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Looking back – looking forward: A novel multi-time slice weight-of-evidence approach for defining reference conditions to assess the impact of human activities on lake systems

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1. Abstract

Lake ecosystems are sensitive recorders of environmental changes that provide continuous archives at annual to decadal resolution over thousands of years. The systematic investigation of land use changes and emission of pollutants archived in Holocene lake sediments as well as the reconstruction of contamination, background conditions, and sensitivity of lake systems offer an ideal opportunity to study environmental dynamics and consequences of anthropogenic impact that increasingly pose risks to human well-being. This paper discusses the use of sediment and other lines of evidence in providing a record of historical and current contamination in lake ecosystems. We present a novel approach to investigate impacts from human activities using chemical-analytical, bioanalytical, ecological, paleolimnological, paleoecotoxicological, archaeological as well as modeling techniques. This multi-time slice weight-of-evidence (WOE) approach will generate knowledge on conditions prior to anthropogenic influence and provide knowledge to (i) create a better understanding of the effects of anthropogenic disturbances on biodiversity, (ii) assess water quality by using quantitative data on historical pollution and persistence of pollutants archived over thousands of years in sediments, and (iii) define environmental threshold values using modeling methods. This technique may be applied in order to gain insights into reference conditions of surface and ground waters in catchments with a long history of land use and human impact, which is still a major need that is currently not yet addressed within the context of the European Water Framework Directive.

Keywords: EU WFD · Lakes · Weight-of-evidence approach · Reference conditions · Dioxin-like activity · Sediment quality triad approach

2. 1 Introduction

Lake ecosystems are particularly sensitive to anthropogenic changes in the hydrological cycle and by large-scale water pollution because they act as terminal sinks for all matter that affects water quality. In particular, a large number of lakes exist in formerly glaciated regions of Europe (e.g., Scandinavia, northern Germany, Poland, perialpine regions), which archive environmental changes from the end of the Pleistocene (i.e., postglacial, calibrated 15 kiloannum before present) to the Holocene (11.7 kiloannum before present) over various temporal scales (Downing et al. 2006; Wessels 1995). Valuable information about the historical, present and potentially future anthropogenic impacts (e.g., land use change, contamination, etc.) can be gained from signals continuously archived in lake sediments, with annual to decadal resolution (Cohen 2003).

Lake systems respond to short-term and long-term changes that affect the fluxes in energy, water and matter, such as modifications to topography, vegetation and soils, climate change, and the input of wastewaters into the system (i.e., signal generation; Smol 2009). Many of these fluxes (i.e., signals) are coupled with each other in a complex manner and can result in gradual or immediate changes in the lake system (e.g., eutrophication caused by nutrient and chemical inputs archived in sediment; signal recording; Fig. 1). These sudden or gradual changes in lake sediment composition and respective signal generations are mostly related to changes in (1) climate parameters such as precipitation, temperature, wind, and frequencies of singular and secular hydrological events; (2) human activities including land use, agricultural techniques, drainage of swamps, settlements and infrastructure, wastewater, industrial activities, and diffuse and point source pollution; and (3) nutrient inputs within the catchment area. In most central European systems, the impacts from human activities on aquatic environments began with the establishment of first settlements approximately 6000 years ago (e.g., Kalis et al. 2003; Litt 2003; Zolitschka et al. 2003). Early human settlements can create measurable signals (e.g., pollen record, nutrient and pollution profile, etc.) in the lake sediment by direct or indirect alteration of matter fluxes such as those due to deforestation, burning, and inflow of wastewater. However, intensities of such anthropogenic influences varied over time and are intercalated by periods of recovery from disturbance (e.g., migration periods, 30 years' war in the 17th century, etc.; Mainberger et al. 2015; Rösch 1992). Therefore, long-term records and cross-correlation of different lakes will allow for better identification and separation of human-induced signals from natural variability.

Signals of long-term environmental change may also be recorded within the catchment (e.g., degraded soils, colluvial and alluvial sediments), however, these records are generally much more incomplete compared to lake sediments, which act as a final trap of particulate material from a catchment area over hundreds and thousands of years (depending on the lifetime of the lake; Dale 2009; Grimalt et al. 2004; Renberg et al. 2000;

Yang and Rose 2005). Lake sediments are generated by direct sedimentation of particulate matter derived from river input and/or eolian transport (detrital),r by settling of precipitated particles from the lake water (authigenic, e.g., shells, organic matter, calcite), or as a result of biological productivity (biogenic). The transport and deposition of detrital material in lakes mostly depend on land use, soil erosion, weathering processes and engineering measures of waterways. The occurrence of authigenic and biogenic material depends on nutrient supply, which increases with agriculture and discharge of sewage water from settlements, and biogeochemical cycling (Meyers and Ishiwatari 1993). After deposition, most particulate matter is transported and transformed at the lake bottom by processes such as decomposition, resuspension by waves and/or bioturbation (Carper and Bachmann 1984; Huettel et al. 2003). These post-depositional processes may amplify, modify, attenuate, shift or erase original signals generated by environmental changes. Thus, it can be difficult to obtain an ideal undisturbed lake sediment core for high-resolution paleolimnological studies with annual stratification ("varves") for the entire lifetime of a lake. Best chances are provided by sediment records from low energy profundal sections in the center of the lake.

In order to reduce risks and manage the impacts of environmental contamination, land use, and climate change on lake systems, we need to gain a better understanding of the sensitivity of our environment and the background conditions prevailing prior to impacts caused by human settlements. Laboratory experiments and thorough monitoring of recent changes alone are not sufficient to understand or validate predictions of the long-term behaviour of complex landscape mosaics of terrestrial, semi-terrestrial and aquatic systems, such as the ones typical for Central Europe. This paper discusses some of the tools and knowledge currently available to assess signals from human activities in lake systems (sections 2-4). We present a multi-time slice weight-of-evidence (WOE) approach (Fig. 2) with multiple lines of evidence (cf. Chapman and Hollert 2006), including paleolimnology and paleoecotoxicological tools, to elucidate historical pollution, identify reference conditions, and improve process understanding of human activities (detailed discussion in section 5). This discussion paper aims at directing terrestrial-aquatic ecosystem research toward a holistic approach and recommends the investigation of modern systems from a historical perspective. This approach considers historical land use and industrial practices since the Neolithic age that have moved the system from natural background conditions to modern human-affected conditions. Such an integrated analysis allows for the evaluation of the extent and duration of disturbances in respective lake ecosystems. In particular, it will (i) create a better understanding of the effects of anthropogenic disturbances on biodiversity, (ii) assess water quality by using quantitative data on pre-historical and historical pollution and persistence of pollutants archived over thousands of years in sediments, and (iii) define environmental threshold values using modeling methods, thus offering a means to refine land use management strategies by defining pre-impact conditions, sensitivities, and recovery rates. Such knowledge can also closely support and assist in fulfilling future water quality goals, especially originating from the European Water Framework Directive (EU WFD, 2000/60/EC). The WFD declares water pollution as a key issue of European Environmental Policy and demands that all European water bodies should be returned to "good ecological status" by the years 2015-2027. "Good Ecological Status", however, deviates only slightly from "undisturbed conditions", which may be derived from paleo-data. Thus, by combining paleoecological, bioanalytical-ecotoxicological, chemical-analytical, geochemical, archaeological, and modeling techniques, it may be possible to establish the link between legacy and current anthropogenic impacts, as well as assist in predicting future impacts on lake systems. It is therefore a promising approach to comprehensively reconstruct and eventually understand the complexity of environmental changes caused by human activities. The individual research foci and tasks are described in the following sections, leading to the overall description of the multi-time slice WOE approach.

3. 2 Paleolimnological and paleoecotoxicological tools and records

4. 2.1 Past human activities

The development of agriculture across many parts of Europe, generally between the 8th and the 4th millennium cal. B.P., led to massive population growth. Since this time, people have altered the natural landscapes on a large scale through deforestation, modifications of the woodland structure and composition, introduction of new species, and through impacts on geomorphology and soils (Berglund 1991; Dotterweich 2008). Additional impacts from agriculture include soil erosion from tillage, as well as soil deterioration and acidification by nutrient loss with the harvest (Heathcote et al. 2013). In many agricultural systems, extensive burning of biomass and animal husbandry also played an important role in landscape alteration and management. Each of these past human activities contributes to distinct patterns that can be viewed in the paleolimnological records of lake sediments (Battarbee and Bennion 2011). Vegetation changes as well as burning processes from deforestation and agriculture can be evaluated by the pollen record and the amount of charred micro-particles archived in chronological order in the sediment (Clark et al. 1989). The pollen production within lakes is often weak and restricted to some limnic macrophytes, which allows for a direct reflection of the vegetation, landscape and land use of the terrestrial surroundings of lakes. Since the beginning of the extensive plough agriculture during the Bronze Age, the strength of human impact and land use change is correlated directly to the degree of deforestation, which is expressed by the percentage of terrestrial non-arboreal pollen (Kalis et al. 2003; Rösch 2012). Soil erosion is recorded in sediments by an increasing amount of minerogenic material and by changes in the chemical composition (e.g., by an increase of Ti; Berglund 1987; Cohen 2003; Lehmkuhl et al. 2014). Within the last decade the development, application and discussion of compositional statistics (compositional data analysis; CoDA; cf. Egozcue et al. 2003; Filzmoser et al. 2009; Van den Boogaart, K Gerald and Tolosana-Delgado 2013) and transformation of multivariate geochemical datasets has advanced paleoclimatic and paleoenvironmental reconstruction (cf. Dietze et al. 2012; Hartmann and Wünnemann 2009; Stauch et al. 2017; Yu et al. 2016). Recently developed multivariate and statistical methods also allow for precise calibration of the pollen record in terms of land cover (Broström et al. 2008; Gaillard et al. 2008; Sugita 2007a; Sugita 2007b). All these reconstructions must be based on a sound chronology and an age model of lake sediments. Here, various techniques are available which are usually combined: radiocarbon dating (^{14}C), lead dating (^{210}Pb), marker horizons (^{137}Cs from bomb tests and Chernobyl), and varve counting (Aitken 2014; Bonk et al. 2015). The application of mass spectrometry in radiocarbon dating has significantly reduced the amount of carbon required. Thus, the selection of organic material of terrestrial origin from the sediment enables reliable spatiotemporal models. As a result of the abovementioned analytical techniques and paleo-records, major environmental shifts archived in lake sediments (i.e., signal generation) based on dated sediment slices can be correlated with historical events dated by well-documented historical, meteorological or archaeological data (e.g. agricultural records, history of local industrial activity, artefacts and materials, flooding events reported in historical accounts) to gain insight into the influence of human impacts.

5. 2.2 Records of pollutants

As human populations continued to grow and advance, industrialization including mining and burning of fossil fuels began to develop. The now “civilized” humans also contributed to effects on the aquatic environments through pollution, ranging from local pollution by lakeside dwellings (e.g., sewage) to the global distribution of mining emissions, each archived in the sediment layer. Imprints of human activities are recorded by abiotic and biotic proxies, serving as indicators of past environmental conditions. Histories of mining activities and burning of fossil fuels can, for example, be derived from $^{206}Pb/^{207}Pb$ isotope ratios, providing the ability to identify (i.e., fingerprint) sources and release of pollution in lake sediments (Abbott and Wolfe 2003; Bränvall et al. 2001; Engstrom et al. 2007) or from fly-ash particles (e.g., spheroidal carbonaceous particles, SCP; Rose et al. 2002). Mercury (Hg) has been used as a proxy for domestic sewage, however, controversy persists to whether Hg levels in sediments archive an accurate record of past accumulation rates due to the potential influence by microbial and diagenetic processes; i.e., chemical, physical and biological changes that occur within the sediment, that can enrich Hg in surface layers of sediment cores and be mistaken as a signal of anthropogenic pollution (Muir et al. 2009; Rasmussen 1994; Rydberg et al. 2008; Smol 2009). In contrast, polycyclic aromatic hydrocarbons (PAHs) were introduced early in history through open burning and natural wildfires, however, industrialization has significantly increased the concentrations of PAHs in ecosystems through the combustion of organic material resulting in good correlation between PAH concentrations in sediment cores and industrial energy consumption (Lima et al. 2005). The combustion of organic material has lead to the production of a wealth of organic compounds that previously had little or no presence in the environment, with many of them persistent and bioaccumulative in the environment, i.e., persistent organic pollutants, such as polychlorinated biphenyls and PAHs. Thus, sedimentary pollutant profiles potentially allow us to track the trajectories and patterns of deposition of many pollutants.

6. 2.3 Ecological effects of pollutants

Aquatic organisms can provide information about the vulnerability of ecosystems, the critical loads of pollutants, and they can document ecosystem degradation (Hübener et al. 2009). In the 1980s, many paleolimnological studies addressed acidification by using diatom assemblages (Battarbee and Charles 1987; Hinderer et al. 1998). Nutrient loading and eutrophication became another popular topic (Cohen 2003; Meriläinen et al. 2000), and thus the quantitative assessment of eutrophication trends with diatoms developed rapidly during the last few decades (Smol and Stoermer 2010). However, the ecotoxicological effects of well-characterized pollutants on organisms used in paleolimnology have only recently been investigated (Doig et al. 2015; Harris et al. 2006; Lucas et al. 2015). Metals and herbicides, for example, although well-studied from a toxicological perspective, have only recently begun to be examined from a paleolimnological perspective using diatoms (Larras et al. 2013; Marcel et al. 2013). Specifically, Cattaneo et al. (2004) reported that Cu pollution led to a taxonomic shift in diatom species, deformation of diatom frustules, and a reduction in size, even though there was no decline in the number of species due to Cu pollution. It was also suggested that teratological forms of diatom cell walls may act as indicators of ecosystem health because their presence is related to the magnitude of environmental stress (Falasco et al. 2009). The quantitative total phosphorous (TP) reconstruction approach has also been established as TP is often the most important factor influencing diatom communities within a calibration data set (Anderson 2000; Hall and Smol 1992). Additionally, paleo-ecotoxicological information can be obtained from cladoceran diapausing eggs (ephippia), which have been shown to preferentially accumulate some maternally derived metals such as cadmium, chromium and molybdenum from urban or industrial sources (e.g., smelting and fossil fuel combustion; Wyn et al. 2007). Both the geochemical and the isotopic composition of calcitic ostracod shells also have been observed to provide an indication of metal pollution and paleo-environmental reconstruction (Holmes 2001; Schwalb 2003). Thus, diatoms and other bioindicators have a great potential to monitor the quality of lake water and efficiency of ecosystem management measures, such as liming, decrease in acidification, and speed of re-oligotrophication in lake systems.

By using different types of bioindicators from a variety of habitats, for example benthonic and planktonic diatoms or infaunal and epifaunal ostracods, processes in different components of a lake system can be analysed. Transfer functions can be established that consequently can be applied to fossil species assemblages archived in sediments by relating modern species assemblages, including diatoms, chironomids and ostracods to environmental parameters (e.g., water chemical composition, water depth, etc.) of their habitats (Hall and Smol 1992; Pérez et al. 2013). Additionally, species assemblages themselves may also be a direct and useful bioindicator of pollution and environmental stressors. These environmental reconstruction approaches serve for deriving quantitative parameters including trophic level, pH, conductivity, temperature and water depth, and it provides a tool to assess current water quality by establishing background conditions or a reference state from a time when humans did not yet affect their environment. Therefore, ecologically and statistically sound environmental reconstructions are required (Juggins 2013), and their reliability needs to be improved with new approaches (e.g., dynamic adjustment of training sets (Hübener et al. 2008), compositional data approaches (Van den Boogaart, K Gerald and Tolosana-Delgado 2013). Regardless, new advances in paleolimnological and paleoecotoxicological research, including morphological studies, may offer crucial insight into the ecological consequences of pollutants over time.

7. 2.4 Bioanalytical tools and paleoecotoxicology

As an interdisciplinary field of research, ecotoxicology deals with the interactions between environmental chemicals and biota, thereby focusing on adverse effects at different levels of biological organization (Fent 2004). Toxic effects of anthropogenic compounds in biota and ecosystems are investigated in close connection to their environmental chemistry and fate in the environment (Fent 2003; Fent 2004). Aquatic sediments act as a sink of anthropogenic pollutants, but they can also act as a source via remobilization (e.g., during resuspension and flood events) and can thus cause adverse effects in the environment, as well as for human health (Brinkmann et al. 2013; Hollert et al. 2007; Schüttrumpf et al. 2011; Wölz et al. 2008; Wölz et al. 2009). Consequently, sediments can be used to assess hazardous impacts and underlying toxicants using different analytical techniques, such as biological or chemical analyses, or the combination of both (e.g., effect-directed analyses; EDA). Bioanalytical tools include *in vitro* and *in vivo* bioassays as well as biomarkers, which provide information about the toxicity or biological response of environmental samples or contaminants. Wernersson et al. (2015) discusses some of the common bioanalytical tools that could be used in different monitoring programmes to link the chemical and ecological status required for assessments of waterbodies by the EU WFD. Furthermore, ecotoxicological investigations of historical sediments provide the opportunity to characterize, assess and compare the burden caused by human activity before and during certain time periods of intensive anthropogenic impact on lake-catchment areas. Biomarkers such as the lipid biomarker fraction of the organic matter in lake sediments can be used to reconstruct historical changes in a lake system including changes in primary productivity, sedimentary sources, climate, anthropogenic influences, diagenetic alterations and recovery rates (Brandenberger et al. 2008; Lu and Meyers 2009; Meyers and Ishiwatari 1993; Zhou et al. 2005). This helps to define at which time natural and undisturbed conditions occurred in a lake system and when the system became impacted. By combining bio- and chemical-analytical, ecotoxicological, geochemical and archaeological data, it might also be possible to narrow down or even identify the source of contamination.

A proof-of-concept study was carried out by our group to demonstrate that the use of multiple lines of evidence with sediment layers across different time periods (i.e., multi-time slice) can be used to identify pollution signatures in lake systems. Multiple slices of sediment were examined from different sediment cores collected from a lake, Stadtsee, in Bad Waldsee, Germany, a key area of human settlement for the past 6000 years (e.g., region was settled since the Late Neolithic according to archaeological data). Comprehensive data regarding archeology and pollen spectra was available for the sediment cores and the dating of the sediment cores was performed through comparison of the pollen record with other, absolutely dated pollen profiles of the same region (Fischer et al. 2010). The activity of the enzyme ethoxyresorufin-*O*-deethylase (EROD) was analyzed from different sediment slices according to the protocols provided elsewhere (Heger et al. 2012; Seiler et al. 2006). The rainbow trout liver cell line (RTL-W1) EROD bioassay is an approved biomarker for dioxin-like contamination and Ah receptor agonists (so called dioxin-like activity) that provides a sensitive indication of cellular changes at the enzyme level. The investigation demonstrated that bioanalytical approaches could be adapted for minute quantities of sample, in the mg quantity range. Furthermore, the resulting activity of the EROD enzyme (Fig. 3) showed large differences among the different limnic archives expressed as biological (i.e., bioassay-derived) toxicity equivalent quotient (BEQ or TEQ) values (Eichbaum et al. 2016). The BEQ values represent the strength of effect expressed relative to the concentration of a reference substance. The greater the BEQ value, the stronger the contamination of the sediment layer. Segments from the High Middle Ages (10th - 12th century AD) revealed dioxin-like activities six times greater than found for uncontaminated horizons. The resulting BEQ values from the sediment cores represent a toxicity equivalent to 2,3,7,8-Tetrachlorodibenzo-p-dioxin (TCDD), a highly potent environmental contaminant. The determined BEQs of 200 to 700 pg TCDD/g sediment already exceeds the 100 pg/g threshold for playing grounds from the German Federal Soil Protection Act (BBodSchG 1998) and approaches the threshold of 1000 pg/g for residential/recreational areas. The BEQ values correspond well with maxima of charcoal and pollen of culture indicators from within the analyzed sediment core samples (Fischer et al. 2010). This proof-of-concept demonstrates that biomarkers such as the EROD induction can also be suitable for small quantities of samples (as available in some lake sediment cores) with low or medium load of pollutants. As a result, bioanalytical tools such as biomarkers should be considered as a useful tool as part of paleoecotoxicological studies. By combining charcoal concentrations, changes in the diversity of trapped pollen, and bioassays, pollution profiles were identified in different sediment layers ranging from the Middle Ages to pre-industrial activities. Our findings provide initial support that multiple lines of evidence from different time slices are suitable for the investigation of environmental dynamics and consequences of anthropogenic impacts.

8. 2.5 Fate of pollutants

Long-term persistence and availability of environmental contaminants associated with soils and sediments under global change conditions is a key issue in environmental risk assessment. Multiple processes on different temporal and spatial scales influence particle and contaminant patterns, as well as sorption and desorption processes and, thus, the availability of potential toxicants for organisms in ecosystems, including humans. Rising temperatures have a direct influence on all chemical reactions, as well as transport and partitioning phenomena, such as diffusion and sorption processes (Schwarzenbach et al., 2003). Other direct and indirect impacts of climate change, including change of the carbon cycle, amount of precipitation and related extreme events, as well as land-use changes and modification of human activities, may have an even greater influence on the availability of pollutants. These types of direct and indirect impacts can modify the quantity and quality of amorphous organic matter (e.g., lignins, polysaccharides, lipoproteins, amino acids, lipids, humic/fulvic acids) and carbonaceous organic matter (e.g., black carbon, kerogen, and coal) in sediments, thereby influencing concentrations and availability of contaminants in the sediment (Cornelissen et al. 2005; Lamon et al. 2009; Lehmann et al. 2002; Lücke et al. 2003). To unravel the complex processes associated with climate change and pollution, analyses of lake sediments that have accumulated over centuries and millennia will help us to understand the availability of sediment-associated compounds and to assist in the assessment of future contaminant behaviour. Combined, these analyses will assist in predicting environmental risks to the biosphere.

While some pollutants have been emitted since pre-historic times, such as pyrogenic polycyclic aromatic hydrocarbons and polar derivatives thereof, as well as human faecal sterols, synthetic organic chemicals have only been produced since industrialization and emitted over the last century. Considering the different time frames, the analysis of both the historical and recent pollutants archived in sediment may be used to understand their bioavailability and fate under different environmental and climatic conditions. It has been shown that the aging of contaminated sediment particles over years and decades reduces bioavailability (Harkey et al. 1995), although there is not yet information available for longer periods of time. It may be hypothesized that bioavailability and toxicity of historical pollution is reduced by the diffusion and binding of organic compounds to the matrix of organic and carbonaceous particles and coating, as well as the increase of carbonaceous carbon relative to degradable organic carbon. However, this hypothesis still must be tested and confirmed because the decay of organic material carrying persistent organic pollutants may also have the opposite effect and increase the bioavailability and toxicity of contaminants. Additional studies are also required on factors that may influence the bioavailability of pollutants archived in sediment, such as physical-chemical properties, aging and conditions of aging. In-depth analysis of lake sediment cores integrating proper dating, carrier particle identification and characterization together with pollutant pattern analysis and desorption experiments may help to address these issues and relate them to knowledge on climate conditions and historic land-use.

9. 3 Integrating dynamic lake models into paleolimnology

As paleolimnology is based on linking biogeochemical signals in sediments to the ecological state of the lake and its catchment, existing modeling approaches for paleolimnological data are dominated by statistical techniques. While the relationships between the large number of variables in paleolimnological studies may be effectively analysed by such static modeling approaches, the dynamic processes mediating these signals often remain undetectable. Those paleolimnological signals related to fluxes of carbon, nutrients, and bioactive substances are, however, formed by ecosystem dynamics that, in turn, are driven by climatic, hydrological, and ecological processes. In that sense, the lake is not only a passive sampler that is archiving signals from its environment, but it is also a reactor that is dynamically transforming energy and matter in a variety of ways (see Fig. 1). We therefore recommend the introduction of dynamic ecosystem models (e.g., Mooij et al. 2010) as a new tool into paleolimnology in order to establish a mechanistic framework for studying the dynamic processing of matter and energy within lakes. By such a framework, external forcings and the biogeochemical transformation processes can be mechanistically linked to paleolimnological signal formation.

Dynamic lake ecosystem models simulate nutrient and carbon cycling in lakes by accounting for the major processes involved in sediment-water interactions, water and gas exchange, population dynamics, and the ecological food web. Since major driving variables of these models are time-series of meteorological data and hydrological inputs from the catchment (mainly water, nutrients and carbon components), these models provide interfaces to climatic conditions and catchment characteristics. Prominent examples of lake ecosystem models include papers on Lake Zürich (Omlin et al. 2001), Lake Kinneret (Bruce et al. 2006), Lake Washington (Arhonditsis and Brett 2005), and Lake Constance (Rinke et al. 2010). There models are practically used, for example, in water quality management of lakes, such as the evaluation of effects from anthropogenic stressors, including climate change or eutrophication (Gal et al. 2009; Mooij et al. 2010). Lake models usually consist of two interacting submodels: first, a physical lake model simulating thermodynamics and hydrodynamics of the waterbody, and second, a physical model is coupled to an ecological model simulating biogeochemical and community dynamics within the ecosystem.

To demonstrate the contributions that ecosystem models can deliver to paleolimnological studies, the ecosystem changes during the warming phase after the last glaciation is an excellent example. The warming is expected to induce discontinuous changes in the mixing of a given lake (e.g., mixis type of lake from cold-monomictic over dimictic to warm-monomictic or even oligomictic; Boehrer and Schultze 2008). The changes in mixis type correspond with major shifts in plankton succession and primary productivity. Lake models can predict the critical warming intensities necessary to induce these shifts in a given lake system and the timing of these critical warming intensities in climatological temperature reconstructions can be compared to corresponding shifts in paleolimnological records in that lake.

10. 4 A holistic framework to model lake ecosystems in a social-ecological context

Since the connection between human activity and climate change became evident, it has become clear that social-ecological systems are complex adaptive entities which are tightly connected to human society (Leuteritz and Ekbia 2008; Muradian 2001; Walker et al. 2004). The awareness of interactions between ecology and society resulted in the development of the concept of social-ecological systems (e.g., Stockholm Resilience Centre), which has recently been integrated in numerous research programs (see e.g., UFO-Project at www.humtec.rwth-aachen.de, Schlüter et al. 2014). In brief, the concept of social-ecological systems is that humans both influence and are influenced by ecosystem processes in dynamic feedback loops (Cumming et al. 2006). Thus, catchment conditions in lake ecosystems, determined by sociological development, have an influence on lake ecology and the subsequent signal formation in the sediments (Angeler et al. 2011). Causal feedback loops from lake systems to the catchment and society often exist due to the influence of lake-use on social conditions (e.g., by the provision of fish). In light of such interacting influences, lake ecosystems and catchment areas are considered as self-organized social-ecological systems (Dearing and Zolitschka 1999). Due to industrial development and the resulting land use changes since the beginning of the industrial revolution, society has become more and more independent from lake systems and the feedback loop from lake to society has become weaker. Such a scale mismatch (Cumming et al. 2006) can have disastrous consequences for lake systems such as mismanagement of natural resources and eutrophication. Thus, in terms of systems theory, the information flow from society to lake ecosystems persisted or was even increased while the information flow from lake systems to society was reduced (the term information flow can be exchanged with entropy flow, energy flow, flow of matter etc.). The importance of such information and energy-related flows for the self-organization of complex systems has long been recognized (Prokopenko et al. 2009).

In population ecology the reduction of resilience due to human impacts has been shown previously, e.g., this effect is strongly connected to the destabilization of feedback loops (Ottermanns et al. 2014) in systems with strong non-linear dynamics. Destabilizing feedbacks can also result in a decrease in social-ecological resilience (Cumming et al. 2006). This feedback loop can be reconstructed for some systems, since currently lake ecosystems are assigned a specific value for society, called ecosystem services (e.g., recreation, etc.; Bergstrom et al. 1996; Bingham et al. 2000; Postel 1997). The process of adaptive co-management (Folke et al. 2002; Olsson et al. 2004) provides a possibility to react to such environmental feedback and direct these coupled social-ecological systems into sustainable trajectories thereby enhancing their resilience (Berkes et al. 2008; Gunderson 2003). The question of how strongly such changes took place in the history of lake ecosystem dynamics (Arrayás et al. 2000), in extreme cases resulting in discrete phase transitions (e.g., plankton or fish population dynamics; Medvinsky et al. 2002), should be integrated into an assessment of human impact on lake systems. This integration would allow for a better understanding of how dependent lake ecosystems were in the past on catchment conditions in order to derive reference conditions, which will aid in the determination and prediction of future scenarios of human impact on lake systems (Croke et al. 2007; Rotmans and van Asselt 1999). Thus, we propose an integrative modeling approach to enable an integrated assessment based on a more holistic principle in order to predict the future development of lake ecosystems within their catchment and sociological context.

In an integrative assessment approach, mechanistic modules can be used to elucidate questions for which we already have theoretical knowledge about the processes (e.g., nutrient cycling in lake sediments), whereas statistical modules can be used to answer questions for which we must rely on empirical evidence (e.g., complex food web interactions in lake-catchment systems) (Kendall et al. 1999). In large-scale modeling approaches, it is important to address challenges to integrate variables from the different scientific disciplines (ecology, ecotoxicology, hydrology, geomorphology, archeology, paleolimnology, sociology, chemical analysis etc.), from different domains (spatial and temporal), on different scales (short-term processes such as population growth, as well as long term processes including climate change), of different nature (metric, ordinal or nominal) and of different uncertainty (objective quasi-experimental and subjective domain knowledge). A wide range of techniques are needed to tackle such challenges, including multivariate statistics (ordination, structural equation models), time series analysis (frequency-domain, time-domain), pattern recognition (support vector machines, neural networks) and dynamical systems theory (attractor reconstruction). Additionally, special attention must be given to the integration of different methods and types of evidence (quantities from empirical evidence and qualities from expert evidence). As such, Bayesian approaches are promising tools to incorporate probabilistic knowledge (Croke et al. 2007; Ticehurst et al. 2007), which is indispensable when predicting future development under uncertainty.

The complexity of catchment-related processes within transformation of climate and human impact signals (Fig. 1) must consider spatial and temporal variations of archived attributes. Integrative data modeling with multivariate time series statistics use these spatial and temporal variations in order to obtain qualitative and quantitative information about transformation processes from catchment characteristics to paleolimnological records (e.g., Hartmann and Wünnemann 2009). Given a sufficiently large dataset, the development of testable causal hypotheses regarding spatial and temporal interactions of processes is possible by integrating knowledge gained from paleoecology, ecotoxicology, chemical analysis, geochemistry and archeology. On the one hand, resulting hypotheses can be tested against observational data in a statistical manner (e.g., Ottermanns et al. 2011). On the other hand, dynamic simulation models can be used to test the hypotheses against the theoretical appraisal regarding biogeochemical transformation processes and the driving mechanisms of paleolimnological signal formation. If expectations do not meet the simulation results, hypotheses have to be rejected or the model structure must be improved. In this way, results from statistical evaluation feed back into dynamical lake models (e.g., in form of time-series models or model validation).

This integrated approach tends to combine theory-based models with data-based models in a hybrid manner, interrelating theories and data. The application of this idea of integrated approach has been demonstrated in recent research to large-scale aquatic ecosystems, such as the Yangtze Three Gorges Dam reservoir (e.g., Yangtze-Project at www.yangtze-project.de; Scholz-Starke et al. 2013). It was concluded that the combination of theoretical models, empirical data, and expert knowledge is in accordance with the concept of Integrated Environmental Modelling (IEM; Argent 2004). This is an important methodology of environmental management and decision-making (Jopp et al. 2011) which allows for the extrapolation and transfer of results to other locations, to different scenarios, and into the future.

11. 5 A multi-time slice weight-of-evidence approach

For the evaluation of the ecological status, the EU WFD requires the identification of type-specific reference conditions for surface water bodies (Bennion and Battarbee 2007). However, according to results of various research projects carried out all over Europe, it is nearly impossible to find sampling sites that represent uncontaminated reference conditions. To overcome this shortcoming, the WFD requests to establish reference conditions on modeling or expert judgment using data from historical, paleoecological and other investigations (EC 2006). It has also been recognized that paleolimnology is a pivotal approach for defining pre-anthropogenic reference conditions (Bennion and Battarbee 2007; Hübener et al. 2009; Smol 2009). For example, paleolimnological studies of 14 dimictic calcareous lakes located in the northern German lowlands (Hübener et al. 2015) demonstrated that the temporal onset of anthropogenic impact is lake-specific and, therefore, the timing for reference conditions is variable and depends on catchment to lake volume ratios. Thus, impacts on lake systems are complex and need to consider not only temporal and spatial variables, but also additional lines of evidence in order to gain comprehensive insight into historical and present environmental shifts. Complementary tools and procedures are needed to translate paleoenvironmental and paleolimnological records (i.e., combination of the biological, chemical and physical state of the environment/waterbodies at the time of deposition, established by the sedimentary record) into quantitative dimensions of the respective long-term environmental change and to identify the driving forces of such impacts (e.g., climate change, intensity of land use, soil treatment, emission rates and sources of pollutants, density and type of settlements). Knowledge of long-term environmental changes as well as frequencies of extreme events and their impacts on aquatic ecosystems have the potential to help define options for lake management and restoration.

In the context of recent ecology and ecotoxicology, the Sediment Quality Triad (SQT) approach is one of the most successfully applied conceptual frameworks to acquire comprehensive knowledge and ecological relevance regarding sediment contamination. The SQT is a weight-of-evidence (WOE) approach originally consisting of three lines of evidence (Chapman 1990): (i) sediment chemistry to determine chemical contamination; (ii) sediment bioassays to determine toxicity; and (iii) benthic community structure to determine the status of resident fauna arguably most exposed to any sediment contaminants. To date, these three original components serve as the primary basis for the SQT, providing a screening-level ecological risk assessment (ERA) of contaminated sediments (Chapman and McDonald 2005). Nevertheless, the SQT was never intended to be limited to only three specific lines of evidence. Shortly after its development, Chapman (1986) conducted a

SQT study in which he substituted bottom fish histopathology for benthic infaunal community structure. Recently, Chapman and Hollert (2006) addressed whether the SQT could become a tetrad, a pentad, or possibly even a hexad based WOE approach, proposing additional lines of evidence, such as *in situ* assays, mechanism-specific endpoints and whole sediment assays in order to achieve a more complete overview of the state of aquatic ecosystems. Hecker and Hollert (2009) also suggested the inclusion of EDA as an additional line of evidence in WOE studies in order to identify the pollutants responsible for the effects in the laboratory and the field. Gerbersdorf et al. (2011) proposed a "triad plus x" approach combining advanced methods of ecotoxicology, environmental microbiology and engineering science.

Based on the tools and knowledge available to assess historical, current and future impacts, we propose the use of a multi-time slice WOE approach (Fig. 2) that utilizes the previously discussed lines of evidence from many interdisciplinary fields. The goal of the multi-time slice WOE approach is to provide a comprehensive overview of how the environment was altered by human activities over the last millennia as a basis for future predictions. Within this new conceptual framework, the classical SQT approach will be applied in order to investigate the toxicological effects of well-defined time slices of sediment samples, but expanded further using interdisciplinary methods from the areas of archaeology, paleolimnology and paleoecotoxicology. Sediment geochemistry will provide knowledge of the type of past human activities (Section 2.1), paleolimnological records of pollutants (Section 2.2) and the fate of pollutants (Section 2.5). Investigation of the benthic community structure will also be supported by data on ecological effects of pollutants (Section 2.3). Additionally, bioassays and other bioanalytical tools (Section 2.4) will support paleoecotoxicological investigations into the effects of pollutants from the cellular to ecosystem level. Statistical modeling will then be used to (i) integrate data from paleoecology, ecotoxicology, chemical analysis, geochemistry and archeology, (ii) connect results to all integrated research tasks, (iii) help identify reference conditions, (iv) improve process understanding, (v) elucidate patterns of contaminations on spatial and temporal scales, and (vi) extrapolate the findings to multiple conditions. The multi-time slice WOE approach therefore goes far beyond pure paleolimnological investigations within the WFD as proposed previously (Bennion and Battarbee 2007; Bennion et al. 2011). We aim to identify and define key methods to describe lake system changes and their impact on the environment, rather than only producing additional data for statistical evaluation in terms of a multi-proxy analysis. The multi-time slice WOE approach will allow for a better understanding of the impact of humans on lake ecosystems, and may be used in future studies in order to gain insights into reference conditions in the same catchment area – a so far not solved but urgent need in the context of the European WFD.

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14. Compliance with Ethical Standards

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15. Conflict of interest

The authors declare that they have no conflict of interest.

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17. Figure captions

Fig. 1 Theoretical framework illustrating the history of signals in catchment-lake systems.

Fig. 2 Schematic of the proposed multi-time slice weight-of-evidence approach. The classical sediment quality triad approach (encompassed in triangle) is amended with archaeological, paleolimnological and limnological methods. This provides valuable insight into human impact, defines reference conditions and eventually allows for derivation of environmental quality standards (EQS) as required by the EU WFD. The cylindrical slices represent a stratified sediment core sample.

Fig. 3 Results of a proof-of-concept study from the Hollert lab (ecotoxicology) and Rösch lab (vegetation history, archeology) investigating dioxin-like activities (bioassay-derived toxicity equivalent quotient; BEQ) using a modified cell-based EROD assay with sediment extracts from lake sediment core slices, Bad Waldsee Stadtsee (BWS, Germany). The age of the sediment, dated from the pollen record, from left to right is Modern Age (most probably older than 18th century AD), High Middle Ages (10th to 12th century AD), and Iron Age (ca. 1050 BC – 1 BC).