

Final Report:

**Leibniz Graduate School
Aquatic boundaries and linkages in a
changing environment
(AQUALINK)**

Leibniz-Institute: Leibniz-Institute of Freshwater Ecology and Inland Fisheries (IGB)

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Abstract

The international Leibniz Graduate School AQUALINK was an interdisciplinary doctoral training and research programme focusing on aquatic boundaries and linkages in a changing environment. These boundaries are characterized by steep physical and biogeochemical gradients and disproportionately high reaction rates (hot spots), and thus, responsible for the regulation of matter transport in aquatic ecosystems. The integrity and functional capability of these ecosystems are especially determined by boundaries and transition zones between freshwater systems and their terrestrial environments.

The Leibniz Graduate School AQUALINK was structured in the following complementary sub-topics: boundaries between aquatic and terrestrial systems, internal boundaries in aquatic systems, and revitalisation and management. Over a 3-year period, doctoral candidates will receive training towards accomplishing an original doctoral thesis and international publications on the aforementioned topics.

Two Leibniz institutes (IGB and ZALF) and three international partners (University of Aberdeen, Eawag, University of Southern Denmark) have promoted the training of PhD students and their skills in various fields by summer schools, by cross-wise supervision, by usage of modern methods and infrastructure as by guest stay at cooperating institutions. The graduate school aimed at the process studies at different spatial and temporal scales and the development of conceptual and mathematical models with the focus on changing water level fluctuations and the transport across boundaries, including also the development of freshwater management strategies.

Zusammenfassung

Die internationale Leibniz Graduiertenschule AQUALINK war ein strukturiertes, interdisziplinäres Ausbildungs- und Forschungsprogramm, welches sich auf Transport- und Transformationsprozesse in Grenz- und Übergangszonen zwischen dem Gewässer und seiner terrestrischen Umgebung und innerhalb der Gewässer fokussierte. Diese Zonen sind durch überproportional hohe Reaktionsraten (hot spots) gekennzeichnet und somit für die Regulation von Stoffflüssen in aquatischen Ökosystemen verantwortlich. Deren Integrität und Funktionsfähigkeit wird entscheidend von Grenz- und Übergangszonen zwischen dem Gewässer und seiner terrestrischen Umgebung und innerhalb der Gewässer bestimmt.

Die Leibniz Graduiertenschule AQUALINK war in drei komplementäre Bereiche unterteilt: Grenz- und Übergangszonen zwischen aquatischen und terrestrischen Systemen, interne Grenzzonen in Gewässern, sowie Revitalisierung und Gewässermanagement. Mit der Ausbildung von Doktorandinnen und Doktoranden wurde das Ziel verfolgt, den wissenschaftlichen Nachwuchs auf die aktuellen und zukünftigen Herausforderungen zum Schutz und Erhalt aquatischer Ökosysteme vorzubereiten.

Zwei Leibniz-Institute (IGB und ZALF) und drei internationale Partner (University of Aberdeen, Eawag, University of Southern Denmark) unterstützten die Ausbildung der Doktoranden und ihrer Fähigkeiten auf verschiedenen Gebieten durch die Organisation von Sommerschulen, durch institutsübergreifende Betreuung, durch die Nutzung moderner Methoden und Infrastruktur sowie durch Gastaufenthalte in den kooperierenden Einrichtungen. Ein zentrales Thema innerhalb von AQUALINK rankte sich um die die Frage, wie sich aktuelle klima- und nutzungsbedingte Wasserstandsänderungen auf die Transport- und Transformationsprozesse in den Grenzzonen auswirken. Die Forschungsansätze dazu reichten von Prozessstudien auf verschiedenen Skalen über mechanistische und stochastische Modellierung bis zur Entwicklung von Sanierungs-, Revitalisierungs- und allgemeinen Gewässermanagementstrategien.

Hypotheses and research foci

Aquatic interfaces are of increasing interest in aquatic science because they play a key role in the interlinking of inland waters to the terrestrial environment as well as in the regulation of matter and energy fluxes within aquatic ecosystems. Therefore, the processes that occur at aquatic interfaces not only control the ecological state of freshwater ecosystems but also act as sinks and sources for matter fluxes at the landscape level. Aquatic interfaces are often characterised by steep physical and biogeochemical gradients that lead to disproportionately high reaction and turnover rates. These steep gradients are formed through the hydraulic connectivity between contrasting environments. Although aquatic interfaces play a pivotal role in controlling the ecological state of freshwater ecosystems and their resilience to environmental changes, they occupy a comparably small area or volume. The localisation and quantification of their turnover capacities at various spatial scales is a crucial prerequisite for predicting the development of aquatic systems under changed climate and land use conditions as well as for the refinement of sustainable management strategies.

Research on aquatic interfaces is of increasing importance for sustainable water management. However, this will require the fast transfer of new scientific knowledge into practice. On the other hand, the sites manipulated for management measures are ideal as “field labs” for empirical research for understanding the function and efficiency of aquatic interfaces. A promising direction in the research on aquatic interfaces is the focus on their function as a “control point” in a changing world. The impact of climate change is of extremely high interest, particularly with regards to the consequences of increases in the temperature on the sediment-water exchange, changes in the mixing regime, and fluctuations in the water level of lakes.

In summary, the overall hypotheses of the AQUALINK Graduate School are formulated as:

- Fluxes of matter and energy in aquatic systems are predominantly determined by processes at reactive boundaries.
- The different types of boundaries play a key role in aquatic ecosystems, both in quantity and in quality (“hot spots” and “hot moments”).
- Because of the intensities of degradation and transformation of solutes in and across these interfaces they control the biogeochemical milieu of aquatic ecosystems (despite of their small extension).

Aquatic interfaces perform several ecosystem services, and the maintenance of “healthy” and “functional” interfaces should be included when proposing management strategies (e.g., as basis for the definition of critical loads and thresholds for regime shifts). The focus of ecological engineering is often on modifying processes in aquatic interfaces such that water quality targets are achieved in the most efficient manner.

The AQUALINK project was structured in the following three research topics with 8 PhD projects (Figure 1).

Topic 1: Boundaries between aquatic and terrestrial systems

- Hydraulically and temperature controlled exchange between surface water and groundwater
- Regime dynamics of aquatic-terrestrial boundaries and their changes
- Climate change and the ecohydrology of connections between landscapes and riverscapes

Topic 2: Internal boundaries in aquatic systems

- Influences of water level fluctuations on sediment water interactions
- Gas exchange across aquatic boundaries
- Effects of groundwater seepage on benthic primary producers and their nutrient retention in lakes

Topic 3: Revitalisation and management

- Influence of river restoration on groundwater recharge
- Lake Management under changing water levels,

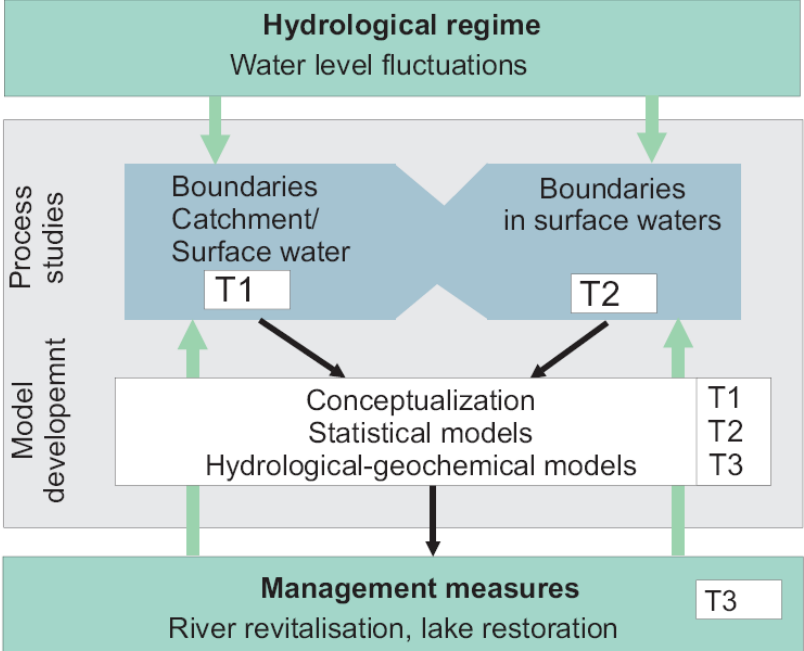


Fig. 1: Structure of the AQUALINK Graduate School

Results

Topic 1: Boundaries between aquatic and terrestrial systems

Sub-project: **Approaches to identify groundwater discharge towards and within lowland surface water bodies on different scales**

PhD: Franziska Pöschke

PIs: Gunnar Nützmann, Jörg Lewandowski, Mario Schirmer

Although the point sources of nutrient input into surface water were significantly reduced in the past decades, there is still an ongoing eutrophication in the lakes and rivers in the North-Eastern German Lowlands. This is mainly related to the diffuse entering pathways. One relevant pathway might be the groundwater. However, the characterization of subsurface flow and its interaction with the surface water in lowlands is difficult. The main reasons for that are the heterogeneously deposited unconsolidated rocks, in which complex and nested aquifer systems (local, intermediate, and regional) establish. Hence, groundwater discharge occurs on different scales, whereas each scale is dominated by different drivers.

The aim of the presented thesis is to apply different methods to determine the parts of lowland aquifer systems which contribute mass, substances and energy into specific surface waters. Therefore, two approaches were used (I) a hydrogeological and (II) a limnological/hydrological one.

The focus of the hydrogeological approach was set on the characterization of groundwater flow systems on different spatial scales. On a small scale (102 m) a principle component analysis was conducted on time series of water level measurements in a river and in the adjacent groundwater of the floodplain. According to different responses of the groundwater on surface water induced pressure waves, areas with different hydraulic connectivity were identified. For the same aquifer, the small scale nutrient (≤ 101 m) distribution in the near-surface groundwater was also investigated. A close linkage between the nutrient distribution and the small-scale topography within the floodplain was detected. Both studies illustrate that comparatively “easy-to-measure” data in a high spatial and temporal resolution are sufficient for the detection of subsurface preferential flow paths as well as spots of potential nutrient sources. On a larger scale (103 m) a study was conducted which investigated the impact of groundwater leakage on the characteristics of a local flow system. Therefore, a simple 2D numerical groundwater model (steady state) was set up for the subsurface catchment of a lake. The model could illustrate that leakage lead to a decrease of the amount of groundwater, which enters the lake and, an increase of the length of the groundwater flow paths. Hence, groundwater leakage needs to be considered for the determination of local groundwater flow systems.

The limnological approach based on the hypothesis that physical and chemical differences between groundwater and lake water can be used to identify areas, where groundwater is exfiltrating. The presented studies tested if it is possible to use temperature measurements at the lake surface (thermal infrared imaging and in situ measurements) in spring, when the warmer groundwater is floating on the colder lake water. However, the comparison of the temperature measurements with hydrogeological and lake data, indicate that in the present case the observed temperature pattern are the result of lake internal processes. However, the aerial detection of groundwater spots should be possible at least for lakes with small volumes and intense groundwater discharge.

Sub-project: **Systems dynamics of aquatic-terrestrial interfaces**

PhD: Christian Lehr

PIs: Gunnar Lischeid, Dörthe Tetzlaff, Gunnar Nützmann

At the Freienbrink research site, hydraulic connectivity between the Spree river and the adjacent aquifer was investigated for a four years period (Lehr et al. 2015), including a former oxbow. The oxbow became reconnected to the stream and the clogging layer in the

oxbow was excavated. We expected increasing hydraulic connectivity between oxbow and aquifer after restoration of the stream. A principal component analysis of time series of groundwater heads and stream water levels was performed. The first component captured the propagation of the pressure signal induced by stream water level fluctuations throughout the adjacent aquifer. Thus it could be used as a measure of hydraulic connectivity between stream and aquifer that revealed a complex spatial pattern. After restoration of the stream no change of hydraulic conductivity was observed for the former shortcut. There is some evidence that the pattern of hydraulic connectivity at the study site is by far more determined by the natural heterogeneity of hydraulic conductivities of the floodplain sediments rather than by the clogging layer in the oxbow.

The Pleistocene landscape in North Europe, North Asia and North America is spotted with thousands of natural ponds called kettle holes. They are biological and biogeochemical hotspots. Due to small size, small perimeter and shallow depth biological and biogeochemical processes in kettle holes are assumed to be closely linked to the dynamics and the emissions of the terrestrial environment. In the Quillow catchment in the Uckermark region, about 100 km north of Berlin, Germany, 62 kettle holes have been regularly sampled since 2013 (Lischeid et al. 2017). Spatial patterns of kettle hole water concentration of (earth) alkaline metals and chloride were fairly stable, presumably reflecting solute concentration of the uppermost aquifer. In contrast, spatial patterns of nutrients and redox-sensitive solutes within the kettle holes were hardly correlated between different sampling campaigns. Correspondingly, effects of season, hydrogeomorphic kettle hole type, shore vegetation or land use in the respective catchments were significant but explained only a minor portion of the total variance. It is concluded that internal processes mask effects of the terrestrial environment. There is some evidence that denitrification and phosphorus release from the sediment during frequent periods of hypoxia might play a major role (Lischeid et al. 2017).

Numerous discussions with layperson as well as with scientists from related disciplines had revealed some widespread misconceptions about the linkages between surface runoff, vadose zone, groundwater, lakes and streams. Thus PhD students of the Institute of Landscape Hydrology at ZALF set up a physical model of a complex aquifer in a sandbox with fully glazed sidewalls as a didactic model to be shown to laypersons. However, in addition to that, and following the invitation of the then President of EGU, the model was presented at the General Assembly 2014 of the European Geosciences Union as well. Here it attracted the attention of numerous hydrologists as well as of scientists of other earth science disciplines. Based on the questions, comments and requests during the conference two videos have been compiled and published (Lehr et al. 2017), including a construction manual and an explanation of some experiments to be performed with the model.

Sub-Project: Spatial organisation of groundwater - surface water interactions in an upland catchment: integrating hydrometric, tracer and modelling approaches

PhD: Maria Blumstock

PIs: Dörthe Tetzlaff, Chris Soulsby, Gunnar Nützmann

In the upper latitudes of the Northern Hemisphere, such as the UK, Groundwater (GW) dynamics play a crucial role in runoff generation and hydrologic connectivity between hillslopes and streams, especially during prolonged periods of low flows. GW-surface water interactions are usually very complex and highly variable on a spatial and temporal scale. Despite the considerable importance of baseflow conditions, studies in montane terrains have traditionally focused on high flows. Understanding the interrelations between landscape characteristics and runoff generation, and the role of landscape heterogeneity in modifying stream hydrochemistry, are vital for quantification and prediction of dominant hydrological processes, also with respect to possible future land use and climate changes.

The Bruntland Burn catchment intersects two dominant bedrock units (granite and metasediments) and is overlain by thick deposits of diverse drift deposits. Typical of these montane headwaters are thick layers of peat soils fringing the stream channel, maintaining high hillslope-stream connectivity. Saturated conditions in the near-stream area are

sustained through GW seepage from more freely draining soils further upslope, with water moving vertically and recharging deeper flow paths.

This thesis presents a study of the heterogeneous nature of GW-surface water interactions at the hillslope and catchment scale. Groundwater fluctuations were measured by 14 shallow GW wells, installed within three contrasting hydrogeological units. The hydrological monitoring network was complemented by four multi tracer surveys of the whole stream network. By analysing stream water samples for major anions, cations and stable isotopes we were able to capture the increased influence of GW to the stream during a 10 year return period drought.

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The Bruntland Burn catchment was shown to have highly dynamic GW stores, with each landscape unit translating into different rainfall-runoff processes. It was further shown that soil characteristics were the strongest predictors for variability in GW dynamics. Each soil type was characterised by a unique storage-discharge relationship and threshold response with a certain GW level above which lateral flow dominated. On the lower hillslope, predominating lateral flow and little recharge to depth is supported by hydrologically responsive soils. Connectivity between the steeper slopes and the valley bottom however, needed persistent wet periods to overcome storage thresholds. Here, vertical flow paths recharging deeper GW dominated, with GW levels falling below the soil layer into the underlying drift.

GW dynamics were characterised by estimating the lag time in relation to stream flow. GW responses varied seasonally, with GW on the upper hillslope showing peak responses up to several hours after stream flow, and GW responses usually a few hours prior to the stream responses in the lower slopes. Landscape controls on the spatial organisation of GW dynamics were strongest during baseflows and for small events. During the wettest periods, the combination of the catchment being close to maximum saturation and limited storage weakened such controls, and GW responded a few hours prior to discharge throughout the catchment on average. This was also reflected by relatively well-mixed homogeneous stream chemistry, reflecting near-surface sources of stream flow draining from acidic riparian peat soils. Increasingly heterogeneous hydrochemical patterns emerged during dry spells, with temporal changes in stream chemistry being highest in areas of high peat coverage. As flows declined and GW dominance of flow increased, GW discharged through the stream bed delivering water from deeper, older GW sources. This reflected winter recharge and longer times for chemical weathering reactions within the minerogenous material. The high spatial variability of the stream chemistry reflects the diverse geochemical signature of the bedrock geology and the distribution of GW stores in drift deposits. The largest sources of GW appeared to be located in the drift situated in the lower catchment where the most marked increase in weathering-derived ions was observed. Depleted, non-fractionated isotope signatures confirmed deeper inflows. However, in some places the GW discharged across valley bottom wetlands and was subjected to evaporative losses and fractionation from the surface of the peat soils.

Geochemical tracers and stable isotopes have proved to be useful for investigating geographical source areas and time scales of runoff. To further support empirically based findings, field data were integrated into a modelling framework to simulate GW-surface water interactions along a representative 2D hillslope transect in Modflow-NWT. To better inform model structure, geophysics (ERT) was used as a basis for defining the hillslope domain and associated boundary conditions. The model was calibrated to measured head values along the hillslope transect over a 12 month period, followed by a 17 month model test period. The

model set-up enabled us to quantify deep and shallow GW flow paths through particle tracking analysis. It was shown that shallow pathways have a ~5 times shorter residence time, maintaining high water tables in the riparian peatlands, than deeper flow paths discharging through the drift and directly into the stream. This model study showed that the hydrogeology underlying the study site is very complex with a diverse range of GW stores and corresponding, resulting in complex spatial patterns of runoff generation. It was further shown that the largest sources of GW were located in drifts in the lower catchment, which were 1.5 the annual precipitation.

This study has improved the understanding of GW-surface water interactions on a hillslope and catchment scale. It was found that relatively well mixed near-surface sources of stream flow predominated in wetter conditions, whilst baseflows are variable and reflect a diverse range of GW stores, especially in the drift. The application of a modelling approach has demonstrated the potential to represent catchment processes in montane headwater catchments dominated by peat. Finally, this work illustrated the utility of a basic model to predict GW flow paths, highlighting how water and solutes are stored and released in montane headwater catchments. Through integrating hydrochemical and hydrological information, such as attained from ERT surveys, GW level measurements and from tracer studies, more reliable insights into catchment dynamics in cold climate environments were gained.

Topic 2: Internal boundaries in aquatic systems

Sub-project: Redox Processes of Organic and Mineral Geochemical Phases at Aquatic Interfaces

PhD: Maximilian Lau

PIs: Michael Hupfer, Henning Jensen, Jörg Gelbrecht

In this subproject, biogeochemical electron exchange reactions at aquatic interfaces were explored. The work focuses on the kinetic and thermodynamics as well as the ecological implications of reactions that involve redox-active moieties in natural organic matter (NOM). The research builds on earlier findings that confirmed that NOM may act as terminal electron acceptor (TEA) for electrons released in microbial respiration. This property was identified to derive from Quinone moieties that are ubiquitously found in NOM from terrestrial and aquatic environments and that may undergo reversible reduction to the respective hydroquinone. Previous methodological advances allowed for a rapid, direct and precise quantification of the electron accepting and donating properties of Quinone moieties in dissolved NOM (DOM) by mediated electrochemical analysis.

In the project, the previously established mediated electrochemical analysis was adapted and used in the characterization of redox properties of particulate natural samples that contain both redox active iron and organic matter (henceforth referred to as “geochemical phases”). For the first time, the reduction of geochemical phases in sediments of lakes and wetlands was directly monitored. Measurements confirmed that microorganisms transferred electrons that were released during microbial respiration to the organic and inorganic electron acceptors in the particulate phase. Particulate organic matter in the sediments was found to provide a capacity to accept or donate electrons of $650 \mu\text{mol e}^- \text{gC}^{-1}$. This value considerably exceeds values found in many previous studies that relied on the indirect quantification of the electron accepting capacities using chelated Fe(II) as redox probe. Analysis of the combined electron-accepting properties of NOM- and Fe-bearing geochemical phases revealed pH-dependent electron fluxes between NOM and Fe species. This finding is in line with earlier studies that suggested overlapping Eh distributions of individual redox-active sites in NOM and Fe in clay minerals. It further points towards a more vivid electron exchange between conjoined redox-active species within heterogeneous matrices like sediments.

Aiming for the spatiotemporal analysis of the dynamics that organic and inorganic TEA species (i.e., nitrate, sulfate, Fe- and Mn oxyhydroxides) in freshwater sediments are subject to, a mesocosm experiment was set up to simulate changes in oxygen availability at the sediment surface. At sediment surfaces oxygen is either consumed in aerobic respiration or when previously reduced species are oxidized. In the latter process, these species regenerate their electron-accepting capacity. The use of mediated electrochemical analysis allowed for the quantification of the redox state of the geochemical phases during their reduction and re-oxidation. Hence, the electron fluxes initiated by the oxic re-generation of the TEAs nitrate, sulfate, Fe(III), Mn(IV) and quinoid moieties in NOM were directly monitored instead of modeled from the species' one-dimensional distribution profiles in interstitial waters. With this set of direct methodologies, the redox-driven biogeochemical processes triggered by system disruptions at the sediment-water interface were closely observed.

Ecosystem dynamics initiate redox cycling

Many aquatic ecosystems undergo recurring fluctuations in oxygen availability. The associated disruptions in redox conditions can cause cyclic reduction and re-oxidation of redox-active species on different timescales. In lakes, oxygen budgets are coupled to the dynamics of benthic redox processes. In seasonally stratified lakes, extended sediment volumes are exposed to oxic conditions (dissolved $O_2 > 1 \text{ mg L}^{-1}$) only upon lake overturn. A combined field and laboratory study of Lake Scharmützelsee showed that this seasonal mixing event introduces a finite amount of oxygen to the hypolimnion and that about 50% of the subsequent sediment oxygen consumption is exclusively associated with the re-generation of TEA species. These reduced species previously formed in the sediment when microorganisms decomposed organic matter during anaerobia.

While lake's overturn can completely de-stratify lakes and mix large quantities of epi- and hypolimnetic waters, small-scaled dynamics in temperature and oxygen availability may confine discrete parts of the water column where physicochemical conditions oscillate. In the studied lake Große Fuchskuhle, a transient thermocline cyclically introduces oxygen to hypoxic hypolimnetic waters close to the pelagic redox interface. In the said lake which is influenced by an adjacent bog area, high concentrations of DOM meet low abundance of inorganic electron accepting species. In this and comparable systems, organic TEAs may represent an important constituent of the total pelagic electron acceptor capacity. Due to the rapid and reversible redox reactions of dissolved NOM, reduced organic TEAs are re-generated upon dislocation to oxic parts of the water column. Results show that diurnal fluctuations of oxycline depth shape a micro-environment selecting for microbial species that are released from TEA limitations by DOM in oxidized state. Pelagic microbial communities subjected to identical amounts of DOM that are in different oxidation states differed by more than 50% after one day.

This work substantiates earlier findings that suggested that NOM may be an important TEA species in many aquatic and terrestrial ecosystems. NOM reduction in microbial respiration was shown to directly affect critical system parameters as bacterial activity, oxygen budgets and aquatic biodiversity.

Both the microbial reduction and subsequent abiotic oxidation of (hydro-)quinoid moieties in NOM are sufficiently fast for a relevant interaction with oxycline fluctuation. Given that organic TEAs are cyclically regenerated, a significant share of ecosystem respiration could be linked to NOM reduction.

This study adds to the new and important findings on the role of electron exchange reactions in NOM-rich environments. As of today, linking the chemistry of aquatic turnover processes with the microbiological and physical conditions at redox interfaces remains challenging. In conclusions, by providing several cases from aquatic environments, this thesis contributes to the mechanistic underpinning of NOM reduction in microbial respiration. The results clearly prompt for further research - especially regarding the competitive inhibition of other respiration pathways, including the reductive production of the potent greenhouse gas methane.

Sub-project: Gas exchange across boundaries in lakes: sediment-water, hypolimnion-epilimnion, water – atmosphere; field and lab studies to identify methanotrophic activities by means of proteomics

PhD: Nina Pansch (nee Ullrich)

PIs: Mark Gessner, Peter Casper, Henning Jensen

Methane (CH₄) is an important greenhouse gas produced in anoxic sediments of lakes, which have recently been recognized as an important source of methane emitted to the atmosphere. However, microbial methane oxidation (methanotrophy) at the oxic/anoxic interface can be key in controlling these emissions. Indeed, up to 99% of CH₄ dissolved in the water column can be oxidized by methanotrophs, depending on lake characteristics, suggesting that methanotrophy can play a major role in mitigating methane emissions to the atmosphere. The subproject 'Inner Boundaries – Greenhouse Gas Exchange across Aquatic Interfaces' of AQUALINK focused on this process and the associated microbes and enzymes.

We applied a proteomic approach to determine enzyme expression patterns of methanotrophs in response to methane enrichment of lake water. In a small-scale incubation experiment with natural bacterial communities, we compared enzymes involved in methane metabolism between control and methane-enriched hypolimnetic water simulating high (~1 mM) and low (~0.001 mM) methane concentrations at oxyclines in lakes. Methane was effectively consumed when the supply was high, reducing oxygen levels from 0.40 mM (12.9 mg/L) to 0.09 mM (3.0 mg/L), well below those in the controls. The dominant key enzyme of microbial methane oxidation, particulate methane monooxygenase (pMMO), was identified in both enriched and control flasks. However, enzymes potentially involved in methane metabolism via the RuMP pathway and serine cycle were essentially restricted to the enriched flasks. All enzymes had best sequence matches with so-called type I methanotrophs, which differ in the metabolic pathways to use methane from type II or type X methanotrophs, for which no compelling indication was found in our proteomic analysis, even though four enzymes of the serine cycle were identified. Overall, our proteomics approach provides convincing evidence that a suite of genes required for methanotrophy are quickly expressed when the presence of both methane and oxygen creates conditions characteristic of oxyclines in lakes.

Microbial aerobic methane oxidation in lakes is restricted to narrow oxic-anoxic interfaces called oxyclines. Two fundamentally different types can be distinguished: oxyclines at the sediment-water interface are located just beneath the sediment surface in lakes with well oxygenated hypolimnetic water, whereas oxyclines are shifted upwards in the water column of stratified lakes during summer when the hypolimnion becomes anoxic. We investigated methanotrophs at these oxyclines in five contrasting lakes in north-eastern Germany to elucidate the factors shaping the methanotrophic communities in such distinct conditions. High-resolution vertical water-column profiles of oxygen and methane were taken to locate the precise depth of the oxycline before taking water samples at six discrete depths. Potential and in-situ methane oxidation rates were determined by ¹³CH₄ incubation experiments. Additionally, proteomic analyses were applied to determine enzyme expression patterns in relation to lake characteristics. The proteomic analysis revealed a total of 194 proteins and showed that γ -Proteobacteria were clearly dominant, although α -Proteobacteria also occurred at all sites. Two key enzymes for methane oxidation, MMO and MDH, were detected in a few sites. The upward shift of the oxycline in the water column during summer stratification greatly impacted methane oxidation capacities and in-situ oxidation rates. Expression patterns of enzymes involved in methanotrophy were also changed. These results suggest that CH₄ emissions to the atmosphere vary notably as a function of oxycline location in the lake sediment or water column.

Both types of oxyclines occur in a single lake during summer stratification and hypolimnetic oxygen depletion. Oxyclines in the water column above anoxic hypolimnia move to the sediment in shallow areas of lakes, such as the littoral zone, where the entire water column is fully oxygenated and the oxycline forms just beneath the sediment surface. The oxyclines of six lakes representing a nutrient gradient from oligotrophic to eutrophic conditions were

sampled in north-eastern Germany. To differentiate between active and inactive methanotrophic communities, we used high-throughput sequencing (HTS) of the DNA and RNA of two functional genes (*pmoA*, *mmoX*) specific to methanotrophs. Quantitative PCR (qPCR) was used to reveal the ratio of methanotrophs to the overall bacterial community. Furthermore, we applied a proteomic approach to determine enzyme expression patterns in response to lake characteristics and location of the oxycline. The mass spectrometric analysis revealed a total of 1850 identified proteins, 87 of which could be assigned to methanotrophic genera. Two of them had best matches with α -Proteobacteria and 85 to γ -Proteobacteria, together representing seven different genera. Ten enzymes were identified as methane monooxygenases (MMO), which is the key enzyme of methane oxidation. Three of these MMOs could be clearly identified as particulate MMOs (pMMO), two as soluble MMOs (sMMO) within the γ -Proteobacteria, and five MMOs as “subunit B of MMO”.

Sub-project: **The effect of groundwater on benthic primary producers in an oligotrophic lake**

PhD: Cecile Perillon

PIs: Sabine Hilt, Henning Jensen, Gunnar Nützmann,

In littoral zones of lakes, multiple processes determine lake ecology and water quality. Lacustrine groundwater discharge (LGD), that most frequently takes place in the littoral zones, can transport or mobilize nutrients from the sediments and there, contribute significantly to lake eutrophication. Furthermore, littoral zones of lakes are the habitat of benthic primary producers, namely submerged macrophytes and periphyton that play a key role in the lake food web and for lake water quality. Groundwater-mediated nutrient-influx can potentially affect the asymmetric competition between submerged macrophytes and periphyton for light and nutrients. While rooted macrophytes have superior access to sediment nutrients, periphyton can negatively affect macrophytes by shading. LGD may thus facilitate periphyton at the expense of macrophytes, however, studies on this hypothesized effect are missing.

This PhD project in AQUALINK aimed at determining how LGD influences periphyton, macrophytes and their interaction. Laboratory experiments were combined with field experiments and measurements in an oligo-mesotrophic hardwater lake. In the first study, a general concept was developed based on a literature review on the existing knowledge regarding potential effects of LGD on nutrient, inorganic and organic carbon loads to lakes and their effects on periphyton and macrophytes. The second study includes a field survey and experiment on effects of LGD on periphyton in an oligotrophic, stratified hardwater lake (Lake Stechlin). It shows that LGD, by mobilizing phosphorus from sediments, significantly promotes epiphyton growth, in particular at the end of summer when phosphorus concentrations in the epilimnion are low. The third study focuses on potential effects of LGD on submerged macrophytes in Lake Stechlin. It revealed that LGD may have contributed to an observed change in macrophyte community composition and abundance in shallow littoral areas of the lake. Finally, a laboratory experiment was conducted mimicking conditions of a seepage lake. Groundwater circulation was shown to mobilize nutrients from the sediments, which significantly promoted periphyton growth. At high periphyton biomass, macrophyte growth was negatively affected confirming the initial hypothesis.

More generally, this thesis shows that groundwater flowing into nutrient-limited lakes, may import, or mobilize nutrients which at first promote periphyton, before provoking radical change in macrophyte populations and finally changing lakes trophic state. Hence, the eutrophying effect of groundwater is delayed and, at moderate nutrient load, partly dampened by benthic primary producers. The present research emphasizes the importance and complexity of littoral processes, and the need to further investigate and monitor benthic environment. As present and future global change can significantly affect LGD, the understanding of these complex interactions is a pre-requisite for a sustainable management of lake water quality.

Topic 3: Revitalization and management

Sub-project: **Investigation of Groundwater-Surface Water Interactions with Distributed Temperature Sensing (DTS)**

PhD: Anne-Marie Kurth

PIs: Mario Schirmer, Jörg Lewandowski, Gunnar Nützmann

Goal of this part of the AQUALINK project was to undertake field and lab studies to investigate processes at the groundwater-surface water interface (GSI) using the distributed temperature sensing technology. The research focused on the effect of heterogeneity in flow at the Niederneunforn study site (Switzerland) and additional field sites. Groundwater-surface water interactions are a vital necessity for aquatic ecosystems as they control the water temperature, the availability of nutrients, dissolved oxygen and the water quality in the hyporheic zone. A lack of groundwater-surface water interactions may result in the deterioration of ecosystem health and functioning. Studies between 1997 and 2008 have shown that 22 % of Swiss water courses were severely degraded, e.g. engineered or covered. As a consequence, river restoration was made a legal obligation, stipulating the restoration of 4000 km of degraded rivers and streams over the course of the next 80 years. For this thesis a review of Swiss river restoration data between 1979 and 2012 for 13 of the 26 Swiss cantons was performed.

Results indicated that restoration activities had steadily increased since 1979, with an average restoration rate of 9.8 km/year. An analysis of the restoration techniques revealed interesting geographical trends. In western Switzerland, more sustainable combinations of restoration measures, such as bioengineering or water quality improvements, were favoured. Cantons in central and eastern Switzerland, on the other hand, preferred single restoration measures with a higher degree of mechanical intervention. In general, the evaluation of restoration effects was only reported for less than 10 % of all investigated restoration projects. These mainly focussed on the number of flagship species, such as trout.

None of the investigated projects tested whether river restoration had re-established groundwater-surface water interactions. Hence, this thesis aims at investigating the effects of river restorations on groundwater-surface water interactions. A number of techniques are commonly used to investigate groundwater-surface water interactions, including geochemical, hydrogeological and physical approaches. In the present study a combination of approaches is employed, with the main focus being on the physical parameter of water temperature. The latter is investigated with Distributed Temperature Sensing (DTS). DTS is used to measure temperature differences between ground- and surface water in surface water bodies. So far, the existing DTS methods have enabled the investigation of groundwater-surface water interactions under gaining conditions in small brooks. In order to investigate the effect of river restoration on groundwater-surface water interactions, however, a method applicable in both gaining and losing conditions, and which is suitable for water courses of all sizes is required. For this purpose, a new methodology, the PAB approach, has been developed, which combines passive (P) and active (A) DTS methods with the burying (B) of the fibre-optic cable in the subsurface. This approach enables long-term distributed investigations of groundwater-surface water interactions under gaining and losing conditions in water courses of all sizes. The active DTS method in the PAB approach, however, requires the direct presence of an operator controlling the heating of the fibre-optic cable. In order to circumvent this limitation and enable long-term temperature measurements with the PAB approach in remote areas, an autonomous DTS system (ADTS system) has been developed. This system combines several advantages, such as remote control, automated data transfer, and automated heating. By aid of the ADTS system and the PAB approach, the effect of river restoration on groundwater-surface water interactions has been investigated in an urban stream.

Results indicate that the installation of gravel islands increased the rate of surface water downwelling. Generalising the results, it may be assumed that such changes to river morphology will have a positive effect on the rate of groundwater-surface water interactions. Therefore, river restoration may be successful in enhancing groundwater-surface water

interactions. Concerning the newly-developed DTS method and measurement system, it could be shown that the combination of the ADTS system and the PAB approach is a powerful tool for the investigation of groundwater-surface water interactions. In future river restoration projects, this tool should be employed for evaluating its success in re-establishing groundwater-surface water interactions.

Sub-project: Effects of changes in water level, land use and regime shift on lake sediments

PhD: Anna-Marie Klamt

PIs: Henning Jensen, Sabine Hilt, Michael Hupfer

Sediments provide various functions in lake ecosystems. They are an important part of the nutrient cycle since they are sinks and sources for nutrients. Further, they represent chronological sequences of deposited material and are thus valuable archives which contain information about past environmental conditions in and around lakes.

This PhD project investigated how water level changes affect (recent) littoral sediments and how land use changes and trophic regime shifts are reflected in sedimentary profiles and have affected lake ecosystems.

The studies in Klamt et al. (2016) comprised lab experiments in which intact sediment cores from the Danish soft water lake Skærsø (a Lobelia lake) were used to simulate a drawdown and a subsequent re-flooding event. The results showed that overall drying did not lower the initial high phosphorus (P) binding capacity of the littoral sediments. This was confirmed by a small scale long-term drying (348 days) and rewetting (228 days) experiment. The findings have implications for lake management since it is concluded that drawdowns can be an appropriate restoration measure for endangered Lobelia lakes, however, only provided that the desired isoetid plant species spread during the desiccation period.

The paleo studies in Klamt et al., 2017 investigated how changes in the dominant catchment vegetation (occurring during the last ca. 200 years) were reflected in the sediments of a Danish soft water lake (Grane Langsø) and how these changes influenced the lake ecosystem. A novel combination of paleo proxies (pollen, microfossils, metals, different P forms, organic matter, carbon and nitrogen contents) revealed that 1) the reduction of deciduous trees in the watershed seemingly reduced the calcium (Ca) supply to the lake and thereby its buffering capacity. This development was accompanied by decreased abundances of Ca-dependent species and subsequent increases in acidophilic species. 2) The sedimentary contents of organic matter, non-reactive P and humic-bound P were evidently higher in sediments deposited during the time when deciduous trees were abundant, which is probably linked to a stabilising effect by Ca. 3) An erosion event clearly reduced the amounts of microfossils of isoetid species and characeans, indicating a reduction in their maximum distribution depth because of lower water transparency. Overall, these results are of importance within lake management by convincingly showing how land use changes may (irreversibly) affect environmental conditions and species composition in soft water lakes and the storage of organic matter and P in their sediments.

In the third part of the PhD project a regime shift from macrophyte to phytoplankton dominance was revealed in the sediments of the German hard water lake Gollinsee with a combination of proxies including sediment color, TOC:TN (total organic carbon:total nitrogen) ratios and stable C isotopes. Beyond that, it was found that total P retention in the sediments had increased after the shift from macrophyte to phytoplankton dominance by more than 4-times, which was mainly caused by increased retention of biogenic P (NaOH-NRP). The proportion of permanently buried P (mainly Ca bound P) was reduced from 81 to 67 % of total P. Therefore, it is assumed that relatively lower Ca-P-co-precipitation in combination with increased sedimentation of bioavailable P forms enhanced internal P cycling and maintains phytoplankton dominance. This positive feedback loop is likely the fate of many eutrophied lakes that underwent a regime shift.

The use of sequential chemical extraction of various P forms in paleolimnological studies, as in the second and third manuscript, is quite uncommon. One outcome of the thesis is that the sediment stratigraphy of various P forms may provide valuable information about past changes in lakes and watersheds even if deposited P is known to undergo post-depositional mobility. Further information may be gained by combining the chemical extraction with ³¹P-NMR analyses.

PhD dissertations

Franziska Pöschke: Approaches to identify groundwater discharge towards and within lowland surface water bodies on different scales. Humboldt-Universität zu Berlin, 2016.

Maria Blumstock: Spatial organisation of groundwater - surface water interactions in an upland catchment: integrating hydrometric, tracer and modelling approaches. University of Aberdeen, 2016.

Maximilian Lau: Redox processes of organic and mineral geochemical phases at aquatic interfaces. University of Greifswald, 2016.

Cecile Perillon: The effect of groundwater on benthic primary producers in an oligotrophic lake. University of Potsdam, 2017.

Anne-Marie Kurth: Investigation of groundwater-surface water interactions with distributed temperature sensing (DTS). University of Neuchatel, 2016.

Anna-Marie Klamt: Effects of changes in water level, land use and regime shift on lake sediments. University of Southern Denmark, 2016.

The PhD dissertations of Nina Pansch and Christian Lehr are still pending but in preparation.

Publications (PhD candidates of AQUALINK are in bold)

Lehr, C., Pöschke, F., Lewandowski, J., Lischeid, G. 2015. A novel method to evaluate the effect of a stream restoration on the spatial pattern of hydraulic connection of stream and groundwater. *Journal of Hydrology*. 527, 394 – 401.

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Workshops and summer schools

1. Opening workshop, 17.09. – 20.09.2012, Berlin and Groß-Väter, kick-off meeting and discussing the different PhD projects
2. Workshop, 22.08. – 23.08.2013, Aberdeen, report of the PhDs, internal organization of the Graduate School (homepage, workshops, guest scientists, research visits/trips, common publications)
3. International summer school in Aberdeen, 25.08. – 30.08.2013, organized by Dörthe Tetzlaff and colleagues once a year with outstanding international scientists in the field of hydrology, ecohydrology and landscape ecology.
4. International summer school for “Lake management and lake restoration” (course arranged by Aqualink and CLEAR), Biological Institute, Field Station in Søgaard (DK), 02.08.- 9.08.2014, organized by Henning Jensen, Kasper Reitzel and Michael Hupfer, Lectures are given by Danish experts and by IGB Aqualink participants in the fields of lake restoration, impact of catchment on water quality, macrophytes, biomanipulation, groundwater; field trip and exercises
5. Workshop, 19.08. – 21.08.2015, Neuglobsow, lectures of guest scientists (Peter Engesgaard, Stefan Krause), reports of the PhDs, broad discussion about the idea of Aqualink and what follows – news from research activities about aquatic interfaces, the idea of a special issue in *Limnologica*.

In the minutes from the last workshop in 2015 we wrote:

With a Special Issue of LIMNOLOGICA we want to highlight aquatic water interfaces and their functions in the water cycle and in an eco-hydrological perspective based on a cross-disciplinary view. The development of the AQUALINK idea was joined to the so called “cross-cutting research topic” at IGB with the same title that were established in order to focus the research on different boundaries between terrestrial and aquatic systems. In practice, a central topic was the question how to quantify the exchange between terrestrial and aquatic ecosystems which are manifested for example in water table fluctuations followed by drying and re-wetting of large areas. These processes are primarily driven by the hydrological

regimes of surface and subsurface waters, and thus, affected by climate change and changes in land use, but have also effects on socioeconomic issues, conflicts, water management strategies, interactions between ground and surface waters, the chemical and the ecological status of waters, and others. Another important issue was the problem of high and low waters as permanent issue of concern.

In the meantime there are several other initiatives dealing with interfaces in aquatic systems, mainly in hyporheic zones (EU ITN projects Interfaces and HypoTRAIN in 2014 and 2015). But focusing on a metropolitan area as a case study there was also hypothesized that urban water interfaces play key roles in the transformation and transport of water, matter, and/or energy in the urban water system, especially because natural and technical system components are mixed (DFG Graduate School "Urban Water Interfaces")

Special Issue of LIMNOLOGICA

The interfaces between terrestrial and aquatic ecosystems have recently been the subject of several interdisciplinary special issue publications as well as key seminal review and vision papers. We started with the idea of a "special issue" within the international well-known journal *Limnologia*, edited by a group of scientists, which are involved in the AQUALINK project:

Michael Hupfer and Gunnar Nützmann (IGB)

Peter Engesgaard (University of Copenhagen, Dept. Geosciences and Natural Resources Management, Copenhagen Denmark)

Henning Jensen (SDU), and

Stefan Krause (School of Geography, Earth and Environmental Sciences, University of Birmingham, Edgbaston, UK).

This special issue makes new contributions to limnology and water science by presenting 15 papers that investigate the complex interactions between the physical, biogeochemical, and ecological processes occurring at aquatic interfaces across a range of different spatial and temporal scales. The special issue will be published with 15 contributions in *Limnologia* in 2018 as Volume 68.

As shown in the special issue an improved understanding and a quantification of the matter flux across various reactive zones in the landscape needs both empirical investigations at the field and laboratory scales and new modelling approaches. Currently, the gap between ever-more detailed basic research at the very small scale and the use of these findings to elucidate the quantitative role of aquatic interfaces at the landscape level seems to be increasing. The upscaling of these processes remains a largely unresolved challenge and can only be tackled by forging a closer link between studies on the processes and modelling. Additionally, groundwater and lake water models are still poorly linked. There is a need for development and application of reactive transport model that can help improve our understanding at the aquatic interfaces by linking hydrological and biochemical processes and for better-linked regional-scale groundwater-surface water models that can help predict the turnover and export of matter in and from a catchment. More systematic research need to be paid to the previously disregarded role of groundwater as a source of nutrients in lakes. One of the central problems related to research on aquatic interfaces is the spatial heterogeneity. Neglecting this heterogeneity can lead to the over- or underestimation of the quantitative importance of matter transport. Therefore, there is an urgent need to identify the effects of the small-scale spatial heterogeneity of aquatic interfaces on the exchange fluxes as well as transformation. In addition, the monitoring of processes at such interfaces requires the determination of patchiness through novel and innovative sensing technologies that can cover different spatial heterogeneities and short-term temporal fluctuations. These include using automatic sensors loggers and remote sensing methods for recording data in real-time and to monitor rapid flow alterations or changes in water quality. Research on aquatic interfaces is of increasing importance for sustainable water management. However, this will require the fast transfer of new scientific knowledge into practice. On the other hand, the

sites manipulated for management measures are ideal as “field labs” for empirical research for understanding the function and efficiency of aquatic interfaces. A promising direction in the research on aquatic interfaces is the focus on their function as a “control point” in a changing world. The impact of climate change is of extremely high interest, particularly with regards to the consequences of increases in the temperature on the sediment-water exchange, changes in the mixing regime, and fluctuations in the water level of lakes. The transient nature of interfacial dynamics is still poorly understood; however, this understanding is essential for resource management and tackling future scenarios. When studying the complex interactions between the various processes inherent to aquatic interfaces, one must consider this issue from the perspective of different disciplines. Therefore, the main challenge with regards to research on aquatic interfaces is to move past the traditional boundaries between various research disciplines.

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SPECIAL ISSUE: Aquatic interfaces and linkages: An emerging topic of interdisciplinary research
Guest Editors: Michael Hüppler, Peter Engesgaard, Henning Jensen, Stefan Krause, Gunnar Nützmann

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Assessing net-uptake of nitrate and natural dissolved organic matter fractions in a revitalized lowland stream reach
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Cover-picture: Rewetted peatlands act as important semi-aquatic interface for nutrient retention between rivers and the terrestrial environment (River valley Trebeltal in NE Germany) (Photo: Dominik Zak)