifiable chemicals. The seven exposure categories are discussed in detail in the Results section. The categorization procedure is given in Fig. 1. The compounds are categorized using three filters: (a) distribution behavior between different environmental media, (b) compound degradability, and (c) input dynamics. If the required chemical property data are not available, the selected compound properties are estimated with publicly available QSPRs, such as EPI SuiteTM (U.S.EPA 2007). If structurally similar compounds are lacking in the training set, then the application of a QSPR is not possible. In this case, the chemicals are not classifiable and assigned to category VII. The three different filters are discussed in detail below.

2.2.1 Filter 1: distribution between media

Filter 1 distinguishes between chemicals that are mainly present in the water phase, which are the main focus of this work, and volatile or strongly sorbing chemicals. To estimate the chemicals' distribution in a representative surface water, we use a phase-equilibrium Mackay-type model (Mackay 2001). Four compartments were considered: water, air, sediment, and suspended particles. Assuming phase equilibrium, the water-phase fraction of a chemical, ϕ_W [dimensionless], is calculated as shown in Eq. 1:

$$\phi_W = \frac{1}{1 + v_{AW} \times D_{AW} + v_{SW} \times \rho_S \times D_{SW} + v_{PW} \times \rho_P \times D_{PW}} \quad (1)$$

Table 1 Potential relevant compound classes and their major input pathways into surface waters

Point sources (municipal waste water, industrial inputs)		Diffuse sources (agriculture, atmospheric transport, waste deposits, and contaminated sites)	
Biocides	Fungicides Herbicides Insecticides Microbicides	Pesticides	Fungicides Herbicides Insecticides Growth regulators
Biocide–metabolites		Pesticide–metabolites	
Pharmaceuticals	Analgetics Antibiotics Antidepressants Anticonvulsants Beta blockers Contrast agents Lipid lowering agents	Veterinary–pharmaceuticals	Anthelmintics Antibiotics
Pharmaceutical–metabolites		Veterinary–pharmaceutical–metabolites	
Estrogens		Mycotoxins Phytoestrogens	
Personal care products	UV filters Musk fragrances		
Industrial–chemicals	Additives Flame retardants Solvents Surfactants Anticorrosive/complexing agents Cooling and insulating liquids Plasticizers	Industrial chemicals	Combustion products Formulation additives

where D_{AW} [liter water per liter air] is the air–water partition coefficient; D_{SW} [liter water per kilogram sediment] is the sediment–water partition coefficient; D_{PW} [liter water per kilogram particles] is the particle–water partition coefficient; v_{AW} , v_{SW} , and v_{PW} [dimensionless] are the volume fractions; ρ_S [kilogram per liter] is the density of the sediment; and ρ_P [kilogram per liter] is the suspended particle density. For ρ_S and ρ_P , the general multimedia model default values of 1.4 and 2.4 kg/L are assumed, respectively (Mackay and Paterson 1991). For the volume fraction, generic values that represent an average medium-sized Swiss lake and a sediment depth of 1 cm are assumed: $v_{AW}=200$, $v_{SW}=10^{-3}$, and $v_{PW}=10^{-5}$ (Schwarzenbach et al. 2003; Zennegg et al. 2007).

The partitioning between particles and water and sediment and water is approximated by the adsorption of chemicals into organic carbon (OC). D_{SW} and D_{PW} are given in Eq. 2:

$$D_{SW} = D_{OC} \times f_{OC,S}; \quad D_{PW} = D_{OC} \times f_{OC,P} \quad (2)$$

where D_{OC} [liter water per kilogram OC] is the OC–water partition coefficient, $f_{OC,S}$ [kilogram OC per kilogram sediment] is the fraction of OC in the sediment, and $f_{OC,P}$

[kilogram OC per kilogram particles] is the fraction of OC in suspended particles. In suspended particles, $f_{OC,P}$ can vary between 0.3 and 0.03 (Schwarzenbach et al. 2003). As an approximation, we assume an average value for the OC fraction in suspended particles of $f_{OC,P}=0.1$. The total OC fraction in sediments in most middle European lakes varies between 0.01 and 0.05 (Hollerbach 1984; Zennegg et al. 2007). For our generic model, we use the average OC fraction of sediments of the Swiss Lake Greifensee between 1906 and 1987, $f_{OC,S}=0.03$ (Zennegg et al. 2007). D_{OC} [liter water per kilogram OC] is calculated by Eq. 3 (Mackay 2001):

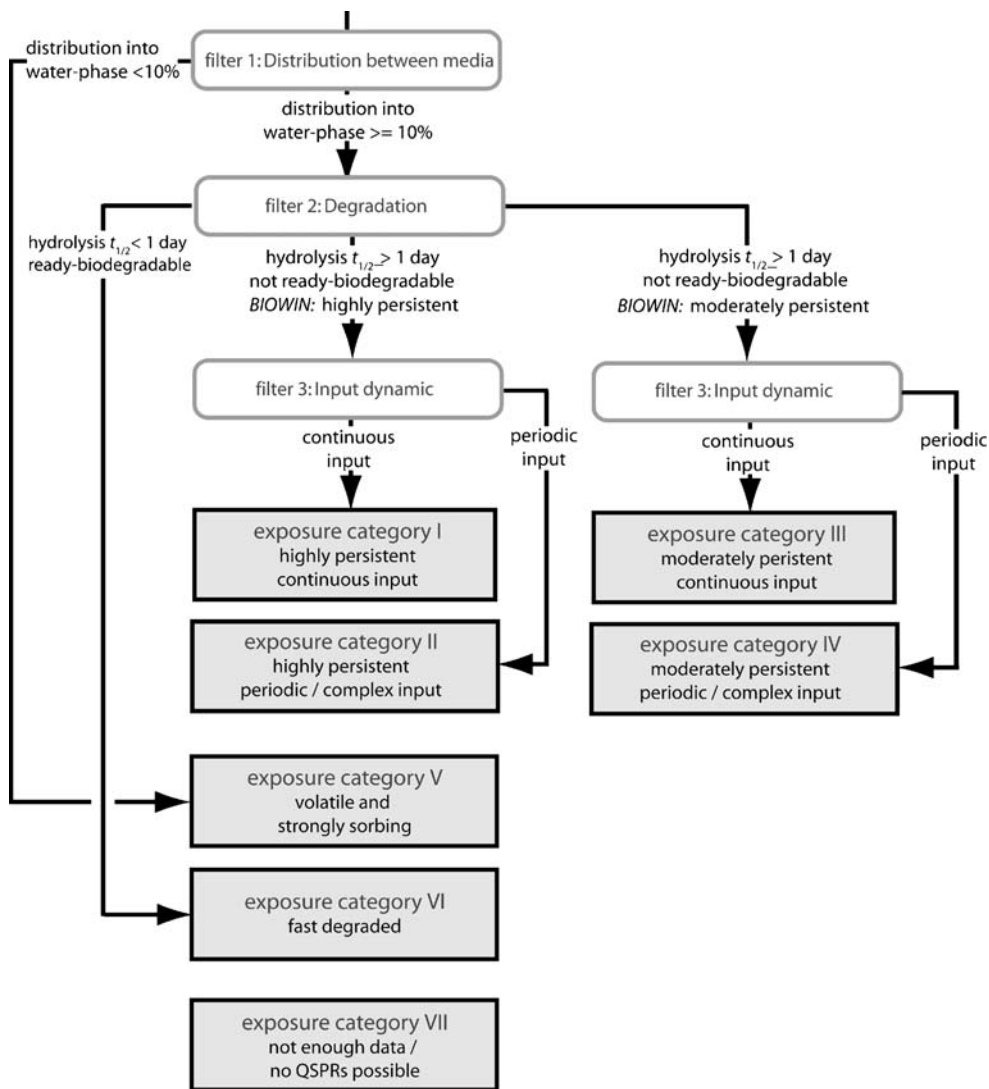
$$D_{OC} = (1 - \alpha) \times K_{OW} \times 0.41 \quad (3)$$

where $(1-\alpha)$ [dimensionless] is the neutral fraction, which can be calculated from the acidity constant, pK_A , and the pH: $\alpha = 10^{-pK_A} / (10^{-pH} + 10^{-pK_A})$ (Schwarzenbach et al. 2003). For surface waters, an average pH value of 7 is assumed. For neutral molecules, α is equal to 0.

Similarly, the partition coefficient between water and air, D_{AW} , is calculated with the dimensionless Henry coefficient, K_{AW} , and α :

$$D_{AW} = (1 - \alpha) \times K_{AW}. \quad (4)$$

Fig. 1 Categorization procedure and resulting categories for potential aquatic pollutants based on physical–chemical properties and input dynamics



Using Eq. 1 and the proposed model framework, the water-phase fraction ϕ_w of a chemical can be determined in direct relationship to its $\log D_{OW}$ and $\log D_{AW}$ values. The threshold for water-phase chemicals was set to $\phi_w = 0.1$ for filter 1. This means that chemicals are considered for water-phase monitoring if they are predicted to be present in the water phase at levels greater than 10% under average conditions. Chemicals that distribute mainly to solids need a different kind of assessment than that focused on in this work. We set the cutoff value relatively conservative ($\phi_w = 0.1$) to avoid that chemicals, considering parameter and model uncertainties and natural variations, are erroneously excluded from water-phase monitoring.

$\phi_w = 0.1$ corresponds approximately to the threshold values for $\log D_{OW}$ and $\log D_{AW}$, given in Eq. 5:

$$\begin{aligned} \log D_{OW} &< 5.9 \\ \log D_{AW} &< -1.4 \end{aligned} \quad (5)$$

The threshold value of $\log D_{AW}$ is in agreement with the value that has been assumed by Baun et al. (2006) for urban stormwater discharge. They identified compounds with a $\log D_{AW} \geq 1.4$ as chemicals with a low potential to occur in the water phase.

For polar chemicals, recent publications show that a better estimation of sorption to OC can be achieved with poly-parameter linear free energy relationships (ppLFERs). However, the most significant deviation between K_{OC} estimated with ppLFER models and K_{OW} -based models is in a $\log K_{OC}$ range between -1 and 2 (Nguyen et al. 2005), which is not relevant for the cutoff values needed in this work. Because of the better data availability of $\log K_{OW}$ values and because QSPRs for the solvation parameters needed for ppLFERs show a considerable uncertainty (Götz et al. 2007), we do not consider ppLFERs here.

If experimental K_{OC} data or solvation parameters for the selected compounds are available, these values may be preferred over the estimated K_{OC} values based on K_{OW} . This

is particularly the case for chemicals present at pH 7 as cations or as dipolar ions. The sorption of cationic and dipolar chemicals cannot accurately be described with the proposed K_{OW} -based model in Eq. 3 and an alternative approach is preferred. However, using a K_{OW} -based approach to estimate sorption of cationic chemicals underestimates the sorption, which might incorrectly assign the chemical as a water-phase chemical but not the other way, which is in agreement with the precautionary principle.

2.2.2 Filter 2: degradation

Filter 2 differentiates between readily degradable, moderately persistent, and highly persistent chemicals. We used ready biodegradability and hydrolysis data to identify fast degradation. If no information was available on these two processes, we applied the precautionary principle within the methodology and assumed the chemical was neither readily biodegradable nor rapidly hydrolyzed. We assumed the chemicals did not undergo photolysis transformations because of a lack of data (applying the precautionary principle). We set the cutoff value for fast hydrolysis to $t_{1/2}=1$ day. A chemical with a half-life of 1 day could occur in small creeks near the source, but would not likely be found in lakes or larger rivers. Such rivers, such as the Rhine in Switzerland, cover an average flow distance of 20 to 50 km per day during base-flow conditions (FOEN 2008a, b). This means that a chemical with a half-life of less than 1 day is efficiently degraded during transport in rivers and is not likely to enter groundwater or downstream connected surface waters in relevant amounts. However, if such chemicals are used in very high amounts and continuously enter surface waters at different locations along a river, they may be found in surface waters even if they are constantly degraded.

Chemicals that are not readily biodegradable and that do not undergo fast hydrolysis are assumed to be moderately to highly persistent. To distinguish between moderately and highly persistent, we use QSPRs. To estimate the chemicals' biodegradation half-life in water, we applied the BIOWIN survey model from EPI Suite™ (U.S.EPA 2007). BIOWIN is based on an expert survey and uses a group contribution approach to predict biodegradability on a scale from 1 to 5. EPI Suite™ converts the results from the BIOWIN Survey Models into eight water half-life categories: <1.75, 180 days; 1.75–2.25, 60 days; 2.25–2.75, 37.5 days; 2.75–3.25, 15 days; 3.25–3.75, 8.7 days; 3.75–4.25, 2.3 days; 4.25–4.75, 1.3 days; and >4.75, 0.2 days. Fenner et al. (2006) have shown that BIOWIN can clearly differentiate between highly persistent chemicals, such as polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane (DDT), aldrin, and dieldrin (BIOWIN biodegradability on a scale of 1–5, <1.75), and moderately

persistent chemicals, such as atrazine, metolachlor, and diclofenac (BIOWIN biodegradability on a scale of 1–5, ≥ 1.75). The correlation between the half-life categories lower than 60 days ($\text{BIOWIN} \geq 1.75$) and experimental half-lives was very weak. Therefore, we differentiate here between highly persistent chemicals ($\text{BIOWIN} < 1.75$) and moderately persistent chemicals ($\text{BIOWIN} \geq 1.75$) only.

2.2.3 Filter 3: input dynamics

A substance can be released into surface water bodies continuously or as a regularly or randomly repeated pulse input. Consumption, transport and transformation mechanisms, and specifically input pathways of a compound directly influence the input dynamic and consequently the temporal concentration pattern in surface waters for most water-phase chemicals. Thus, filter 3 was established to consider input dynamics.

We distinguish between continuous inputs and complex input dynamics. The concentrations of chemicals that are continuously released are approximately independent of the season and, in the base flow of rivers, show quite constant concentrations. Substances that are present over the whole year, even if they are degraded, are labeled “pseudo-persistent” (Daughton 2004). The concentrations of the chemicals in rivers depend on weather conditions because of varying dilution with varying river discharge. Characteristic examples of continuously released chemicals are consumer products and pharmaceuticals, which enter surface waters continuously through STPs. Generally, chemicals that are released through diffuse sources always have complex input dynamics, whereas chemicals that are released through point sources can have continuous or complex input dynamics. In Table S3 of the Supporting Information input dynamics for all considered compound classes are given.

Other microcontaminants show complex input dynamics, which are due to seasonal application and/or to rain-event driven mobilization (e.g., pesticides, biocides in material protection). Nonpolar highly persistent chemicals, such as most POPs, show an even more complex input pattern; besides direct release into surface waters, atmospheric long-range transport, and subsequent deposition, rerelease from capped landfills, sediments, and contaminated soils can be of importance (Schneider et al. 2007).

3 Results

3.1 Probability to enter natural surface waters

Based on the different exposure categories that contain chemicals with similar distribution behavior, persistence, and input dynamics, we qualitatively estimate the probabilities

Table 2 Properties of the seven different exposure categories

Exposure category	Distribution into water phase	Persistence	Input dynamics	Potential to occur in surface waters ^a
I	≥10%	High	Continuous	Very high
II	≥10%	High	Complex	Very high
III	≥10%	Moderate	Continuous	High
IV	≥10%	Moderate	Complex	High
V	<10%	nc	nc	Moderate–low
VI	≥10%	Low	nc	Moderate–low
VII	One of the attributes above is not known			Unknown

Estimated potential of the chemicals of the different exposure categories to occur in natural surface waters

nc not considered

^aRelative comparison between different exposure categories: Only comparable if similar amounts are used and similar input pathways are assumed. Related to the water phase only

that the chemicals in the different categories will be detected in natural surface waters if they are used in similar amounts and have similar limits of detection, as shown in Table 2. Chemicals of exposure categories I and II have a very high potential, chemicals of categories III and IV have a high potential, and chemicals of categories V and VI have a moderate to low potential of occurring in surface waters. The potential of chemicals in category VII cannot be assessed.

3.2 Exposure categories

The different compound classes of the candidate substances that are contained in categories I–IV are shown in Fig. 2. The different exposure categories are discussed in detail below.

Exposure category I (high persistence, continuous input)

Exposure category I contains highly persistent chemicals that partition into the water phase at a level greater than 10% and that are continuously released into surface waters. Of the chemicals on the candidate list, those assigned to exposure category I include the highly persistent pharmaceuticals, such as contrast media, macrolide and fluoroquinolone antibiotics, pentachlorobenzene, perfluorooctanoic acid (PFOA), and perfluorooctane sulfonate (PFOS). Generally, these chemicals enter natural surface waters through STPs. Macrolide antibiotics and contrast media that are nonvolatile and only weakly sorbing to solids are not eliminated in STPs and can be detected in surface waters all over Switzerland (Göbel et al. 2005). Similarly, PFOA and PFOS are widely used as surfactants and are entering the environment mainly through STPs. Contrast media, PFOA and PFOS are conserved during their transport in rivers (Brauch et al. 2006; Huset et al. 2008). The widely used perfluorinated surfactants are of importance for water pollution issues because of their concurrent potential to bioaccumulate and high solubility.

A monitoring concept for compounds of exposure category I can relatively easily be established. Because of the low variability of their concentrations, time proportional composite samples or, in some cases, even grab samples may be sufficient. In Switzerland and other European countries, daily flow proportional samples of sewage effluents are taken on STPs that treat more than 5,000 habitants. These samples could be used for investigating average MP concentrations in sewage effluents and, thus, to estimate total environmental burden of MCs through STPs. However, the dynamics in consumption and thereby the environmental impact are region specific. Only some of

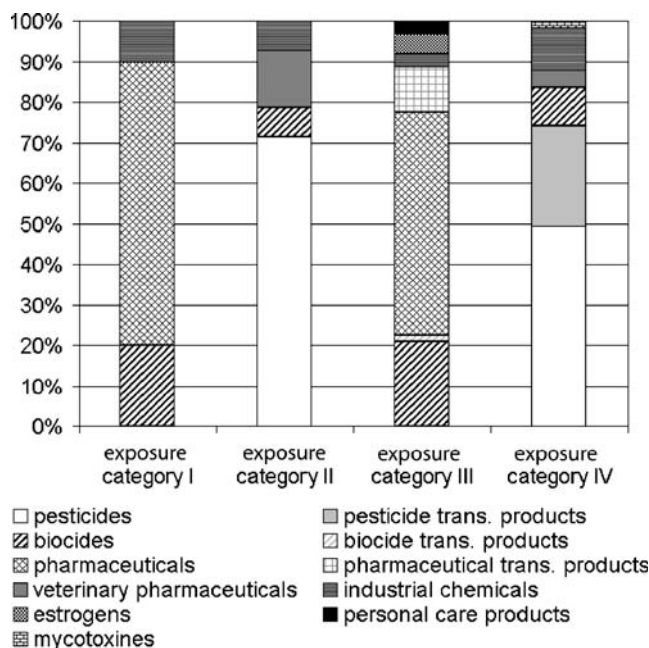


Fig. 2 Distribution of different compound classes to the potentially relevant exposure categories for exposure categories I–IV

these chemicals occur regularly in measurable concentrations in surface waters. High concentrations of these chemicals in natural surface waters are usually associated with a high fraction of urban wastewater. Generally, chemicals that are assigned to exposure category I have a high priority and should be considered for potential water protection measures.

Exposure category II (high persistence, complex input patterns) Exposure category II contains highly persistent chemicals that partition into the water phase at a level greater than 10% that have a complex input pattern. Category II comprises mainly industrial chemicals and antiquated non- and weakly polar pesticides, such as aldrin, dieldrin, DDT, and lindane (see Table S1 in the Supporting Information). These chemicals typically undergo atmospheric long-range transport and are ubiquitous in the environment (Wegmann et al. 2007). Many of the chemicals of category II can also be found globally in sediments and soils. To evaluate specific regional and national measures, such as additional treatment steps in STPs or improvements in agricultural practice, the POPs and POP-like compounds, such as dieldrin, lindane, or PCBs, are not appropriate because of their ubiquitous and nonregion-specific presence in the environment. Additionally, many of these chemicals have already been phased out in Switzerland and most European countries (e.g., aldrin, dieldrin, DDT, and lindane; European-Parliament 2004). Monitoring these kinds of compounds needs a more comprehensive approach than water-phase monitoring alone, such as the approach of Bogdal et al. (2008). Bogdal et al. have assessed the input pathways of DDT, brominated flame retardants, polychlorinated biphenyls, and polychlorinated naphthalenes with monitoring the water phase, sediments, and the atmosphere.

Exposure category III (moderate persistence, continuous input) Exposure category III contains chemicals with a moderate persistence and that continuously enter surface waters. Generally, pharmaceuticals, pharmaceutical transformation products, and biocides (excluding biocides from material protection) are assigned to category III. Some of these compounds can be found very frequently in relatively constant concentrations in surface waters. Even MCs with relatively short half-lives can establish a steady-state presence because their environmental degradation is continually being balanced by inputs via STPs. These compounds, such as atenolol, diclofenac, ibuprofen, and quaternary ammonium compounds, are pseudo-persistent. The monitoring of these chemicals is, similarly to exposure category I, relatively easy to handle because of their relatively constant concentrations over the whole year.

Exposure category IV (moderate persistence, complex input patterns) Exposure category IV contains chemicals with a

moderate persistence and that have complex input dynamics. The most important substance classes of this group are pesticides and biocides from material protection. The release of pesticides into surface waters is coupled to rain events and to seasonal use, whereas the release of biocides is coupled to rain events only. These chemicals show normally much more complex concentration patterns in surface waters than compounds of exposure category III. In contrast to the highly persistent chemicals of category II, atmospheric long-range transport and remobilization from sediments and soils is less important than direct input pathways. For pesticides, it has been reported that very high concentrations are observed during rain events or shortly after application periods, whereas the concentrations in the base flow are lower (Leu et al. 2004). Chemicals of exposure category IV require a more complex monitoring concept to determine the environmental exposition accurately. However, to evaluate the impact of agricultural management, these chemicals have to be monitored as well as continuously released chemicals. Stamm et al. (2006) propose a monitoring concept that allows for an estimation of the environmental exposure of agricultural pesticides with relatively few samples. To minimize the number of samples, they exploit the fact that pesticide losses are primarily occurring during and after the application period (Leu et al. 2004). Furthermore, they have shown that the pesticide concentrations are strongly correlated to the discharge in smaller creeks after application. Thus, if the discharge is measured, the analysis of a few samples during the period after application and, for comparison, a measurement of the concentration in the river base-flow are sufficient to extrapolate to the total environmental exposition (Stamm et al. 2006). However, because pesticide use is region specific, information about regional use is needed, to establish an effective monitoring concept.

Furthermore, pesticide transformation products are assigned to this category. To a certain extent, their concentrations follow the dynamics of their parent compounds, even if their dynamics is less distinct. For metolachlor ethanesulfonic acid (ESA), it was shown that it was present in the base flow during the whole harvest season, whereas the parent compound metolachlor was only present immediately after the application period. Furthermore, mobile transformation products such as metolachlor ESA enter the surface waters through groundwater recharge, which leads to a certain basic level concentration independent of the season (Huntscha et al. 2008). However, compared to continuously released substances, the concentration dynamics of pesticide transformation products are certainly more pronounced.

Exposure category V (strongly sorbing or volatile) Exposure category V contains chemicals that partition into the water phase at a level less than 10%. These are either quite

volatile or strongly sorbing chemicals, or both. Strongly sorbing chemicals are entering the water compartment through particle-bound atmospheric deposition, overflows from sewer systems, or via preferential flow pathways and surface runoff from agricultural land (Stamm et al. 1998). However, if entering surface water, they are likely to be bound to suspended particles and may be found in river and lake sediments, as is the case for some chlorinated hydrocarbons, PAHs, and higher chlorinated PCBs (Zennegg et al. 2007). Significant sorption to particles strongly influences the chemical’s bioavailability, hydrolysis, and biodegradation. The main degradation path of many volatile chemicals is through reaction with hydroxyl radicals in the atmosphere. Thus, this environmentally relevant group of chemicals needs a completely different assessment and monitoring strategy (e.g., sampling of sediments, particles, or fish) than water-phase chemicals and is not further investigated here. However, this group of chemicals contains many hazardous pollutants and is of high relevance for the environment.

Exposure category VI (rapidly degraded) Exposure category VI contains chemicals that are rapidly degraded either through biological degradation or hydrolysis. Thus, these compounds are generally present in lower concentrations in the environment. However, some of these compounds can be found in the environment, if they are used in very high amounts. An additional challenge in monitoring chemicals that undergo fast hydrolysis is the storage of the water samples. After taking water samples, the samples have to be prepared and analyzed in a very short time, which makes it

very difficult to do a quantitative analysis. Furthermore, the storage of standards for quantitative analysis is problematic.

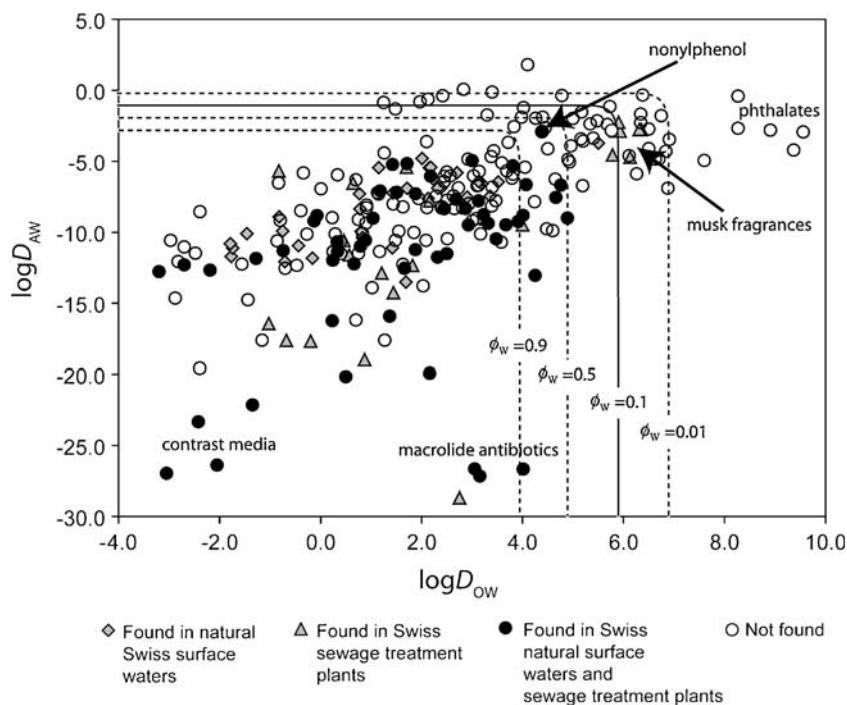
Exposure category VII (not sufficient data available) Exposure category VII contains chemicals that cannot be assessed because of missing data and because the application of QSPRs is not possible. This category acts as a “safety net” and identifies substances with a need for further investigation. From the selected candidate list, it was possible to apply the QSPR EPI Suite™ (U.S.EPA 2007) to estimate the required properties for all considered candidate substances if experimental data were missing. Thus, none of the candidate substances are assigned to exposure category VII.

4 Discussion

4.1 Differentiation between water-phase chemicals and strongly sorbing and/or volatile chemicals

In Fig. 3, the octanol–water partition coefficients (D_{OW}) and the air–water partition coefficients (D_{AW}) of the candidate substances are shown. Additionally, the predicted water-phase fraction contours for $\phi_W=0.01, 0.1, 0.5,$ and 0.9 , calculated with the phase equilibrium model presented above, are shown. D_{OW} s of the candidate substances comprise 14 orders of magnitude and D_{AW} s about 30 orders of magnitude. More than 95% of the candidate substances that were found in Swiss surface waters are within the selected cutoff value $\phi_W=0.1$.

Fig. 3 Chemicals of the candidate substance list that are found in both surface waters and STP effluents, surface waters only, STP effluents only, and in none of them. Sorted according to their $\log D_{OW}$ and $\log D_{AW}$ values. The selected cutoff value for water-phase chemicals is 0.1



Exceptions are polycyclic musk fragrances, which were detected in the water phase of STP effluents, even if they are bound to a high fraction on particles under environmental conditions. In natural surface waters, they are generally present at much lower concentrations than in sewage effluents (Fromme et al. 2001). Polycyclic musk fragrances have not been previously investigated in Swiss surface waters. Polycyclic musk fragrances are rapidly removed from the water phase through sorption to particulate OC and subsequent deposition to sediments and through volatilization (Peck and Hornbuckle 2004). Other compounds on the candidate list that have a predicted distribution into the water phase of less than 10% have not been found or measured in the water phase of Swiss surface waters or sewage effluents. Compounds with a ϕ_w lower than 0.1 are mainly found in sewage sludge, in lake, and in river sediments and bound to dissolved particles (Zennegg et al. 2007). These chemicals, however, are of environmental importance, specifically because of their high potential to bioaccumulate, but have to be treated separately and need different monitoring and evaluation concepts than water-phase chemicals.

Within the selected area for water-phase relevant compounds, there are many compounds that are not detected in surface waters so far. However, these could have several reasons: (a) they are not used in relevant amounts, (b) they are rapidly degraded in surface waters, or (c) they cannot be measured with sufficiently low quantification limits.

4.2 Evaluation of exposure categories I–IV based on some representative chemicals using Swiss consumption data

Beside the property and system-related potential exposure, the actual occurrence of any chemical in the environment is dependent on how they are produced, used, and consumed. To evaluate the categorization methodology, we compare average measured water-phase concentrations and consumption data of chemicals on the candidate list.

To illustrate the potential of a chemical to occur in surface waters, we use a consumption-to-concentration ratio Q , which is the annual consumption (kilogram per year) divided by the average measured concentration (nanogram):

$$Q \left(\frac{\text{kg} \cdot \text{L}}{\text{ng} \cdot \text{year}} \right) = \frac{\text{use}(\text{kg}/\text{year})}{\text{av. conc.}(\text{ng}/\text{L})} \quad (6)$$

Q [kilogram liter per nanogram per year] is a descriptive factor without physical–chemical significance. Generally, we would expect Q to be the lowest for categories I and II, higher for III and IV, and the highest or no measurable concentrations for the exposure categories V and VI.

In most cases, consumption, sales, and production data of the chemicals are not publically available. If regional sales

data are available, a chemicals' nationwide consumption can be estimated and extrapolated using the total number of inhabitants (for pharmaceuticals) or agricultural areas (for pesticides). However, for many chemicals, it is not possible to estimate their use and consumption. Therefore, we are able to evaluate the categorization methodology only with some representative chemicals from different categories, for which reliable consumption data are available or can be estimated. In Table 3, consumption data for Switzerland gathered from various national reports and averaged monitoring data from Swiss surface waters (mainly rivers) are shown.

For chemicals that are widely used and that have no inputs from production processes, we assume a qualitative correlation between annual consumption and averaged environmental concentrations. The consumption to concentration ratios Q for a range of substances is given in Table 3.

For evaluation of the exposure categories, these ratios are compared within the same compound classes: the pharmaceuticals amidotrizoic acid and erythromycin that are assigned to exposure category I show a significantly lower ratio than the other pharmaceuticals of exposure category III. This difference is expected because of the higher metabolization rates of carbamazepine, diclofenac, ibuprofen, and iopromide, which are metabolized to about 80%, whereas amidotricic acid and erythromycin are excreted approximately unmetabolized (Lienert et al. 2007). However, this difference explains only about 10% of the difference in the ratio Q , whereas the remaining difference is probably due to different degradation in the environment. This means that higher concentrations for pharmaceuticals of exposure class I are expected if the same amount is used than for pharmaceuticals of exposure class III. This supports the assignment of amidotrizoic acid and erythromycin to a higher exposure category due to their higher persistence when compared to other moderately persistent pharmaceuticals.

From the considered biocides with available use data, most substances are assigned to exposure category III or IV. Permethrin is in exposure category V because of the high $\log D_{OW}$ value of 6.5. Monitoring data confirm that permethrin is found very rarely in the water phase in surface waters: From 328 measurements across Switzerland, it was found in only one sample (FOEN 2008a, b). Permethrin was measured with solid-phase extraction–gas chromatography–mass spectrometry with a limit of quantification (QL) of 0.05 $\mu\text{g}/\text{L}$. However, the fact that permethrin was found only once in a concentration higher than the QL stays in agreement with the categorization of permethrin as a compound that does not partition or only partitions in small amounts to the water phase within the surface water compartment. All other biocides in Table 3 were found much more frequently and show quite similar consumption/concentration ratios. These findings support the differentia-

Table 3 Comparison between consumption data and averaged surface water monitoring data (mainly river water) for some representative chemicals in Switzerland

	Exposure category	Yearly consumption (kg/year)	Monitoring data ^f (av. conc. ng/L)	Ratio, <i>Q</i> (kg L/ng/year)
Pharmaceuticals				
Amidotrizoic acid	I	487 ^a	149	3.2
Erythromycin	I	110 ^b	20 ^e	5.5
Carbamazepine	III	4,000 ^c	66	60
Diclofenac	III	4,326 ^c	12	360
Ibuprofen	III	23,151 ^c	20	1,100
Iopromide	III	6,933 ^c	13 ^h	530
Biocides				
Carbendazim	III	26,096 ^b	19	1,400
Diuron	IV	23,625 ^b	51	460
Terbutryn	IV	27,127 ^b	19	1,400
Irgarol	IV	24,223 ^b	5	4,800
Permethrin	V	13,607 ^b	Not found ⁱ	–
Pesticides				
Atrazine	IV	37,000 ^d	47	790
Mecoprop	IV	11,000 ^d	25	440
Isoproturon	IV	31,000 ^d	100	310
Industrial chemicals				
Benzotriazole	III	16,000 ^c	300	53

^a Blüm et al. (2005)

^b Bürgi et al. (2009)

^c IMS (2005); Lienert et al. (2007)

^d SGCI (2006)

^e Giger et al. (2006)

^f Averaged monitoring data from Swiss surface waters (FOEN 2008a, b)

^g Data from different rivers in Zürich (AWEL 2008)

^h Measurement data in the lake of Geneva (Edder et al. 2007)

ⁱ Not detected and quantified in 327 of 328 water samples

tion between water-phase chemicals (for most biocides, exposure category III and IV) and strongly sorbing chemicals (exposure category V). The ratios of the evaluated pesticides are similar to those of the biocides. In many cases, the same substances are applied in both urban areas and agriculture and it is often difficult to differentiate the origins of the measured concentrations.

Industrial chemicals, biocides, and pharmaceuticals enter surface water mainly via STPs. The investigated biocides show higher consumption to concentration ratios than the pharmaceuticals of the same category. This is probably due to the fact that the investigated biocides are mobilized mainly during rainfall events. In contrast to pharmaceuticals, biocides are not directly entering the sewer system. The mobilization of biocides from material protection is transport limited (mobilization during rain events), whereas the input of pharmaceuticals is mainly source limited.

4.3 Swiss relevant water-phase chemicals

For policy guidance and monitoring programs in surface waters in Switzerland, we recommend the consideration of four criteria: (a) exposure categories according to the proposed categorization methodology, (b) data on annual consumption if available, (c) already observed occurrence in surface waters, and (d) the availability of a state-of-the-art analytical method. For mandatory monitoring, it is important that the selected compounds can be measured with standard analytical methods, which can be carried out by commercial or governmental labs. In contrast, for investigative monitoring, all available measurements, including special analysis, should be considered. Table 4 summarizes the chemicals that meet these criteria in Switzerland. Chemicals that were found in more than 50% of the investigated samples are highlighted.

Table 4 Potential relevant microcontaminants in the water phase of surface waters for Switzerland

Exposure category I	Exposure category III	Exposure category IV
Amidotrizeic acid 12/48 ^a	4-Acetamidoantipyrindine	2,4-D
Azithromycin 1/35	5-Methylbenzotriazole	2,4-Dimethylphenyl-formamide
Clarithromycin 31/56	Amoxicilline	2,6-Dichlorobenzamide
Erythromycin 6/18	Atenolol 39/57	Alachlor
	BAC-C12	Atrazine 1290/1389
	BAC-C14	Azoxystrobin
	BAC-C16	Bentazon
	BAC-C18	Benzothiazole
	Benzoisothiazoline	Bisphenol A 37/59
	Benzotriazole 325/328	Carbofuran 4/23
	Bezafibrate 9/48	Cyproconazole
	Carbamazepine 136/524	Deoxynivalenole
	Carbendazim 25/42	Desethylatrazine 522/631
	Ciprofloxacin 2/37	Diethyltoluamide
	Clindamycin 9/39	Dimethenamid
	Caffeine 7/8 (tracer)	Dimethoat 14/332
	Diazinon 359/1193	Dinoseb
	Dichloroethylisothiazolinon	Diuron 74/656
	Diclofenac 143/193	<i>N,N</i> -Dimethylaminosulfanilid
	Estradiol 10/86	Ethofumesate
	Estron 31/110	Irgarol 9/860
	Ethinylestradiol 4/93	Isoproturon 130/686
	Fenofibrate	Linuron 15/348
	Fluoxetine HCl	Mecoprop 67/148
	Ibuprofen 82/193	Metalaxyl
	Iohexol	Metamitron
	Iomeprol	Metamitron-desamino
	Iopamidol	Metazachlor
	Iopromide	Metobromuron 8/336
	Ioxitalaminic acid	Metolachlor 98/336
	Mefenamic acid	Metolachlor-ESA
	Metoprolol 14/39	Metolachlor-OXA
	Naproxen 88/193	Napropamide
	Norfloxacin	Nonylphenol 443/480
	Octylisothiazolinon	Octylphenol
	Paracetamol	Oxadixyl
	Propranolol 5/39	Penconazol
	Sotalol 33/56	Primicarb 30/346
	Sulfadiazine 2/20	Propachlor 28/328
	Sulfamethoxazole 34/56	Propiconazol
	Sulfapyridine 14/20	Simazine 265/505
	Triclosan	Sulfamethazine 1/20
	Trimethoprim 19/56	Sulfathiazole
		Tebutam
		Terbutylazine 176/350
		Terbutryn 126/1196

All listed chemicals were observed in Swiss rivers. Chemicals that are above the quantification limit in at least 50% of the investigated samples are marked bold

^a Number of quantified samples/ number of investigated water samples. If no numbers are given, there are only a few observations available that are not recorded in the national database of the Swiss Federal Institute for the Environment (FOEN 2008a, b)

Besides the information included in Table 4, additional criteria should be considered to select model substances for water policy guidance. For the further evaluation of the relevance of these chemicals, ecotoxicological data are needed. The costs of an adequate sampling strategy depend on the input dynamics and the linked concentration dynamics in surface waters. For a monitoring program, the chemicals shown in Table 4 that are in exposure category I and III are easier to handle and, thus, we recommend preferential selection of them for monitoring surface water quality in urban areas if the resources needed to monitor more complex concentration dynamics are not available. However, chemicals of exposure category II and IV are of environmental importance, in particular in smaller creeks in agricultural areas and in weakly diluted urban-originated creeks.

5 Conclusions

The classification methodology proposed here provides a systematic overview of potential MCs. The exposure classification simplifies and supports the selection of compounds for further investigation, monitoring campaigns, and for the evaluation of water protection measures. Generally, compounds of exposure categories I–IV are potentially relevant for surface water quality concerning organic microcontaminants and can be monitored with water-phase sampling. Their occurrence in surface water, however, depends on specific use patterns and may vary by nation or even regionally. Furthermore, the information provided by the presented methodology is only suitable for an exposure-based assessment. To assess the hazard or risk of these compounds, water quality criteria and ecotoxicological data have to be considered.

Information about the compound and feasible analytical methods are necessary to include new potentially hazardous chemicals into mandatory monitoring programs. Ecotoxicological data should be included in a detailed risk assessment of the compounds in categories I–IV. In addition, analytical methods should be developed for these compounds, if not yet existing. Furthermore, the handling of strongly sorbing chemicals should be further investigated and monitoring strategies (monitoring in biota as proposed by WFD) for these kinds of chemicals explored.

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