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Setting environmental quality criteria for water and sediment in The Netherlands: a pragmatic ecotoxicological approach

In 1987, a project was begun to prepare environmental water quality criteria for the National Water Management Plan 1989–1994. In setting the new Netherlands *Environmental Water Quality Objectives 2000* (EQO), the No Observed Effect Concentration (NOEC) from standard chronic toxicity tests with aquatic species served as a starting point, in combination with a number of new elements that had not been included in other risk-extrapolation methods until then. These new elements were: (1) the equilibrium partitioning of compounds between the dissolved fraction in water and the fraction adsorbed in suspended solids or sediment; (2) the joint toxic action of related substances on aquatic species; and (3) the impact of persistent bioaccumulating chemicals on predator species feeding on the aquatic environment.

The use of the equilibrium partitioning theory attempts to incorporate the biological availability of compounds in the criteria. The dissolved concentration is at this time the best estimation of the bioavailable fraction. Criteria in sediment and suspended solids can be derived from the dissolved concentration using equilibrium partitioning. In this way, the control of criteria can be restricted to the matrix where measurements are most efficient. The combined toxicity was included, because there is enough evidence to show us that mixtures of related compounds at low concentrations present a risk, even though none of them is present at toxic levels. The third item, predator species, was included because it is literally cited in the verbal definition of the EQO. A set of EQO's was defined following these lines for 8 metals and 88 organic compounds in water, suspended solids and/or sediment.

ENVIRONMENTAL CRITERIA FOR WATER AND SEDIMENT

Water quality criteria aim, among other things, at protecting the aquatic environment. With our present state of knowledge, it is impossible to set criteria that will guarantee an absolute protection of the ecosystem. Therefore, these criteria will remain a subject of public discussion, dominated by a struggle between economic interests and environmental considerations. A clear and widely supported protocol, that translates ecotoxicological and chemical data into practical standards is needed to achieve the greatest possible involvement of all parties concerned in attaining the proposed goals. The major requirements for such an approach are:

(1) The use of state-of-the-art knowledge on ecotoxicology and environmental chemistry as a starting point: generally accepted scientific concepts will not be subject to intensive discussions.

(2) The derivation of criteria from the available data by a well described procedure, consisting of logical steps that are as clear to outsiders as they are to those who constructed them. As scientific knowledge is often limited, a number of these steps will have to be arbitrary.

(3) Setting criteria for those compounds that we know presents a real problem at current concentration levels. Criteria for non-problem compounds are not urgent and monitoring non-problem compounds diverts the available funds from more problematic environmental impacts, and will not stimulate the effort needed to improve the environment.

Extrapolation from laboratory data

Ecotoxicological data from the laboratory are the most favoured approach to estimate risks of chemicals in the aquatic environment. Different methods were evaluated recently by the Dutch Health Council [1]. The first protocols had an empiric character and were based on acute (short-term) toxicity and, in later stages, on the lowest chronic No Observed Effect Concentration (NOEC) from toxicity studies with aquatic species in the laboratory. The US Environmental Protection Agency (EPA) [2] derived its standards from acute and chronic toxicity studies with species from three trophic levels, using arbitrary factors to account for missing data and variability between species. A few years later, Slooff and co-workers [3] proposed a risk extrapolation based on the regression analysis between laboratory and micro-ecosystem toxicity data. Recent developments focused more on the variability between species, estimating risks for an aquatic ecosystem from laboratory tests on a limited number of species using a statistical method [4, 5]. A number of points were

not, however, addressed by these methods, such as the bioavailability, combined toxicity and biomagnification.

A solid basis and some new elements

In 1987, a project was started to prepare the water quality criteria for the National Water Management Plan 1989–1994 [6]. These criteria are aimed at the protection of the environment, and are complementary to standards for drinking water and human health. In setting the new Dutch Environmental Water Quality Objectives (EQO) for the year 2000 [7], the NOEC from standard chronic toxicity tests with aquatic species served as a starting point. The NOEC is widely accepted and also used by all risk-assessment methods stated above. For the EQO it was combined with a number of new elements:

(1) The equilibrium partitioning of compounds between the dissolved fraction in water and the fraction adsorbed to suspended solids or sediment.

(2) The joint toxic action of related substances on aquatic species.

(3) The impact of persistent bioaccumulating chemicals on predator species feeding on the aquatic environment.

The use of the equilibrium partitioning theory is an attempt to incorporate the biological availability of compounds in the criteria. The dissolved concentration is at this time the best estimation of the bioavailable fraction. Criteria in sediment and suspended solids can be derived from the dissolved concentration through the equilibrium partitioning. In this way, the control of criteria can be restricted to the matrix where measurements are most efficient. The combined toxicity was included, because there is enough evidence to show us that mixtures of related compounds at low concentrations present a risk, even though none of them is present at toxic levels. The third item (predator species) was included because it is literally cited in the verbal definition of the EQO, stating that it should provide "a quality of surface waters such that it ... allows the development of an aquatic ecosystem, that will not only support the life of higher taxonomic groups such as fish, but also will protect ecological interests outside the water, such as specialized predators feeding on aquatic species" [7]. A set of EQO's was defined following these lines for 8 metals and 88 organic compounds in water, suspended solids and/or sediment.

SELECTION OF RELEVANT COMPOUNDS

A first step in reviewing the criteria was to select a limited number of compounds for the EQO based on environmental occurrence and toxicity. The aim of this selection was to reduce the EQO's to a manageable number that includes most of the compounds known to us and that are estimated to present an environmental hazard. In this way, the effort of water quality management can focus on the most problematic substances, cutting the costs of monitoring. The reduction was achieved by using the ratio between the acute toxicity to fish or crustaceans and the concentrations measured in, or assumed to be present in, surface waters, in combination with the bioaccumulating potential (Fig. 1).

To start with, a list of nearly 287 substances was drawn up. This was a compilation of "black lists" from the European Community, the US EPA and The Netherlands Dangerous Substances Act, completed by commonly used pesticides, bulk chemicals handled by Rotterdam harbour, and compounds known to be released into the aquatic environment by various sources [6]. Obviously, this list only represents the "tip of the iceberg", as many toxic compounds in the environment are still unknown to us because we cannot measure them. Substances were selected for EQO's if the ratio between environmental concentrations and acute toxicity was greater than 0.001, or if this ratio was higher than 0.0001, in combination with a calculated bioconcentration factor higher than 1000 (Fig. 1).

When data were missing, the acute toxicity was estimated from the relation between "minimum toxicity" and K_{ow} by a QSAR (Quantitative Structure–Activity Relation) [8]. K_{ow} is the partition coefficient between octanol and water. K_{ow} not only correlates with the bioaccumulative potential of many organic chemicals, but also with the "minimum toxicity" of organics in acute toxicity tests with fish or water fleas. If information on concentrations in water was missing, the ratio between the estimated production or emission in the EC or the Netherlands (translated into an aqueous concentration) and the acute toxicity was used. This was the case for many pesticides.

Only fourteen compounds were found to present a negligible aquatic problem. This group included a number of volatile (halogenated) compounds with a low toxicity (naphthalene, isopropylbenzene, 1,2 dichloropropane, etc.), and some problem chemicals from the past (α-, β- and δ-HCH). Insufficient data or sometimes even a total absence of accessible data, hampered a proper priority assessment for 147 compounds [6].

FROM TOXICITY TESTS TO ENVIRONMENTAL CRITERIA

Protecting aquatic ecosystems

The philosophy of the EQO is to protect the essential structure of aquatic ecosystems, *including terrestrial species that depend on them for their food*. The EQO is a national minimum objective, that has to be achieved

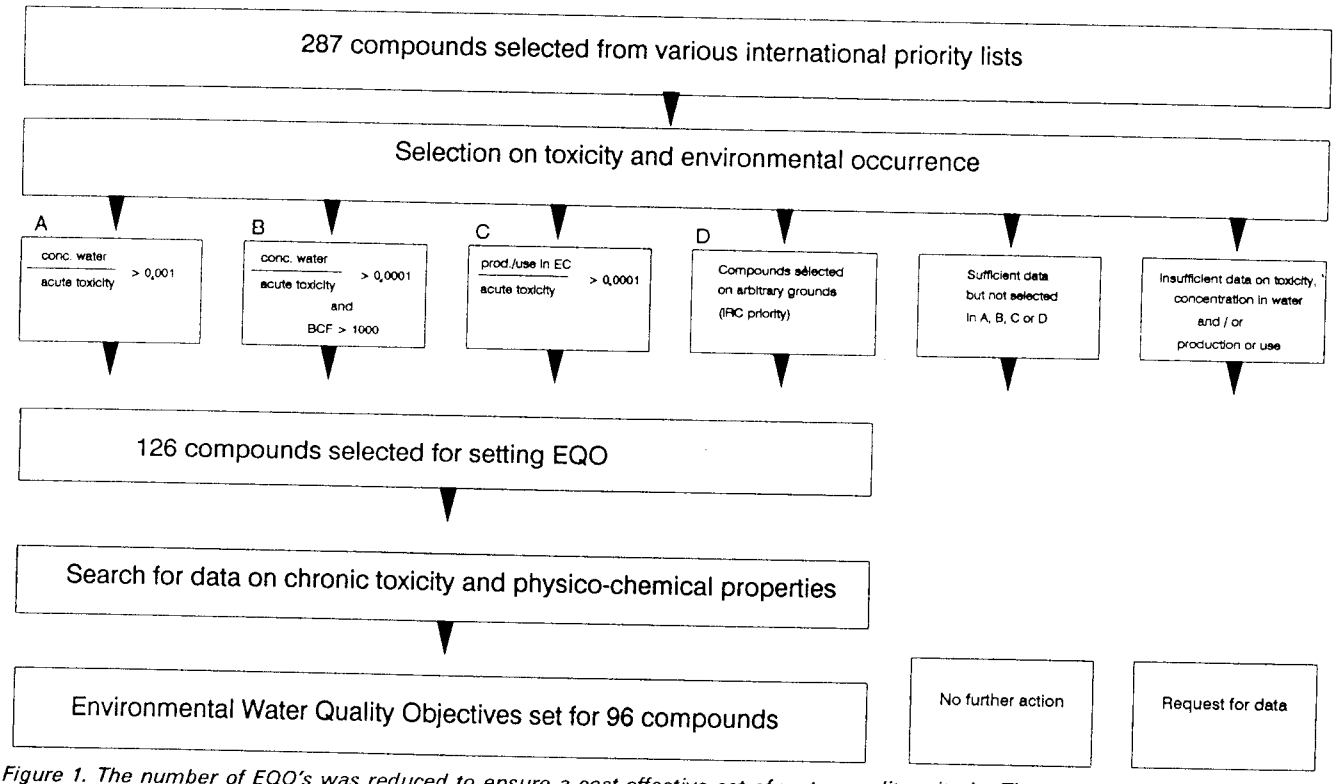

Figure 1. The number of EQO's was reduced to ensure a cost-effective set of water quality criteria. The selection was based on the ratio's between toxicity criteria and concentration measured or expected to be present in surface waters. The selection scheme resulted in a final list of 96 compounds for which EQO's were drawn.

by the year 2000. Locally more stringent standards may be defined, if necessary, to protect vulnerable ecosystems. From this it is clear that the EQO is an interim goal, that may be subject to further review in a few years using new developments in risk assessment.

EQO's have to be derived from toxicological and chemical information that is currently available. This is why only data were used from standardized toxicity tests, from at most four taxonomic groups. Evaluations were restricted to assays with algae, crustaceans and fish, and if available, also from molluscs, a rough caricature of an ecosystem. If a compound was estimated to present a hazard to predators by accumulation in the food chain, the evaluation was extended to toxicity data in mammals or birds, or existing standards for wildlife or human consumption were used. The steps from lowest NOEC to EQO only include the selection of the most sensitive element in the aquatic ecosystem, a possible correction from the nominal NOEC to the dissolved concentration, the application of an extrapolation factor accounting for joint toxic effect of related compounds (Fig. 2). An EQO is finally expressed as total concentration in water, or as concentration in suspended matter and sediment, according to the environmental partitioning of the compound in question.

The lowest chronic no-effect level in the most sensitive species

The absence of long-term effects in the most sensitive species from a selection of trophic levels in the ecosystem, is widely accepted as the starting point for setting water quality criteria. Sticking to standard tests, the effects that can be considered, are limited to increased mortality, growth retardation and reduced reproduction. The potential impact of such overall effects in natural populations will not be subject to much discussion. The use of more refined endpoints is out of the question, because it is not always possible to relate them to effects on populations [9]. Often also these physiological parameters are not measured in standardized tests, and the interpretation of the results in relation to ecological relevance may not be undisputed [10].

Although the NOEC is not the most accurate method of describing the "true" No Effect Concentration (NEC), it was used for calculating the EQO's. The NOEC is the highest concentration tested that was not significantly different from the control. The ideal NEC would be a concentration between the NOEC and LOEC (Lowest Observed Effect Concentration), but could eventually turn out to be lower than the NOEC if, for instance, the statistical evaluation of the test results was not adequate. The quality of a NOEC also depends on the effects that were studied. Moreover, the gap between NOEC and LOEC usually varies between a ratio of 1.8 and 10, according to the amplitude of the dilution steps. Nevertheless, given the published studies available and the interlaboratory differences that are observed in practice, the lowest NOEC is the best alternative at this time.

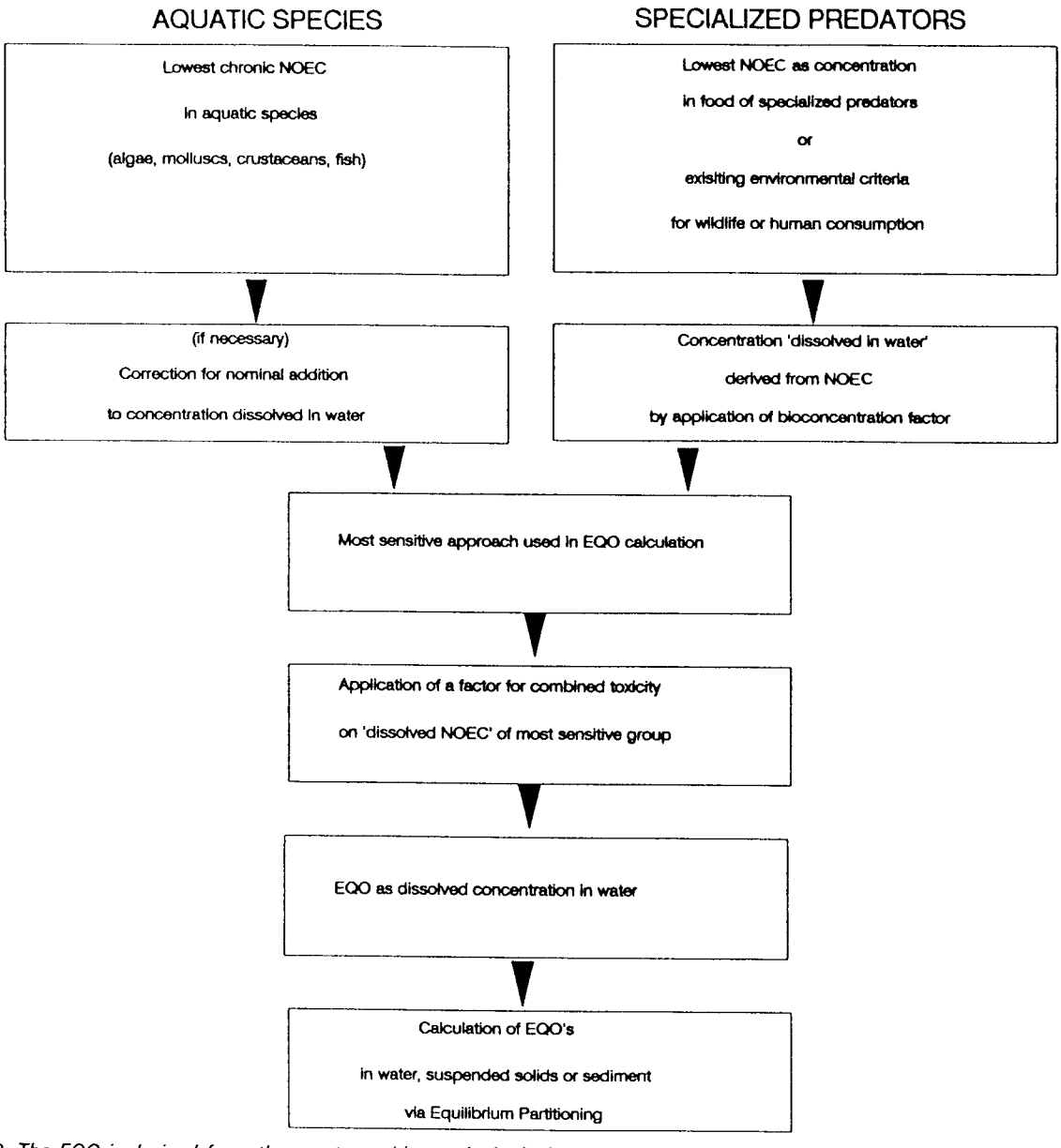

Figure 2. The EQO is derived from the most sensitive toxicological route in a number of subsequent steps. The procedure first determines which element of the ecosystem is at risk. The EQO is then calculated from the lowest No Effect Concentration in long-term chronic assays, taking into account the fraction that is biologically available and the possible combined effect of related chemicals.

Protecting specialized predators

Many bird species, and a number of mammalian predators, are dependent upon aquatic ecosystems for their food, which will mainly consist of fish and/or shellfish. Persistent hydrophobic compounds and some heavy metals accumulate in aquatic species. In this way, predator birds and mammals are subject to increased exposure to toxic substances. The NOED (No Observed Effect Dose) in food from chronic toxicity studies with related species, were used to calculate the EQO's. If wildlife or food quality criteria were available and turned out to be lower than the NOED, the EQO was derived from these standards. From these data, the corresponding "dissolved concentration" in water was calculated by dividing them by the bioconcentration factor (BCF). As BCF values from experimental studies vary over a wide range, a theoretical BCF was used for organic compounds with a log K_{ow} lower than 6.5, assuming a fat content of 5%, according to the relation [8]:

$$BCF = K_{ow}/20$$

The final EQO, for these compounds always stated in sediment (and suspended solids), was derived from the "dissolved concentration" using equilibrium partitioning.

Incomplete data

Many data sets referred to in literature are incomplete. In chronic toxicity studies, the concentrations of chemicals were often not measured during the test.

In order to integrate the concept of equilibrium partitioning of chemicals in the water, the NOEC must be stated as the dissolved concentration in water. If measured data were not given in a report, a "dissolved" concentration was estimated arbitrarily from the nominal addition to the test medium.

Where chronic toxicity studies were missing for the most sensitive species, a NOEC was sometimes calculated from acute toxicity data with an extrapolation factor. If sufficient acute to chronic ratios were known from related compounds, the "worst case" acute/chronic ratio was applied. If no information was available, and the need for an EQO was urgent, the lowest acute LC50 value (lethal concentration for 50% of test animals) of the most sensitive taxonomic group was divided by 100, in analogy to extrapolation factors used in other protocols.

Combined toxicity

A toxic substance will never be on its own in the environment. Joint toxic action of different compounds will occur, even at very low levels [11]. Additivity is the most common interaction. The acute toxicity of chemicals is very often additive, even when different modes of action are concerned. Additivity has also been demonstrated in mixtures of related compounds at chronic exposure levels, whereas compounds with different toxic endpoints will probably show a less-than-additive response [12–14]. A recent study in enclosure experiments, simulating an ecosystem, confirmed the additive response of eight heavy metals at chronic-exposure doses (Van der Gaag et al., unpublished results).

EQO's aim at the protection of the structure of an ecosystem at a maximum tolerable level. If criteria would have been set at the NOEC level of each compound, an effect might happen in an ecosystem even if all criteria are met. Therefore the potential additivity of compounds with related toxic endpoints was integrated into the extrapolation factor that was applied to the most sensitive NOEC level. The principle of additivity was applied within each group of compounds that showed structural similarities or related mechanisms of action according to the Toxic Units (TU) principle, which assumes that no effect will occur if the sum of the quotients of the concentration in water and the NOEC is lower than 1:

$$\sum_i \frac{\text{concentration } i \text{ in water}}{\text{NOEC } i} < 1$$

The ideal situation would be to set a criterion at the "dissolved" NOEC level, and to apply the TU after monitoring when the evaluation is carried out. This approach is at present quite utopian, and also would assume that *all* compounds with related modes of action are being monitored. A first correction based on production or use was applied in an arbitrary way to account for the related compounds for which no EQO was set:

$$\sum \frac{\text{concentration in water}}{\text{NOEC}} < \frac{\text{production/use compounds with EQO}}{\text{production/use total group of related compounds}}$$

From this point, three alternatives have been used. If the occurrence of a group of compounds is very widespread, and their uses not related, the EQO was calculated by dividing the NOEC corrected for production/use by the number of compounds for which criteria were set in the group. This approach was followed for industrial chemicals with a narcotic mode of action (solvents, many organohalogens, such as chlorobenzenes and chloro-anilines) and for the classical organochlorinated pesticides, such as DDT and derivatives, the "drins", lindane, etc.

Other groups of compounds, such as PCB's and polycyclic aromatic hydrocarbons, show a pattern of environmental occurrence. For these groups the EQO's were adapted to this pattern of environmental occurrence.

The TU approach as such was maintained within a number of groups of pesticides. The background for this decision is that the application of a pest control agent always involves the choice between alternatives, and that its use is often restricted in time. In this way, different pesticides may "take turns" to fill the EQO over the year.

Equilibrium partitioning: the link between sediment and water

The bioavailable fraction, as dissolved concentration in water, is the cornerstone of the EQO's. The equilibrium partitioning theory was used to derive the concentrations in sediment and suspended matter from the dissolved concentration [15]. The final EQO is set in the most suitable matrix (Table 1). This translation enhances the efficiency and lowers the costs of monitoring programs, as measurement is performed in the most appropriate environmental compartment: either dissolved in water, or adsorbed in suspended matter or sediment. For instance, it makes no sense to measure extremely low dissolved concentrations of heavy metals, PCB's or dioxins in water, when most of it is adsorbed in suspended solids.

The equilibrium partitioning for heavy metals was based on median empiric K_d values measured in surface waters in the Netherlands. The K_d is the distribution coefficient of metals between water and suspended solids. For organics it was derived from $\log K_{ow}$ values. EQO's were set in suspended solids and/or sediments for poorly soluble compounds. For comparison,

TABLE 1. Selection of some Environmental Quality Objectives. EQO's are calculated for different matrices: dissolved in water, in water with 30 mg dw/l suspended solids, and in sediment. For most metals, and for lipophilic compounds such as dieldrin, the final EQO has been set as a value in sediment (or suspended solids), because this is the most appropriate matrix for measurements [6]

	Lowest NOEC[1] (μg/l)	EQO		
		Dissolved (μg/l)	Total[2] (μg/l)	Sediment (mg/kg dw)
Atrazin	2	0.08	0.08	1.5
Azinphos-methyl	0.1	0.02	0.02	0.1
Cadmium	0.1	0.03	0.2	2
Copper	5	1.3	3	35
Dieldrin	0.12	0.0005	0.003	0.04
Lindane	2.2	0.1	0.1	0.01
Pentachlorophenol	1.8	0.05	0.02[3]	0.2
Tributyl tin	0.04	0.01	0.01	0.0015
Zinc	26	6.5	28	477

[1] Based on chronic NOEC values for aquatic species (algae, crustaceans, molluscs and fish) available mid 1988.
[2] Assuming 30 mg/l dw of suspended solids.
[3] The former water quality criterion of pentachlorophenol was lower than the one calculated using the EQO approach, and was maintained according to the stand still principle.

values for "total water" (including suspended solids) are calculated for an "average" surface water in The Netherlands. This assumes a concentration of 30 mg dw of suspended solids/l, with an average content of 40% particles smaller than 2 μm and 10% organic carbon. In sediment, the organic carbon contents amounts to 5%, the clay and silt fraction to 25%.

COMPARISON WITH OTHER METHODS

At a national level, the EQO has been positively welcomed both by local water authorities and industry, even though some of the criteria mean that strenuous efforts will have to be made to realize them within the planned period. The chosen approach may have facilitated the acceptance of the EQO, the outcomes most of the time fall within the same ranges as provided by other protocols, except when it comes to the bioaccumulating substances (Table 2).

TABLE 2. Comparison based on identical data sets of ecotoxicological risk assessment with the EQO method, the EPA method [2] and the RAE method [5, 19]. The final differences between these approaches are relatively small, except for situations where biomagnification is involved, such as with dieldrin. For comparative reasons, the values are expressed as dissolved concentrations in μg/l

	Lowest NOEC[1]	Method		
		EQO	EPA	RAE
Atrazin	2	0.08	0.2	0.8
Azinphos-methyl	0.1	0.02	0.01	0.07
Cadmium	0.1	0.03	0.01	0.16
Copper	3	1	0.3	1.7
Dieldrin	0.12	0.0005	0.012	0.05
Lindane	2.2	0.1	0.22	0.55
Pentachlorophenol	1.8	0.4	0.18	2
Tributyl tin	0.04	0.01	0.004	0.01
Zinc	5	1.5	0.5	1.6

[1] Based on chronic NOEC values for aquatic species (algae, crustaceans, molluscs and fish) available mid 1990 [19].

Evidently no single method has managed to cope with all the different aspects that are involved in risk extrapolation, giving each method its own weaknesses and strengths. The "oldest" approaches were handicapped by the lack of data, and therefore invested in the translation from scarce acute toxicity data to a multispecies NOEC for ecosystems [2, 3]. As more chronic toxicity data became available, a further attempt to integrate the interspecies variation was developed by Kooijman [4] and refined by Van Straalen and Denneman [5]. The EQO concept contains a number of elements that have not yet been handled by other protocols: environmental partitioning, combined toxicity and bioaccumulating potential (Table 3).

TABLE 3. Approaches used for risk assessment. The different approaches for risk assessment have tried to include a number of factors that should be considered when setting environmental quality criteria. None of the methods proposed was complete in its considerations

	EQO	EPA	RAE	BQZ
Lack of data	±	+	+	+
Persistence	[1]	–	–	+
Single species to ecosystem	–	+	+	+
Biomagnification	+	[2]	–	–
Combined toxicity	+	–	–	–
Laboratory to field	–	–	–	–
Environmental partitioning	+	–	–	–

EQO = Environmental Quality Objectives (this paper); EPA = EPA risk assessment method [2]; RAE = risk assessment for ecosystems [4, 5]; BQZ = BundesLänder QualitätsZiele [16]; – = not explicitly considered; + = included in the risk-assessment protocol.

[1] Persistence is not explicitly included in the scheme. However, the selection of compounds for the setting of EQO is based primarily on environmental occurrence. This means that the persistence of a compound will always be sufficient to cause increased concentrations in rivers in the Netherlands.

[2] The EPA has not accounted for biomagnification in its water quality criteria, but only set criteria in this respect for human consumption.

EPA: emphasis on missing data

The U.S. EPA was one of the first policy-making agencies to derive criteria for aquatic species from toxicity data. In essence, the EPA primarily focused on the lack of available data. Extrapolation factors are high if data are lacking. If a criterion is derived from only one acute toxicity test, a factor of 1000 is applied. If three LC50 values are available from different taxonomic groups (algae, crustaceans and fish), the factor amounts to 100. If the chronic NOEC values from the most sensitive species are known, a factor of 10 is applied to account for the variability between species [2]. The German "Qualitätsziele" recently introduced by the Bundes-Länder ArbeitsKreis (BLAK-QZ) [16] follow a quite similar approach, but were completed by the introduction of a fourth taxonomic group (bacteria), and by the application of an extra factor of ten if a compound is persistent, or if toxic metabolites may be produced in the environment. The pragmatic $10 \times 10 \times 10$ approach from EPA and BLAK-QZ is supported by the experience of Slooff et al. [3], based on interspecies variation and correlations between laboratory data and micro-ecosystem studies: for many compounds the $10 \times 10 \times 10$ approach will remain on the safe side, but the acute/chronic ratios will increase, with many compounds showing specific modes of action.

New concepts based on interspecies variation

During the EQO preparation, a new risk extrapolation method, the Risk Assessment for Soil Ecosystems (RAE), was introduced aiming at determining the concentration level in the environment that will protect 95% of the species [5, 17]. The concept was derived from the ideas postulated by Kooijman [4] for the assessment of risks for aquatic species. This approach introduces a number of promising new concepts, that were not used for the EQO, because the scientific discussions on the values and performance of this method had just started [1]. However, a number of pitfalls still have to be sorted out. A 95% protection level, based only on toxicity data for aquatic species, will not protect specialized predators feeding on fish and shellfish from aquatic environments. Problems may also arise when assessing the risk of compounds with specific modes of action [18]. A practical problem may also concern the availability of sufficient data. The statistical reliability of the method increases with the number of taxonomic groups that are included in the calculation. At present, the application of the RAE method in standard protocols for risk assessment is still hampered by the lack of toxicity data for many compounds.

MONITORING STRATEGIES: A PERMANENT SEARCH FOR THE MOST PROBLEMATIC COMPOUNDS

It is impossible to set EQO's for every compound that might happen to occur in water. First of all, only a part of the organic pollutants present in water can be identified. Secondly, even if technically feasible, an efficient *routine* control would be out of the question for economic reasons. This is why an effort was made to reduce the number of EQO's by selecting the compounds that present the most realistic environmental problem. Unfortunately, the largest reduction in numbers was not realized by selection based on ecotoxicological data and environmental concentrations, but resulted from a lack of relevant data for almost 50% of the compounds considered. Although in this way many substances remain outside the field of our attention, EQO's help us to set controllable goals for what we do know. Measures taken to reduce the discharge of the identified pollutants will also reduce pollution by many of the unidentified compounds in their wake. EQO's that have been set for problematic substances also will draw more attention towards environmental affairs than an objective for a non-problem compound that has already been achieved before the standard was set.

Efficient monitoring programs and political attention for the environment therefore ask for a permanent updating of monitoring lists, and for a weeding of criteria that are included in official regulatory settings requiring frequent routine monitoring. Once environmental concentrations have fallen far below the monitoring standard, criteria could be moved from a *routine* monitoring list to a "historical" list. In this way, EQO's could contribute to a better environment, even if the number of compounds that is actively and routinely monitored, is limited to a small part of the total pollution.

FUTURE DEVELOPMENTS

Much more data needed

All methods involved are hampered in their use by a lack of data. There is an urgent need for test results from chronic toxicity assays with aquatic species and of data needed to predict environmental fate. QSAR's can help us in setting the priorities, but test data from the most problematic compounds are needed to establish reliable water quality criteria. Another gap in our knowledge is our poor understanding of the possible secondary ecological consequences of toxic chemicals in the aquatic ecosystem.

Integration of complementary approaches

Ecotoxicology and environmental chemistry are fields of science that are rapidly evolving. This is especially true with regard to risk-assessment. Clearly the pragmatic choices made by the end of the eighties to set EQO's will turn obsolete by 2000, the year in which the objectives have to be met. It is also clear that up to now none of the methods for risk-assessment has covered all the different aspects of this field. However, the analysis of the different approaches shows that they are in many ways complementary (Table 3), although an exception has to be made to the way in which the differences in laboratory experiments and the field have to be incorporated. But the environmental partitioning theory can be linked to the outcomes of the different methods, and the same applies to the combined toxic effects of substances. Integration and refinement of these items deserve more attention in the next few years. In the long term, the development of ecotoxicological risk-assessment methods should focus on indicating the bottom line: the concentration that will not interfere with the evolution towards a sustainable economical and ecological development.*

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* More information on the Netherlands EQO's is available in the English summary of the Netherlands National Policy Document on Water Management, titled "Water in The Netherlands: a time for action" (The Hague, 1989), and in the report "Perspectives for aquatic organisms" (Stortelder et al., 1989), available at RIZA.