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Validation of an ecosystem modelling approach as a tool for ecological effect assessments

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Abstract

In ecotoxicology, derivation of a "safe" environmental concentration is usually achieved by the use of extrapolation factors or by statistical extrapolation from a set of single species toxicity data. These approaches ignore ecological interactions between species in the field. An ecology-based alternative to this pragmatic approach can be ecosystem modelling, which can account for ecological interactions. However, it is largely unexplored how well the predictions of these models quantitatively agree with large-scale experimental studies. Therefore, we evaluated the capacity of a flexible ecosystem model to predict population and ecosystem-level no observed effect concentrations (NOECs) of 7 organic toxicants. These NOECs were compared with population and ecosystem-NOECs observed in 11 micro- and mesocosm studies. For each of the latter studies, the model was customized to account for the specific ecological interactions within these systems and combined with appropriate single-species toxicity data from literature. Population-NOEC predictions were accurate, or at least protective, for 60, and 85% of all considered model populations, respectively. For all 11 studies, a protective ecosystem-NOEC could be derived, being accurate in 7 cases, and conservative in 4 cases. In general, it can be stated that this type of models can serve as an ecology-based alternative to current extrapolation techniques in EEAs and water quality standard setting.

Keywords: Water quality criteria setting; Dynamic ecosystem modelling; Toxicant stress; Uncertainty propagation

1. Introduction

Ecological effect assessment (EEA) aims at assessing or predicting potential effects of a chemical substance on the structure and function of ecosystems. These higher-level effects are usually estimated through extrapolation of single-species effect data. However, these approaches are based on largely unproven hypotheses and are therefore heavily criticized (Versteeg et al., 1999; Forbes and Calow, 2002). One of the most crucial hypotheses is the representation of the community sensitivity as a set of independent species sensitivities (Wagner and Lokke, 1991). Possible ecological interactions between populations in communities (e.g. Sommer et al., 1986; Preston and Snell, 2001) are thus ignored. Since effects on these ecological interactions combine with direct toxicant effects on populations to determine effects at the community and ecosystem level (Chapman et al., 2003; Fleeger et al., 2003), knowledge about such interactions should be incorporated in EEAs to more accurately estimate these higher level effects of chemicals.

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Large scale experimental studies, i.e. micro-, mesocosm and field enclosure studies, are capable of accounting for direct and indirect toxicant effects resulting from ecological interactions (e.g. Hoagland et al., 1993; Clements and Kiffney, 1994; Joern and Hoagland, 1996). Unfortunately, these types of studies are very resource-demanding and can thus not be used for routine evaluation of chemical toxicity. Especially in view of REACH (Registration, Evaluation and Authorisation of Chemicals; http://ecb.jrc.it/REACH/, a EU-legislation aiming at environmental risk assessments for approximately 30 000 substances) there is a clear need for alternative, less resource-demanding methodologies to predict the effects of chemicals on ecosystem structure and function.

Mathematical models have been proposed as an alternative approach to incorporate ecological interactions in environmental effect assessments and water quality standard setting (e.g. Pastorok et al., 2003). Such models integrate toxic effect sub-models in ecosystem models to simulate effects of toxicants on ecosystems. Toxic effect sub-models vary in complexity and range from highly realistic toxicokinetic models (Bartell et al., 1988; Traas et al., 2004a) to rather simple external concentration-effect functions (Bartell et al., 1999; Traas et al., 2004b). In terms of feasibility, use of the latter is preferable, since these sub-models only require a limited set of single-species toxicity test results. In contrast, toxicokinetic sub-models are often characterized by a large number of uncertain parameters (Sijm and Vanderlinde, 1995). In a previous paper (De Laender et al., in press), we demonstrated that population- and ecosystem-level no observed effect concentrations (NOECs) of copper in microcosms could be predicted accurately using an ecosystem model with logistic external concentration-effect functions as toxic effect sub-models. In that paper, the ecosystem model was customized to reflect the ecological interactions within these microcosms. The toxicity data used in that paper were: lethal concentrations (LC_{50}) for invertebrates and macrophytes, effect concentrations (EC_{50}) for phytoplankton and macrophyte growth rates, and published default slope values of concentration response functions (Smit et al., 2001). Given these minimal data requirements, the idea of applying this type of model for assessing effects and setting water quality standards seems appealing. However, the use of this model as an ecology-based alternative to current statistical extrapolation approaches requires a validation of its predictive capacity for a wider range of toxicants and ecosystems. Naito et al. (2003) have performed such a validation by comparing model-predicted ecosystem-NOECs with those observed in different artificial ecosystems. However, these authors did not account for the specific ecological interactions in the considered ecosystems, but rather examined if one generic model-ecosystem could be used to simulate effects in a range of large-scale experimental studies. Predicted population-level effects were only qualitatively compared with the observations.

This paper presents a validation study, based on the quantitative comparison of predicted population- and ecosystem-NOECs with those observed in 11 experimental community-level studies (micro-, mesocosms and enclosure studies). In these studies, the effects of 7 different organic toxicants were examined. We examined if this approach resulted in an adequate agreement between predictions and observations, both at the population- and ecosystem-level.

2. Material and methods

2.1. Ecosystem model – concept

A dynamic ecosystem model was constructed in an object oriented framework using the software package WEST® (World wide Engine for Simulation, Training and Automation, Hemmis NV; Kortrijk, Belgium). The model consists of a set of objects, and each object describes the growth of a model population in terms of its biomass concentration using differential equations. By connecting different objects and defining feeding relationships between them, a customized food web can be designed. The number of populations that can be modelled is unlimited and available objects are: phytoplankton, macrophytes, zooplankton, planktivorous fish and piscivorous fish. Intrinsically identical objects (e.g. phytoplankton) can be differentiated by parameter tuning (e.g. spring vs. summer populations). All equations are based on Park (1974) and USEPA (2000a). Equations for phytoplankton, zooplankton, and organic matter/nutrient cycling together with a description of parameters and variables are given in Fig. 1. Equations for fish are not explicitly given in this figure, as they only differ from the zooplankton equations by the food source they consider. The food source for planktivorous and piscivorous fish is zooplankton and planktivorous fish, respectively. Equations for macrophytes are identical to those for phytoplankton, but do not contain the rates 'consumption by zooplankton' and 'sinking' and are thus not explicitly given in Fig. 1. Equations of temperature limitation and temperature correction are identical to those proposed by USEPA (2000a) and are thus not given in Fig. 1.

Dynamic driver variables, also called 'forcing functions' are photoperiod, temperature, and toxicant concentration. The latter allows to simulate dynamic exposure patterns, i.e. time-varying toxicant exposure. Daily values of the dynamic driver variables are contained in an input file which is read by the ecosystem model during simulation.

2.2. Large-scale experimental studies

Community-level studies describing toxicant effects on populations' biomass and/or abundance were taken from literature. To maximize the amount of information on the effects of the toxicants on the different populations, we selected studies where observed population dynamics exhibited at least one of the following five seasonal events

a

phytoplankton equations

$$\frac{dBio_{phytoplankton}}{dt} = photosynthesis - respiration - excretion - mortality - sinking - consumption_{zooplankton}$$

$$photosynthesis = PS_{max} \cdot PS_{limit} \cdot Bio_{phytoplankton}$$

$$PS_{limit} = Nutr_{limit} \cdot Temp_{limit} \cdot Light_{limit}$$

$$Nutr_{limit} = \min(N_{limit}, P_{limit})$$

$$N_{limit} = \frac{N}{(N + K_N)}$$

$$P_{limit} = \frac{P}{(P + K_P)}$$

$$Light_{limit} = 2.718 \cdot \frac{Photoperiod}{Extinction \cdot Depth} \cdot \left(\exp\left(-\frac{Light}{L_m} \cdot \exp(-Extinction \cdot Depth) \right) - \exp\left(-\frac{Light}{L_m} \right) \right)$$

$$Extinction = Extinction_{water} + Extinction_{phytoplankton} + Extinction_{macrophytes} + Extinction_{DOM} + Extinction_{POM}$$

$$Extinction_{phytoplankton} = Ecoeff_{phytoplankton} \cdot Bio_{all\ phytoplankton}$$

$$Extinction_{DOM} = Ecoeff_{DOM} \cdot DOM$$

$$Extinction_{POM} = Ecoeff_{POM} \cdot POM$$

$$Light = \frac{Solar}{2}$$

$$respiration = Resp0 \cdot \exp(TempResp \cdot Temperature) \cdot Bio_{phytoplankton}$$

$$excretion = Exc \cdot photosynthesis \cdot (1 - Light_{limit})$$

$$mortality = (Mort + ExcessT + Stress) \cdot Bio_{phytoplankton}$$

$$ExcessT = \frac{\exp(Temperature - T_{max})}{2}$$

$$Stress = 1 - \exp\left(-Emort \cdot (1 - Nutr_{limit} \cdot Light_{limit})\right)$$

$$sinking = \frac{Sed}{Depth} \cdot SedAccel \cdot Bio_{phytoplankton}$$

$$SedAccel = \exp\left(ESed \cdot (1 - Light_{limit} \cdot Nutr_{limit} \cdot Temp_{limit})\right)$$

$Bio_{phytoplankton}$	biomass concentration of phytoplankton (mg L^{-1})	N_{limit}	nitrogen limitation (-)
$consumption_{zooplankton}$	consumption rate by zooplankton (mg L^{-1} d^{-1})	$Nutr_{limit}$	nutrient limitation (-)
Depth	depth of the reservoir (m)	P	phosphorus (mg L^{-1})
DOM / POM	dissolved (D) and particulate (P) organic matter (OM)	Photoperiod	fraction of the day with light (-)
$Ecoeff_i$	extinction coefficient of source i (m^{-1} mg^{-1} L)	photosynthesis	photosynthesis rate (mg L^{-1} d^{-1})
Emort	coefficient for stress-related increased mortality (-)	P_{limit}	phosphorus limitation (-)
ESed	factor for accelerated sinking (-)	PS_{limit}	photosynthesis limitation (-)
Exc	excretion / photosynthesis ratio (-)	PS_{max}	maximum photosynthesis rate (d^{-1})
ExcessT	increased mortality factor (d^{-1})	Resp0	intrinsic respiration (d^{-1})
excretion	excretion rate (mg L^{-1} d^{-1})	respiration	respiration rate (mg L^{-1} d^{-1})
Extinction	extinction of sunlight (m^{-1})	Sed	intrinsic sinking or sedimentation rate (m d^{-1})
$Extinction_i$	light extinction by source i (m^{-1})	SedAccel	accelerated sedimentation (-)
K_N	constant for nitrogen limitation (mg L^{-1})	sinking	sinking rate (mg L^{-1} d^{-1})
K_P	constant for phosphorus limitation (mg L^{-1})	Solar	light intensity (cal m^{-2} day^{-1})
Light	light intensity (cal m^{-2} day^{-1})	Stress	increased mortality resulting from stress (d^{-1})
$Light_{limit}$	light limitation (-)	Temperature	water temperature (°C)
L_m	optimal light intensity (cal m^{-2} day^{-1})	$Temp_{limit}$	temperature limitation (-)
Mort	intrinsic mortality (d^{-1})	TempResp	coefficient for increased respiration ($°C^{-1}$)
mortality	mortality rate (mg L^{-1} d^{-1})	T_{max}	maximum water temperature for photosynthesis (°C)
N	nitrogen (mg L^{-1})		

Fig. 1. (a) Equations of the object describing phytoplankton growth (upper panel) and a list of used parameters and variables (lower panel). (b) Equations of the object describing zooplankton growth (upper panel) and a list of used parameters and variables (lower panel). (c) Equations of the object describing organic matter and nutrient cycling (upper panel) and a list of used parameters and variables (lower panel).

b

zooplankton equations

$$\frac{dBio_{zooplankton}}{dt} = consumption - defecation - respiration - excretion - mortality - predation$$

$$consumption = consumption_{phytoplankton} + consumption_{POM}$$

$$consumption_{phytoplankton} = \sum_{i=1}^{p} consumption_{phytoplankton_i}$$

$$consumption_{phytoplankton_i} = C_{max} \cdot SatFeeding_{phytoplankton_i} \cdot Temp_{limit} \cdot Bio_{zooplankton}$$

$$SatFeeding_{phytoplankton_i} = Pref_{phytoplankton_i} \cdot \frac{Food_{phytoplankton_i}}{(Helping\ variable + FHalfSat_{phytoplankton_i})}$$

$$Food_{phytoplankton_i} = Bio_{phytoplankton_i} - MinBio_{phytoplankton_i}$$

$$Helping\ variable = \sum_{i=1}^{p} Food_{phytoplankton_i} \cdot Pref_{phytoplankton_i} + Pref_{POM} \cdot Food_{POM}$$

$$Food_{POM} = POM - MinPOM$$

$$defecation = defecation_{phytoplankton} + defecation_{POM}$$

$$defecation_{phytoplankton} = \sum_{i=1}^{p} defecation_{phytoplankton_i}$$

$$defecation_{phytoplankton_i} = EgestionCoeff_{phytoplankton_i} \cdot consumption_{phytoplankton_i}$$

$$defecation_{POM} = EgestCoeff_{POM} \cdot consumption_{POM}$$

$$respiration = StandardRespiration + DynamicAction$$

$$StandardRespiration = Resp0 \cdot Temp_{limit} \cdot Bio_{zooplankton}$$

$$DynamicAction = Resp \cdot (consumption - defecation)$$

$$excretion = Excr \cdot respiration$$

$$mortality = Mort \cdot Bio_{zooplankton} + \exp\frac{(Temperature - T_{max})}{2} \cdot Bio_{zooplankton}$$

$Bio_{phytoplankton\ i}$	biomass concentration of phytoplankton population i (mg L^{-1})
$Bio_{zooplankton}$	zooplankton biomass concentration (mg L^{-1})
C_{max}	maximum ingestion rate for given zooplankton (d^{-1})
consumption	consumption of resources (mg L^{-1} d^{-1})
$consumption_{resource\ i}$	consumption of resource i (mg L^{-1} d^{-1})
defecation	defecation of ingested resources (mg L^{-1} d^{-1})
$defecation_{resource\ i}$	defecation of resource i (mg L^{-1} d^{-1})
DynamicAction	the additional cost for the processing of consumed resources (mg L^{-1} d^{-1})
$EgestionCoeff_{resource\ i}$	fraction of consumed resource i lost through egestion (-)
Excr	constant relationship between excretion and respiration (-)
excretion	excretion of dissolved organic matter (mg L^{-1} d^{-1})
$FHalfSat_{resource\ i}$	half saturation constant for consumption of resource i (mg L^{-1})
$Food_{resource\ i}$	concentration of resource which is available for consumption by zooplankton (mg L^{-1})
Helping variable	helping variable (mg L^{-1})
$MinBio_{resource\ i}$	minimum resource concentration at which given consumer begins consuming (mg L^{-1})
MinPOM	minimum POM concentration to begin feeding (mg L^{-1})
Mort	intrinsic mortality (d^{-1})
mortality	intrinsic mortality (mg L^{-1} d^{-1})
POM	particulate organic matter concentration (mg L^{-1})
predation	consumption of zooplankton by planktivorous fish (mg L^{-1} d^{-1})
$Pref_{resource\ i}$	preference of given consumer for resource i (-)
Resp	fraction of energy lost to dynamic action (-)
Resp0	intrinsic respiration (d^{-1})
respiration	respiratory losses (mg L^{-1} d^{-1})
$SatFeeding_{resource\ i}$	kinetic factor to express feeding saturation (-)
StandardRespiration	standard respiration, i.e. when the organisms in a population are in a resting state (mg L^{-1} d^{-1})
Temperature	water temperature (°C)
$Temp_{limit}$	limitation due to suboptimal water temperature (-)
T_{max}	maximum temperature which is tolerated by the considered zooplankton population (°C)

Fig. 1 (*continued*)

c

organic matter and nutrient equations

$$\frac{dDOM}{dt} = excretion_{phytoplankton\ and\ macrophyte} + excretion_{zooplankton\ and\ fish} - decomposition_{DOM}$$

$$decomposition_{DOM} = DecayMax_{DOM} \cdot Temp_{corr} \cdot DOM$$

$$\frac{dPOM}{dt} = mortality_{phytoplankton\ and\ macrophyte} + mortality_{zooplankton\ and\ fish} - decomposition_{POM} - sedimentation_{POM} - consumption_{POM}$$

$$decomposition_{POM} = DecayMax_{POM} \cdot Temp_{corr} \cdot POM$$

$$sedimentation_{POM} = \frac{Sed}{Depth} \cdot POM$$

$$\frac{dSOM}{dt} = defecation + sedimentation_{POM} - decomposition_{SOM} + sinking_{phytoplankton}$$

$$decomposition_{SOM} = DecayMax_{SOM} \cdot Temp_{corr} \cdot SOM$$

$$\frac{dNH_3\text{-}N}{dt} = (decomposition_{DOM} + decomposition_{POM} + decomposition_{SOM} + respiration_{phytoplankton\ and\ macrophyte} + respiration_{zooplankton\ and\ fish}) \cdot Org2Ammonia - nitrification - NH_3\text{-}N\ assimilation_{phytoplankton\ and\ macrophyte}$$

$$nitrification = K_{nitri} \cdot \frac{Area}{Volume} \cdot Temp_{corr} \cdot NH_3\text{-}N$$

$$NH_3\text{-}N\ assimilation_{phytoplankton\ and\ macrophyte} = (photosynthesis_{phytoplankton\ and\ macrophyte}) \cdot Org2Ammonia$$

$$\frac{dNO_3\text{-}N}{dt} = nitrification - denitrification$$

$$denitrification = K_{denitri} \cdot \frac{Area}{Volume} \cdot Temp_{limit} \cdot NO_3\text{-}N$$

$$\frac{dPO_4\text{-}P}{dt} = (decomposition_{DOM} + decomposition_{POM} + decomposition_{SOM} + respiration_{phytoplankton\ and\ macrophyte} + respiration_{zooplankton\ and\ fish}) \cdot Org2Phos - PO_4\text{-}P\ assimilation_{phytoplankton\ and\ macrophyte}$$

$$PO_4\text{-}P\ assimilation_{phytoplankton\ and\ macrophyte} = (photosynthesis_{phytoplankton\ and\ macrophyte}) \cdot Org2Phos$$

Area	surface area of reservoir (m^2)
$consumption_{POM}$	consumption of POM by all zooplankton (mg L^{-1} d^{-1})
$DecayMax_{DOM}$	maximum rate of DOM conversion to nutrients (d^{-1})
$DecayMax_{POM}$	maximum rate of POM conversion to nutrients (d^{-1})
$DecayMax_{SOM}$	maximum rate of SOM conversion to nutrients (d^{-1})
$decomposition_{DOM}$	conversion of DOM to nutrients (mg L^{-1} d^{-1})
$decomposition_{POM}$	decomposition of POM (mg L^{-1} d^{-1})
$decomposition_{SOM}$	decomposition of SOM (mg L^{-1} d^{-1})
defecation	defecation of all zooplankton and fish (mg L^{-1} d^{-1})
denitrification	conversion of NO_3-N to N_2 (mg L^{-1} d^{-1})
Depth	depth of the reservoir (m)
DOM	dissolved organic matter (mg L^{-1})
$excretion_{phytoplankton\ and\ macrophyte}$	excretion (mg L^{-1} d^{-1})
$excretion_{zooplankton\ and\ fish}$	excretion (mg L^{-1} d^{-1})
$K_{denitri}$	maximum rate of nitrification (m d^{-1})
K_{nitri}	maximum rate of nitrification (m d^{-1})
$mortality_{phytoplankton\ and\ macrophyte}$	mortality (mg L^{-1} d^{-1})
$mortality_{zooplankton\ and\ fish}$	mortality of all zooplankton and fish (mg L^{-1} d^{-1})
NH_3-N	ammonia-nitrogen (mg L^{-1})
NH_3-N $assimilation_{phytoplankton\ and\ macrophyte}$	assimilation of NH_3-N (mg L^{-1} d^{-1})
nitrification	conversion of NH_3-N to NO_3-N (mg L^{-1} d^{-1})
NO_3-N	nitrate-nitrogen (mg L^{-1})
Org2Ammonia	default conversion factor between organic matter and NH_3-N (-)
Org2Phos	default conversion factor between organic matter and PO_4-P (-)
$photosynthesis_{phytoplankton\ and\ macrophyte}$	photosynthesis (mg L^{-1} d^{-1})
PO_4-P	phospate-phosphorus (mg L^{-1})
PO_4-P $assimilation_{phytoplankton\ and\ macrophyte}$	assimilation of PO_4-P (mg L^{-1} d^{-1})
POM	particulate organic matter
$respiration_{phytoplankton\ and\ macrophyte}$	respiration (mg L^{-1} d^{-1})
$respiration_{zooplankton\ and\ fish}$	respiration (mg L^{-1} d^{-1})
Sed	sedimentation velocity rate (m d^{-1})
$sedimentation_{POM}$	sedimentation of POM (mg L^{-1} d^{-1})
$sinking_{phytoplankton}$	sinking of phytoplankton (mg L^{-1} d^{-1})
SOM	settled organic matter
$Temp_{corr}$	temperature correction
Volume	volume of water in reservoir (m^3)

Fig. 1 (*continued*)

Table 1
Overview of the mean of the used, log transformed toxicity data ($\mu g\,l^{-1}$) in the model to represent model population sensitivities in the different studies

No.	Spring		Summer		Macro	Small		Large		Fish		Toxicant	References
	Phyto1	Phyto2	Phyto1	Phyto2		Zoo1	Zoo2	Zoo1	Zoo2	Plank.	Pisc.		
1	–		–			1.48[a]		4[a]	0.71	4.95	4.95	Diflubenzuron	Miura and Takahashi (1974), Julin and Sanders (1978), Hansen and Garton (1982), Mayer and Ellersieck (1986), Liber et al. (1994), USEPA (2000a,b)
2	2.52	2.13	2.41			–		–				Atrazine	Kallqvist and Romstad (1994), Kotrikla et al. (1997), Carrasco and Sabater (1997), Tang et al. (1997), Berard et al. (1999), Rojickova-Padrtova and Marsalek (1999), Benhra et al. (1997), Okamura et al. (2000)
3	–		–			−0.05[b]		−0.05[c]		0.28[c]		Esfenvalerate	Fairchild et al. (1992)
4	–		–			−0.05[b]		−0.05[c]		0.28[c]		Esfenvalerate	Fairchild et al. (1992)
5	1.01		1.01	2.74	1.32	–	–	–				Metribuzin	Fairchild et al. (1994), Fairchild et al. (1998), USEPA (2000a,b)
6	–		–			4.93[b]		4.93	0.25			Azinphos-methyl	Dortland (1980), Guzzella et al. (1997), USEPA (2000a,b)
7	–		–			3.85		3.60	0.16			Fenthion	Roux et al. (1995), Kaur and Ansal (1996)
8	–		–			4.93[b]		4.93	0.25	0.68-0.74		Azinphos-methyl	Dortland (1980), Guzzella et al. (1997), USEPA (2000a,b)
9	2.52	2.13	2.41			–		–				Atrazine	Kallqvist and Romstad (1994), Kotrikla et al. (1997), Carrasco and Sabater (1997), Tang et al. (1997), Berard et al. (1999), Rojickova-Padrtova and Marsalek (1999), Benhra et al. (1997), Okamura et al. (2000)
10	2.70		1.81			–		–		–	–	Atrazine	Kallqvist and Romstad (1994), Kotrikla et al. (1997), Carrasco and Sabater (1997), Tang et al. (1997), Berard et al. (1999), Rojickova-Padrtova and Marsalek (1999), Benhra et al. (1997), Okamura et al. (2000)
11	1.14		1.59	1.59	0.40	–		–	–			Linuron	USEPA (2000a,b)

Phytoplankton ('phyto') and macrophyte values are growth-EC_{50}'s; the other values are LC_{50}'s. With 'plankt. fish', 'pisc. fish', and 'zoo', planktivorous fish, piscivorous fish, and zooplankton are indicated. '–' indicates that no proof of effects of the considered toxicant at the tested concentration range was found. Hence, these populations were assumed to be not directly affected by the toxicant in question. All $L(E)C_x$ values were characterized by a lognormal distribution. A default of 10% was arbitrarily chosen. In the case of planktivorous fish in study 8, a uniform distribution, of which the upper and lower limits are given, was chosen to represent uncertainty. Slope values were derived from: $L(E)C_{50} \times L(E)C_5 - 1 = \exp(1.6449 \times Sm)$; and $slope = \ln(5 \times 95 - 1) \times (\ln(L(E)C_5) - \ln(L(E)C_{50})) - 1$ (Smit et al., 2001). Sm was characterized by a uniform distribution between 0.75 and 2, 0.45 and 0.7, 0.25 and 0.4 for phytoplankton, zooplankton, and fish, respectively (Smit et al., 2001).

(Sommer et al., 1986): (1) a bloom of spring phytoplankton, (2) a bloom of small zooplankton, resulting in a 'clear water phase', (3) a bloom of summer phytoplankton, followed by (4) a bloom of larger zooplankton and (5) a biomass increase of fish. As a compromise between data availability and experiment duration, a 40 day experiment duration was taken as a cut-off value for inclusion in this analysis. The selected studies represent a wide range of different ecosystems, i.e. from relatively simple planktonic systems to systems which include planktivorous and piscivorous fish Considered studies are: Boyle et al. (1996; study 1), Hamilton et al. (1988; study 2), Webber et al. (1992; study 3), Fairchild et al. (1992; study 4), Brock et al. (2004; study 5), Sierszen and Lozano (1998; study 6), Hanazato and Kasai (1995; study 7), Tanner and Knuth (1995; study 8), Juttner et al. (1995; study 9), Denoyelles

Table 2
Result of species grouping into model populations where "ns" stands for "not specified" and "[a]" denotes that the considered population contains all phytoplankton or zooplankton species not included in the other model phytoplankton or zooplankton populations, respectively

No.	Spring		Summer		Macro	Small		Large		Fish	
	Phyto1	Phyto2	Phyto1	Phyto2		Zoo1	Zoo2	Zoo1	Zoo2	Plank.	Pisc.
1	ns		ns			Rotifers		Copepods	Cladocerans	Bluegill	Bass
2	Diatoms	Chryptophyta Chrysophyta	Dinoflagellates Chlorophyta			Rotifers		Copepods Cladocerans			
3	ns		ns			Copepod nauplii Rotifers		Cladocerans Copepods		Bluegill	
4	ns		ns			Rotifers		Copepods Cladocerans		Bluegill	
5	*Fragilaria ulna*		*Gomphonema* sp.	*Anabaena cylindrica*	*Myriophillum spicatum*	*Chydorus sphaericus* *Lecane* sp. *Mytilana ventralis* *Polyarthra remata*	*Polyarthra remata* *Trichocerca*	Other[a]			
6	ns		ns			Rotifers		Copepods	Cladocerans		
7	Cryptophyceae Centrales		Chlorococcales Volvocales			Rotifers		Copepods	Cladocerans		
8	ns		ns			Rotifers		Copepods	Cladocerans	Bluegill	
9	Diatoms	Cryptophyceae	Other[a]			Copepod nauplii Rotifers		Cladocerans Copepods			
10	*Cryptomonas* sp. *Mallomonas* sp.		*Peridinium* sp.			Rotifers		Cladocerans Copepods		Bluegill	Bass
11	*Cocconeis*		*Chlamydomonas*	*Phormidium foveolarum*	*Elodea nuttallii*	Rotifers		Copepods	Cladocerans		

Table 3A
Parameter values which are constant in all the constructed ecosystem models

	Spring phyto	Summer phyto	Macrophyte	Reference
Phytoplankton				
Emort	0.04	0.04	0.01	USEPA (2000a)
ESed	1.1	1.1		Wetzel (2001)
KT	5	5	5	Kitchell et al. (1972)
Q10	2	2	2	DeNicola (1996)
Resp0	0.02	0.01	0.01	Hewett and Johnson (1992), USEPA (2000a), Collins and Wlosinski (1983)
Sed	0.15	0.15	0.15	Collins and Wlosinski (1983)
TempResp	0.065	0.065	0.065	USEPA (2000a)
T_{max}	30	40	40	Collins and Wlosinski (1983)
T_{opt}	8	20	20	Collins and Wlosinski (1983)
T_{ref}	2	10	10	Collins and Wlosinski (1983)
XM	5	5	5	Kitchell et al. (1972)

	Small zoo	Large zoo	Plank. fish	Pisc. fish	
Zooplankton and fish					
Excr	0.17	0.17	0.05	0.05	Scavia and Park (1976)
KT	5	5	5	5	Kitchell et al. (1972)
$MinBio_{resource\ i}$	0.05	0.05	0.1	0.25	USEPA (2000a), Walz (1995)
$Pref_{resource\ i}$	~	~	~	~	
Q10	2	2	2	2	DeNicola (1996)
Resp	0.25	0.18	0.172	0.172	Hewett and Johnson (1992), USEPA (2000a)
Resp0	0.03	0.014	0.04	0.04	Hewett and Johnson (1992), USEPA (2000a) Collins and Wlosinski (1983)
TempResp	0.065	0.065	0.065	0.065	USEPA (2000a)
T_{max}	34	34	36	36	Collins and Wlosinski (1983)
T_{opt}	26	26	27	27	Collins and Wlosinski (1983)
T_{ref}	5	5	2.5	2.5	Collins and Wlosinski (1983)
XM	5	5	5	5	Kitchell et al. (1972)

	Value	
OM and nutrient cycling		
Area/volume	1	#
$DecayMax_{DOM}$	0.29	USEPA (2000a)
$DecayMax_{POM}$	0.29	USEPA (2000a)
$DecayMax_{SOM}$	0.04	USEPA (2000a)
Depth	1	Micro- or mesocosm
$Ecoeff_{DOM}$	0.03	USEPA (2000a)
$Ecoeff_{macrophytes}$	0.05	Le Cren and Lowe-McConnell (1980)
$Ecoeff_{phytoplankton}$	0.014	Collins and Wlosinski (1983)
$Ecoeff_{POM}$	0.12	Verduin (1982)
$Extinction_{water}$	0.016	Wetzel (1975)
$K_{denitri}$	0.1	Di Toro (2001)
K_{nitri}	0.135	Effler (1996)
K_O	0.1	Bowie et al. (1985)
KT	5	Kitchell et al. (1972)
Org2Ammonia	0.079	Redfield (1958)
Org2Phos	0.018	Redfield (1958)
pH_{max}	8.5	Lyman et al. (1982)
pH_{min}	5	Lyman et al. (1982)
Q10	2	DeNicola (1996)
Sed	0.15	Collins and Wlosinski (1983)
T_{max}	60	Collins and Wlosinski (1983)
T_{obs}	25	Collins and Wlosinski (1983)

"#" denotes that a constant area/volume ratio of 1 was used in all exercises. "~" denotes that these values are given in the text of this paper.

et al. (1982; study 10), Cuppen et al. (1997; study 11); van den Brink et al. (1997; study 11). To account for the species present in the experimental ecosystems, a different ecosystem model was constructed for each of the considered studies.

2.3. Toxic effect sub-model

Given their high accuracy in predicting population- and ecosystem-NOECs in a previous exercise (De Laender et al., in press), ecosystem models were equipped with logistic

Table 3B
Parameters receiving a different value per study

	Spring phyto1	Summer phyto1	Small zoo1	Large zoo1	Large zoo2	Plank. fish	Pisc. fish
Parameter in study 1							
C_{max}			4	1.8	1.8	1.3	1.2
$EgestionCoeff_{resource\ i}$		0.2	0.2	0.2	0.1	0.1	
Exc	0.025	0.02					
$FHalfSat_{resource\ i}$			0.5	1	1	5	5
K_N	0.05	0.002					
K_P	0.01	0.002					
L_m	48	100					
Mort	0.06	0.02	0.06	0.03	0.03	0.0001	0.001
PS_{max}	3.8	1.8					

	Spring phyto1	Spring phyto2	Summer phyto1	Small zoo1	Large zoo1
Parameter in studies 2 and 9					
C_{max}				4	1.8
$EgestionCoeff_{resource\ i}$			0.2	0.2	
Exc	0.025	0.025	0.02		
$FHalfSat_{resource\ i}$				0.5	1
K_N	0.05	0.05	0.002		
K_P	0.01	0.01	0.002		
L_m	50	50	100		
Mort	0.06	0.06	0.02	0.06	0.03
PS_{max}	4	4	1.8		

	Spring phyto1	Summer phyto1	Small zoo1	Large zoo1	Plank. fish
Parameter in studies 3 and 4					
C_{max}			4	1.8	1.2
$EgestionCoeff_{resource\ i}$		0.2	0.2	0.1	
Exc	0.025	0.02			
$FHalfSat_{resource\ i}$			0.5	1	5
K_N	0.05	0.002			
K_P	0.01	0.002			
L_m	50	100			
Mort	0.06	0.02	0.04	0.03	0.0001
PS_{max}	4	1.8			

	Spring phyto1	Summer phyto1	Summer phyto2	Macro	Small zoo1	Small zoo2	Large zoo1
Parameter in study 5							
C_{max}					4	4	1.8
$EgestionCoeff_{resource\ i}$				0.2	0.2	0.2	
Exc	0.025	0.02	0.02	0.4			
$FHalfSat_{resource\ i}$					0.5	0.5	1
K_N	0.02	0.002	0.002	0.002			
K_P	0.005	0.002	0.002	0.002			
L_m	48	100	100	100			
Mort	0.06	0.02	0.02	0.001	0.06	0.06	0.03
PS_{max}	3.8	1.8	1.8	0.2			

	Spring phyto1	Summer phyto1	Small zoo1	Large zoo1	Large zoo2
Parameter in study 6 and 7					
C_{max}			4	1.8	1.8
$EgestionCoeff_{resource\ i}$		0.2	0.2	0.2	
Exc	0.025	0.02			
$FHalfSat_{resource\ i}$			0.5	1	1
K_N	0.05	0.002			
K_P	0.01	0.002			
L_m	48	100			
Mort	0.06	0.02	0.04	0.03	0.03
PS_{max}	3.8	1.8			
PS_{max}	3.8	1.8			

(*continued on next page*)

Table 3B (*continued*)

	Spring phyto1	Summer phyto1	Small zoo1	Large zoo1	Large zoo2	Plank. fish
Parameter in study 8						
C_{max}			4	1.8	1.8	1.2
$EgestionCoeff_{resource\ i}$		0.2	0.2	0.2	0.1	
Exc	0.025	0.02				
$FHalfSat_{resource\ i}$			0.5	1	1	5
K_N	0.05	0.002				
K_P	0.01	0.002				
L_m	48	100				
Mort	0.06	0.02	0.04	0.03	0.03	0.0001
PS_{max}	3.8	1.8				

	Spring phyto1	Summer phyto1	Small zoo1	Large zoo1	Plank. fish	Pisc. fish
Parameter in study 10						
C_{max}			4	1.8	1.1	0.1
$EgestionCoeff_{resource\ i}$		0.2	0.2	0.1	0.1	
Exc	0.025	0.02				
$FHalfSat_{resource\ i}$			0.5	1	0.25	0.25
K_N	0.05	0.002				
K_P	0.01	0.002				
L_m	48	100				
Mort	0.06	0.02	0.04	0.03	0.0001	0.008
PS_{max}	4.5	1.5				

	Spring phyto1	Summer phyto1	Summer phyto2	Macrophyte	Small zoo1	Large zoo1	Large zoo2
Parameter in study 11							
C_{max}					4	1.8	1.8
$EgestionCoeff_{resource\ i}$				0.2	0.2	0.2	
Exc	0.025	0.02	0.02	0.3			
$FHalfSat_{resource\ i}$					0.5	1	1
K_N	0.05	0.002	0.002	0.002			
K_P	0.01	0.002	0.002	0.002			
L_m	48	100	100	100			
Mort	0.06	0.02	0.02	0.001	0.04	0.03	0.03
PS_{max}	4.5	1.5	1.5	0.2			

Note that these values are the result of the qualitative calibration discussed in the text of this paper.

concentration-effect functions as toxic effect sub-models. The ecosystem model uses these sub-models to calculate photosynthesis rate (PS_{max}) of phytoplankton, and mortality rate (Mort) of zooplankton and fish as a function of toxicant concentration 'tox'. At every time step, the value of 'tox' is read from the ecosystem model input file and used to calculate PS_{max} and Mort.

$$PS_{max} = \frac{PS_{max,0}}{1 + \left(\frac{tox}{EC_{50,PS_{max}}}\right)^{slope}} \quad (1)$$

$$Mort = \frac{1}{time} \cdot \ln\left(1 + \left(\frac{tox}{LC_{50}}\right)^{slope}\right) \quad (2)$$

with LC_{50} is the lethal concentration for 50% of the tested population, as established in a single-species test ($\mu g\,l^{-1}$), time is duration of that single-species test in which the LC_{50} was derived (d), EC_{50} is the concentration resulting in a 50% reduction of the photosynthesis rate of the tested phytoplankton population, established in a single-species test ($\mu g\,l^{-1}$), slope is slope of the considered concentration-effect function (–).

Appropriate values for those parameters were collected from literature (Table 1). In case no EC_{50} for photosynthesis inhibition was available, an EC_{50} for phytoplankton growth inhibition was taken.

2.4. Ecosystem model simulations – species grouping

Consistent with the seasonal events described by Sommer et al. (1986), species were grouped into one of the following model populations: spring phytoplankton, summer phytoplankton, small zooplankton, large zooplankton, planktivorous fish, piscivorous fish, macrophytes. Grouping was based on their single-species sensitivity for the considered chemical and their feeding characteristics. Large bodied cladocerans and copepods were both categorized as 'large zooplankton', rotifers and small cladocerans as 'small zooplankton'. In cases where the single-species sensitivity of cladocerans and copepods is different, they were modelled as two separate model populations. The cut-off value for body size of zooplankton was 0.7 mm. Phytoplankton species were modelled as spring or summer populations, depending on (1) the species given in Sommer

et al. (1986) or (2) on the reported dynamics in the paper of the considered study. In Table 2, the result of this grouping into model populations is given.

2.5. Ecosystem model simulations – parameterization

Parameters for which the default values from USEPA (2000a) were used are given in Table 3A. The ecosystem model for every study was qualitatively calibrated to obtain a plausible succession of planktonic events as described above by adjusting the parameters given in Table 3B. During this calibration, parameter values were constrained in several ways. Changes larger than 20% from default values proposed by USEPA (2000a) were not allowed. Maximum photosynthesis rates (i.e. PS_{max}) of spring phytoplankton were set higher than those of summer phytoplankton (Knisely and Geller, 1986; Muller and Schlegel, 1999). Saturation constants K_P and K_N were set lower for summer phytoplankton species than for spring phytoplankton species to account for the competitive advantage summer phytoplankton species have to grow in low-nutrient conditions (Sommer et al., 1986). Large zooplankton populations and fish have slower growth kinetics than small zooplankton populations, i.e. lower ingestion and mortality rates (Collins and Wlosinski, 1983; Leidy and Ploskey, 1983).

Planktivorous fish were assumed to exclusively feed on large-bodied zooplankton (Sommer et al., 1986). Large zooplankton was assumed to feed on spring and summer phytoplankton (Sommer et al., 1986). Small zooplankton was assumed to feed on spring phytoplankton only. Indeed, summer phytoplankton is often colony-forming or large-celled, which renders them unsuitable for ingestion by small zooplankton (Sommer et al., 1986). To model these interactions, preference factors of small zooplankton for spring phytoplankton were set to 1, i.e. excluding ingestion of summer phytoplankton. Preference factors of large zooplankton were set to 0.5 and 0.5 for summer and spring

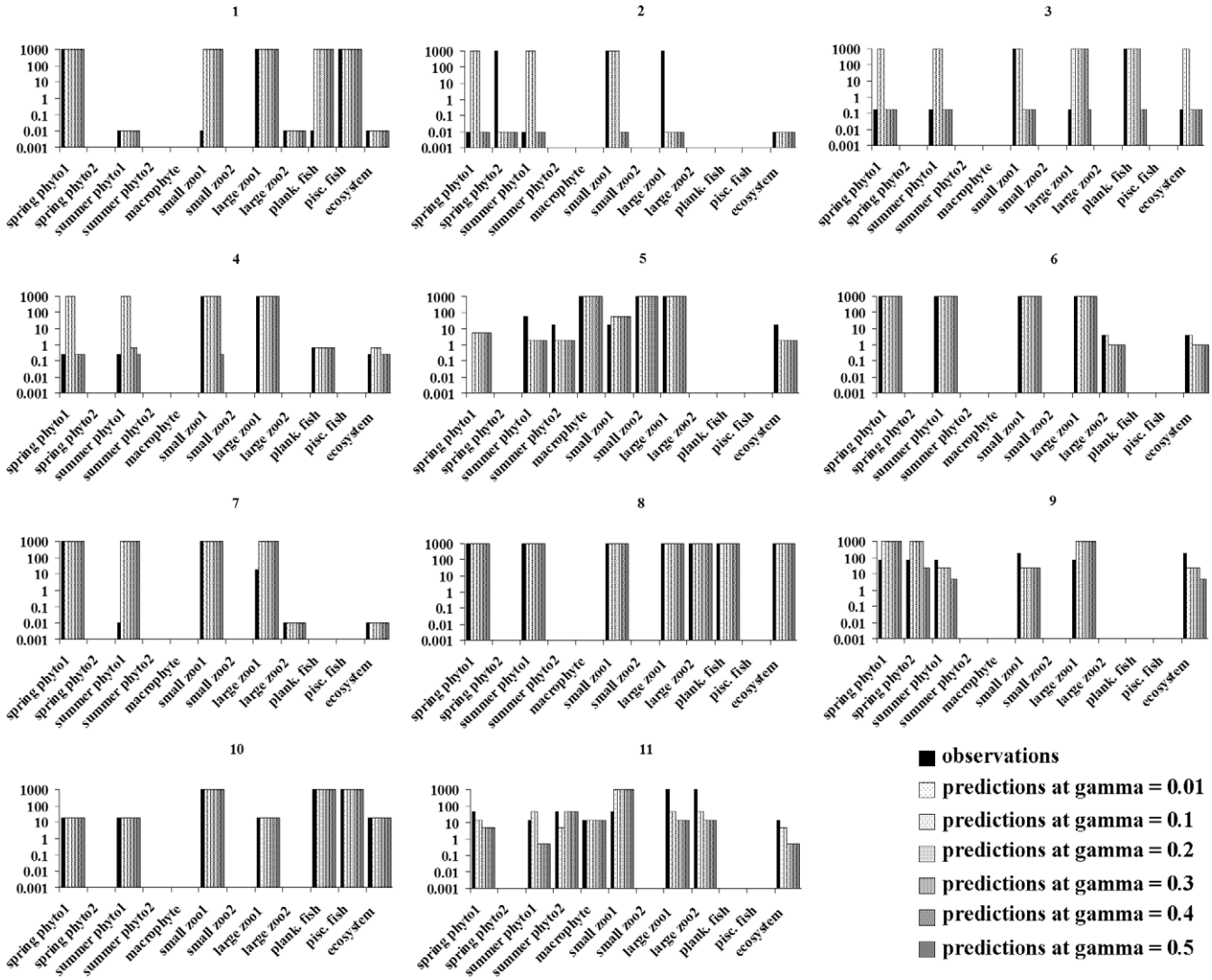

Fig. 2. Observed NOECs and NOEC-predictions at different γ-values. * The NOEC of $<1.8\ \mu g\ l^{-1}$ for *Fragilaria ulna* was not included because of the low abundance of this species. Moreover, this NOEC was explicitly reported as unreliable by Brock et al. (2004).

phytoplankton. Preference factors of planktivorous fish for large zooplankton was set to 1, i.e. excluding ingestion of small zooplankton. The preference factor of piscivorous fish was set to 1 for planctivorous fish.

By setting the constraints on parameter values and the ecological interactions as described above, a qualitative calibration was performed using initial conditions of 0.01, 0.005, 0.001 mg l^{-1} for phytoplankton, zooplankton, and

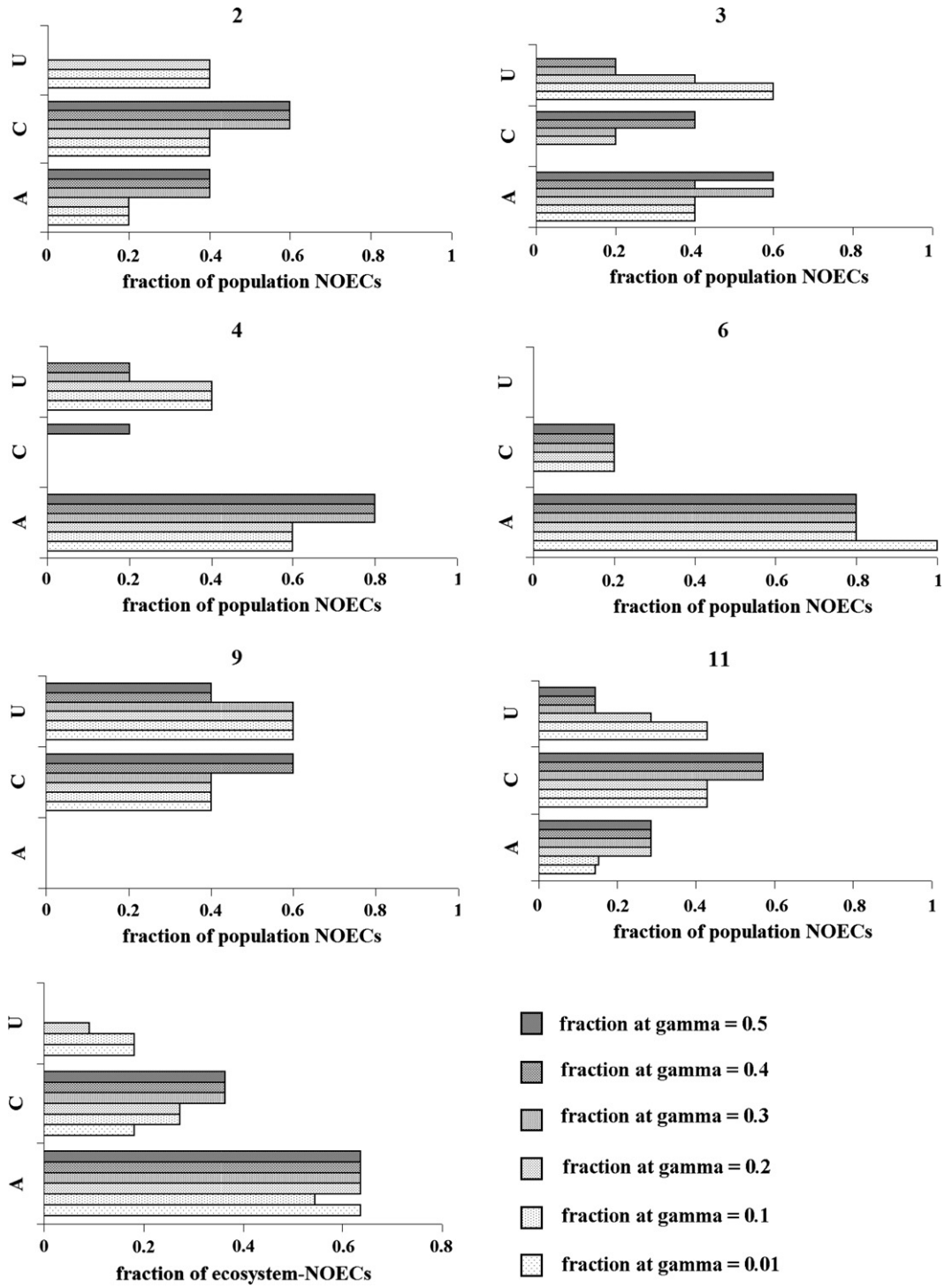

Fig. 3. (Upper 6 panels) Model performance in predicting population-level no observed effect concentrations in 6 different studies (2, 3, 4, 6, 9, 11) at gamma-levels ranging from 0.01 to 0.5. Performance is expressed as the fraction of model populations for which the NOEC was predicted accurately (A), conservatively (C), and underprotective (U). Population-NOECs in the other 5 studies were independent of gamma and are given in the text. (lower panel) Model performance in predicting ecosystem-level no observed effect concentrations at gamma-levels ranging from 0.01 to 0.5. Performance is expressed as the fraction of accurate, conservative, and underprotective ecosystem-NOECs amongst the 11 studies.

fish, respectively. In Fig 1, supporting documents, an example is shown of the dynamics of a system with phytoplankton, zooplankton, and fish, after qualitative calibration. Parameter values resulting in such dynamics are listed per study in Table 3B. The as such obtained dynamics are used as the reference dynamics.

We then simulated the exposure of the customized ecosystems to the same toxicant concentrations as those used in the respective experimental study. Starting date, exposure concentration and duration, and number of exposure events in the simulations were identical to those reported in the respective large-scale experiments (Table 1, supporting document). To compare a model populations' biomass in a control treatment with that at different exposure concentrations, its biomass concentration in both treatments was averaged over the complete exposure period. The relative difference (RD) between control and treatment population's biomass was calculated by

$$\mathrm{RD}_{\mathrm{tox},i} = \frac{X_{\mathrm{tox},i} - X_{\mathrm{ref},i}}{X_{\mathrm{ref},i}} \quad (3)$$

with $\mathrm{RD}_{\mathrm{tox},i}$ the relative difference of the average biomass concentration of population '*i*', when exposed to a toxicant concentration 'tox', with its biomass concentration in the control treatment, $X_{\mathrm{tox},i}$ is the time-averaged biomass concentration of population *i*, when exposed to a toxicant concentration 'tox', $X_{\mathrm{ref},i}$ is the time-averaged biomass concentration of population '*i*' in the control treatment, i.e. the reference value.

RD-values were calculated for each population at every toxicant concentration. As 20% has been suggested as the minimum detectable difference in population characteristics in the field (Suter, 1993), a RD-value of −0.2 or lower is considered as an observable decrease of a population. Similarly, a RD-value of 0.2 or higher can be considered as an observable increase of a population biomass. In the context of EEA, both increases and decreases of phytoplankton biomass are considered undesirable. For macrophytes, invertebrates, and fish, only biomass decreases are considered as undesirable.

2.6. Derivation of predicted population-no observed effect concentrations (NOECs)

To account for variability of the used toxicity data (Table1), the customized ecosystem models were run in a Monte-Carlo setting (Cullen and Frey, 1999). Characteristics of the statistical distributions describing this variability are also given in Table1. Using latin hypersquare sampling, 100 simulations per concentration were run. The number of runs (100) was determined by the rule of convergence (Melching, 1995). Each of these 100 simulations was compared with its reference simulation, i.e. with the control treatment, yielding 100 values of RD per model population and exposure concentration. From these RD-values, one NOEC was derived per considered population. The highest exposure concentration at which less than $100\,(1-\gamma)$ % of the simulated RD-values were smaller than −0.2 was defined as the NOEC_{γ} for decrease. Similarly, the highest concentration at which less than $100\cdot(1-\gamma)$ % of the simulated RD values were larger than 0.2 was defined as the NOEC_{γ} for increase. The influence of the γ-level was investigated for γ-values between 0.01 and 0.5. All NOECs on a population level are termed 'population-NOEC' in the rest of this paper.

Predicted population-NOECs were compared with those observed in the 11 experimental studies. Only NOECs describing effects on populations biomass or abundance were considered. Other reported NOECs (e.g. number of offspring for fish) were not included. Because NOECs were not always provided as such in the 11 considered studies, they had to be derived based on the reported results. In case NOECs were derived per sampling date, the NOEC with the highest frequency of occurrence was taken. If the frequency of occurrence was equal for two or more NOEC-values, the lowest value was chosen. The same strategy was followed if species belonging to the same model population had different NOECs. In study 5, the NOEC of $<1.8\ \mu g\ l^{-1}$ for *Fragilaria ulna* was not accounted for because of the low abundance of this species. Moreover, this NOEC was reported as unreliable by Brock et al. (2004). A detailed overview of the effects occurring in the considered studies can be found in the original research papers. An overview of the main effects is also provided in the supporting document.

In Section 3, predicted population-NOECs which are higher than, equal to, or lower than observed population-NOECs, will be termed 'underprotective', 'accurate', and 'conservative', respectively.

2.7. Derivation of predicted ecosystem-NOECs

The ecosystem-NOEC was defined as the lowest population-NOEC. As such, it is assured that when exposing an ecosystem to a concentration equal to its NOEC, no model populations' mean biomass will be adversely affected. The used terminology (i.e. 'accurate', 'conservative', and 'underprotective') for the description of predictions, relative to observations is the same as in the case of population-NOECs.

3. Results and discussion

3.1. Predicted population-NOECs

Predicted and observed NOECs for the different model populations at $0.01 \leqslant \gamma \leqslant 0.5$ are summarized in Fig. 2. For studies 1, 5, 7, 8 and 10, predicted population-NOECs were independent of γ and were equal to the observed ones for 57, 50, 67, 100, and 100% of the model populations, respectively. When a population-NOEC is independent of γ, this means that for this population, at all concentrations, the upper simulated 50% confidence region of RD did not

encompass −0.2, nor did the lower simulated 50% confidence region encompass 0.2.

In the 6 remaining studies (2, 3, 4, 6, 9, and 11), predicted population-NOECs were up to a factor 7 lower at $\gamma = 0.5$ than at $\gamma = 0.01$. As such, the agreement with observed population-NOECs also varied with changing γ for these 6 studies (Fig 3). For 2 out of these 6 studies (study 9: Juttner et al., 1995; study 11: Cuppen et al., 1997 and Van den Brink et al., 1997) the proportion of accurate population-NOEC predictions is never higher than 30%, regardless of γ. Amongst the other 4 studies, the percentage of accurate predictions tends to increase with increasing γ, while the percentage of underprotective predictions decreases. Study 6 is the only case in which applying a low γ results in somewhat better population-NOEC predictions than applying a high γ. At $\gamma = 0.01$ and 0.1, model performance in predicting protective population-NOECs is low, as indicated by the high number of underprotective predictions for studies 3 and 9. At higher γ-values, the percentage of accurate population-NOECs in studies 2, 3, 4, 6, 9, and 11 was only marginally higher, and even slightly lower in the case of study 6. However, the percentage of underprotective population-NOECs decreased at increasing γ, resulting in a higher proportion of conservative population-NOECs. This conservatism can be regarded as the consequence of reducing the number of underprotective population-NOECs.

In Hanratty and Liber (1996), the use of an LC_{50} estimate for *Daphnia magna* of 4.5 $\mu g\,l^{-1}$ to represent cladoceran sensitivity is claimed as the reason for the disagreement between predicted and observed population-level effects of diflubenzuron. In the present paper, the poor population-NOEC predictions by the model in studies 9 (Juttner et al., 1995) and 11 (Cuppen et al., 1997; Van den Brink et al., 1997) did not seem to originate solely from the less representative toxicity data. For study 9, an EC_{50} of a green algae assemblage was used to represent the sensitivity of all phytoplankton not included in the diatom and Cryptophyceae model populations. In study 11, the sensitivity of the summer phytoplankton was represented by a single EC_{50} of blue-green algae. However, a field-derived LC_{50} had to be used for small zooplankton in absence of laboratory-derived LC_{50}s in study 1, and in studies 3, 4, 6, and 8, the LC_{50} of small zooplankton was assumed equal to that of large zooplankton. Still, 57, 60, 80, 80, and 100% of population-NOECs were predicted accurately (at $\gamma = 0.5$) in study 1, 3, 4, 6, and 8, respectively.

Next to the used single-species toxicity data, the aggregation of species into model populations within the model is equally important. An example of this can be found in Traas et al. (2004b), where cladocerans and copepods are modelled as one population, despite their different sensitivity for chlorpyrifos, the chemical evaluated. These authors acknowledge that this aggregation results in poor predictions of the effects on this aggregated model population.

Another possible source of disagreement between model predictions and observed NOECs may be type II errors, typically associated with observations from large-scale studies (Brett and Goldman, 1996). The apparent absence of observed effects on certain populations in an experimental setting may as such result from the high variability of the observations. Hamilton et al. (1988) report that in some large-scale experimental settings, reductions up to 50% are the smallest significant difference. This percentage will mostly depend on the used sampling techniques. For example, in study 3 (Webber et al., 1992), passive fish trapping techniques characterized by a large variability were used. Based on these measurements, no significant effects on fish biomass were observed, i.e. the $NOEC_{fish\ \ biomass} \geqslant 0.69\ \mu g\,l^{-1}$ ($\geqslant$the highest treatment). Whether adverse effects were truly absent or whether this was an artefact of the trapping method, is difficult to establish. The predicted NOEC for fish biomass reduction (0.18 $\mu g\,l^{-1}$) in study 3 should be considered in this context.

The main advantage of an ecosystem model approach versus currently used (statistical) extrapolation (e.g. the species sensitivity distribution, SSD) approaches is that the former can account for ecological interactions. These interactions can give rise to indirect toxicant effects which can not be predicted from single-species toxicity data alone, but which may be assessed through ecosystem model simulations. Indeed, in 8 out of the 11 cases in which populations were observed to experience indirect effects in the original study, these indirect effects were correctly predicted by our ecosystem model. In studies 1, 3 and 4, phytoplankton biomass was found to be higher in ponds treated with an insecticide (study 1: diflubenzuron; study 3 and 4: esfenvalerate) than in the control ponds, although the available data did not suggest that phytoplankton is directly stimulated by these chemicals. Boyle et al. (1996), Webber et al. (1992) and Fairchild et al. (1992) attribute these increases to decreased grazing activity of directly affected zooplankton. These increases were predicted correctly by the ecosystem model in all three studies (Fig 2).

A reduction of small zooplankton (*Chydorus sphaericus*, *Lecane* sp., *Mytilana ventralis*, *Polyarthra remata*) in field enclosures treated with the photosynthesis-inhibiting herbicides metribuzin (study 5) was observed by Brock et al. (2004). Because metribuzin is a herbicide with a very specific mode of action, it is unlikely to have affected zooplankton in a direct way. Instead, an indirect effect, i.e. a reduction of its food source (spring phytoplankton), may explain this observation. The same mechanisms was suggested by Denoyelles et al. (1982, study 10), who observed a decrease of large zooplankton when exposed to the herbicide atrazine in experimental ponds. These authors demonstrated that this decrease was the result of a decrease in food abundance, i.e. in phytoplankton density. These indirect effects were predicted by the ecosystem model in both study 5 and study 10.

Although the indirect effects observed in studies 9 and 11 were governed by similar mechanisms as those described in the previous paragraph, they were not predicted by our

ecosystem model. However, in both studies model accuracy was generally low (Fig 3) as not only indirect but also direct toxicant effects were predicted inaccurately.

Of all population-NOEC predictions considered in this paper, $\geqslant 55\%$ were estimated accurately at all tested γ-levels (0.01–0.5). Raising the γ-level from 0.01 to 0.5 increased the proportion of accurate population-NOEC predictions to 60%. A concurrent increase of conservative predictions (from 15 to 26%) was observed and underprotective predictions were reduced from 29 to 14%. An increase in γ can thus reduce the number of underprotective NOECs and increase the number of conservative NOECs. Note that $\gamma = 0.5$ corresponds to the median of the 100 RD-values. This allows to rephrase our definition of NOEC for decrease of a population to "the highest concentration at which the median of the RD-values of that population is higher than -0.2." Similarly, the highest concentration at which the median of the RD-values of that population is smaller than 0.2 is the NOEC for increase of that population.

The impact of the underprotective population-NOEC predictions on the resulting ecosystem-NOEC will depend on how these NOECs relate to those of the other model populations in the ecosystem. If the NOEC of the most sensitive population is overestimated, i.e. the effect is underestimated, the resulting ecosystem-NOEC will also be too high. Hence, it can not be used as a "safe" concentration for the considered ecosystem. Overestimation of a population-NOEC which is not the lowest observed in the study, will not impede an accurate estimation of the ecosystems' NOEC. Which of these two cases dominates in this validation paper, will be discussed in the next paragraph.

3.2. Predicted ecosystem-NOECs

The predicted ecosystem-NOECs of studies 1, 5, 7, 8, and 10 were independent of the γ and were accurate for studies 1, 7, 8, and 10. For study 5, a conservative ecosystem-NOEC was predicted which was 10 times lower than the observed NOEC. Predictions of the other 6 ecosystem-NOECs varied with changing γ-level. The percentage of accurate predictions increases with increasing γ, and can reach 63% at $\gamma = 0.5$. As for the predicted population-NOECs, the percentage of underprotective ecosystem-NOECs decreased with an increasing number of conservative estimates at $\gamma = 0.5$ (Fig 3). At $\gamma = 0.01$, nearly 20% of the predicted ecosystem-NOECs were underprotective. At $\gamma = 0.5$, the model predicted the same ecosystem-NOEC as the observed value for 7 of the 11 considered studies (i.e. in 63% of the studies). Predicted ecosystem-NOECs were never higher than the observed values at this γ-level, i.e. they were never underprotective. This indicates that the 14% of underprotective population-NOEC predictions at $\gamma = 0.5$, as derived in the previous paragraph, did not result in underprotective ecosystem-NOEC predictions. Hence, the model populations for which the predicted NOECs were too high were not the most sensitive populations in the considered studies. The NOECs of the most sensitive model populations were predicted accurately, or were conservative. This agrees with the finding that this type of ecosystem models predicts effects more accurately at low toxicant concentrations, i.e. the concentration range in which the most sensitive populations are affected, than at intermediate concentrations (Bartell et al., 1992). Based on our simulations, or on literature, it is impossible to explain this phenomenon with a true causal relationship.

For studies 5, 6, 9 and 11, a conservative ecosystem-NOEC was predicted which was 4–30 times lower than the observed value. In a similar validation study with the comprehensive aquatic systems model (CASM), Naito et al. (2003) found that most of the predicted ecosystem-NOECs were a factor 100 lower than the observed ones. The lower factor found in the present study (10–20) may result from customizing the considered ecosystems, i.e. from the inclusion of the relevant model populations. The CASM model features one specific ecosystem, the Lake Suwa ecosystem. It was tested if this shallow lake ecosystem could be used as a model for other experimental systems. It is logical that the latter approach results in less accurate estimates than the methodology established in the current paper. Because of the importance of indirect effects, resulting from a combination of inherent sensitivities and ecological interactions, implementing the relevant populations is crucial, from an ecological point of view.

The incorporation of ecological interactions by applying the presented ecosystem model resulted in rather accurate predictions of ecological effects of chemicals, both on a population- and ecosystem-level. It should be recognized though that the proposed modelling approach can only increase ecological realism to a certain extend. Morphological and behavioural changes in zooplankton, altering their vulnerability to fish predation, and reduction of stress tolerance of populations in time are examples of insecticide effects which are not included in this modelling approach (for examples, see references in Hanazato, 2001). It should be clear that such phenomena can not be accounted for by the proposed modelling technique.

In general, it can be stated that this type of models can serve as an ecology-based alternative to current extrapolation techniques in EEAs and water quality standard setting allowing to predict protective population- and ecosystem-NOECs in 85 and 100% of the cases, respectively.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.chemosphere.2007.09.052.

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