Integrated Environmental Assessment and Management — Volume 5, Number 1—pp. 50–61

Toward an Integrated Assessment of the Ecological and Chemical Status of European River Basins

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(Received 17 September 2008; Accepted 2 October 2008)

Special Series

EDITOR'S NOTE:

This is 1 of 12 papers prepared by participants attending the workshop "Risk Assessment in European River Basins—State of the Art and Future Challenges" held in Liepzig, Germany on 12–14 November 2007. The meeting was organized within the framework of the European Commission's Coordination Action RISKBASE program. The objective of RISKBASE is to review and synthesize the outcome of European Commission FP4–FP6 projects, and other major initiatives, related to integrated risk assessment–based management of the water/sediment/soil environment at the river basin scale.

ABSTRACT

Here, recommendations to improve ecological and chemical status assessments in accordance with the European Water Framework Directive (WFD) are made on the basis of experience gained from the MODELKEY project database, linking existing biological and chemical monitoring data of 3 case study river basins (Elbe, Scheldt, and Llobregat). The data analysis within and across river basins revealed major obstacles to be tackled, including scarcity of matching ecological and chemical monitoring sites for cause–effect relationships as well as a general lack of stressor-specific metrics for single biological quality elements (BQE) to enable a comprehensive risk assessment of all predominant stressors, including toxicity. An example of such a metric, which is recommended for the BQE of benthic macroinvertebrates, is the trait-based species-at-risk index (SPEAR) that correlated well with a respective measure for toxic stress, referred to as toxic units, based on simple mixture toxicity concepts. Surprisingly, the assessment of chemical status of a total of 695 monitoring sites for 2000 to 2004 showed that environmental quality standards (EQSs) were exceeded for at least 1 of the currently 41 priority pollutants (PPs) in 92% to 98% of the cases in all 3 of the river basins, which, according to definition, indicates potential effects on ecological status. A comparison of compliance with EQSs for 41 PPs with a respective effect threshold (derived for benthic macroinvertebrates) revealed that the rather conservative concept of chemical status is most likely not protective in all cases. Furthermore, to account for the many other compounds that are detected frequently in European surface waters and that may also have ecotoxicological effects, we introduced a provisional predicted no-effect concentration that is in accordance with the EQS methodology and is suggested to identify potential emerging compounds for which no or insufficient toxicity data exist. In conclusion, this study aims to support the implementation of the WFD by drawing conclusions from the analysis of heterogeneous data sets of various member states and by introducing new tools to move toward an integrated European assessment of ecological and chemical status.

Keywords: Water Framework Directive Environmental quality standards Predicted no-effect concentration Toxic units Species-at-risk index (SPEAR)

INTRODUCTION

Freshwater is one of the most valuable resources on earth, and its protection and conservation for future generations pose a major challenge to our society (Millennium Ecosystem Assessment 2005). Standing to this challenge, the European Commission enacted the Water Framework Directive (WFD) to manage European river basins in a sustainable way (CEC 2000). For the first time, the WFD mandates all member states (MS) to protect the aquatic ecosystems as a whole and across borders. The main objectives of the WFD are to achieve "good ecological and chemical status" for surface water bodies by 2015 but also has a general "no deterioration" provision to prevent deterioration of the current status.

For 2004, the WFD required all member states to assess the actual status of their water bodies. These assessments were summarized in Article 5 Status Reports. The analysis of these reports revealed that 40% of all European surface water bodies will probably not meet the WFD quality targets by 2015 (classified as "at risk" according to WFD terminology), another 30% have "insufficient data" to assess the actual status, while only 30% are currently classified as not at risk of degradation (CEC 2007). In Germany, even 62% of the national water bodies were classified as being at risk of failing to achieve good ecological status, while only 12% will

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Published on the Web 10/9/2008.

probably achieve good ecological status. At the same time, 9% of the German water bodies are likely to fail to achieve the good chemical status, and another 28% have insufficient data to assess chemical status (BMU 2006). For the Elbe River basin, as investigated in the present analysis, the report states that 6% of its water bodies are likely failing to achieve the good chemical status, and another 24% of its water bodies have insufficient data to assess chemical status, respectively (Borchardt et al. 2005). This implies that 70% of the Elbe water bodies were classified as of good chemical status based on the available information at that time. The European Commission, however, also noted that the information on the relative importance of diffuse and point sources of pollution as reported by the MS were often insufficient to perform an adequate pressures and impacts analysis on the European level (CEC 2007).

The preliminary conclusions and interpretations from the Article 5 environmental analysis suggested that agriculture (including nutrient loading) is the largest factor responsible for the deterioration of water bodies, followed by hydromorphological degradation (CEC 2007). It is, thus, believed that on the European scale, improvement of hydromorphological conditions is essential to achieve the good ecological status. However, stream conservation and restoration measures taken to improve the hydromorphology in the past often failed to rehabilitate the expected stream biodiversity (Brooks et al. 2002; Lepori et al. 2005; Suren and McMurtrie 2005; Jähnig et al. 2008). On the other hand, elevated levels of numerous chemicals (such as pesticides) are detected frequently in European surface waters (e.g., Pietsch et al. 1995), indicating that chemical stress may be one of the driving forces for an insufficient ecological status (Brack et al. 2005). Several studies have shown clear effects of chemical stress on stream-dwelling communities, especially for pesticides that are deliberately released into the environment (Schulz and Liess 1999; Leonard et al. 2000; Friberg et al. 2003; Liess and von der Ohe 2005; Schäfer et al. 2007).

In general, aquatic ecosystems are subject to various pressures (Furse et al. 2006) whose effects may also be altered by climate change, and achieving good ecological status requires European water managers to undertake an integrated assessment of all available information (von der Ohe et al. 2007b). For example, habitat characteristics, which would be useful to explain natural variability of the aquatic community structure, are difficult to quantify and often lacking in electronic form. However, the joint consideration of all information is crucial to eliminate confounding factors, such as the co-occurrence of other natural stressors (e.g., floods or droughts), which may mask the adverse effects of anthropogenic stressors (Schäfer et al. 2007). Hence, the ecological and chemical monitoring programs should be combined to allow the identification of all stressors that may be responsible for the deviation from good ecological status. As river basins often cover several member states, the management of these catchments requires also an integrative assessment approach to allow for a similar protection of all water resources. Furthermore, to reveal the relevance of single anthropogenic or natural stressors, the use of stressor-specific indicators are recommended (von der Ohe et al. 2007b), which, however, are still limited (Hering et al. 2004). Only an integrated assessment would allow selecting those management options that will be most effective to improve water quality (von der Ohe et al. 2007b; Brack et al. 2008).

With this paper, we aim to assist water managers by providing generic tools toward an integrated risk assessment of predominant stressors. However, in order to apply these tools, the available monitoring data of different river basins need to be harmonized first in order to allow an integrative risk assessment applicable all over Europe. For that purpose, monitoring data of 3 European river basins were compiled by the European Integrated Project MODELKEY: MODELs for assessing and forecasting the impact of environmental KEY pollutants on freshwater and marine ecosystems and biodiversity (Brack et al. 2005). The aim of the present study is to support the implementation of the WFD by drawing conclusions from the analysis of the MODELKEY monitoring data set and by introducing new tools to allow for a more integrated and integrative assessment within and among all basins.

River basins, database, and data handling

The MODELKEY database (von der Ohe et al. 2007b) used compiled existing monitoring data (which was sampled before the WFD requirement) of 3 case study river basins that are part of the Article 5 Status Reports. The data were provided by regional water authorities that are responsible for the implementation of the WFD and who act as end users of the MODELKEY project: The Landesbetrieb für Hochwasserschutz und Wasserwirtschaft Sachsen-Anhalt (LHW, Magdeburg) and the Sächsisches Landesamt für Umwelt und Geologie (LfUG, Dresden) provided data on two federal states of Germany in the Elbe River basin, the Agencia Catalana de l'Aigua (ACA, Barcelona) in Spain on the whole Llobregat River basin and the Vlaamse Milieumaatschappij (VMM, Erembodegem) in Belgium on the Flemish part of the Scheldt River basin, respectively.

The Elbe River, with a total length of 1091 km and a catchment area of about 140000 km^2, is a large central European river that flows through the Czech Republic and Germany into the North Sea. Heavily industrialized areas were spread along the main river and its tributaries; however, recently, many industrial plants have been closed down, so aquatic ecosystems are expected to recover. In the German part, intensive agriculture is the main land use in the catchment. The sampling sites were distributed over the Elbe River as well as the catchments of the Saale, Mulde, and Schwarze Elster tributaries and were restricted to the current sampling sites of the surveillance monitoring program.

The Llobregat River is 170 km long and is the second-longest river in Catalonia (Spain), which discharges into the Mediterranean Sea and covers a catchment area of about 4948 km^2. It has been highly polluted by industrial and urban wastewaters as well as by surface runoff from agricultural areas, while salt-mining activities has caused an increase in the salinity of the water. The river experiences periodic floods and droughts that lead to frequent morphological variations in the riverbed and to modifications in its banks that may influence the ecological status of the water bodies. The sampling sites represent a longitudinal profile of the main river as well as its two major tributaries, the Cardener and the Anoia.

The Scheldt River, with a total length of 350 km and a catchment area of about 21000 km^2, flows through France, Belgium, and the Netherlands into the North Sea. The Scheldt is an important waterway, and the port of Antwerp, the second largest in Europe, lies on its banks. It is known as one of the most polluted river systems within western

Table 1. Overview of monitoring data in the MODELKEY database, consisting of the number of subbasins, the number of ecological and chemical monitoring sites, the number of chemicals analyzed and detected above the limit of quantification (>LOQ), including the list of 41 priority pollutants (PPs), as well as the availability of respective effect data (LC50) and the number of sites "achieving good chemical status" separately for each river basin and as total[a]

Subcategory	Scheldt	Elbe	Llobregat	All basins
Subbasins	4	4	3	11
Ecological monitoring sites	807	46	34	887
Chemical monitoring sites	482	169	44	695
Monitoring sites with organics analyzed	90	106	44	240
Monitoring sites "achieving good chemical status"	7	14	1	31
Matching chemical and ecological monitoring sites (with organics analyzed)	392 (68)	39 (20)	29 (29)	460 (117)
Original chemical names	361	332	207	871
Chemical compounds (with different CAS number)	352	233	194	467
Chemical compounds > LOQ	246	188	82	346
LC50 for *Daphnia magna*	197	121	94	233
LC50 for *Selenastrum capricornutum*	91	67	44	106
LC50 for *Pimephales promelas*	76	49	51	101
PPs analyzed	37	37	35	40
PP > LOQ	36	33	28	39
PP > EQS	27	19	26	37

[a] CAS number = Chemical Abstract Service number; PP analyzed = number of PPs that have been analyzed by water authorities; PP > LOQ = number of PPs above the limit of quantification; PP > EQS = number of PPs above the respective environmental quality standard.

Europe, but quality is improving by the installation of wastewater treatment plants in Belgium during the past and current decade. However, many of the tributaries and smaller catchments have a ditchlike character due to the generally low elevation, which may affect the aquatic communities. The sampling sites are evenly distributed over 4 subbasins in the Flemish part of the Scheldt, namely, the Boven Schelde, Beneden Schelde, Nete, and Dijle Zenne.

A total of 62628 entries on species abundances were included in the biological data table, of which the majority (32151 entries) was related to the Biological Quality Element (BQE) of benthic macroinvertebrates, recorded at 887 monitoring sites (Table 1). The German data were sampled according to the DIN 38410–1 normative with semiquantitative results at the species level (DIN 1992). In the Llobregat River, samples were collected from the riffle and pool habitats using an adaptation of the kick-net method. Results obtained were qualitative, on the taxonomic level of the family (Alba-Tercedor and Sanchez-Ortega 1988). In the Scheldt River, quantitative results were obtained at the genus level, expressed as number of individuals per dredge using a Van Veen dredge engine (De Pauw et al. 1992). Hence, the biological sampling procedures were comparable only within but not across river basins.

The physicochemical data table contained even 2455642 data entries, but only 869228 data entries were related to chemical concentrations in the water phase for the most recent years 2000 to 2004, referring to a total of 695 monitoring sites. In order to obtain the chemical concentrations in surface water, water samples were taken by scoop. In the Elbe and Scheldt rivers, these whole-water samples were taken to account for compounds with high K_{OC} that bind to the suspended matter fraction. In the Llobregat River, water samples were filtered before the analysis to obtain dissolved concentrations. Surprisingly, only 460 of these monitoring sites had matching biological and chemical data (i.e., for metals only), and the number was further reduced to only 127 sites with additional data on organic toxicants, which allowed for an integrated assessment of the impacts of chemical stress at those sites (Table 1).

One first problem to be tackled before including the chemical monitoring data into the MODELKEY database concerned the heterogeneous nomenclature of the pollutants for which data were provided (e.g., by 2 federal authorities) that would have prevented an integrative assessment within and among all MS of a river basin. Hence, a European standard for the reporting of chemical data (e.g., including the CAS number) would improve data assessments dramatically. A step in this direction was taken with the Water Information System for Europe (WISE) initiative that aims to compile the monitoring data reported by MS. However, as the MS are required to submit only those data on priority pollutants (PPs) (to assess chemical status) and of a short list of river basin–specific compounds (defined by each MS), it is questionable if these data are sufficient to assess the actual ecotoxicological stress stemming from all toxicants present.

Generally, the same difficulties apply for the existing monitoring data on BQE that were also provided from

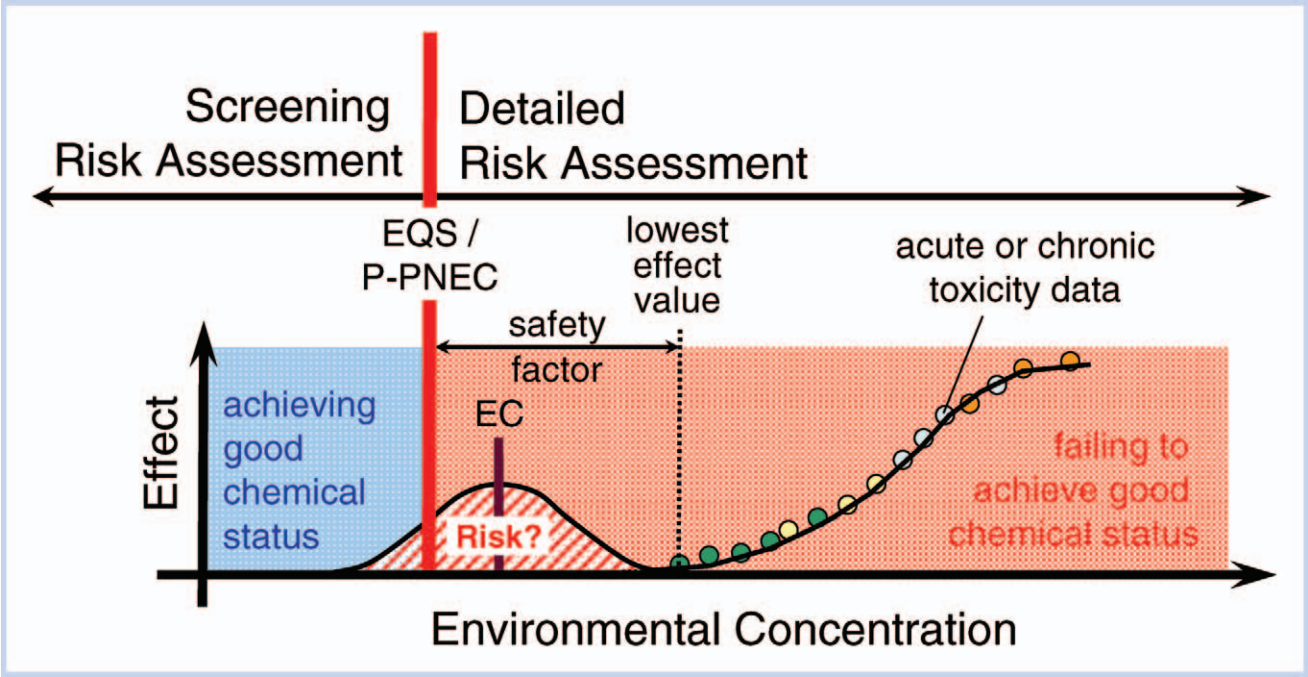

Figure 1. Screening versus detailed risk assessment with regard to the Water Framework Directive "chemical status." The chart shows the estimation procedure for "environmental quality standards" (EQS) and "provisional predicted no-effect concentrations" (P-PNECs), representing safe levels of chemicals in the environment. A clear exceedance of these threshold levels anticipates potential effects on "ecological status." EC stands for an environmental concentration of a pollutant above its EQS value but well below the lowest effect value. The bell-shaped curve indicates the uncertainty of the measured concentration and the potential risk for the aquatic community that might arise from this compound.

different MS or federal authorities. The application of European-wide assessment tools requires the identification of the same taxa in different MS. Hence, to achieve consistency, the macroinvertebrate data of all basins were classified and adjusted taxonomically according to the operational taxa list of the AQEM assessment system (AQEM Consortium 2002) that was developed to support the WFD, before inclusion in the MODELKEY database.

Assessment of chemical status: Screening versus detailed risk assessment

The assessment of chemical status is based on currently 33 priority substances and 8 priority hazardous substances, both referred to as PPs, as listed in Annex X of the WFD. These individual pollutants or groups of pollutants were proposed by the Commission, as they are expected to pose significant risks to or via the aquatic environment. According to Article 16 and Annex V of the WFD, "achieving good chemical status" was defined as compliance of the 41 PPs with certain concentration levels, referred to as environmental quality standards (EQSs). In case of exceedance of these compound-specific thresholds (CEC 2000), the classification of a water body will change to "failing to achieve good chemical status." The aim behind these EQS values was to derive thresholds below which no effects on the aquatic communities were expected. This rather conservative approach could be described as a kind of screening risk assessment, aiming at safe levels of chemicals in the environment (Figure 1). In contrast, a clear exceedance of these threshold levels anticipates potential effects on the aquatic communities to be protected. Hence, in case a water body is classified as "failing to achieve good chemical status," a detailed risk assessment is recommended to quantify the effects that would be expected on the ecological status (Figure 1).

Assessment of chemical status for 3 case study river basins—In order to assess the chemical status according to the WFD requirements, the monitoring data of 3 case study river basins of the MODELKEY project were analyzed with regard to the 41 PPs only. The analysis was restricted to the water phase, as there were only few scattered data on concentrations in sediment and biota available. This may have underestimated the real risk to a water body, especially for the more hydrophobic compounds that bind at the solid-phase fraction.

The analysis showed that all PPs, with the exception of PP 7 (C10–13 chloralkanes), had actually been monitored between 2000 and 2004, although not in all river basins (Table 1). Interestingly, 37 PPs were also detected above their EQS values in at least one of the river basins, justifying their selection as PPs on a European scale.

Surprisingly, a comparison of the data on PP from all monitoring sites with respective EQS values revealed that the majority of the investigated sites (92%–98%) would have to be classified as "failing to achieve good chemical status" with respect to one or more PPs. Only 31 of the total 695 chemical monitoring sites reported concentrations below the EQSs for all PPs and in all years (Table 1). For the Elbe River and its tributaries, 92% of the 169 chemical monitoring sites analyzed here were classified as "failing to achieve good chemical status," which conflicts with the former assessment of the Article 5 Status Report, stating that about 70% of all Elbe water bodies were likely to achieve good chemical status (Borchardt et al. 2005). Although the numbers of sampling sites considered for both assessments do not agree, these ratios could be taken as indication. A possible explanation for the deviating assessments is the rather low final EQS values for the 41 PPs, which had not been set in 2004. Hence, compliance with respective thresholds could not have been checked then.

In order to analyze which compounds are responsible for so many sites classified as "failing to achieve good chemical status," we counted the number of sites at which a certain PP was exceeding the EQS values. Interestingly, only a few compounds were responsible for the frequent exceedance in all 3 river basins. Cadmium (PP 6) was most often responsible for exceeding EQSs in the Elbe River, with exceedances at 154 sites. In the Llobregat River, benzo[*g,h,i*]perylene and indeno(1,2,3-*c,d*)pyrene (both PP 28) exceeded the EQSs at 43 sites, while in the Scheldt River, cadmium (PP 6) and lead (PP 20) exceeded EQSs at 460 and 401 sites, respectively. These 4 compounds alone account for more than 90% of the water bodies that did not comply with good chemical status.

Another obstacle for the assessment of chemical status concerned data on chemical concentrations that were reported as below the limit of quantification (LOQ). In the database, these values were reported with the addition "<" before the concentration value. As can be seen from Table 2, in some cases, the LOQ was above the rather low EQS values. Thus, values that were reported as below the LOQ and that accounted for half of this value in the assessment will automatically lead to an exceedance of respective EQS values. Hence, a second analysis was performed, taking only those PPs into account that were detected above the LOQ in the respective river basin (Clarke 1998). The result merely improved for the Llobregat River, where the number of sites classified as "achieving good chemical status" increased from only 1 to 13 sites because of PP 28 (polyaromatic hydrocarbons), while the result did not change for the other 2 basins.

Moreover, the analytical methods used to determine chemical concentrations in water and hence affecting the assessment of chemical status differed widely among MS. The risk assessment of chemicals measured in whole water samples, as, for example, in the Scheldt and Elbe river basins, was hampered by the fact that the more hydrophobic compounds were likely to be adsorbed to the suspended particle fraction, which most likely reduced the bioavailability (Carriquiriborde et al. 2007). However, this method could be regarded as worst-case scenario for a screening risk assessment like the chemical status assessment. In contrast, chemical measurements in filtered water samples, as, for example, in the Llobregat River basin, seemed to be more appropriate for a detailed risk assessment in order to support the assessment of ecological status. In general, the assessment of environmental pollutant concentrations in environmental samples that were taken and processed differently hamper an integrative assessment on the river basin scale despite precise analytical measurements. Hence, it is recommended to use consistent analytical methods to allow for river basin–specific cause–effect relationships in order to link ecological and chemical status.

EQSs for use in screening risk assessments—The compound-specific threshold values (EQSs) mentioned previously were commonly derived from laboratory toxicity data with the aim to ensure no harm for the environment due to exposure to that chemical. The EQS Guideline required at least 3 acute values from 3 trophic levels, considering fish, algae, and macroinvertebrates (Fraunhofer Institute 2005). If available, chronic data for each trophic level were preferred over acute data. After selecting the lowest effect data, a safety factor was applied to account for species differences and data uncertainty (Figure 1), varying from a factor of 10 to 1000, depending on data availability. At this stage, official EQS values were in place only for the 41 PPs. However, MS are required to designate river basin specific compounds for which respective EQS values have to be derived. An example for such a list of compounds is available for the Danube River basin (International Commission for the Protection of the Danube River 2003). In general, this procedure will allow for a better screening risk assessment in the future and may generate synergies for compounds that are identified in different river basins across Europe. This requires that the respective toxicity data for these compounds are available in order to allow for identification of potential effects. However, this is often not the case.

For example, a total of 467 compounds had been measured in the 3 case study river basins, of which 346 had been clearly detected above the LOQ such that all may be of potential concern (Table 1). Yet toxicity data with regard to the BQE of benthic macroinvertebrates (i.e., *Daphnia magna*) were available for only approximately 50% of the compounds, 22% for fish (i.e., *Pimephales promelas*), and 23% for algae (i.e., *Selenastrum capricornutum*), respectively. The use of existing toxicity data for approximately 25% to 50% of the compounds was hampered by the EQS requirement to cover all 3 trophic levels. Therefore, the number of compounds with sufficient toxicity data sets was reduced to 43, or 9% of all compounds. This calls for a practical solution to allow for a screening risk assessment of those compounds that were frequently detected above the LOQ but that did not have sufficient toxicity data. Hence, a pragmatic approach is suggested in the following paragraph to cope with the current limitations of data scarcity.

Provisional predicted no-effect concentrations—For those compounds where a first evaluation of the data availability revealed the absence of toxicity data for 1, 2, or all 3 trophic levels, a methodology to derive a provisional predicted no-effect concentration is suggested, referred to as provisional predicted no-effect concentration (P-PNEC). This effect threshold will allow for the provisional assessment of concentration levels detected in the environment and to identify potential emerging compounds across Europe.

In this context, the European REACH Directive (Registration, Evaluation, and Authorization of Chemicals) is expected to initiate the risk assessment for a huge inventory of compounds (CEC 2003). This directive will eventually provide the necessary toxicity data to derive EQS for many chemicals. However, this directive also faces the challenge of assessing hundreds of thousands of compounds. Hence, the use of reliable computer models (e.g., quantitative structure–activity relationships [QSARs]) were encouraged to support the REACH process by closing data gaps or providing additional evidence through prediction of compound properties based on information of known compounds (e.g., through read-across methods using large data sets) (Netzeva et al. 2008). In the following, a stepwise procedure is described to derive respective P-PNEC thresholds (Table 3).

As a first step, missing acute toxicity data are predicted for chemicals that are expected to have a narcotic-like mode of action (MOA) from respective baseline QSARs that represent the minimum toxicity (e.g., von der Ohe et al. 2005), expected from the lipophilicity of the compound. For these compounds, it is assumed that acute toxicity could be estimated with reasonable accuracy from existing QSARs for all 3 trophic levels (e.g., Galassi and Vighi 1981; Veith et

al. 1983; Hermans et al. 1984). Furthermore, it is also possible to estimate missing toxicity data from existing experimental values for similar compounds, using read-across methods (Schäfer et al. 2008). Precondition for the application of any QSAR is however, that the assessed compound is in the chemical domain as well as the application domain of the respective model (von der Ohe et al. 2007a). Only then is the QSAR is expected to allow reliable predictions. In cases where a structural alert is present (indicating excess toxicity of the compound by employing simple structural features as molecular descriptors), it is recommended to test the respective compound for the trophic level with the highest expected effects (von der Ohe et al. 2005).

In the next step, the test data with the highest toxicity is chosen, either predicted or available from literature. Subsequently, a respective safety factor with regard to the EQS methodology will be applied (Fraunhofer Institute 2005), using safety factors of 10 (where 3 chronic values are available from literature) up to 1000 (with at least 3 acute data available). In case 1 or 2 data have to be predicted but the lowest acute value is a literature value or predicted from read-across, we also suggest using a factor of 1000. In cases where all 3 acute data are predicted from baseline QSARs, a safety factor of 10000 is suggested to account for both differences in species sensitivities and polar narcosis, as covered by the narcotic-like mode of action classification (von der Ohe et al. 2005). Therefore, the P-PNEC values should allow for similar protection as compared to EQS values that are based on only 3 acute literature values. However, specific chronic or indirect effects of some pollutants could not fully be excluded by using only acute data on few standard test organisms, which holds for the EQS approach as well. Therefore, the P-PNEC values should be used for screening of emerging compounds as well as to identify potentially affected water bodies and be replaced by proper EQS values in cases of clear exceedance.

Assessment of ecological status: Relationships of probable causes versus observed effects

The ecological status of a water body is preliminarily assessed by the most affected BQE, based on computed metric values that represent the actual status of the present communities (CEC 2000). The classification of at least "good ecological status" depends on BQE-specific class boundaries between good and moderate status (Figure 2). These class boundaries were established in an intercalibration process to guarantee similar status assessments among MS that apply different local metrics (CIS WG 2004), which holds, for example, for the 3 case study river basins investigated here.

In order to assess the potential effects of the prevailing pressures on all trophic levels, stressor-specific metrics for all BQE would be required. Moreover, it is possible to draw the correct conclusions on responsible stressor from these metrics only if reliable cause–effect relationships between the metric and a respective environmental stress measure exist. Moreover, these relationships should be derived only for similar ecoregions or water body types. As sampling procedures differ widely among MS, these relationships will often be restricted to river basins or MS and require an intercalibration of type-specific class boundaries of these metrics.

Figure 2 shows a model relationship between a stressor-specific metric and a BQE-specific stress measure. In this example, the effect on the metric increases linearly with increasing stressor intensity. Hence, sites with high status correlate with low stressor intensities, while sites with bad status refer to high stressor intensities. Such relationships would allow confirming the observed community effects (expressed as metric values) with prevailing environmental conditions. Hence, in cases where 1 or more EQS or P-PNEC values are exceeded, a detailed risk assessment of the chemical pollution is recommended to support the ecological status assessment with regard to the chemical status of a water body (Figure 1).

BQE-specific toxic units—For the purpose of quantifying toxic stress related to organic pollutants, BQE-specific toxic units (TUs) are recommended, based on the TU approach by Peterson (1994). A restriction to organic compounds was made because of different community effects that would be expected from organic and metal compounds, at least for invertebrate species (von der Ohe and Liess 2004). To derive respective TUs, the measured compound concentrations were scaled to the inherent toxicity of each compound towards a standard test organism (e.g., *D. magna*) and optionally added to an overall toxicity measure:

$$\mathrm{TU} = \log \sum_{i=1}^{n} \frac{C_i}{\mathrm{LC50}_i} \quad (1)$$

where i is the compound, C_i is the measured environmental concentration of compound i, $\mathrm{LC50}_i$ is the respective acute lethal concentration in a standard toxicity test (48 h for *Daphnia*, 48–96 h for algae, and 96 h for fish), and n is the number of compounds considered. The endpoint of the acute LC50 was chosen because it was representative for observed acute effects (Liess and von der Ohe 2005) and comparable effect data were available for many compounds (von der Ohe and Liess 2004). Concentrations below the LOQ (reported values labeled with "<") were considered with half the LOQ in the computation if concentrations in other samples were detected at least once above the LOQ in the respective basin. Compounds that were never detected above the LOQ were excluded from the computation of TUs in order to avoid overestimation of risks by including compounds that were likely to be absent (Clarke 1998).

For the purpose of stressor identification, it is recommended to use BQE-specific TUs, as the effects of specific toxicants, such as insecticides or herbicides, often act specifically on certain trophic levels. Hence, as the classification of ecological status is based on the most affected BQE (CEC 2000), BQE-specific TUs may be more indicative of effects responsible for the deviation from the good ecological status than compliance with EQSs that were derived from the most sensitive trophic level. For this purpose, we used the reference species with the most effect data available to derive BQE-specific TUs (i.e., *D. magna* for macroinvertebrates) (von der Ohe and Liess 2004), *S. capricornutum* for algae, and *P. promelas* for fish.

To identify the contribution of individual compounds to the overall toxicity, the sum of TUs ($\mathrm{TU}_{\mathrm{sum}}$) of all compounds as well as the maximum TU ($\mathrm{TU}_{\mathrm{max}}$) for every single compound were calculated. The former related to the model of concentration addition that assumed an additive behavior of all components. This model was successfully applied earlier, for example, to confirm toxicant effects of polycyclic aromatic hydrocarbons (Boxall and Maltby 1997). In contrast, $\mathrm{TU}_{\mathrm{max}}$ accounted for the minimum expected effect based on the most potent toxicant in a mixture. For compounds where no toxicity data were available, the acute data were estimated

Table 2. Overview of the 41 priority pollutants (PPs), CAS numbers, annual average environmental quality standards (AA-EQS), and maximum annual concentration EQS (MAC-EQS) values as well as the annual average and annual maximum concentrations at the most impacted monitoring site of the Scheldt, Elbe and Llobregat river basins within the years 2000 to 2004. All concentrations are given in μg/L. "<" indicates that concentrations are less than the limit of quantification. n.a. = not applicable

Nr	PP name	CAS nr	AA-EQS	MAC-EQS	Scheldt		Elbe		Llobregat	
					Average	Max	Average	Max	Average	Max
1	Alachlor	15972-60-8	0.3	0.7	3.5	10	0.11	0.33	0.10	0.20
2	Anthracene	120-12-7	0.1	0.4	0.61	0.97	0.65	4.5	0.10	0.20
3	Atrazine	1912-24-9	0.6	2.0	3.2	15	0.24	0.60	0.10	0.20
4	Benzene	71-43-2	10	50	0.63	5.9	<1.0	<1.0	1.1	1.5
5	Pentabromodiph enylether	32534-81-9	0.0005	n.a.	n.a.	n.a.	<0.003	<0.003	n.a.	n.a.
6	Cadmium and its compounds	7440-43-9	0.08–0.25	0.45–1.5	30	324	240	310	16[a]	16[a]
7	C10–13 chloroalkanes	85535-84-8	0.4	1.4	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
8	Chlorfenvinphos	470-90-6	0.1	0.3	1.8[a]	1.8[a]	n.a.	n.a.	0.04[a]	0.04[a]
9	Chlorpyrifos	2921-88-2	0.03	0.1	0.13	0.16	0.008	0.07	0.20	0.28
10	1,2-Dichloroethane	107-06-2	10	n.a.	4.8	15	1.7	10	16[a]	16[a]
11	Dichloromethane	75-09-2	20	n.a.	31	88	4.3	15	63	120
12	Di(2-ethylhexyl) phthalate	117-81-7	1.3	n.a.	12[a]	12[a]	n.a.	n.a.	n.a.	n.a.
13	Diuron	330-54-1	0.2	1.8	6.2	18	0.23	0.84	n.a.	n.a.
14	Endosulfan	115-29-7	0.005	0.01	0.08	0.15	n.a.	n.a.	0.14	0.27
15	Fluoranthene	206-44-0	0.1	1	2.2	4.1	12	82	0.10	0.20
16	Hexachlorobenzene	118-74-1	0.01	0.05	0.04[a]	0.04[a]	0.02	0.15	0.03	0.05
17	Hexachloro butadiene	87-68-3	0.1	0.6	0.02[a]	0.02[a]	0.02	0.10	0.29	0.5
18	Hexachloro cyclohexane	608-73-1	0.02	0.04	0.07	0.39	0.03	0.08	0.03	0.18
19	Isoproturon	34123-59-6	0.3	1.0	1.6	8.1	0.16	0.86	n.a.	n.a.
20	Lead and its compounds	7439-92-1	7.2	n.a.	3650	24300	331	2175	270[a]	270[a]
21	Mercury and its compounds	7439-97-6	0.05	0.07	1.9	15	7.6	49	<500	<500
22	Naphthalene	91-20-3	2.4	n.a.	4.1	19	1.5	8.3	5.0[a]	5.0[a]
23	Nickel and its compounds	7440-02-0	20	n.a.	436	3010	33	85	529[a]	529[a]
24	Nonylphenol	25154-52-3	0.3	2.0	<0.03	<0.03	1.2	3	4.0	18
25	Octylphenol	1806-26-4	0.1	n.a.	1.4	1.8	0.16	0.29	0.26	0.56
26	Pentachlorobenzene	608-93-5	0.007	n.a.	0.04[a]	0.04[a]	5.0[a]	5.0[a]	0.05[a]	0.05[a]
27	Pentachlorophenol	87-86-5	0.4	1	0.09	0.10	25	25	0.26[a]	0.26[a]
28	Benzo[*a*]pyrene	50-32-8	0.05	0.1	0.18	1.2	0.48	2.5	<0.020	<0.020
	Benzo[*b*]fluoranthene	205-99-2	$\Sigma = 0.03$	n.a.	0.23	1.7	0.29	1.2	<0.020	<0.020
	Benzo[*k*]fluoranthene	207-08-9		n.a.	0.27	0.76	0.15	0.60	<0.020	<0.020

Table 2. Continued

Nr.	PP name	CAS nr	AA-EQS	MAC-EQS	Scheldt		Elbe		Llobregat	
	Benzo[*ghi*]perylene	191-24-2	$\Sigma = 0.002$	n.a.	0.17	1.4	0.30	1.5	<0.020	<0.020
	Indeno[1,2,3-*cd*]pyrene	193-39-5		n.a.	0.19	1.4	0.34	1.5	<0.020	<0.020
29	Simazine	122-34-9	1	4	1.2	3.6	0.06	0.41	0.10	0.20
30	Tributyltin compounds	688-73-3	0.0002	0.0015	n.a.	n.a.	0.21	0.35	n.a.	n.a.
31	Trichlorobenzenes	12002-48-1	0.4	n.a.	3.6	6.5	1.2	1.2	1.8	2.6
32	Trichloromethane	67-66-3	2.5	n.a.	2.4	6.9	1.4	6.1	0.84	1.6
33	Trifluralin	1582-09-8	0.03	n.a.	0.64[a]	0.64[a]	0.003	0.005	0.10	0.20
34	Para-para-DDT	50-29-3	0.01	n.a.	0.001	0.002	0.68	1.9	0.125	0.250
35	Aldrin	309-00-2	0.01	n.a.	0.05[a]	0.05[a]	0.0016	0.0023	0.025	0.050
36	Dieldrin	60-57-1	0.01	n.a.	0.02[a]	0.02[a]	<0.005	<0.005	0.025	0.05
37	Endrin	72-20-8	0.01	n.a.	0.01[a]	0.01[a]	<0.002	<0.002	0.125	0.25
38	Isodrin	465-73-6	0.01	n.a.	0.008[a]	0.008[a]	0.005[a]	0.005[a]	0.025	0.05
39	Carbontetrachloride	56-23-5	12	n.a.	1.3	2.3	0.01	0.07	<0.05	<0.05
40	Tetrachloroethylene	127-18-4	10	n.a.	4.7	18	0.92	7.7	5.1	9.9
41	Trichloroethylene	79-01-6	10	n.a.	n.a.	n.a.	34	150	20[a]	20[a]

[a] Only 1 annual measurement available.

from respective baseline QSARs. In another study, this methodology was applied for the first time for all 3 trophic levels, using read-across methods and baseline QSARs to predict missing acute LC50s (Schäfer et al. 2008).

Stressor-specific cause–effect relationships—In order to establish reliable cause–effect relationships between community metric values and stressor intensities, stream-dwelling macroinvertebrates are probably best suited, as they are still the most commonly used organism group and quality metrics for several stressors already exist (e.g., for organic pollution [DIN 1992] or acidification [Braukmann and Biss 2004]). However, although identified as one of the major pressures on the European scale, the indication of morphological degradation gradients is still difficult (Völker and Borchardt 2007). With respect to chemical stress, the trait-based species-at-risk index (SPEAR) was introduced to detect adverse effects of pesticides on stream-dwelling macroinvertebrates of small agricultural streams (Liess and von der Ohe 2005), representing the ratio of physiologically sensitive species in the macroinvertebrate community. For this analysis, an adaptation of the SPEAR index was used, which is recommended to assess the effects stemming from a chronic exposure to organic toxicants, as

Table 3. Stepwise procedure to derive provisional predicted no-effect concentrations (P-PNECs). n.a. = not applicable

Step	Procedure	Yes	No
1	Are acute test data for all three trophic levels available?	Go to step 2	Go to step 3
2	Are additional chronic test data for any trophic levels available?	Derive EQS	Derive EQS
3	Does the compound comply with narcotic-like mode of action, or are similar compounds available from read-across?	Go to step 4	Testing required
4	Predict missing acute data from baseline quantitative structure–activity relationship or from read-across for missing trophic levels	Go to step 5	n.a.
5	Is there a chronic effect value for the most affected trophic level available from literature?	Go to step 7	Go to step 6
6	Is there a lowest acute effect value available from literature or from read-across?	Go to step 8	Go to step 9
7	Apply a safety factor according to environmental quality standards methodology	n.a.	n.a.
8	Apply a safety factor of 1000 to lowest effect data to derive P-PNEC	n.a.	n.a.
9	Apply a safety factor of 10000 to the lowest predicted acute data to derive P-PNEC	n.a.	n.a.

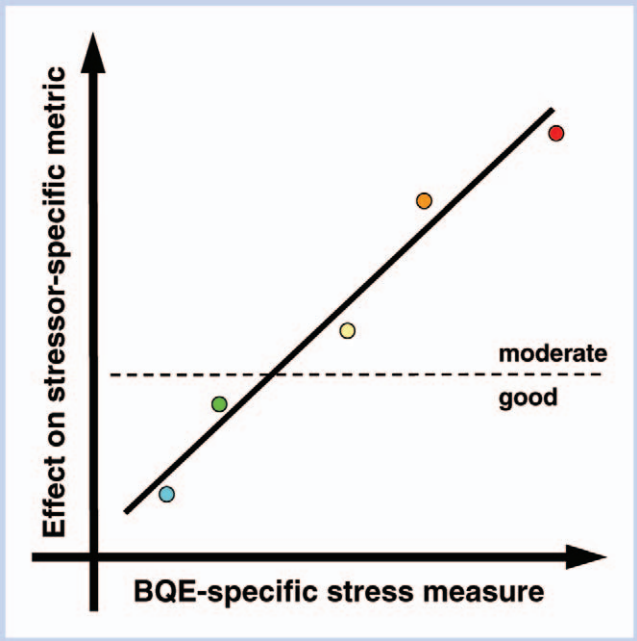

Figure 2. Generic chart of a cause–effect relationship with regard to the Water Framework Directive "ecological status." In order to validate the cause of increased environmental effects in the aquatic community (*y* axis), stressor-specific metrics are correlated to environmental pressures, expressed as BQE-specific stress measure (*x* axis). For illustration, a class boundary between good and moderate ecological status is given. The blue and green dots represent examples of sites that have been correctly classified as of "high and good ecological status," while the yellow, orange, and red dots represent site that are most likely affected by the environmental cause assessed, resulting in "moderate, bad and poor status" classifications, respectively.

typical for large rivers (von der Ohe et al. 2007b). This metric was assumed to be applicable in different biogeographical regions across Europe (Schäfer et al. 2007; von der Ohe et al. 2007b), as it could be adjusted to the taxonomic resolution of all sampling data. However, high average SPEAR values at a monitoring site indicate only the absence of toxic pressure and not necessarily the absence of any pressure.

This stressor-specific metric was also used to derive an example for a cause–effect relationship between ecological and chemical status. The logarithm of the maximum (TU_{max} *D. magna*) and sum (TU_{sum} *D. magna*) TUs were used to represent toxic stress resulting from organic toxicants, as outlined previously. Note that the lower end of the toxicity range was set to a log(TU) of −4, corresponding to 1/10000 of the acute LC50 that was assumed to be a protective concentration level, where no effects on the community were expected (compare P-PNEC). To account for interannual (or seasonal) variation in both chemical exposure and the characteristics of the aquatic communities, the average for the community metric (SPEAR) and the maximum of the toxic pressure (TU) were calculated per site (Figure 3).

The Llobregat was chosen to derive a stressor-specific cause–effect relationship, as for this river basin the most appropriate data set was available: Several annual records existed for the years 2001 to 2004 for macroinvertebrate samples and organic toxicants measured in filtered water samples, both of which were monitored from the same water agency. For the analysis, at least 3 consecutive years with comparable lists of analyzed compounds were required. Hence, from a total of 29 matching ecological and chemical status monitoring sites (compare Table 1), 3 sites were omitted because chemical measurements were available for only 1 or 2 y within the time frame. In contrast, 2 pristine sites in the Llobregat headwaters without chemical data were included in the analysis whose log(TU) was set to −4, as outlined previously.

Previous studies revealed that disturbed communities are positively influenced by undisturbed stream sections upstream (Hatakeyama and Yokoyama 1997; Liess and von der Ohe 2005; Schäfer et al. 2007), which were expected to act as source pools allowing for recolonization of the downstream communities. To account for the absence or presence of these stretches, referred to as "recovery potential," 2 separate regressions were performed for both measures of toxic pressure (Figure 3).

The 2 correlations of SPEAR with log TU_{Max} *D. magna* (Figure 3a) as well as the 2 relationships with logTU_{Sum} *D.*

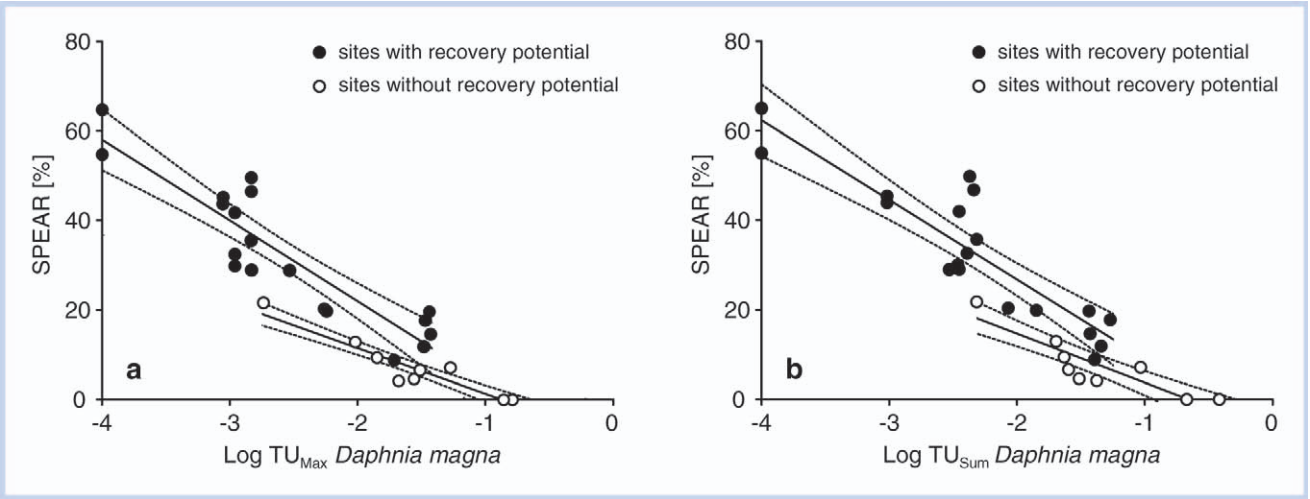

Figure 3. Relationship between (**a**) the maximum toxic units (log TU_{max} *Daphnia magna*) as well as (**b**) the sum of toxic units (log TU_{sum} *D. magna*) for 2001 to 2004 and the average percentage of invertebrate "species at risk" (SPEAR) to be affected by organic toxicants with regard to the BQE of "benthic macroinvertebrates. Twenty-eight monitoring sites in the Llobregat River are significantly differentiated on the presence of recovery sections upstream of the study sites (filled circles, linear regression, $p \leq 0.01$) or absence of such sites (open circles; linear regression, $p \leq 0.01$). Confidence bands show the 95% confidence limit of the respective means.

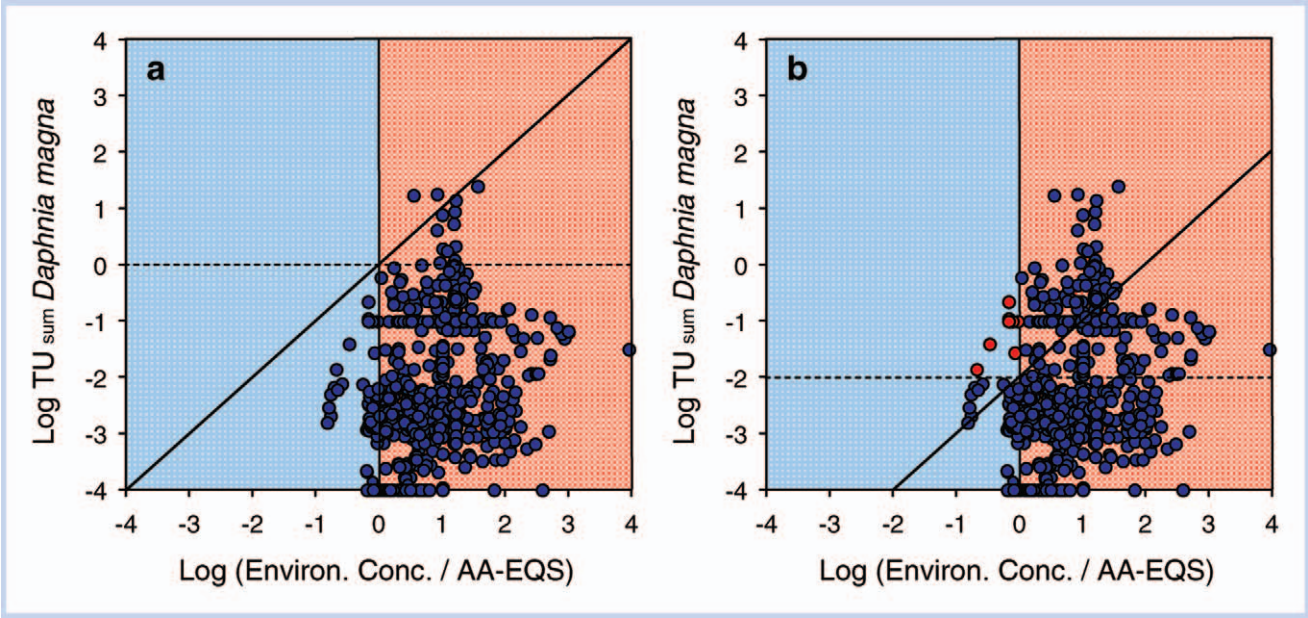

Figure 4. Relationship between the sum of toxic units (log TU_{sum} *Daphnia magna*) per year and site for 2000 to 2004 with regard to the BQE of "benthic macroinvertebrates" and the maximal exceedance of an annual average environmental quality standard (AA-EQS), based on the 41 priority pollutants only. The blue area represents "achieving the good chemical status," while the red area indicates "failing to achieve good chemical status." The solid line indicates the one-to-one relationship for (**a**) the effect endpoint of acute mortality (LC50), while (**b**) considers an effect threshold of 100 times below the LC50, corresponding to a log TU_{sum} *D. magna* of −2. Above this threshold, community effects are expected. The red dots indicate sites that have been classified as "achieving a good chemical status" but that are likely to be affected by chemical stress.

magna (Figure 3b) revealed an increasing impact on the aquatic community with increasing toxic pressure. Interestingly, the correlations for the 2 toxic measures were not significantly different, which indicated that always 1 compound was responsible for most of the TUs (analyses of covariance, $p \leq 0.062$). At the same time, the slopes and intercepts of the 2 regressions for sites with and without recovery potential were significantly different, for both measures of toxic stress (analyses of covariance, $p \leq 0.01$), which indicated higher impacts of toxic stress at sites without recovery potential. For future comparisons, only the 2 linear regressions for log TU_{Max} *D. magna* are reported here. For the 19 sites with recovery potential present, linear regression yielded

$$\mathrm{SPEAR}(\%) = -18.16(\pm 2.06)\log\mathrm{TUmax}D.magna - 14.25(\pm 5.52) \quad (2)$$

where $n = 19$, $r^2 = 0.81$, SE = 6.87, and $F_{1,17} = 78.08$. Interestingly, the slope and intercept of a regression obtained from 2 field studies in Germany and France (Schäfer et al. 2007) were not significantly different (analysis of covariance, $p \leq 0.01$). Hence, it is assumed that invertebrate communities in different basins act similar to toxic stress. This supports the hypothesis that the SPEAR index could be applied across Europe and that similar class boundaries could be used (von der Ohe et al. 2007b). For the 9 sites without recovery potential, linear regression yielded

$$\mathrm{SPEAR}(\%) = -10.76(\pm 1.43)\log\mathrm{TUmax}D.magna - 9.73(\pm 2.41) \quad (3)$$

where $n = 9$, $r^2 = 0.87$, SE = 2.41, and $F_{1,7} = 56.3$. note that the estimated TUs are mostly in the sublethal range, as a log TU of 0 would correspond to an effective concentration similar to the acute LC50.

Applying the suggested SPEAR (%) class boundary value of 29% for the good ecological status (von der Ohe et al. 2007b), a total of 11 sites were classified to be not at risk with respect to organic toxicants, all of which had log(TU) below −2, corresponding to 1/100 of the acute LC50. Therefore, −2 log(TU) is suggested as threshold for toxic effects expected from organic compounds toward benthic macroinvertebrates. However, all these sites had undisturbed stream sections upstream that partially compensated for the effects, in contrast to those sites without recovery potential present.

TUs versus compliance with EQS

In order to directly link ecological status to chemical status assessments, the 2 measures that stand in the present study for these different concepts of the WFD (TUs and compliance with the EQS, respectively) were plotted against each other (Figure 4). For this purpose, all site/year combinations for the 3 basins and the years 2000 to 2004 were compiled. The requirement was that at least 1 PP as well as 1 organic toxicant was measured, resulting in a total of 793 combinations. For the first analysis (Figure 4a), a one-to-one relationship between the acute toxic effect toward the BQE of benthic macroinvertebrates (i.e., log TU_{sum} *D. magna*) was correlated to the compliance of the most potent PPs with EQS (i.e., the logarithmic quotient of the environmental concentration and the respective EQS), both effect levels equaling a value of 0. On the one hand, this first relationship revealed that the EQSs for the measured PPs were rather protective, except for 2 samples above the regression line, for which the maximum log TU_{sum} *D. magna* indicated higher potential effects than estimated from the EQS assessment of PPs alone. On the other hand, these samples were already classified as "failing to achieve good chemical status" and hence were at least not misclassified.

However, the correlations of SPEAR versus TU described previously revealed effects at concentrations 100 times below the acute LC50 (Figure 3a and b). Therefore, the one-to-one relationship (intersecting at safe levels for both stress

measures) would have to be shifted to a log(TU_{sum}) of −2, as community effects would be expected already above this threshold. In this case, for 9 sites that were classified as "achieving good chemical status," the estimated TUs indicated potential effects on the invertebrate community, stemming from all organic toxicants. In other words, compliance with single-compound EQSs, even though rather conservative safety factors had been applied, are probably not protective in all cases. This, however, is not surprising, as the concept of chemical status is based on few PPs, whereas the TU approach considers toxicity of all chemicals detected at the monitoring sites. Therefore, the classification of "achieving good chemical status" cannot be assumed to prevent toxic stress in general (Brack et al. 2008).

A similar result was obtained for the assessment of all chemical compounds measured in surface water of the Scheldt River basin, including the toxic pressure that resulted from dissolved metals and using more sophisticated mixture toxicity concepts (De Zwart et al. 2008). In that study, negative effects were expected at many sites that were classified as "achieving good chemical status" based on effect estimations of the whole community to all toxicants. Moreover, a recent study in the Elbe River indicated potential effects from organic toxicants with regard to all trophic levels despite classification of those sites as "achieving good chemical status" (Schäfer et al. 2008). All studies, including the present analysis, highlight the importance of an updated risk assessment of the available chemical monitoring data in all river basins across Europe.

Recommendations and perspectives

This paper provides recommendations on handling, storage, and evaluation of monitoring data that may help to better achieve the objectives of the WFD. We recommend using standardized chemical codes, for example, provided by a central compound database (like WISE), to ensure a consistent reporting and storage of chemical data (e.g., CAS numbers) instead of using various MS names. In this context, the species identification numbers for macroinvertebrates, as introduced from the AQEM project, are a good example that should be transferred to other BQEs. This would facilitate the application of new scientific assessment tools and allow for comparing assessment results across borders.

The use of stressor-specific metrics that yield similar results in different ecoregions, such as the SPEAR index, are recommended over multimetric indicators, which correspond to a general degradation gradient but do not allow identification of the predominant stressors responsible for the deviation of the good ecological status. The observation that chemical concentrations exceed EQSs for 1 or more PPs at the majority of chemical monitoring sites, corresponding to the classification of "failing to achieve good chemical status," suggests higher potential effects of chemical exposure on ecological status than currently expected. Therefore, the authors suggest considering the application of the current EQS values for refined chemical status assessments that might be already overdue. We also recommend including the risks of sediment bound compounds, which could not be considered in this study.

Consequently, chemical status and toxic stress in general should be considered as of similar relevance as morphological degradation and eutrophication for the assessment of ecological status by applying basic mixture toxicity concepts (i.e., TUs) with regard to different BQE. With regard to the assessment of chemical status, the use of P-PNECs is suggested for compounds for which no or insufficient toxicity data are available, allowing for the identification of potential emerging compounds at the European level. The P-PNEC values could also be applied to both target analysis to preliminarily identified screening compounds to prioritize those with the highest potential concern. Finally, integrated risk assessment tools and decision support systems (as provided by the MODELKEY project) that cover many different stress factors simultaneously may permit assigning an ecological status in a more comprehensive way and help unravel potential confounding factors. It is only then that effective management measures can be identified that enhance the ecological status in a sustainable and cost-efficient way.

Acknowledgment—The presented work was supported by the European Commission through the Integrated Project MODELKEY (contract 511237-GOCE). We would like to thank Ralf B. Schäfer, Emma L. Schymanski, and Leila I. Cruzat, who provided valuable suggestions that improved the manuscript. Furthermore, we would like to thank Ward de Cooman and the Flemish Environment Agency for kindly providing data used for the analyses. Note Digitale Daten des Landesbetriebes für Hochwasserschutz und Wasserwirtschaft Sachsen-Anhalt—Mit Genehmigung des LHW Gen. LHW/5.4/003/2006.

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