# Credible Targets for Environmental Management

## Evaluating Reconstructions of Reference Values for Ecological Status in Surface Waters


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# Credible Targets for Environmental Management. Evaluating reconstructions of reference values for ecological status in surface waters


## Abstract

In the year 2000, countries of the European Union agreed to harmonise water management by implementing the European Water Framework Directive (WFD). The WFD uses good ecological status as the overall goal, which is defined as minor deviation from an undisturbed state. Reference conditions provide the means of estimating water status and ecological integrity, which are the focal point of decision-making processes. When determining reference conditions, the WFD prescribes the use of all available knowledge so as to reduce the predictive uncertainties. While the goal of achieving conditions similar to an undisturbed state can be desirable, the use of reference conditions as a target creates inherent complexity on multiple levels. This thesis focuses on evaluating the implications of different interpretations and implementations of WFD reference conditions for status assessment of Swedish surface waters. Specifically, the objectives were to: (I) define the implications of different reference condition criteria and understand the diversity of ways in which the term is used, (II) compare independent estimates of reference conditions estimated using historical fish archives and biogeochemical modelling for acidification assessment, (III) evaluate recovery from acidification of acid sensitive Swedish lakes and analyse potential confounding factors influencing future management, (IV) develop empirical models to estimate historical and present temporal trends in lake total organic carbon concentrations.

Together and individually these studies have developed methods and models in which predictive uncertainties associated with reference conditions have been reduced. This thesis show how multiple methods can be used to reduce the uncertainties in surface water reference condition estimates as well as presenting approaches to the use of all available information as prescribed by the WFD. Furthermore, this thesis highlight the multiple competing, but equally valid, approaches to estimate reference conditions based on knowledge from different sources. In this thesis, it is argued that representations of the undisturbed state are a moving target and greater acceptance for multiple reference conditions based on all available knowledge is needed.



*Keywords:* EU-WFD, Reference conditions, Undisturbed state, Acidification, MAGIC, TOC, Modelling, Policy, Water management, EQC, Organic Matter



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# Sammanfattning

Medlemsländerna i Europeiska unionen accepterade år 2000 EU ramdirektiv för vatten som syftade till att harmonisera vattenförvaltningen inom unionen. Det övergripande målet för Ramdirektivet är att uppnå god status i alla vattenförekomster. Med god status avses att man bara tillåter en viss avvikelse från ett referenstillstånd vilket kännetecknas av obetydlig mänsklig påverkan. Det nya i detta direktiv var att man lämnade den sektoriella vattenförvaltningen till att samla alla delar under ett direktiv med ekologisk integritet som utgångspunkt. En viktig del i att bedöma referenstillstånd är att använda all tillgänglig kunskap för att minska osäkerheten i klassificeringen av referenstillstånd. Att ramdirektivet för vatten använder sig av referenstillstånd för att sätta miljömål kan, i många fall vara önskvärt. Samtidigt kan det skapa komplexitet på sociala, ekonomiska och vetenskapliga nivåer eftersom ordet i många fall har olika betydelser i olika sammanhang. Syftet med denna avhandling är att utvärdera konsekvenserna av att använda referensförhållanden för att sätta miljömål för svenska ytvatten. Mer specifikt syftar avhandlingen till att: (I) utvärdera konsekvenserna av att använda referenstillstånd i vattenförvaltning, (II) jämföra oberoende bedömningar av referenstillstånd med hjälp av historiska arkiv av fiskförekomst och biogeokemisk modellering för att minska osäkerheter i referenstillståndsbedömningar, (III) utvärdera återhämtningen från försurning i försurningskänsliga sjöar i relation till ramdirektivets mål och (IV) utveckla empiriska modeller för att uppskatta historiska och nutida förändringar i halten organiskt kol i sjöar. Dessa studier har tillsammans och individuellt svarat på två viktiga frågor i förhållande till upprättande av referenstillstånd: Dessa studier har minskat de prediktiva osäkerheterna som finns när man ska bedöma historiska tillstånd med hjälp av modeller eller historiska arkiv. Denna avhandling har utvecklat flera metoder såsom lokal kunskap, skriftliga dokument, hydrogeokemisk modellering, miljöövervakning och paleolimnologi har använts för att minska osäkerheterna relaterade till beräkning av referensförhållanden, dvs. användandet av all tillgänglig kunskap. Förutom att minska osäkerheterna i bedömning av referenstillstånd diskuterar denna avhandlingen problemen med att använda sig av referenstillstånd för att sätta miljömål inom vattenförvaltningen. Denna avhandlingen argumenterar för att det finns flera konkurrerande, men lika giltiga referensförhållanden baserade på olika kunskapskällor. Denna avhandling visar på ett behov av en strukturell förändring och acceptans för flera referensförhållanden baserade på all tillgänglig kunskap, vilket skulle skapa ett klassificeringssystem med större acceptans.

# Dedication

تقدیم به پدر ، مادر و برادر عزیزم
که در طی این مسیر همواره حامی و پشتیبان من بوده
اند

*De som fastnade i förnuft och definitioner*
*förspillde sin tid att tvista om vara eller inte vara.*
*O du Kloke, gå och välj druvors vätska,*
*då de tvistande trodde de var russin, fast de var omogna!*

Omar Khayyam

# Contents

# List of Publications

This thesis is based on the work contained in the following papers, referred to by Roman numerals in the text:

I Valinia, S., Hansen, H.P., Futter, M.N., Bishop, K., Sriskandarajah, N. & Fölster, J. (2012). Problems with the reconciliation of good ecological status and public participation in the Water Framework Directive. *Science of the Total Environment,* 433(0), pp. 482-490.

II Valinia, S., Englund, G., Moldan, F., Futter, M.N., Köhler, S.J., Bishop, K. & Fölster, J. (2014). Assessing anthropogenic impact on boreal lakes with historical fish species distribution data and hydrogeochemical modeling. *Global Change Biology,* 20(9), pp. 2752-2764.

III Futter, M.N., Valinia, S., Löfgren, S., Köhler, S.J. & Fölster, J. (2014). Long-term trends in water chemistry of acid-sensitive Swedish lakes show slow recovery from historic acidification. *Ambio,* 43(1), pp. 77-90.

IV Valinia, S., Futter, M.N., Cosby, B.J., Rosén, P. & Fölster, J. (2015). Simple Models to Estimate Historical and Recent Changes of Total Organic Carbon Concentrations in Lakes. *Environmental Science & Technology,* 49(1), pp. 386-394.

Papers I-IV are reproduced with the permission of the publishers.

The contribution of Salar Valinia (SV) to the papers included in this thesis was as follows:

I SV had main responsibility for interviews, data analysis and writing of the MS with support of all authors.

II SV had the main responsibility of compiling data, data analysis and writing of the MS with support from all authors.

III MF had main responsibility of the MS, SV contributed to the reference condition discussion and the assessment of reference pH in relation to recovery from acidification in Swedish lakes. All authors contributed to the writing of the MS.

IV SV, MF and BJC carried out data analysis, PR provided VNIRS reconstructions. SV had main responsibility for the MS writing together with all authors.

# Abbreviations

| | |
|---|---|
| AA | Acid Anions |
| ANC | Acid Neutralizing Capacity |
| BC | Base Cations |
| CLRTAP | Convention on Long-Range Transboundary Air Pollution |
| EQC | Environmental Quality Criteria |
| MAGIC | The Model of Acidification of Groundwater In Catchments |
| SEPA | Swedish Environmental Protection Agency |
| TOC | Total Organic Carbon |
| VNIRS | Visible Near Infrared Spectroscopy |
| WFD | European Water Framework Directive |

# Reference conditions and the implications for water management

In the year 2000, countries of the European Union implemented the Water Framework Directive (WFD). The WFD was revolutionary in many ways. It was the first attempt to harmonize water protection across Europe, and it was built on a classification system that put ecological integrity as the focal point of management. The overall goal of the WFD is to manage water bodies so they achieve good ecological status which is defined as minor deviation from an undisturbed state, this state is determined by a reference condition. In Sweden, reference conditions for acidification assessment are set to the year 1860. While the WFD has dictated management practices in Europe over the past 15 years, it has been criticised for its complex classification system with multiple biological, chemical and hydromorphological parameters. Estimates of the undisturbed state can be ambiguous as the concept can relate to multiple, often contradictory perceptions of when reference conditions occurred (Bishop *et al.*, 2009; Moss, 2008; Stoddard *et al.*, 2006). As the reference condition concept covers a broad range of topics and sources of knowledge, two main questions can be asked about their desirability as the overall target for water management. First, is it scientifically possibly to identify reference conditions with enough accuracy to motivate remediation measures? Second, do reference conditions identify feasible and desirable targets for stakeholders, water managers and scientists?

This project aimed to answer these questions by taking an interdisciplinary approach improving the definition of reference conditions. By exploring potential improvements to both reduce the predictive uncertainties and improve the desirability of reference conditions as a target for environmental management. In the legislative text, the WFD stresses the importance of using all available knowledge when estimating reference conditions. This recommendation may reduce the predictive uncertainties but there are knowledge gaps as to how to facilitate the use of all available knowledge in the classification process. This thesis has taken an important step in developing methods and tools based on multiple sources of knowledge to reduce the predictive uncertainties associated with reference condition estimates. By using multiple sources of knowledge and an interdisciplinary approach, lay knowledge based on historical events leads to an improved understanding of reference conditions and can validate geochemical models and paleolimnology (Paper I). We compared archival records of societally important fish species distribution data and hydrogeochemical modelling on a regional scale in order to reduce uncertainties in acidification assessments (Paper II). We evaluated temporal patterns in water chemistry of acid sensitive Swedish lakes as a means of highlighting the complexity of using reference conditions as the overall goal of water management (Paper III). The role of organic carbon and organic acids in surface water acidification has been heavily debated in the scientific community. So as to better constrain reference condition organic acidity estimates, we developed simple models based on paleolimnology and monitoring data to estimate changes in surface water organic carbon concentrations from 1860 until the present (Paper IV). The methods developed and their validation with different sources of knowledge in (Paper I-IV) can all contribute to reducing the uncertainties in regional and local scale reference condition estimates. We have shown that the use of all available knowledge, including lay and scientific sources, improves our understanding of reference conditions and the effects of anthropogenic pressures on the natural environment. The results of these studies provide a framework for water managers, scientists and decision-makers to use when estimating reference conditions for lakes.

The use of all available knowledge in determining reference conditions will reduce the predictive uncertainties (Paper I-IV), but the question remains as to whether or not reference conditions are desirable and feasible to use as the overall goal for water management. This work has addressed this question in an interdisciplinary manner. Reference conditions can be derived from feelings and emotions about what is important in the natural environment. The reference condition concept is connected to perceptions and knowledge, which in some cases provides multiple potentially competing estimates of reference conditions. We would argue that reference conditions are a subjective state based on multiple forms of knowledge and perceptions. Hence, they are an idealized goal for water management. We have exemplified this subjectivity by showing that reference conditions can be based on memories and lay knowledge (Paper I), historical records and hydrogeochemical modelling (Paper II), monitoring data (Paper III) or paleolimnological reconstruction (Paper IV). Ultimately, a reference condition could specify a state of affairs existing in the recent past and decided by regional monitoring campaigns (1983) to paleolimnological reconstructions of conditions assumed to have existed at the end of the last glaciation (Figure 1). The WFD defines acceptable water quality as having only minor deviations from reference conditions. Thus, multiple interpretations of reference conditions can create conflict between water managers and other stakeholders. A cause for the potential conflict is that reference conditions trigger emotions. They are based on knowledge and perceptions of different time periods.

Biological, chemical and hydromorphological indicators are all used for WFD assessments of ecological status, with a primary focus on aquatic biota. The research in this thesis has focused on chemical parameters for a number of practical reasons. Biological data are in general more expensive to obtain and offer less widespread coverage in both space and time than chemical data. In Paper II, it was showed that independent biological and chemical assessments of reference conditions were highly coherent. This increases the credibility of using chemical parameters to assess deviations from reference conditions due to anthropogenic pressures on surface water when biological data are unavailable.

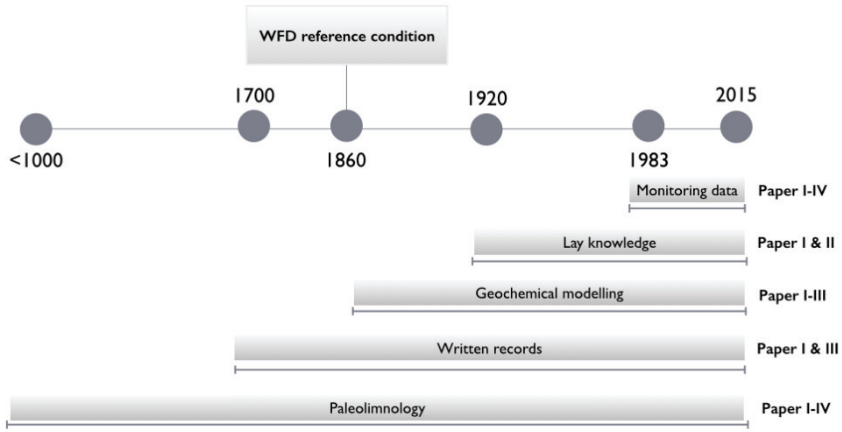


*Figure 1.* Conceptual figure of multiple reference conditions based on different sources of knowledge, the conceptual figure is based on lakes.

This issue of multiple, potentially contradictory reference conditions can be seen at Rotehogstjärnen. This lake has been part of all four papers in this thesis and shows how different sources of knowledge could provide multiple estimates of reference conditions. Lay knowledge of individuals living and working on or near the lake linked reference conditions to childhood memories of a much clearer lake. Monitoring data corroborates the memories of people living near the lake as they show substantial increases in organic carbon concentrations during the last three decades, which would reduce water transparency. Both local residents' conceptualization of reference conditions and the available monitoring data available support a decrease in water clarity. However, in contrast to these two approaches to estimating reference conditions, paleolimnological reconstructions showed that organic carbon concentrations were higher historically and the identified decrease in water clarity was a result of reduced anthropogenic pressures associated with acid deposition. This is directly in conflict with other sources of knowledge as a browner lake is a less acidified lake, hence, what is perceived as a worsening in water quality is actually a recovery from anthropogenic stressors and return to one definition of a reference condition (Figure 1). This creates an inherent complexity as all available knowledge has given us a greater understanding of reference conditions but it becomes very difficult to communicate this further. So to reconcile these differences, there is a need for multiple reference conditions based on different sources of knowledge that are equally valid. This would provide reference condition estimates that are desirable and feasible for water management as well as a basis for water managers to use knowledge from multiple sources and the opportunity to effectively communicate decisions and remediation measures to all involved parties in multiple levels in society. The multiple reference conditions would help reconcile the WFD ambition to use all available knowledge in decision making processes and in management practices. Furthermore, if used appropriately, multiple reference conditions can help democratise environmental decision-making.

# 1 Introduction

Reference conditions are an important part of water management as they provide a direct way of quantifying the effects of anthropogenic activities on the natural environment. Reference conditions provide a baseline to which contemporary conditions can be compared and have grown increasingly important in legislation such as the European Water Framework Directive (WFD) and U.S Clean Water Act. Previous European directives have also used the reference condition concept so it is proven to be a desirable approach for environmental management in different forms. For instance Swedish Environmental Quality Criteria (EQC) are based on the difference between present day and reference conditions.

Sweden has a long tradition of using EQC to assess the impact of anthropogenic activity on surface waters changes and protect them from further deterioration. As early as 1969 the concept of natural conditions was suggested as a part of the Swedish management program which specified high or low concentration thresholds for status assessment. This was intended to harmonize water management across Sweden (SEPA, 1969). This suggestion was never implemented. In 1990 a new set of suggestions for classification were adopted as general recommendations and reference conditions were introduced. The severity of impacts were classified based on deviations from reference value (SEPA, 1990). A new revision to the EQC was made in 1999 to include biological indicators as part of the assessment. This was an important step towards harmonisation of water management in Europe (SEPA, 1999).

After many years of negotiation and preparation, the European Union and its member states implemented the European Water Framework Directive (WFD) in the year 2000 (EC, 2000) as a comprehensive legal framework to improve water quality across Europe. Ecological integrity is the foundation and focal point of the WFD (Futter *et al.*, 2011; Crane, 2003). Implementation of the directive meant that member states moved away from traditional sector-based water management to a more holistic approach. One of the main purposes of using ecological integrity was the ability to manage water bodies using social, environmental and economic criteria (Hatton-Ellis, 2008; Steyaert & Ollivier, 2007). The WFD covers many different areas in one single piece of legislation including: ‘good ecological status’ (WFD, Article 2 & Annex V), public participation (WFD, Article 14), recovery of costs for water services (WFD, Article 9), groundwater protection (WFD, Article 17) and river basin management plans (WFD, Article 13). Sweden implemented the WFD as a whole through *Vattenförvaltningsförordningen* (VFF, SFS, 2004:660) with the Swedish Environmental Quality Criteria (EQC) adapted so as to meet WFD requirements.

The WFD introduced a complex classification system that combined biological, chemical and hydromorphological parameters into one ecological status assessment. Using all these different classification systems, the WFD intended to create a holistic approach to water management, which meant that monitoring should detect ecological change in the catchment (Josefsson & Baaner, 2011). An important aspect of the WFD is that the classification system is based on a wide range of biological communities, rather than a few chemical parameters (Moss, 2007). The WFD included the concept of ‘one-out, all-out’ (Hatton-Ellis, 2008). This means that if a water body fails to meet good ecological status for a single parameter being evaluated, it will not be classified as of good ecological status.

For water management in Europe, the WFD Annex V dictates and drives the monitoring and management of surface and transitional waters (Moss, 2008). The WFD classification system is based on a comparison between contemporary condition and a reference condition (which is assumed to represent environmental conditions which would have existed at a defined point in history). The classification systems in the WFD were similar to those already present in the Swedish EQCs. The classification system is based on five different classes: high, good, moderate, poor and bad status (EC, 2000). The overall goal of surface waters with good ecological status is defined as:

> “The values of the biological quality elements for the surface water body type show low levels of distortion resulting from human activity, but deviate only slightly from those normally associated with the surface water body type under undisturbed conditions” (Annex V, 1.2).

Focusing on the physio-chemical classification system of the WFD, minor deviation from good ecological status is acceptable. To assist member states in defining reference conditions and make the classification harmonious across Europe, the WFD added Common Implementation Strategies (EC, 2003a) as guidance documents for member states to use. Multiple studies have addressed the high level of uncertainty and complexity when assessing surface water status using undisturbed state as the base for management goals (Futter *et al.*, 2011; Hering *et al.*, 2010; Stoddard *et al.*, 2006). The use of a reference condition as a single target has been criticised as an uncertain management goal without taking into consideration different historical anthropogenic pressures. In reality, such reference sites are rarely found in Europe and monitoring data does not extend for such long time periods to cover the near undisturbed conditions. The lack of monitoring data has forced surface water management towards model simulation of historical conditions (SEPA, 2007). Thus, the success of WFD and Swedish environmental objectives depend on these models. As the WFD is reaching 15 years, there are still considerable discussions on how to manage surface water bodies and determine reference states on a regional level. In particular we can determine this state with enough certainty that remediation measures can be motivated to decision makers and stakeholders. Further a discussion should be lifted if reference conditions are desirable or even feasible in surface water management.

The use of a single reference condition has proven to be problematic in surface water management. The 'one out all out' has been criticised. Tools to predict the undisturbed state evolve over time. All available knowledge is not used in setting reference conditions. Stoddard *et al.* (2006) argue that a reference state can have multiple meanings with inherent flexibility and suggests that there should be multiple terms within the classification with narrow definitions to reduce the uncertainties associated with using reference conditions as targets.

This thesis will focus on understanding the implications of reference conditions on surface water management in Sweden. We will combine different reconstructions of pre-industrial conditions with biogeochemical modelling in order to enhance our knowledge of how anthropogenic activities have impacted ecosystem quality and to improve the models used for setting reference conditions. The thesis aims to develop approaches in which the predictive uncertainties are reduced and more reliable reference conditions target values are provided. It also highlights the need to discuss what implications reference conditions have on water management as well as whether the goal is desirable or even feasible on multiple levels in society.

## 1.1 Acidification and recovery of surface waters, a snapshot

Acidification of surface waters has been one of the major environmental problems of the $20^{th}$ century. The effects have been seen in large parts of the world including Europe and North America (Stoddard *et al.*, 1999). Large scale anthropogenic acidification started in the beginning of the 19th century due to large scale industrial revolution in Europe (Mylona, 1993). Long-range transport of acidifying compounds resulted in areas away from the source of pollution been affected by acidification. For instance in Fennoscandia, characterized by thin soils without substantial buffer capacity, the effects manifested themselves as fish kills in surface waters (Leivestad & Muniz, 1976; Almer *et al.*, 1974). Similar effects were observed in North America and the United Kingdom (Schindler, 1988; Beamish & Harvey, 1972). Peak sulfur deposition occurred in 1970-80 in Sweden (Figure 2) and has had substantial effects on biological life and ecosystem functioning. Large fish populations such as Roach (*Rutilius rutilius*), Atlantic salmon (*Salmo salar*) and Brown trout (*Salmo trutta*) were extinct as a result of large-scale regional acidification. In the Swedish national survey of freshwaters (2014) 10% of the surface waters were classified as significantly acidified (Fölster *et al.*, 2014b). With the spatial scale of the effects, acidification was considered a global environmental problem in the 1980s.

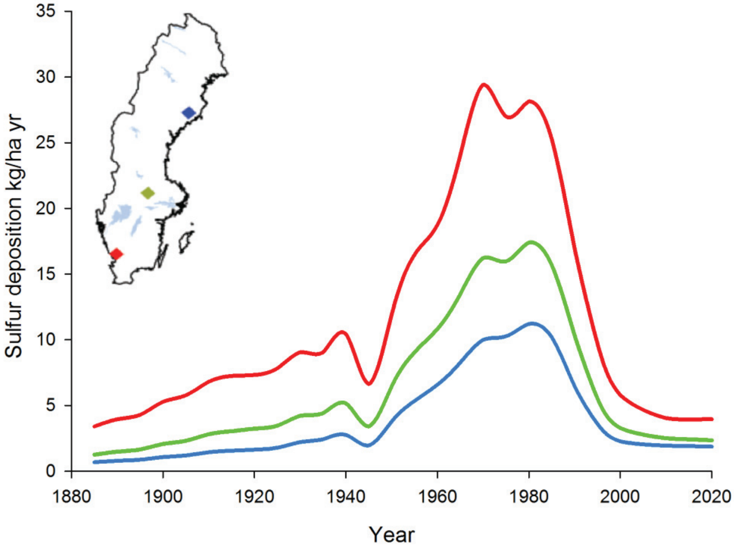


*Figure 2.* Calculated historical trends of sulfur at three EMEP grid squares in southern, central and north eastern Sweden. Data provided from EMEP.

As acidification was considered one of the main environmental problems of the 20th century, it is also a success story when it comes to international cooperation to reduce sulfur emissions. Countries agreed to reduce the sulfur emissions in accordance with the terms of the United Nations Economic

Commission for Europe (UN-ECE) Convention on Long-Range Transboundary Air Pollution (CLRTAP). (UNECE, 2012). A number of associated protocols such as The Oslo Protocol (1994) and The Gothenburg Protocol (1999) were signed and reductions of other pollutants were agreed upon. As a result of the commitments to CLRTAP, the sulfur deposition in large parts of Europe and North America has returned to near pre-industrial levels.

As a result of the substantial decrease in sulfur deposition over Fennoscandia, Europe and North America lakes have started to recover from acidification (Futter *et al.*, 2014; Skjelkvåle *et al.*, 2007; Skjelkvåle *et al.*, 2005; Forsius *et al.*, 2003; Evans & Monteith, 2001; Stoddard *et al.*, 1999). Multiple studies have identified chemical recovery in surface waters in Fennoscandia, Europe and North America (Futter *et al.*, 2014; Skjelkvale *et al.*, 2003; Fölster & Wilander, 2002; Evans *et al.*, 2001; Stoddard *et al.*, 1999). A recent study of acid sensitive lakes in Sweden showed that lakes with pH around the critical low level (pH 5.6, Figure 3) have declined since 1988. These results indicate that even the most acid sensitive lakes in Sweden are slowly recovering to levels not toxic for biological indicators (Fölster *et al.*, 2014b).

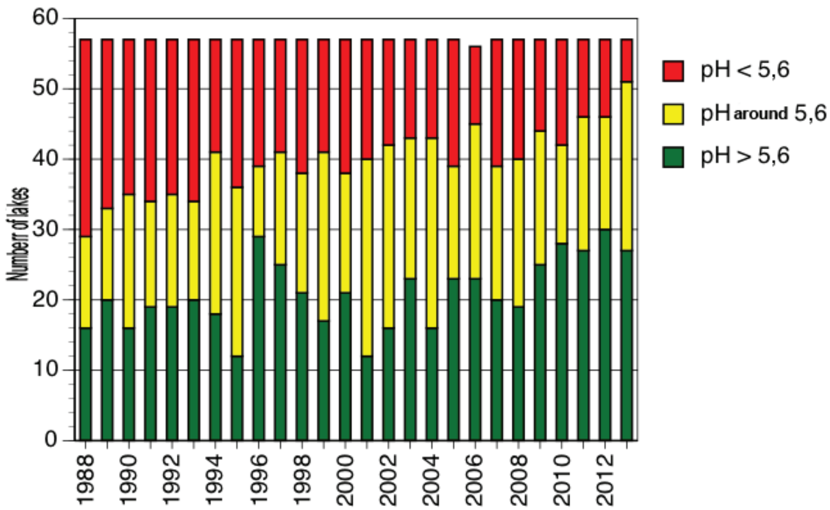


*Figure 3.* Yearly classification of pH in 57 trend lakes with measured pH <5.6 sometime during the monitoring period from 1988 to 2013. The lakes are classified based on four samples taken during each year that had a measurement of pH below 5.6 (red), above 5.6 (green) or both (yellow). Figure from Fölster et al 2014.

Regional assessments show that recovery is occurring but the process is slow. For instance, increases in organic acids (TOC/DOC), sea salt episodes, episodic acidification, and nitrogen deposition inhibits recovery processes (Akselsson *et al.*, 2013; Lofgren *et al.*, 2011). The long-term depletion of base cations (BC: $Ca^{2+}$, $Mg^{2+}$, $Na^{+}$, $K^{+}$) creates higher vulnerability for the soils to re-acidification (Likens *et al.*, 1996). Land use shifts and forestry intensification can delay the recharge of BC pools in soils and even re-acidify surface waters (Zetterberg *et al.*, 2014; Aherne *et al.*, 2008; Renberg *et al.*, 1993).

Even though there has been a significant recovery for chemical parameters in surface waters, the biological recovery, as prescribed by the WFD is more equivocal (Stendera & Johnson, 2008). Indications of biological recovery have occurred but there are more factors such as climate variability (Johnson & Angeler, 2010), spatial factors (Angeler & Johnson, 2012) dispersal difficulties (Yan *et al.*, 2003) that are affecting the recovery processes. Deciding what biological indicator to monitor will affect the recovery rates for the lakes, and using the wrong indicators has been suggested as a potential factor for lack of observed biological recovery (Holmgren, 2014; Johnson *et al.*, 2006).

## 1.2 Chemical classification system for acidification

The classification system of the WFD is based on chemical, biological and hydromorphological indicators. The WFD makes a combined assessment of all these groups to evaluate if the lakes achieve good ecological status. One of the main problems is that multiple areas of classification creates complexity on regional scale assessments, in particular when using management goals based on undisturbed state. This thesis has focused on chemical parameters and specific fish indicators.

In acidification assessment of surface waters, pH is used as the official indicator for anthropogenic influences on the natural environment in Sweden. As total organic carbon (TOC) is a weak acid it will contribute to pH. Surface waters are classified as acidified if the change between reference values and contemporary conditions are larger than 0.4 pH units (i.e. $\Delta$pH< 0.4) (SEPA, 2007). Thus estimates of reference condition pH are dependent on both TOC concentration and its acid dissociation constants.

The WFD has focused on ecosystem effects and this threshold value was chosen due to its effects on biological indicators (Fölster *et al.*, 2007) and the high TOC concentration often seen in Swedish lakes.

## 1.3 Surface water changes in Total Organic Carbon

During the last three decades, monitoring programs in Fennoscandia, UK, Easter North America and Central Europe have identified substantial increases in TOC in surface waters. In boreal surface waters, TOC consists almost exclusively of dissolved material, thus total and dissolved organic carbon are operationally identical. Organic carbon is a key controller of lake ecosystem functions and structure. The terrestrial input of TOC is large to the water column and a small change in composition or concentration can affect metabolic rates, primary and secondary production and the distribution of organisms (Faithfull *et al.*, 2014; Prairie, 2008; Blomqvist *et al.*, 2001). Concentrations of TOC in surface waters control attenuation of solar radiation, heating of the water column and thermal stratification (Snucins & John, 2000), leading to lower rates of photosynthesis and steeper thermal gradients (Solomon *et al.*, 2015). Large-scale recovery from acidification linked to decreases in sulfur deposition (Stoddard *et al.*, 1999) has occurred in many areas reporting increased surface water TOC concentrations. Identified increases in surface water TOC concentrations can cause problems for drinking water treatment plants (Ledesma *et al.*, 2012) with chlorination residues (Lavonen *et al.*, 2013), organic pollutants (Murphy *et al.*, 1990) and toxic metals (Klaminder *et al.*, 2006).

There have been close connections between organic acidity and acidification since the beginnings of acidification research. In the early 1980s some researchers argued that there was no link between acid rain and declines in pH (Krug & Frink, 1983) since sulphate replaced the organic acids in the surface waters, hence declines in organic acidity counteracted the sulfur emissions. This hypothesis was rejected even though researchers have acknowledged its importance (Evans *et al.*, 2008; Driscoll *et al.*, 2003). Studies have shown that the increases in TOC concentrations can retard recovery rates in surface water and in some sense counteract the substantial declines in sulphate deposition (Erlandsson *et al.*, 2010; Evans *et al.*, 2008; Driscoll *et al.*, 2003). Another factor of importance is what role TOC has in reference condition calculations. Studies based solely on surface waters have shown that pH calculations are highly dependent on chosen TOC concentrations on a regional scale (Erlandsson *et al.*, 2011). Modelling exercises in acidified areas in the Czech Republic that include soil and surface water TOC interactions showed that changes in reference pH were highly dependent on assumptions regarding TOC concentrations (Hruška *et al.*, 2014). In defining reference conditions for acidification assessment TOC becomes a key parameter and credible estimates of historical conditions are needed to reduce the uncertainties. The potential drivers for these increases of surface water changes in TOC have been highly debated in the scientific literature for the last 15 years. But no unifying driver has been brought forward, which can be linked to the short monitoring periods and the heterogeneity between different studies (Clark *et al.*, 2010).

Paleolimnological reconstructions have shown the evolution of lake water TOC concentrations since the end of the last glaciation (Figure 4). For much of the past 12.000 years, TOC concentrations were higher than during the period of peak acidification. The current increasing TOC trends in many lakes may thus be a return to reference conditions (Meyer-Jacob *et al.*, 2015; Rosén *et al.*, 2011).

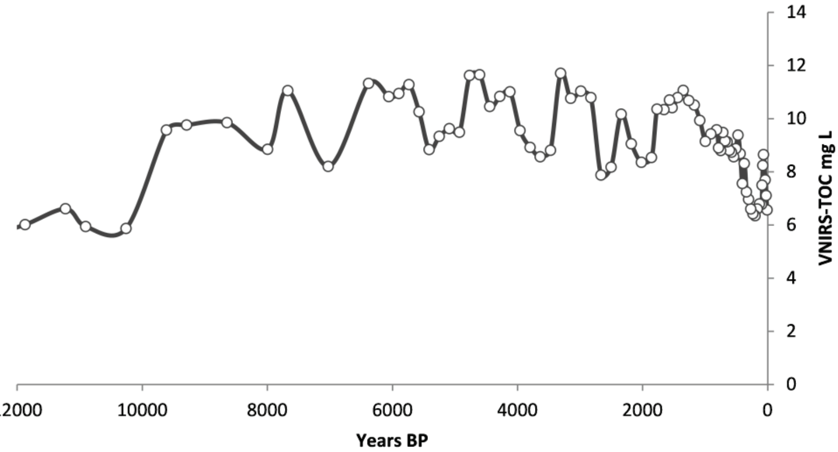


*Figure 4.* VNIRS-TOC reconstruction in Lysevatten from south-western Sweden since last glaciation. Data from (Rosén et al 2011).

### 1.3.1 Changes in atmospheric acid deposition

Monitoring data in in Fennoscandia, eastern North America, central Europe and the UK have connected the changes in TOC to declines of atmospheric acid deposition (Evans *et al.*, 2012; De Wit *et al.*, 2007; Monteith *et al.*, 2007; Evans *et al.*, 2005). Increases in sulfate can inhibit the mobility of TOC by at least two mechanisms – by changing the acidity of soils or changing the ionic strength of soil solutions. Acidifying effects resulting in lower soil pH can cause the solubility of TOC to decrease due to protonation and steric conformation. During recovery from acidification the increasing soil pH increases TOC solubility, leading to increased transport from terrestrial to aquatic systems (Evans *et al.*, 2012; Tipping & Hurley, 1988). Declines in acid deposition can result in lower concentrations of multivalent ions found in soil solution, such as $Ca^{2+}$, $Mg^{2+}$ and Al, as the protons no longer compete for adsorption sites (Hruška *et al.*, 2009). This can also be linked to organic matter coagulation and Al binding. Acidifying effects of sea salt ($Cl^-$) episodes can trigger mobility of TOC from the soil to surface waters (Clark *et al.*, 2011; Clark *et al.*, 2010). Change in soil acidity have proven to change the quality of organic matter in the soil solution (Ekström *et al.*, 2011)

### 1.3.2 Land use shifts and management practices

Long-term paleolimnological reconstructions of TOC have shown that changes in TOC occurred many centuries ago as an effect of land use shifts, early settlement and cultivation (Figure 4) (Meyer-Jacob *et al.*, 2015; Bragée *et al.*, 2013; Cunningham *et al.*, 2011; Rosén *et al.*, 2011). Land use shifts and different management types will affect soil organic matter and its potential transport from terrestrial to aquatic systems as soil and plant litter are the primary sources of organic carbon in boreal aquatic ecosystems (Chantigny, 2003). Management practices can alter the input of organic matter and change the rates of both atmospheric carbon fixation and mineralization (Kalbitz *et al.*, 2000). Afforestation has been shown to increase the leaching of organic matter from the soil (Guo & Gifford, 2002). Other management practices such as nitrogen (N) fertilization, forest liming and mineral fertilization have all shown to have substantial mobilizing effects on boreal soil carbon stocks (Chan & Heenan, 1999; Chantigny *et al.*, 1999; Zsolnay & Görlitz, 1994). Forestry effects on soil carbon stocks have been well studied, for instance the effects of clear cutting (Schelker *et al.*, 2012) and cultivation of forest soil (Qualls *et al.*, 2000) and afforestation (Paul *et al.*, 2002).

### 1.3.3 Changes in precipitation and runoff

Studies have linked increases in precipitation and runoff to changes in TOC export from soils to streams (Lepistö *et al.*, 2013; Ledesma *et al.*, 2012; Eimers *et al.*, 2008a; Erlandsson *et al.*, 2008b; Futter *et al.*, 2007; Worrall & Burt, 2007). Increases in precipitation and runoff may alter water flow paths and bring the water table closer to the soil surface, thereby routing water through more organic rich soil horizons and increasing contact times thereby facilitating greater TOC production. Seasonal variability in runoff and precipitation may affect TOC export as changes in seasonal temperature and flow regimes may alter rates of production and transport processes (Eimers *et al.*, 2008b; Evans *et al.*, 2005; Freeman *et al.*, 2001). The role of precipitation and runoff has also been shown to be sensitive to soil type, for instance in soils with high clay content, TOC concentrations may drop 50-90% from the surface organic layers to subsurface mineral layer (Neff & Asner, 2001). On the other hand, studies have shown a strong dilution effect on TOC with increases in runoff and precipitation (Monteith *et al.*, 2015).

### 1.3.4 Temperature controls on TOC

Temperature controls of TOC are often connected to both biological and physical mechanisms through processes related to primary production, microbial decomposition, increased litter fall, soil respiration and vegetation cover (Kalbitz *et al.*, 2000). Organic matter decomposition should increase exponentially with increasing temperature in accordance with the Arrhenius equation ($Q_{10}$). Meta studies have shown that a median increase of 2.39 for soil respiration per 10 °C increase in temperature (Hamdi *et al.*, 2013). Other studies have linked temperature and elevated $CO_2$ to increases in net primary production and hence higher export of TOC (Freeman *et al.*, 2004). The effects of temperature on TOC has been equivocal in previous studies, increases of TOC have been linked to increasing temperature (Larsen *et al.*, 2011; Weyhenmeyer, 2008; Freeman *et al.*, 2001) but other studies have found weak links to changes in temperature (Clark *et al.*, 2010; Yallop *et al.*, 2010; Evans *et al.*, 2005). Furthermore, studies have shown that temperature alone cannot account for the substantial changes in TOC export (Tranvik & Jansson, 2002).

## 1.4 Fish as an indicator for acidification

Public concern about acidification of surface waters resulted mainly from fish mortality in lakes and streams of acid sensitive species, like Roach (*Rutilius rutilius*), Atlantic salmon (*Salmo salar*) and Brown trout (*Salmo trutta*) (Hesthagen *et al.*, 1999; Rask *et al.*, 1995; Leivestad & Muniz, 1976; Schofield, 1976). The negative effects of acidification on Roach populations were identified in the 1960s (Almer *et al.*, 1974) and for Brown trout in southern Norway as early as the 1920s (Dahl, 1921). The current Swedish classification systems of fish species composition in lakes are based on eight different metrics (EQR 8) including fish biomass indexes estimating anthropogenic effects as well as species richness (SEPA, 2007).

Roach is an acid sensitive fish species that is abundant in large parts of southern and central Sweden that has been identified as an important indicator of acidification. A critical value for disturbance in the reproduction and extinction of Roach occurs at a pH level around 5.5 (Vuorinen *et al.*, 1993; Almer *et al.*, 1974). The lowering of lake water pH will increase the toxicity of Al, which eventually will lead to the extinction of species (Keinänen *et al.*, 2000; Leivestad & Muniz, 1976). It is mainly the inorganic (monomeric) species of Al ($Al_i$) that results in fish mortality (Gensemer & Playle, 1999; Baker & Schofield, 1982; Driscoll *et al.*, 1980). In waters where pH ranges between 4.6-5.8, $Al_i$ levels between 20-75 µg/l are toxic. For instance Hultberg (1988) stated that the Roach community is affected at pH 5.8 $Al_i$ <50 µg/l. Rask *et al.* (1995) showed that 100 % of the Roach population was extinct in Finnish lakes with pH below 5, and over 60 % was extinct in the pH interval between 5.1-5.5.

Due to lack of monitoring data for inorganic forms of Al, pH was used as a proxy for fish mortality. The toxic mechanism of pH changes has been studied comprehensively (Holmgren & Buffam, 2005; Keinänen *et al.*, 2000; Lien *et al.*, 1996; Rask *et al.*, 1995; Vuorinen *et al.*, 1993). When a lake is affected by acidification, one of the main effects noticed is disturbances to fish reproduction. Disturbances to reproduction have consequences for the population age structure as the proportion of older, larger specimens will increase substantially. The reproduction disturbance will lead to decreased hatching and diminished fish stocks, thus less competition for food and higher growth rates (Almer *et al.*, 1974).

# 2 Objectives

The main aim of this thesis was to evaluate reference conditions in accordance with the Water Framework Directive for Swedish surface waters. The thesis has a strong interdisciplinary approach and the outcomes will help in reducing predictive uncertainties associated with estimating reference conditions. This will lead to better estimates as to how climate change, land use and other anthropogenic pressures affect surface water quality. The thesis has four main objectives corresponding to the four papers included.

I Define the implications of reference conditions for water management in accordance with the Water Framework Directive (Paper I).

II Compare independent estimates of reference conditions derived from historical fish archives and biogeochemical modelling with respect to acidification assessment (Paper II).

III Evaluate the recovery from acidification in acid sensitive Swedish lakes and analyse potential limiting factors for future management (Paper III).

IV Develop empirical models to estimate historical and present changes in TOC for Swedish lakes (Paper IV).

# 3 Material and Methods

## 3.1 Site description and monitoring programs

Water chemistry data were obtained from the Swedish national monitoring programs (Fölster *et al.*, 2014a). Data from two main national surveys were used: the Swedish "trend lake" monitoring program and "national lake" survey program. The trend lake program consists of small- to intermediate sized lakes with minor influence from point sources or intensive land use like agriculture or forestry in the catchment. Climatic factors and transboundary pollutants mainly control the water quality in these lakes. The trend lakes are sampled for water chemistry four times each year to cover the seasonal variability. Monitoring of the trend lakes started in 1983 but some parameters such as TOC were sampled from 1988 onwards (Fölster *et al.*, 2014a). Data from the national trend lake program were used throughout the thesis (Paper I-IV). Lakes in the "national lake" survey are sampled once every six years. The survey includes 4800 randomly selected lakes across Sweden greater than 1 ha (800 samples each year). Data from the national lake survey were mainly used in (Paper II). The lakes used in this thesis include a wide range of chemical parameters and cover large parts of Sweden (Figure 5 & 6). Data from the "trend lakes" monitoring program were used in Paper IV, with a bias towards low alkalinity, acid sensitive lakes. Catchment land-use data were obtained from the European CORINE Land Cover database for the year 2000 (SEPA, 2014). Lake depths were obtained from the Swedish Meteorological and Hydrological Institute (SMHI) lake register (http://www.smhi.se/klimatdata/hydrologi/sjoar-och-vattendrag) when available. Depths for the remainder of the lakes were calculated (Sobek *et al.*, 2011). Modelled long-term annual mean runoff 1990-2010 was obtained using the SMHI S-HYPE model (http://vattenweb.smhi.se). Gridded (50x50 km) reconstructions of atmospheric sulfur deposition from 1880 to 2010 were provided by Coordination Centre for Effects (CCE) (Schöpp *et al.*, 2003). Time series data precipitation and temperature from 1901-2013 were extracted from the Centre for Environmental Data Archive (CEDA). The time series are presented at a resolution of 0.5 x 0.5 and data were extracted using Panoply (http://www.giss.nasa.gov/tools/panoply/). Catchment properties, deposition estimates and climate data were mainly used in (Paper IV).

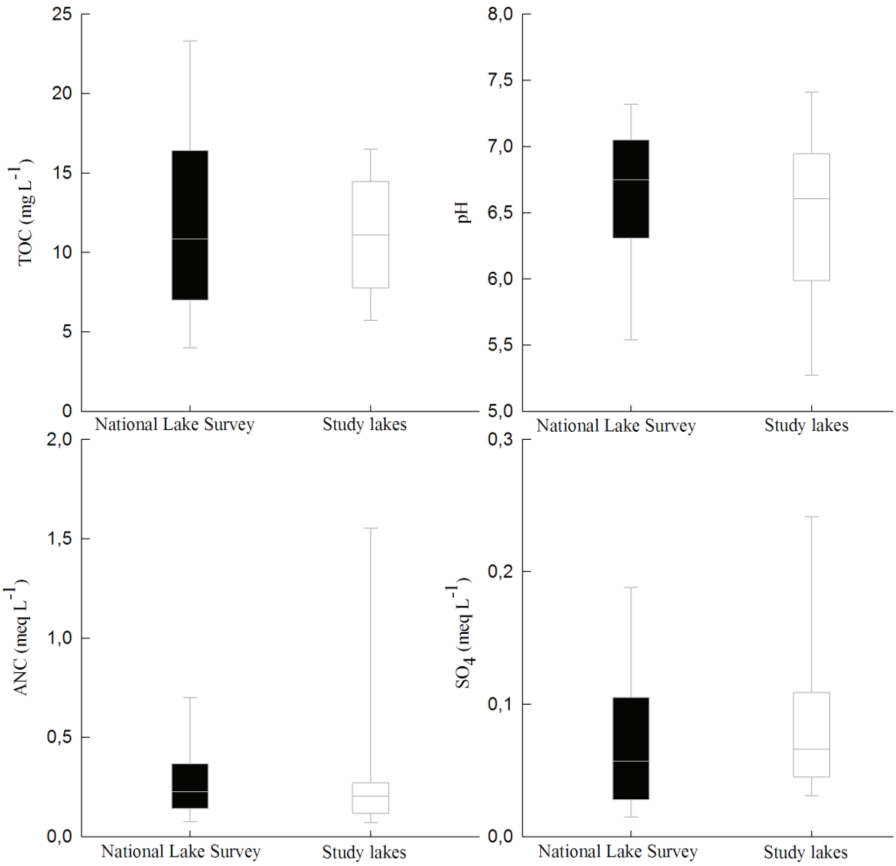


*Figure 5.* Representativeness of chemical parameters in Paper II compared to one lake survey (n=4600). Medians, 5, 25, 75 and 95 percentiles are shown in the figure (from Paper II).

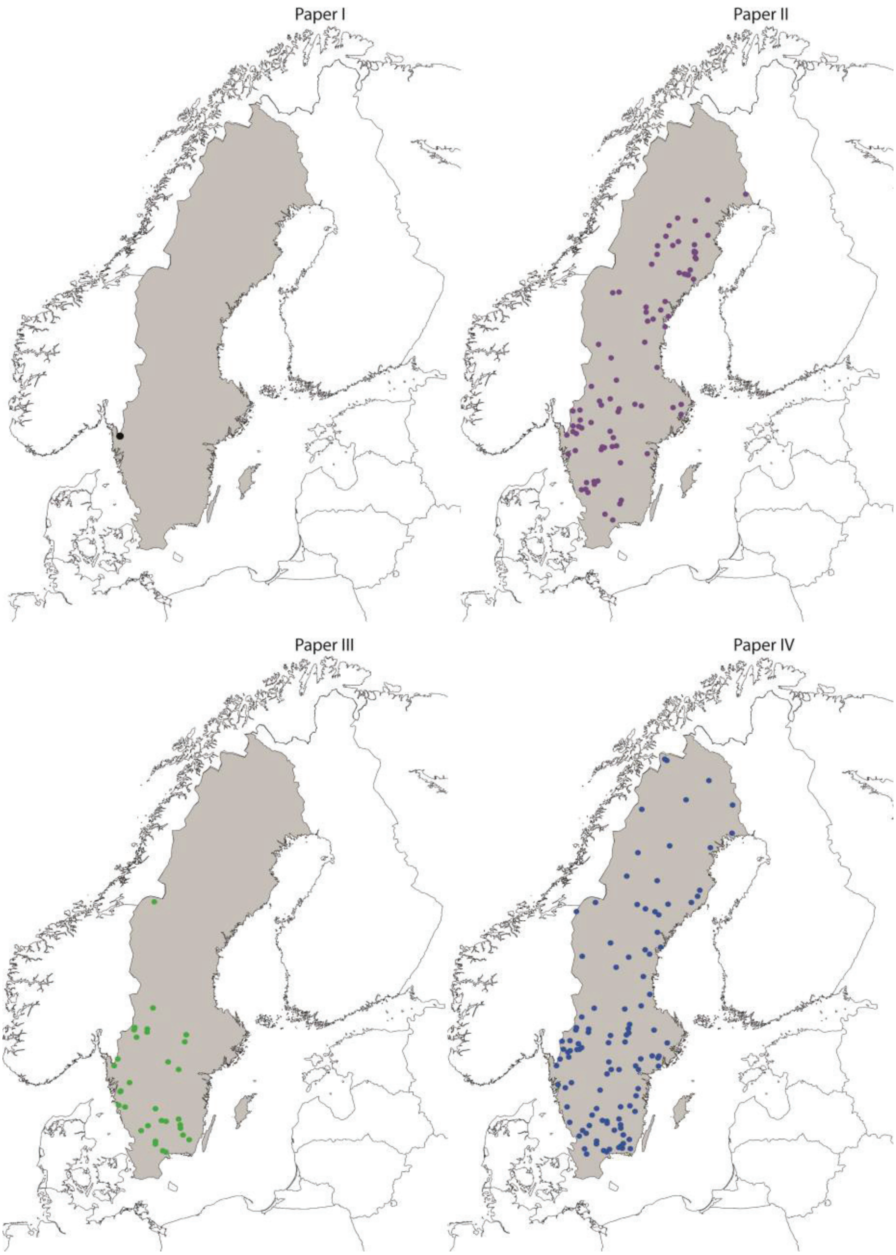


*Figure 6.* Location of lakes used in Paper I-IV. Rotehogstjärnen (Paper I) was used in all four papers.

## 3.2 Interview methodology and round table exercise

Paper I presents a case study conducted at Rotehogstjärnen (58°81 N, 11°61 E) in South-western Sweden (Figure 6). Semi-structured interviews (Kvale, 2009) were conducted with 11 landowners with house properties adjacent to the lake (Table 1) over three field visits during 2011. Contact information was collected from Tanumshede municipality and all landowners were contacted, therefore, no active selection was made. The semi-structured interview form is based on themes and not firm questions. The interview questions were flexible. During the interview, the answers to the questions varied and additional questions were added. The interviews lasted about one hour and were held in Swedish. The main goal was to highlight the different attitudes and perceptions of stakeholders in the Rotehogstjärnen catchment area in relation to reference conditions and public participation.

Table 1. *Information about the respondents in the interviews and the round table event. Modified from Paper I.*

| Interview ID: | Sex | Event |
|---|---|---|
| Land owner 1 | Male | Interview |
| Land owner 2 | Female | Interview and round table |
| Land owner 3 | Male | Interview and round table |
| Land owner 4 | Male | Interview |
| Forest Farmer | Male | Interview and round table |
| Hunter | Male | Interview and round table |
| Governmental worker | Male | Interview and round table |
| Angler | Male | Round table |
| Senior researcher | Male | Round table |
| Researcher | Male | Round table |
| Researcher | Female | Round table |

The round table event occurred during the last visit to the lake where all interviewees and additional stakeholders were invited. The aim of the round table was to have a discussion about undisturbed state and what could be a desired state. An external consultant facilitated the round table event so as to reduce bias from the researchers. The event lasted for four hours and occurred in the Tanumshede municipality. The round table also functioned as a validation of the individual interviews.

The interviews and round table event were recorded and transcribed. The transcription was analysed using meaning coding (Burnard, 1991) in which the interviews and round table were broken down to different themes so as to facilitate more systematic comparisons between respondents. The coding of the interview transcripts was analysed using the Nvivo9 software for qualitative data analysis (Richards, 1999).

The emphasis of the interviews and round table event was on understanding local knowledge, perceptions, values and experiences about lake Rotehogstjärnen. Their memory of historical changes and specific events, what they would desire and what they believed was the undisturbed state. These themes were brought up and additional questions and themes were added during the interview process

Table 2. *Overview of questions and themes discussed during the interviews and the round table event. Modified from Paper I.*

| Questions asked during single interviews and round table event |
|---|
| How do you perceive your lake today? |
| How do you use your lake today? |
| Have you identified any changes in the lake? (e.g. fish stocks, water clarity) |
| Have there been any major changes in the landscape? |
| What was the desirable state of the lake from your viewpoint? |
| What made this state desirable? |
| Do you think that this desirable state is achievable? |
| What was the undisturbed state from your perspective? |

## 3.3 Historical fish database (PIKE)

A wealth of historical fish observations are available in the PIKE database (http://www.emg.umu.se/english/research/research projects/pike/ assembled by G. Englund). The database includes presence/absence data for 55 fish species in c. 18 000 Swedish lakes. The database assembles information from interviews, postal questionnaires, and gillnet surveys performed during the period of 1890-2012. It includes multiple large surveys (c. 7000 lakes) conducted between 1890-1940. The database also includes important information about rotenone treatments, fish stockings, time of extinctions and large changes in the catchment.

Roach (*Rutilus Rutilus)* is a good indicator for acidification in Sweden, as the species is acid sensitive and abundant in large parts of Sweden. At pH levels below 6, Roach fails to reproduce and is extirpated at pH levels below 5.5 (Holmgren & Buffam, 2005). In Sweden, the first effects of anthropogenic acidification were detected from observations of fish species like Roach (Almer *et al.*, 1974). The PIKE database was used in (Paper II) to determine if the species was present historically (prior to significant acidification) and if/when it was extirpated from a lake. To get an overview of the historical database and the effects of anthropogenic acidification, observations were grouped into three time periods: (I) beginning of acidification period (pre 1960), this point in time was used as many areas in Sweden started to acidify before peak acid deposition due to weak buffering capacity. (II) heavy acidification period (1960-1990), this was used as the peak of acidification based on EMEP reconstructions of sulfur deposition (Figure 2). (III) recovery period (1990-present) during which atmospheric deposition declined and signs of small improvements in water chemistry was identified. Lakes with observations from both periods I and II were analysed further. One of the problems with the PIKE database was that different sources of observations were available during the same time period, such as 'directed' surveys for a specific species where low-density populations were reported as undetected during certain gillnet surveys. This sometimes led to contradictory information, thus all single species observations during period I and II were excluded and probabilistic criteria were used when classifying the lake as acidified or not based on the Roach population.

A lake was classified as acidified if the proportion of surveys that reported Roach was ≥75% before 1960 and ≤25% between 1960 and 1990. A lake was classified as non-acidified if more than 90 % of the surveys reported Roach during both periods. Based on this simple acidification criterion, 267 lakes matched, from which 121 were classified as acidified and 146 as non-acidified. In Paper II, 85 lakes were used as they had Model for Acidification of Groundwater In Catchments (MAGIC) reconstructed pH. The 85 lakes covered the natural distribution of Roach in Sweden (Figure 6).

## 3.4 Model of acidification of ground water in catchment (MAGIC)

The MAGIC model typically models the temporal evolution of soil and water chemistry from reference state until present. The model uses observed or estimated atmospheric deposition and land use histories at the modelled site. The model 'hind-casts' based on contemporary conditions and a model run is successfully completed when the reconstructed past leads to a simulation of present conditions that matches observations of soil and water chemistry. The MAGIC model also has the ability to make future predictions based on hypothetical land use, atmospheric deposition and climate. MAGIC was first developed nearly 30 years ago to predict the effects of acid deposition on soils and waters (Cosby *et al.*, 1985). The model has been successfully applied in many sites across Europe, North America and Asia to evaluate the effects of atmospheric acid deposition on surface waters (Cosby *et al.*, 2001). The MAGIC model has been further developed in multiple areas including organic acid buffering (Cosby *et al.*, 1995), aluminium solubility (Sullivan & Cosby, 1998), nitrogen dynamics (Cosby *et al.*, 2001) as well as nitrogen and carbon turnover (Oulehle *et al.*, 2012). Here, MAGIC7 (Cosby *et al.*, 2001) was used to assess the effects of acid deposition on lakes across Sweden.

MAGIC simulates annual or monthly concentrations of major ions in the streams, lakes or soil solution. The focal point of MAGIC is the pool of exchangeable base cations in the soil. As the fluxes from this exchangeable pool change over time due to atmospheric deposition, the chemical equilibrium between soil and soil solution shifts to give changes in surface water chemistry (Cosby & Wright, 1998; Cosby *et al.*, 1985). A mass balance takes into consideration effects of mineral weathering, biological uptake and immobilization, atmospheric deposition, decomposition and mineralization of organic matter and losses to runoff. Equilibrium processes include cation exchange between the soil and soil solution, dissolution-precipitation and speciation of inorganic and organic aluminium, sulfate absorption and speciation of organic and inorganic carbon.

The MAGIC reconstructions used in Paper II are the same reconstructions published by Moldan *et al.* (2013). These regional reconstructions of 2985 lakes are used in the regional assessment of WFD ecological status (SEPA, 2007). Detailed descriptions of input parameters and model performance are presented in (Moldan *et al.*, 2013). Two main parameters were of special interest for the questions posed in Paper II, namely pH and Aluminium (Al) as they are important determinants for fish reproduction and mortality. The reconstructions of pH from the MAGIC model (Moldan *et al.*, 2013) were used in Paper II and III to assess reference pH values for acid sensitive lakes.

### 3.4.1 pH in the MAGIC model

Positive charge equivalents in the soil solution pH are calculated by the charge balance equation (Equation 1). Base cations ($Ca^{2+}$, $Mg^{2+}$, $Na^{+}$, $K^{+}$), protons, and positively charged aluminium species (Equation 1) are the basis for the pH simulation. Negative charge equivalence is due to the presence of strong acid anions ($SO_4^{2-}$, $Cl^{-}$, $NO_3^{-}$, $F^{-}$) bicarbonate, hydroxide, negatively charged aluminium species (Equation 2) and organic acids. Dissociation of organic acids is based on a tripotic model developed by Hruška *et al.* (2003).

Bicarbonate was modelled in equilibrium with 4 times atmospheric pressure ($pCO_2$=2.8). Charge balance was achieved through iteratively adjusting proton concentrations and all proton related reactions including as aluminium solubility, bicarbonate, organic acid equilibrium and aluminium speciation. The main charge balance equation was written as follows (Equation 1)

$$[H^+] = [OH^-] + [HCO_3^-] + 2[CO_3^{2-}] + SAA + \Sigma OA - \Sigma BC - Al_{ekv} \quad \text{(Equation 1)}$$

where SAA, the strong acid anions, are equal to $2[SO_4^{2-}]+[Cl^-]+[NO_3^-]+[F^-]$; $\Sigma BC$ is equal to $2[Ca^{2+}]+2[Mg^{2+}]+[Na^+]+[K^+]+[NH_4^+]$. The organic acids are obtained from the Hruška *et al.* (2003) tri-protic acid calculations:

$$\Sigma OA=[H_2A^-] + 2[HA^{2-}] + 3[A^{3-}] \text{ and } \Sigma AlF=[AlF_4^-] + [AlF_5^{2}] + 3[AlF_6^{3-}] - 2[AlF^{2+}] - [Al\,F_2^+] \quad \text{Equation (2)}$$

MAGIC is calibrated against observed pH by adjusting the solubility constant ($K_{Al}$) of Gibbsite according to the following formula:

$$pH = (1/3)(pAl + K_{Al}) \quad \text{(Equation 3)}$$

pAl is the negative logarithm (base 10) of the $Al^{3+}$ ion activity. There can be large variability in site-specific $K_{Al}$ values. If the $K_{Al}$ used is not representative of the specific site, estimated pAl will not represent in-lake conditions and will affect the pH reconstructions. Detailed description of pH calculations can be found in (Cosby *et al.*, 2001).

## 3.5 Visible Near Infrared Spectroscopy TOC reconstructions

Historical lake water TOC concentrations were reconstructed using Visible Near Infrared Spectroscopy (VNIRS-TOC) with lake sediment cores. VNIRS is sensitive to organic matter quality and the terrestrial input of organic matter ultimately leaves a fingerprint that is reflected in the VNIRS spectra obtained from sediment cores. A NIRSystems 6500 instrument (FOSS NIRSystems Inc.) was used for analysis. Before analyses, sediment samples were freeze-dried and ground to a fine powder and stored for six hours in a climate controlled room to reduce temperature effects. To infer historical TOC concentrations, a transfer function between the VNIRS spectra in the top sediment layer and TOC concentration in the lake was used. The transfer function used in Paper (IV) was initially developed by (Rosén, 2005) and extended by (Cunningham *et al.*, 2011) to include more lakes from southern Sweden. The calibration set included 140 lakes ranging from 0.7 to 24 mg TOC $L^{-1}$ including nemoral, boreal and subarctic sites. Cores were dated using Pb-210 gamma spectrometry (Appleby, 2001). The top 30 cm of each core was analysed with VNIRS at 0.5 or 1 cm resolution. Only the sediment depth back to 1860 or bottom sediment with Pb-210 dating was used in (Paper IV).

## 3.6 Time series analysis and statistical methods

To assess recovery from acidification (Paper III) data from the national 'trend lakes' monitoring program (Fölster *et al.*, 2014a) were used. Data on acid sensitive lakes, based on the criteria of alkalinity <20 μeq $l^{-1}$, were selected and in total 34 lakes (Figure 6) with monitoring data between (1988-2012) were used. Trends were assessed for measured alkalinity, pH, individual and total base cations ($Ca^{2+}$, $Mg^{2+}$, $K^{+}$ and $Na^{+}$), individual and total strong acid anions ($SO_4^{2-}$, $NO_3^{-}$ and $Cl^{-}$), total aluminium and total organic carbon (TOC). Trend significance were assessed using the Mann-Kendall test with a Bonferroni correction for multiple comparisons to avoid type I errors in the analysis. Trend slopes were calculated using Sen's slope. All trend analysis were performed using SQL in Microsoft Access (Paper III).

Stepwise multiple linear regressions (forward selection) were conducted to estimate the annual change in TOC concentrations between 1860-2010 (Paper IV). One model was created using VNIRS-TOC to estimate changes between 1860-1980, and a second model based on national monitoring data were developed to estimate changes between 1988-2012. These models were then combined to predict changes in TOC between 1860-2012. The first step in model building was to evaluate the most significant candidate predictor variables. Model goodness was evaluated using the coefficient of determination $R^2$. For a candidate parameter to be accepted in the final model, two criteria needed to be fulfilled: Inclusion of a candidate parameter had to improve the adjusted $R^2$ by at least 0.01 and had to be significant ($p < 0.05$). To avoid spurious intercorrelation a candidate parameter was only accepted if the VIF (variable inflation factor) value was below 10. The model performance was evaluated using normalized root mean square error.

# 4 Result and Discussion

## 4.1 Perceptions and knowledge on undisturbed state (Paper I)

The use of undisturbed state and reference conditions in water management promotes ecological integrity. A reference condition is an objective base for setting goals, a state without major anthropogenic influence and which represents a desirable natural environment. Reference conditions are subject to an inherent conflict between the objectivity of the goal and the understanding of what a reference condition consists of. Based on the interviews and round table event, undisturbed state was dependent on perception, knowledge and values of the respondents. For instance, the governmental worker stated:

> It is ridiculous, because, what is an undisturbed landscape? We have to go back to the stone age, when we were hunters and gatherers, then we can speak about the natural landscape. Once we settled and began farming the land with grazing and then with agriculture, we have had an influence from the people”

The governmental worker saw the problems with using the undisturbed state as the goal from a management perspective, in particular there are difficulties in deciding when that state occurred.

On the other hand owner 1 had another perception, which was closely connected to a desired state and childhood memories.

> ““It is the memories I have when I was 15-20 years old, you could stand on the shore and look out over the lake and see the pikes far out. I cannot do that today. Something has happened to the lake, and it would be wonderful if it could go back to how it was. That is probably my cherished dream”.

The invocation of childhood memories was a common component in the interviews with the hunter and angler, amongst others.

The different interviewees had historical knowledge and recollections about the lake. As this area has been substantially acidified, the interviewees had good knowledge on when fish mortality occurred, and were told that acidification was the main driver and saw signs of recovery. The landowners also showed good recollection of the changes in water clarity. One of the landowners had identified a worsening of water clarity during recent years and noted that the lake was much clearer historically. Landowner 1 stated:

> "I am going to tell you something that should not be told, just to make the point of how clear the lake was in the past. When I was growing up I had an uncle who fished and hunted a lot. He went down to the lake one day and saw a large pike lying in the water, do you know what he did, he had a shotgun and fired towards the pike and the pike floated up of course. What I want to point out is that the visibility in the lake was much better in the past, in parentheses you are not allowed to do like my uncle did"

The perception of undisturbed state was connected to basic values of clean and healthy water. Stakeholders in the catchment provided valuable knowledge on changes in chemical parameters, fish species distribution and historical events. Using lay knowledge in classifying lakes might prove valuable in reducing the predictive uncertainties associated with estimating reference conditions. Lay knowledge can also improve the understanding and acceptance for what goals should be used in surface water management in accordance with the WFD.

The use of undisturbed state and reference conditions has advantages and disadvantages. Use of the undisturbed state creates the opportunity to move away from the traditional water management paradigm towards goals related to ecological integrity. With ecological integrity as the focal point of management, an objective measure of the magnitude of anthropogenic impacts could be evaluated. Undisturbed state is a simple value-based term that is easy to communicate to politicians, managers and stakeholders. On the other hand as it is strongly based on individual perceptions, undisturbed state can create an inherent conflict as it has multiple meanings. Stoddard *et al.* (2006) presented a conceptual framework in which the term "reference condition" was reserved for natural conditions with alternative classifications that were narrowly defined to reduce uncertainties related to management plans. The ideas presented in (Stoddard *et al.*, 2006) are complementary to the results presented in (Paper I) as they show that what constitutes a reference conditions differs between stakeholders. This creates uncertainties in goal-setting processes and remediation measures. The disadvantages of using reference conditions are connected to defining the undisturbed state for scientists and local authorities even though it might be defined appropriately. The uncertainty increases on a regional scale with a large number of lakes when predictive uncertainties increase substantially. This project highlighted the importance of understanding what is meant by undisturbed state, and to do this by lifting the perceptions of different stakeholder groups. A question of potentially greater importance is how stakeholder knowledge can be better used when defining the undisturbed state and what effects including this knowledge might have in reducing predictive uncertainties (Paper I). The WFD guidance documents have identified the value of involving stakeholders and others in the planning stage, defining goals, and working towards the potential remediation measures (EC, 2003b). Multiple studies have also shown the value of using local stakeholders in the whole management process (Fritsch & Newig, 2011; Gebrehiwot *et al.*, 2010; Rault & Jeffrey, 2008; Newig, 2005). The WFD classification system provides the opportunity to use 'all available' knowledge when assessing ecological status and the effects of anthropogenic stressors. The multiple perceptions of what a reference condition was might create a conflict between stakeholders and managers in defining goals and potential remediation measures if the validity of multiple reference conditions is questioned. The use of multiple approaches, including monitoring data, historical fish surveys, paleolimnology, lay knowledge and geochemical modelling, for a lake that has been subjected to anthropogenic pressure since 1950 provides a holistic overview of reference conditions. With a combination of scientific and lay knowledge, the uncertainties of classification can be reduced as both types of knowledge validate each other (Paper I). When stakeholders are not only informed about management decisions but are actively involved in planning, implementation and follow up, the opportunity is created for successful water quality management. The multiple perceptions of a reference condition might create a conflict between stakeholders and managers in defining goals and potential remediation measures. However, accepting that the use of all available knowledge will provide multiple valid reference conditions could reconcile reference conditions and public participation in the WFD.

## 4.2 Linking biological and chemical assessments of reference conditions (Paper II)

An overview assessment was made to compare chemical and biological assessments of acidification using Roach as a biological indicator and pH estimated using the MAGIC model as a chemical indicator. For MAGIC, three periods were chosen to assess acidification status ($\Delta pH < 0.4$): 1860 as the reference period, 1980 as the peak of acidification and 2010 as contemporary conditions. All 85 lakes showed decreasing pH between 1860 and 1980, and 28 lakes were significantly acidified based on the ΔpH criteria. Of the 27 acidified lakes, 14 lakes had a pH ≤ 5.5 during peak acidification (1980) and Roach was absent in 11 of them (Figure 7a). A similar approach was followed to assess recovery between 1980-2010. All lakes had experienced chemical recovery from acidification and 14 of 85 lakes were significantly acidified, which is a decrease of 50 % compared to 1980. Seven lakes had a pH ≤ 5.5 and Roach was absent in five of these (Figure 7b).

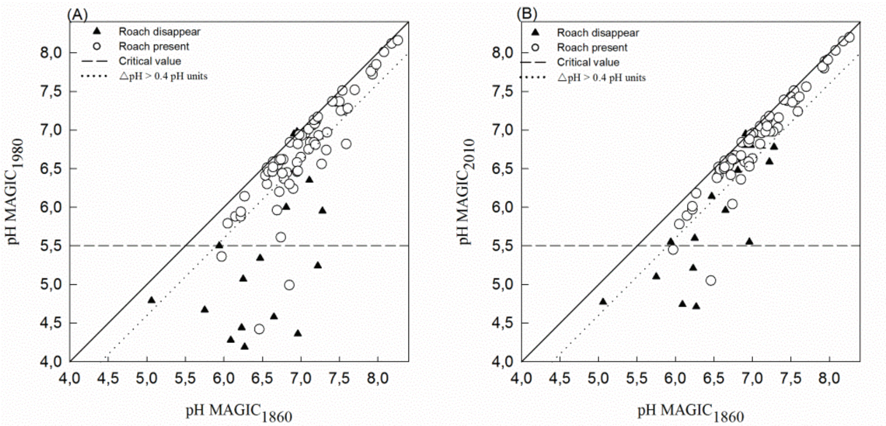


*Figure 7 a-b.* The 85 lakes analysed with MAGIC hind-casts, and historical observations of Roach. On the Y axis pH reconstructed from MAGIC in 1980 (a) and 2010 (b) and on the X axis reconstructed pH 1860. Black triangles (▲) indicate lakes where Roach has disappeared and circles (○) indicate lakes where Roach are present. The dotted line shows the criteria for significant acidification (ΔpH > 0.4), the striped horizontal line indicates the critical value of pH 5.5, and the solid black line is the 1:1 line (Figure from Paper II).

The lakes were further divided into four different categories for a more detailed comparison: (I) Non-acidified lakes with either method, and (II) acidified with both methods. These two categories accounted for 78 of the 85 lakes. The other two categories were for lakes without consistent classification: (III) MAGIC predicted acidification but Roach was present and (IV) MAGIC did not predict acidification but Roach was absent. These two categories had a combined total of seven out of 85 lakes.

In category (I), when neither of the methods showed significant acidification, pH levels were not below 5.5 and most lakes had pH levels around 7. For instance, Västra Solsjön had a minor change in pH during peak of acidification but not large enough to affect the Roach community and the criteria of ΔpH> 0.4 was not met (Figure 8c).

In category (II), in which both methods predicted acidification, substantial changes in surface water pH had occurred since the reference period. For instance Övre Skärsjön had a pH of 4.7 in 1980, and Roach was present until 1950 (Figure 8a). In Bäen, the Roach population survived for a longer time period and was not caught in the 1994 survey but reproduction had been disturbed much earlier. MAGIC showed substantial acidification with a pH of 4.4 in 1980 (Figure 8b). Category (III) showed inconsistency between the classifications, with MAGIC modelling indicating acidification while Roach were present. For instance, in Rotehogstjärnen, Roach has been present since the first survey in 1896 but pH was 4.4 in 1980 and 5.1 in 2010 (Figure 8e). Ulvsjön showed similar patterns; MAGIC predicted significant acidification but Roach was present (Figure 8f). Lakes in category (IV) showed the opposite behaviour; MAGIC did not predict acidification but Roach was absent in surveys. Gransträsket was not acidified according to MAGIC but Roach were absent in surveys from 1980 (Figure 8d).

The two independent methods used to classify reference conditions had high coherency and functioned to validate each other on a regional scale. Previous attempts have been made to compare different methods of inferring historical acidification in Sweden and elsewhere, for instance diatom reconstructions (Erlandsson *et al.*, 2008a; Battarbee *et al.*, 2005; Wright *et al.*, 1986). One of the main problems with diatom reconstructions is the time needed to reconstruct a single lake, in particular for regional scale assessments as the WFD dictates (EC, 2000). The use of historical fish species distribution data opens the possibility for regional scale assessments. The spatial differences in deposition, climate and land use showed that Roach is a stable indicator of acidification across the landscape (Paper II). The tools presented here provided new insights as to how multiple methods can be used to reduce the predictive uncertainties associated with determining reference conditions. On the basis of this comparison a conceptual model (Paper II) was developed to help prioritize management activities. The conceptual model creates the option to use remediation measures for species important to society and a communication tool for managers to use when informing about remediation measures.

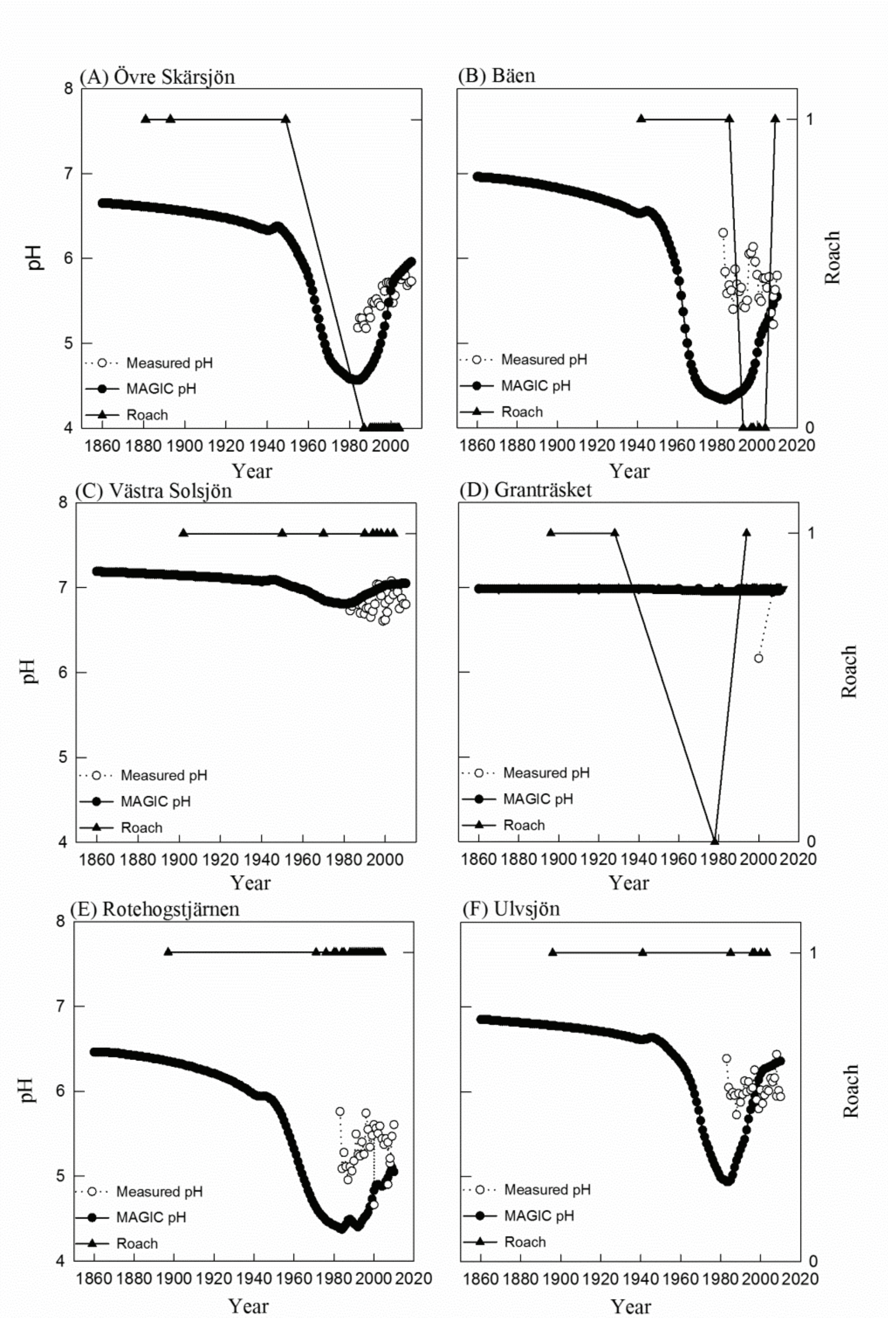


*Figure 8 a-f.* Reconstructed pH from MAGIC and measured pH (left y-axis) as well as presence (1) and absence (0) of Roach (right y-axis) by year on the x axis. (A) Övre Skärsjön. (B) Bäen. (C) Västra Solsjön, (D) Granträsket, (E) Rotehogstjärnen and (F) Ulvsjön (From paper II).

## 4.3 Assessing temporal trends of recovery from acidification in Swedish lakes (Paper III)

In general, the trends in water chemistry of acid sensitive Swedish lakes were consistent with recovery from acidification. Increases were observed in pH and measured alkalinity/acidity. Sulfate concentrations declined in almost all lakes and TOC concentrations increased. No trends were identified for total aluminium concentrations. In all the study lakes, the overall pH increase was approximately 0.5 pH units (n=34). It is noteworthy that the range in pH was 4.3 to >6. Despite the significant increase in pH, little change in alkalinity/acidity was observed (n=31). The year-to-year variability was greater than the magnitude of the upward trend. Most lakes switched between positive and negative alkalinity/acidity. Sulfate concentrations declined in all lakes, which was consistent with declines in atmospheric acid deposition. The overall average decrease was from 0.15 meq $l^{-1}$ in 1988 to 0.05 meq $l^{-1}$ in 2012. Average TOC concentrations ranged substantially, from <2 mg $l^{-1}$ to >20 mg $l^{-1}$ (n=33,) and increased on average from 8.5 mg $l^{-1}$ in 1988 to 14.5 mg $l^{-1}$ in 2012.

Trend analysis (Sen's slope) showed declines in nitrate and sulfate concentrations in most lakes. Chloride concentrations showed both increasing and decreasing trends. $[H^+]$ was declining in most lakes, but there were a few lakes showed increasing $[H^+]$, as a result of declining pH. No direct interpretation of total aluminium trends can be given as the results were equivocal. Concentrations of organic acids have increased over time for all lakes. Strong acids, magnesium and calcium were declining. Sodium had increasing and decreasing trends while base cation concentrations are declining in all lakes. All estimated ANC parameters increased over time (Figure 9).

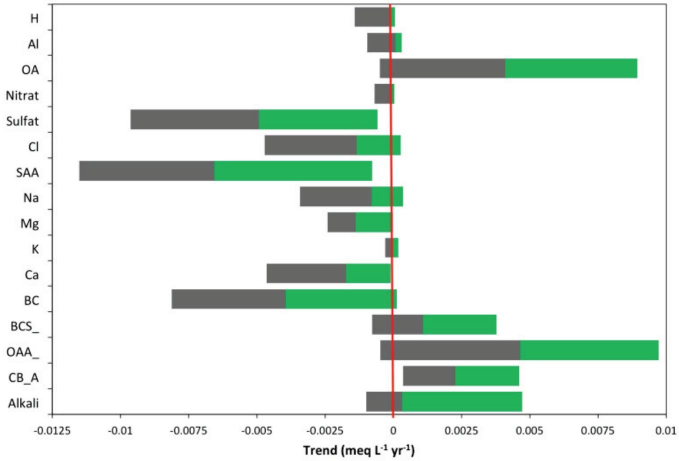


*Figure 9.* Trend summary for all analysed parameters. Trends were estimated using the Sen slope. Trends are presented for all parameters from (1988-2012), except for alk/ac where trends are reported for 1994-2012. Grey bars show values below the median and green bars are above the median. All trends are expressed in mekv/l/yr. From Paper III

The conceptual model developed in Paper II was used to assess the recovery from acidification and its effects on a biological indicator in accordance with the WFD (Figure 10). Reference pH (1860) was compared to two different pH levels indicating acidification (1990) and recovery (2010). The comparison showed that many lakes were recovering from acidification. The solid line 1:1 indicates that measured pH is equal to reference pH. The dashed line below is the ΔpH< 0.4 criteria, the horizontal line indicates reproduction failure for acid sensitive fish species (Roach). In 1990, 22 of 31 lakes were anthropogenically acidified. This number had declined to 17 in 2010 (Figure 10). These acid sensitive lakes in Sweden show recovery from acidification but the legacy has substantial effects on biota, in particular fish reproduction and mortality (Figure 10).

Based on the trend analysis, lakes are recovering from acidification with important parameters like ANC, BC and pH showing signs of recovery. One of the main questions is to what state the lakes are recovering? If recovery from acidification is a return to reference conditions, then most lakes have not recovered (Figure 10). This raises important questions about what a reference condition is and how it is defined. Based on the recovery from acidified conditions (Figure 10), many lakes are far from their reference state and even though lakes are recovering, a small change in water chemistry such as may occur during snowmelt or as a result of storms can have adverse effects on important, acid sensitive fish species.

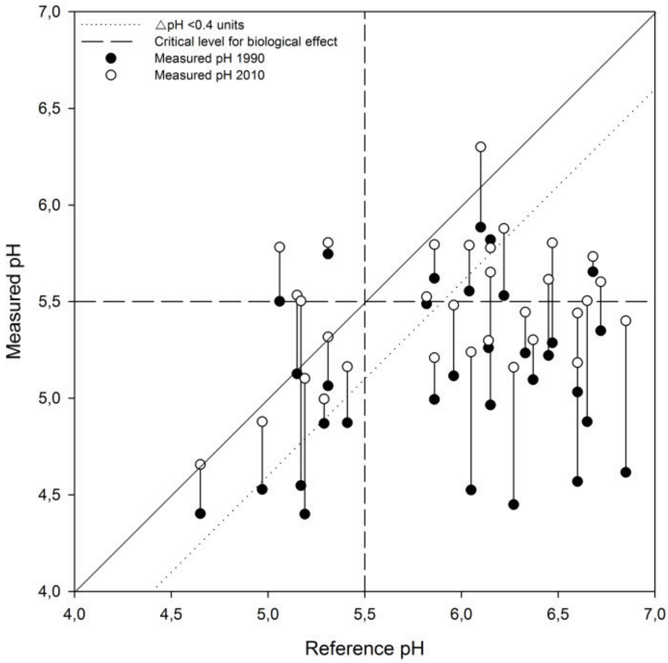


*Figure 10.* Lake recovery from acidification between 1990 and 2010 as indicated by deviation from reference pH. Reference (MAGIC-library estimated) pH is on the horizontal axis and measured pH on the vertical. Any lake >0.4 pH units from the 1:1 line is assumed to have been anthropogenically acidified. From Paper III

## 4.4 Simple models to estimate historical and recent trends of TOC in lakes (Paper IV)

Two models were created to estimate TOC concentrations in lakes. The first model predicted TOC concentration trends between 1860-1980 using VNIRS-TOC data, and the second predicted TOC trends between 1988-2012 using monitoring data.

The model predicting trends in TOC concentrations (VNIRS-TOC, n=14) between 1860-1980 was based on catchment characteristics and EMEP sulfur deposition. Measured water chemistry was left out of the model due to lack of observations during reference periods. The best performing model used three parameters: lake area ($km^2$), log catchment area ($km^2$) and EMEP sulfur deposition in 1980 (mg S $m^{-2}$ $yr^{-1}$) (Equation 6). The model had an adjusted $R^2$ of 0.85 and NRMSE of 12 % for predicting the VNIRS-TOC slope between 1860-1980 (Figure 11).

$\Delta[TOC]_{VNIRS} = 0.098 + 0.058 * [Lake\ area] - 0.069 * [Log\ catchment\ area] - 0.000052 * [Sulfur_{EMEP}\ 1980]$ (Equation 6)

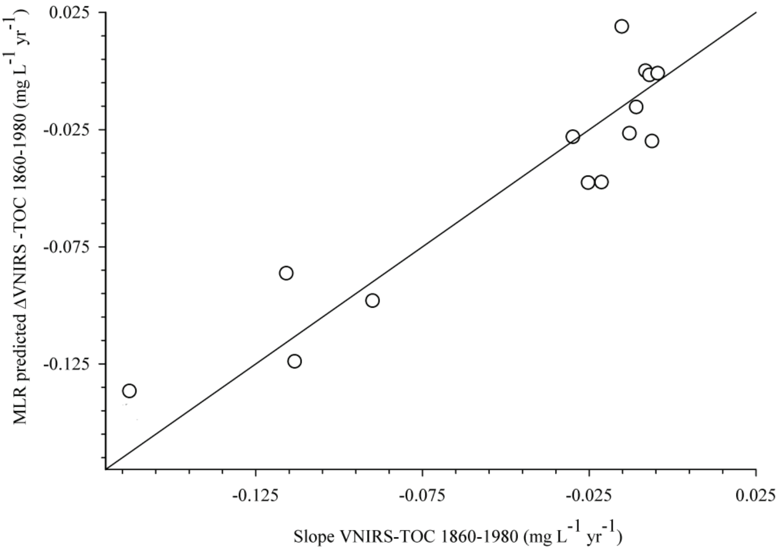


*Figure 11.* The slope of individual VNIRS inferred TOC time series trends plotted against the predicted slope (mg $L^{-1}$ $yr^{-1}$) from the MLR until 1980. The 1:1 line is showed in the graph Adj. $R^2$ 0.85 and NRMSE 12%.

To estimate recent temporal trends in TOC between 1988-2012, a model was constructed based on monitoring data (n=107) water chemistry and catchment characteristics (Equation 7). The best model included five parameters. TOC was predicted with an adjusted $R^2$ of 0.71 and NRMSE of 12% (Figure 12).

$\Delta[TOC]_{Pred\text{-}Mon.} = 1.090 - 0.427*[Runoff] + 0.000053*[Sulfur_{EMEP}\ 1980] - 0.003997*\%\ wetland - 0.160*[pH] + 0.749*[\Sigma BC^*] - 1.292*[SO_4^{2-}*]$ (Equation 7)

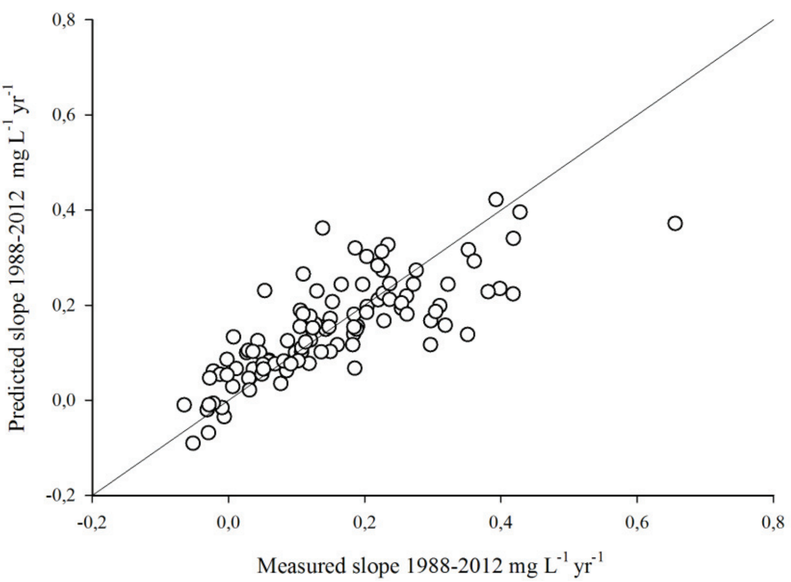


*Figure 12.* Measured TOC slope between 1988-2012 plotted against predicted TOC slope 1988-2012 for trend lakes based on the MLR (n=107). The 1:1 line is shown in the graph. Modified from paper IV.

The simple models developed here can provide new insights into changes in surface water TOC between 1860 and the present on a regional scale. Based on the historical model, catchment characteristics and acid deposition were the most important parameters when estimating reference condition TOC. Catchment characteristics have proven to play an important role in determining lake TOC concentrations (Kothawala *et al.*, 2014) and VNIRS reconstructions from southern Sweden have shown TOC minima during peak acidification (Bragée *et al.*, 2015). Other historical reconstructions have linked changes in TOC to changes in land use (Meyer-Jacob *et al.*, 2015; Rosén *et al.*, 2011). The contemporary models based on long term monitoring data were developed using routine water chemistry available in large parts of Sweden and other parts of the world. By linking the two models, it is possible to obtain a good indication of the historical changes in TOC concentrations and pinpoint potential drivers caused by anthropogenic stressors.

Estimates of reference condition pH made using MAGIC are sensitive to TOC concentrations, and can substantially alter the ΔpH< 0.4 criteria (Erlandsson *et al.*, 2011; Erlandsson *et al.*, 2008a). It is noteworthy that these studies did not take into account changes in soil water TOC, only the changes in surface water TOC concentrations. This can be criticised as changes in the soil BC pool, associated with soil transport, which is the heart of MAGIC, are not linked to assumed changes in TOC. A recent Czech study showed that soil and surface water TOC showed substantial changes between reference conditions and the present, leading in to lower estimated of 1860 pH (Hruška *et al.*, 2014). However, studies in Sweden have not shown increases in soil solution TOC from deeper soil horizons (Löfgren *et al.*, 2010). The use of variable TOC in MAGIC has been based on different contemporary conditions or modelled background concentrations based relationships with ionic strength. Reconstructions of surface water TOC concentrations can be made with VNIRS-TOC on a century to millennial time scale (Meyer-Jacob *et al.*, 2015; Cunningham *et al.*, 2011; Rosén *et al.*, 2011; Rosén, 2005). Different factors have proven to drive changes in TOC on these timescales, mainly land use changes and human settlement on a millennial scale (Meyer-Jacob *et al.*, 2015; Rosén *et al.*, 2011) and acid deposition on a decadal time scale (Paper IV) (Bragée *et al.*, 2015; Bragée *et al.*, 2013). As with most paleolimnological methods, one of the main problems with the VNIRS-TOC method is the amount of time and resources needed to reconstruct a single lake. With the WFD classification system, VNIRS methods become very labour intensive. The simple models in (Paper IV) can help in creating credible reference condition estimates of TOC concentrations based on VNIRS-TOC data and monitoring. These estimates can be used on a regional scale in more complex biogeochemical models like MAGIC and reduce the predictive uncertainties dealing with reference conditions. The strength of the TOC models in (Paper IV) lies in their simplicity. These simple models can be used to make regional scale assessments as the WFD mandates.

# 5 Conclusions and Future Challenges

The main conclusions of this thesis are:

I Multiple approaches to reference conditions are needed. Simple tools for estimating regional scale reference conditions are needed to handle the large number of water bodies. Multiple methods integrating different biological and chemical parameters are needed to validate reference condition estimates. In societally important water bodies all available sources of information, including lay knowledge must be included when formulating reference conditions.

II Public participation and good ecological status can be aligned so as to reduce the uncertainties of determining reference conditions in lakes. The use of all available knowledge will improve the understanding of changes in the catchment and reduce the inherent conflict between reference conditions and water management. The use of multiple reference conditions with all available knowledge can create base for communication on what an appropriate reference condition is for a water body. The alignment also creates the possibility to have remediation measures that are accepted by the public as they have been part of the decision making process.

III Independent estimates of historical fish population and geochemical modelling were compared so as it increases the credibility of using either method when estimating reference conditions and the effects of anthropogenic impacts on surface water acidification. The high coherence between chemical and biological indicators increases the credibility of using chemical parameters when biology is missing. A conceptual model was developed based on geochemical modelling and fish species distribution data to help in prioritizing management efforts on a regional scale.

IV Recovery rates in acid sensitive Swedish lakes were evaluated and the conceptual model developed in III was used to help with prioritizing remediation efforts on a regional scale.
V Simple models were developed to estimate historical and recent trends of TOC in lakes across Sweden. The models can be used to understand historical and future drivers as well to serve as input to more complex biogeochemical models such as MAGIC.

The main aim of this thesis was to reduce the predictive uncertainties and increase credibility of reference conditions estimates in Swedish surface waters in accordance with the European legislation (WFD). By developing tools and methods to reduce uncertainties associated with using reference conditions as to set targets, predictions as to how climate, land use and other anthropogenic stressors affect surface water quality can be obtained. The thesis has often used acidification from atmospheric pollution as an example but in the future the results of this thesis are also relevant for other anthropogenic pressures such as increased forest production (Laudon *et al.*, 2011), climate change (IPCC *et al.*, 2007), and eutrophication (Meyer *et al.*, 1999) that will affect management strategies and prioritization. The efforts made here to understand implications for water management of different reference condition estimates as well as how conceptual and practical tools can be used will help to reduce the predictive uncertainties in future management.

Beyond this thesis, there are two main areas in which I would like to direct future research. More efforts are needed to reduce the predictive uncertainties in estimating reference conditions and discussions on the elusive baseline (Likens & Buso, 2012) should be brought forward. A comprehensive assessment based on varying TOC concentrations in the MAGIC model should be made. Presently, classification of acidification (Fölster *et al.*, 2007) does not take into account this large change in TOC when the $\Delta pH<0.4$ criterion is evaluated. I would like to examine what affect improved estimates of reference condition TOC would have in classification of surface water acidification status on a regional scale based on the model presented in (Paper IV). Secondly, a framework in which multiple reference conditions based on all available knowledge could be developed to propose reference conditions. Multiple sources of knowledge would foster a discussion on what is meant by reference conditions. This discussion would become a framework in which an appropriate reference condition would be chosen based on all available knowledge. By developing this framework, the acceptance of using different kinds of knowledge as a tool to estimate reference conditions would be communicable to stakeholders, managers and decision-makers.

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*Je suis fatigué*

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