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International Cooperative Programme on Assessment and Monitoring Effects of Air Pollution on Rivers and Lakes



Convention on Long-Range Transboundary Air Pollution

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REPORT

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Abstract

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CONVENTION ON LONG-RANGE TRANSBOUNDARY AIR POLLUTION

INTERNATIONAL COOPERATIVE PROGRAMME ON ASSESSMENT AND MONITORING EFFECTS OF AIR POLLUTION ON RIVERS AND LAKES

Proceedings of the 31st Task Force meeting of the ICP Waters Programme in Monte Verità, Switzerland, October 6 – 8, 2015

Prepared at the ICP Waters Programme Centre Norwegian Institute for Water Research Oslo, February 2016

Preface

The international cooperative programme on assessment and monitoring of air pollution on rivers and lakes (ICP Waters) was established under the Executive Body of the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP) in July 1985. Since then ICP Waters has been an important contributor to document the effects of implementing the Protocols under the Convention. Numerous assessments, workshops, reports and publications covering the effects of long-range transported air pollution has been published over the years.

The ICP Waters Programme Centre is hosted by the Norwegian Institute for Water Research (NIVA), while the Norwegian Environment Agency leads the programme. The Programme Centre's work is supported financially by the Norwegian Environment Agency.

The main aim of the ICP Waters Programme is to assess, on a regional basis, the degree and geographical extent of the impact of atmospheric pollution, in particular acidification, on surface waters. More than 20 countries in Europe and North America participate in the programme on a regular basis.

ICP Waters is based on existing surface water monitoring programmes in the participating countries, implemented by voluntary contributions. The ICP Waters site network is geographically extensive and includes long-term data series (more than 25 years) for many sites. The programme yearly conducts chemical and biological intercalibrations.

At the annual Programme Task Force, national ongoing activities in many countries are presented. This report presents national contributions from the 31st Task Force meeting of the ICP Waters programme, held in Monte Verità, Switzerland, October 6-8, 2015.

Hele de toto

Heleen de Wit ICP Waters Programme Centre

Oslo, February 2016

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1. Introduction

The International Cooperative Programme on Assessment and Monitoring of Rivers and Lakes (ICP Waters) is a programme under the Executive Body of the Convention on Long-Range Transboundary Air Pollution. The main aims of the programme are:

- To assess the degree and geographic extent of the impact of atmospheric pollution, in particular acidification, on surface waters;
- To collect information to evaluate dose/response relationships;
- To describe and evaluate long-term trends and variation in aquatic chemistry and biota attributable to atmospheric pollution.

The national contributions on ongoing activities that were presented during the ICP Waters Task Force meeting in Monte Verità, Switzerland, October 6–8, 2015 were grouped thematically. A short summary of each presentation is given in the Minutes (Chapter 7). Selected presentations are reported more extensively in the Proceedings.

List of presentations:

Water chemistry

Recent trends in major ions in subalpine lakes and rivers in Northern Italy, Michela Rogora, Italy

Air Pollution Impacts on Debed River Waters in Different Parts of the Watershed, Marine Nalbandyan, Armenia

High quality long-term monitoring: Swedish experiences with assessments of surface water acidification and recovery, *Jens Fölster, Sweden*

Biological response

Lago Nero - a high alpine system to study the consequences of environmental change for lake ecosystems, *Andreas Bruder, Switzerland*

Comparing biological indicators and geochemical modelling from the beginning of the 20th century, *Salar Valinia, Programme centre*

Water chemistry - dissolved organic carbon

Terrestrial DOC Input and the Recovery of Carbon-Starved Ecosystems, John Gunn, Canada

DOC trend analysis, Heleen de Wit, Programme Centre

Critical Loads

Changes in stream water chemistry under future scenarios of acid deposition and forestry practices (Uhlířská catchment), Jakub Hruška, Czech Republic

Heavy Metals and POPs

Mercury in fish in a TOC-changing world: A model to improve the general dietary regulations and focus on individual lakes versus large-scale catchment advices, *Bjørn Olav Rosseland, Programme centre*

Metals forms distribution in the surface waters of the Kola North under anthropogenic loads, *Marina Dinu*, Russian Federation

Trends in mercury in fish, *Staffan Åkerblom, Sweden* The 2016 Mercury report, *Heleen de Wit, Programme centre*

2. Deposition of acidifying pollutants in Southern Switzerland and its effects on high altitude lakes

Sandra Steingruber

Ufficio protezione aria, Sezione protezione aria, acqua e suolo

1 Introduction

Sulphur and nitrogen oxides from combustion processes and ammonia from agriculture can be transported over long distances, transformed and then loaded on natural ecosystems causing acidification and eutrophication of sensitive ecosystems. Because of its proximity to the emission rich Po Plain and its generally abundant precipitations (MeteoSvizzera, 2012), Southern Switzerland is particularly exposed to deposition of anthropogenic pollutants. In fact, wet deposition in Southern Switzerland is mainly determined by warm, humid air masses originating from the Mediterranean Sea, passing over the Po Plain and colliding with the Alps. In addition, because of the dominance of base-poor rocks with low buffering capacity, in Southern Switzerland many high altitude soils and freshwaters are sensitive to acidification (Zobrist and Denver, 1990).

For this reasons at the beginning of the 1980's the Canton of Ticino started to monitor wet deposition and water chemistry of high altitude lakes. This article resumes the most important results of the monitoring programme.

2 Methods and study site

Rainwater has been sampled at weekly intervals with wet-only samplers first only at Locarno Monti and Lugano later also at other sampling stations. Nowadays, precipitation is collected at 9 sampling sites distributed over the whole area of the Canton of Ticino (Fig. 1). Before 2000 lake surface water was sampled only occasionally, from 2000 to 2005 20 lakes were monitored twice a year (once at beginning of summer, once in autumn) and after 2006 three times a year (once at the beginning of summer, twice in autumn).



Figure 1: Study area with sampling points

Rain- and lake-water have been analyzed for pH, conductivity, Gran alkalinity and the main anions and cations, lake samples also for phosphorus, silica, DOC, aluminum, copper, zinc, lead and cadmium.

For rainwater, monthly, yearly and 5-years mean concentrations were calculated by weighting weekly concentrations with the sampled precipitation volume, while monthly and yearly wet deposition were calculated by multiplying monthly and yearly mean concentrations with the precipitation volume measured at a meteorological sampling station located close to the sampling site.

Trend analyses were performed with the Mann-Kendall test to detect temporal trends in wet deposition and lake water chemistry. For wet depositions a seasonal Mann-Kendall test (Hirsch et al., 1982) was performed on monthly mean concentrations and depositions and a correction among block was considered (Hirsch and Slack, 1984). For lake chemistry a simple Mann-Kendall test was performed on autumn concentrations (Mann, 1945). The two sided tests for the null hypothesis that no trend is present were rejected for p-values below 0.05. Estimates for temporal variations of wet depositions and lake water chemistry were quantified with the seasonal Kendall slope estimator (Gilbert, 1987). All trend analysis was calculated with the CRAN package "rkt 1.3" (Marchetto, 2014).

Since 5-years concentration averages of sulphur, nitrogen and base cations correlated well with the 3 geographic parameters latitude, longitude and altitude (Steingruber, 2015a; Steingruber, 2015b), it was possible to calculate parameters for multiple linear regression equations describing the studied concentrations as a function of the three geographic parameters. To increase the significance of the multiple regression equations, also data collected at Italian sites located close to the Swiss border, next to our precipitation sampling sites, were used in the analysis (provided by CNR-ISE in Verbania Pallanza; for a description of the sites see Rogora et al., 2012). In addition, to facilitate the modeling of rainwater concentrations at very high altitudes, results from the analysis of snow sampled at the Basodino glacier (2650-3100 m) were also considered. It was therefore possible to calculate 1 km x 1 km concentrations maps for the 5-years periods 1988-1992, 1993-1997, 1998-2002, 2003-2007, and 2008-2012. Multiplying these maps with precipitation maps and adding dry deposition maps, deposition maps of total sulphur and nitrogen, total oxidized and reduced nitrogen could be calculated. For a more detailed description of the mapping method see Steingruber (2015a).

3 Results

The Mann-Kendall trend analyses (data not shown; Steingruber, 2015a) revealed that sulphate concentrations decreased significantly at all sampling sites. Concentrations of nitrate also decreased significantly at most sites, although not as much as sulphate. Decreases in concentrations of ammonium are still difficult to observe. Because of the strong influence of precipitation volumes, temporal trends in depositions are more difficult to be observed compared to concentrations. In fact, the Mann-Kendall trend analysis showed again significant decreasing trends in sulphate deposition at almost all sites, while significant decreasing trends in nitrate deposition are detected only at 4 sites (instead of 6) and of ammonium only at 1 site (instead of 3 sites).

Similar to rainwater concentrations, deposition maps of total Sulphur (Fig. 2), total nitrogen (Fig. 3), and total oxidized and reduced nitrogen also showed strong spatial variations with highest depositions at low latitudes and altitudes. Temporal changes of total sulphur and total nitrogen loads are in line with observations of wet deposition of sulphate, nitrate and ammonium. Total deposition of sulphur decreased considerably during the monitoring period. Average deposition of total sulphur decreased by 66% from 111 meq m⁻² yr⁻¹ (1988-1992) to 38 meq m⁻² yr⁻¹ (2008-2012). Deposition of total nitrogen also generally decreased. Average oxidized nitrogen (Nox) decreased from to 78 meq m⁻² yr⁻¹ to 61 meq m⁻² yr⁻¹ (by 22%) and reduced nitrogen (Nred) from 75 meq m⁻² yr⁻¹ to 65 meq m⁻² yr⁻¹ (by 13%). However, nitrogen deposition is still disturbed by precipitation volumes. During particularly wet periods (e.g. 1998-2002), values can increase again to levels similar to those observed at the beginning of the monitoring period. In fact, average deposition of total nitrogen during the different time periods were 152, 125, 148, 117 and 126 meq m⁻² yr⁻¹, respectively. Nox and Nred contributed with about 50% each to the total. The stronger reduction in total deposition of sulphur compared to nitrogen caused also a change in the relative contribution of sulphur and nitrogen to the acidifying load. During 1988-1992, 41% of the potential

acidifying load was determined by sulphur and 59% by nitrogen compounds, whereas at present the contribution of sulphur has almost halved (23% sulphur and 77% nitrogen). A further result of the analysis was that, for both sulphur and nitrogen, wet deposition contributed most to total deposition. From 1988 to 2012, the deposition of the potential acidifying compounds was determined between 67% and 79% by wet deposition.



Figure 2: Deposition maps of total sulphur

Temporal variations of lake water quality (annual median values with their 10th, 25th, 75th and 90th percentile values of pH, alkalinity and concentrations of base cations, sulphate and nitrate) are shown in Fig. 4. In general, concentrations of sulphate, nitrate and base cations decreased, while pH and alkalinity increased. The trend analysis revealed that between 1980's and 2014 sulphate decreased significantly in 15 lakes, nitrate in 13 lakes, while pH and alkalinity increased significantly in 14 lakes. During the 1980's 12 lakes out of 20 had autumn alkalinities below 20 meq m⁻³, while today critical values are measured only in 5 lakes (Gardiscio, Starlarescio, Tomeo, Barone, Sascola). Similar results can be observed for pH. During the 1980's 9 lakes had autumn pH's below 6, while today autumn pH is below this value only in 3 lakes (Gardiscio, Starlarescio, Tomeo). Immediately after snow melt the situation is slightly worse. Two lakes have pH's that are normally above 6 in autumn but can drop below this critical value in spring/beginning of summer (Barone, Sascola). Interestingly, 3 lakes are characterized by significant increasing sulphate trends.



Figure 3: Deposition maps of total nitrogen

Comparing the sulphur loads on lake's catchment thought time with their estimated outflow, it results, that at the beginning of the 1990's (1988-1992), on average 80% of the sulphur content in lakes originated from the atmosphere. Today (2008-2012) the same percentage decreased to 50%, indicating that because of the decrease in sulphur deposition the geogenic sulphur gained in importance. With regard to nitrogen, the lake output has always been lower than the atmospheric input and the relative retention did not change greatly through time. During 1988-1992 the average relative retention was 62% and during 2008-2012 66%, indicating that the decrease in nitrogen deposition was comparable to the decrease in in-lake nitrogen concentrations. Interestingly, the mean relative nitrogen retention in lakes correlated well with the mean catchment slope, suggesting that with increasing mean catchment slopes the relative nitrogen retention decreases, which does not surprise since at lower slopes the accumulation of soils and the diffusion of vegetation might be slightly higher.



Figure 4: Temporal variations of annual median chemical values and their 10th, 25th, 75th, 90th percentiles measured in 20 Alpine lakes (calculated from autumn mean values).

4 Discussion

The correlation with latitude and altitude of rainwater concentrations and depositions reflect the influence of long-range transboundary air pollution moving along a south to north gradient from the Po plain toward the Alps and the distance from pollution sources. The trend analysis showed a significant decrease of both rainwater concentrations and depositions for sulphate and nitrate at most sites and for ammonium at a few sites. Similar results can be observed for depositions of total sulphur, total oxidized nitrogen and total reduced nitrogen. This reflects the decrease of emission reductions. Interestingly, mean depositions of total sulphur and total oxidized nitrogen are still higher in Southern Switzerland than the Swiss averages. In Switzerland, between 2008 and 2012, average depositions of total sulphur, total oxidized nitrogen and total reduced nitrogen were 15, 30 and 68 meg m⁻² yr⁻¹, respectively (Gauss et al., 2014), whereas in the Canton of Ticino the respective values were 38, 61 and 65 meg m^{-2} yr⁻¹. Interesting is also the different contribution of sulphur and nitrogen compounds to the total acidifying load. During the most recent monitoring period (2008-2012), in the Canton of Ticino total sulphur, oxidized nitrogen and reduced nitrogen contributed on average by 23%, 38% and 39%, respectively, to the potential load of acidity, while Switzerland's averages were 15%, 26% and 59% (Gauss et al., 2014). Different is also the contribution of wet and dry deposition to total nitrogen deposition. Between 1988 and 2012, in the Canton of Ticino the contribution of wet to total nitrogen deposition was on average 71%. At the Northern side of the Alps, because of lower precipitation volumes, this percentage is smaller. Thimonier et al. (2005) determined a contribution of 40-60% between 1995 and 2001 in the Jura and the Central Plateau and EKL (2005) reported an average Swiss value for the year 2000 of 51%.

Subtracting minimum critical loads of nitrogen (CLN) of 1km x 1 km sites with sensitive vegetation from total nitrogen depositions, maximum exceedances of CLN in Southern Switzerland can be calculated (FOEN in prep.). In 1990 95% of the sensitive sites had positive exceedance values, ranging between 1 and 39 kg N ha⁻¹ yr⁻¹ (mean 14 kg N ha⁻¹ yr⁻¹), while in 2010 the same percentage decreased to 91%, with values ranging between 1 and 29 kg N ha⁻¹ yr⁻¹ (mean 9 kg N ha⁻¹ yr⁻¹), indicating that for sensitive vegetation the exceedance of CLN is still a problem in Southern Switzerland. Compared to nitrogen deposition, the situation looks better for deposition of acidity. Critical loads of acidity (CLA) for Swiss forests soils have been calculated with the simple mass balance model (Achermann et al., in prep.). During

1988-1992, 81% of the national forest inventory sites had depositions of nitrogen and sulphur that exceeded critical loads. The same percentage decreased to 67% in 1993-1997, 72% in 1998-2002, 40% in 2003-2007 and 43% in 2008-2012.

Similar results were obtained for Alpine lakes. At the beginning of the monitoring period, alkalinity measured in autumn surface water was below the critical limit of 20 meq m⁻³ (Posch et al., 2007) in 60% of the lakes, while at present the same percentage decreased to 20%. However, there might be lakes that will never reach alkalinities above 20 meq m⁻³ and probably never did. For example Laghetto Gardiscio had alkalinities around -10 meq m⁻³ in the 1980's and -5 meq m⁻³ today and pH's around 5.0 in the 1980's and 5.3 today. This lake is the highest lake here studied (2580 m a.s.l.), with a short snow free period, with a very small steep base poor catchment (12 ha), with almost no soil and vegetation. Rainwater, even if once not polluted anymore (pH ca. 5.6), will probably never have the chance to be greatly buffered in that catchment. Next to changing atmospheric depositions, climate change might also influence the time trends of lake chemistry. Increasing sulphate concentrations in 3 lakes (Leit, Morghirolo, Mognola) are probably related to melting of rock glaciers present in the catchments of these lakes (Thies et al., 2007).

To conclude, the Canton of Ticino, compared to the rest of Switzerland is particularly exposed to deposition of atmospheric pollutants because of its proximity to the Po Plain in Italy, an area with high anthropogenic emissions, and because of its rainy climate. In addition, since base poor rocks, mainly gneiss, dominate most of the lithology in Southern Switzerland, many high altitude surface water bodies with small catchments and forest soils are sensitive to acidification. While deposition of acidifying pollutants is nowadays below the critical load at most sensitive sites, deposition of nitrogen is still too high. Further emission reductions of both NO_x and NH₃ in Switzerland and in its neighboring countries are therefore necessary.

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3. Recent trends in major ions in lakes and rivers in northern Italy

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1. Introduction

Beside atmospheric deposition, climate drivers and land cover change may affect long-term trends of ionic species in surface waters (Houle et al. 2010; Mitchell et al. 2013). Several studies put in evidence the prominent role of climate in driving long-term change in solute content and in the ratios between the various ions in surface water bodies (e.g. Rogora et al. 2003; Manning et al., 2013). Particularly important for lakes in the alpine area is the effect of change in snow and glacial cover in lake catchments: thawing of the cryosphere (glaciers and permafrost) may indeed affect the export of chemical compounds to surface waters, with effects on water quality and the biota (Thies et al., 2012, 2013).

In recent times there has been also a growing concern about the effects of increasing chloride concentration in surface waters, due to their potential damage to aquatic life (Corsi et al., 2010). This environmental issue has been widely assessed in lakes and rivers in urban areas or close to major roads in Europe and US, where increasing Cl concentrations has been attributed to the widespread use of NaCl as road de-icers in winter (Kaushal et al., 2005; Novotny et al., 2008; Müller and Gächter, 2012).

In Italy the use of NaCl as road de-icing agent is limited to northern regions and mountain areas. Na and Cl represent a minor part of the dissolved ions in surface water in Italy, and temporal changes of their concentrations have never been assessed, due to the low availability of long-term data series, of adequate analytical quality. However, road salt may be a source of Cl to surface waters in the northern part of the country, especially in mountain catchments (Rogora et al., 2005).

Here we focus on trends in major ions, with special attention towards Na and Cl, in both Lake Maggiore and the ICP Waters sites located in its watershed, with the aim to discuss the possible causes of the observed changes. We used chemical data (surface water and atmospheric deposition) covering a 25-30 year period, collected within national and international projects. In particular, data on Lake Maggiore and its tributaries have been collected in the framework on long-term studies funded by the CIPAIS (International Commission for the Protection of Waters between Italy and Switzerland). A detailed analysis of the Na and Cl trends and budget in Lake Maggiore and in the other deep subalpine lakes can be found in Rogora et al. (2015).

2. Study area

Lake Maggiore is a big and deep lake (surface area 212 km², max depth 370 m) located in North Western Italy, in the so-called subalpine Lake District. Together with lakes Lugano, Como, Iseo and Garda, it forms the most important water district in Italy, located south of the Alps, in one of the most densely populated and highly productive area of the country. These lakes represent a strategic water supply for agriculture, industry, fishing and drinking and are important resource for recreation and tourism. In terms of water volume, they represent about 80% of the total freshwater volume in Italy (Salmaso et al., 2014).

As regards chemical trends in Lake Maggiore, the focus have been mainly on nutrients and eutrophication related problems (Mosello et al., 2001). Trends in major ions such as Ca, Mg, Na, K, Cl have rarely been addressed.

Lake Maggiore have been sampled at the deepest point (Ghiffa station) since the beginning of the monitoring. Here we used averaged concentrations of major ions, obtained from 13 samplings at different depths along the water column, weighted on the volume of each lake layer. The main tributaries of the lake (13 rivers) and the outlet have been systematically monitored since 1978 to evaluate algal nutrients and main ion loadings into the lake (Mosello et al. 2001)

In the watershed of Lake Maggiore, further study sites have been regularly monitored for water chemistry in the framework of studies dealing with atmospheric deposition and its effect on surface water. These sites are subalpine streams and high altitude lakes (above 2000 m a.s.l.), for which data are available since the early 1980. Some of these sites are part of the ICP Waters network and of the LTER (Long-Term Ecological Research) Italian network. With respect to Lake Maggiore, alpine lakes have been studied with a lesser frequency and only during the ice-free period (1-2 samplings per year in late summer/early autumn).

Both lake and river samples have been analysed for the main chemical variables at the laboratory of CNR Institute of Ecosystem Study, in Verbania Pallanza, Italy. For details on the analytical methods and quality controls, see http://www.idrolab.ise.cnr.it/

3. Results and discussion

We assess long-term trends of major ions in Lake Maggiore for the period 1988-2015 (Fig. 1). The data put in evidence an increase in total ion concentrations and electrical conductivity. Most of the increase, in absolute value, was due to Ca^{++} and HCO_3^{-} ions, followed by Na⁺ and Cl⁻. Although the increases of Na and Cl were lower in absolute value with respect to those of calcium and bicarbonate, their percent of increase in the two periods was the highest among all ions (26% for Na and 39% for Cl, with respect to 3-8% for the other ions). The rate of increase over the whole period was 1.23 and 1.41 µeq L⁻¹ y⁻¹ for Na⁺ and Cl⁻, respectively (Rogora et al., 2015).



Fig. 1 – Long-term trends of monthly concentration of major ions and conductivity in Lake Maggiore (average values on the water column).

Concentrations of Na and Cl also increased in the main tributaries of Lake Maggiore, with steeper slope in recent years (after 2001). Positive trends of Na and Cl were detected in all seasons, but with peak values in winter. Furthermore, significant positive trends were found both in major rivers with wide catchment and in small mountain streams (Fig. 2). These observations, coupled with the similar rate of increase and the synchronous temporal pattern of Na and Cl lead to the hypothesis that their main source is sodium chloride from roadsalt.

We extended the analysis of Na and Cl trends also to other sites in lake Maggiore watershed: positive trends were found for instance at the ICP Waters sites Lake Mergozzo, a small subalpine lake, and Rivers Pellino and Pellesino, a couple of small subalpine streams draining into Lake Orta (Fig. 3). These results confirmed that the increase of Na and Cl is a widespread phenomenon in the study area and affect different type of ecosystems.

Thanks to the availability of data on chemical loads, a mass budget was calculated for Lake Maggiore, to assess the equilibrium concentrations of Na and Cl in lake water with respect to external loads. Furthermore, an analysis of the relative contribution of different Cl sources (atmospheric deposition, population, road salt) to the total input to the lake was performed. Details on this analysis are provided in Rogora et al. (2015). The results was that road salt is the primary source of Cl to the lake, representing more than 40% of the total input, while population and atmospheric deposition contributed for 23 and 13%, respectively.



Fig. 2 – Long-term trends of Cl concentrations (monthly data; left panels) and distribution of values in the various months (right panels) in two tributaries of Lake Maggiore: a subalpine river, with a wide catchment (River Ticino) and a small mountain stream (River Cannobino).



Fig. 3 – Trends of Na and Cl concentrations at the Italian ICP Waters sites Lake Mergozzo (annual data at winter overturn) and River Pellino (monthly data).

4. Concluding remarks

As regards Na and Cl trends, we hypothesized that the increasing use, without specific regulation, of NaCl as road de-icer is probably the main responsible for the observed changes. At present these changes do not represent a threat for water quality or for the biota. Measured concentrations of Na and Cl in both rivers and lakes are well below the recommended limits for drinking water, according to the European regulations. However, considering the number of sites and the relevant volume of water involved, these trends are indicators of an important chemical change, which warrants further investigation.

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4. Metal form distribution in the surface waters of the Kola North, impacted by atmospheric deposition

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1. Introduction

Forms of finding metals in natural waters are important information about the level of toxicity of a natural object. According to numerous published data, the most dangerous form of migration of heavy metals (except mercury) is an ionic form. However, the study of metals distribution forms in each water object is a task that requires a huge physical-chemical work. Understanding the patterns of element distributions in surface water and the reasons for the increase in their concentrations at the regional and global level is one of the most urgent problems facing the environment. Enrichment of surface water by metals is the result of both natural processes and human activities. The anthropogenic impact in the discharge of trace elements in the environment has increased dramatically over the last century, which is associated with the ever-increasing volumes of extracted metals and their dispersal in the environment. We investigated more than 50 lakes at different distances from industrial plants, lakes with high humus content, and anthropogenic acidification of lakes (Fig. 1). The aim of our research was to investigate the distribution of the metals by the forms on the Kola Peninsula in terms of different anthropogenic load.



Figure 1. Location of the investigated lakes

2. Methods

Analyses was conducted based on methodology from ICP-water monitoring and ionic-change resin (Fig. 2) in laboratory of Kola scientific center (Apatite).





Figure 2. Laboratorial preparation and method. All measurements were carried out at the Institute of methods of Microelectronics Technology and High Purity Materials, Russian Academy of Sciences (Chernogolovka).

When we analyzed the results, we use the following gradation of lakes. Firstly, location lakes the distance from the pollution source:

- 1. Lakes without the direct source of pollution
- 2. Lakes near the source of pollution
- 3. Lakes on excision the source of pollution
- Secondly, nature of chemical composition of lakes (features of acidification of lakes):
- 1. Nature acidification
- 2. Anthropogenic acidification

Thirdly, chemical properties of elements

- 1. Alkali, alkaline earth metals
- 2. Subgroup of chrome
- 3. Subgroup of lanthanides, actinides
- 4. Ions of iron and aluminum
- 5. Subgroup of stanium

3. Results and discussion

Lakes, without direct source of pollution

Four lakes on the Kola Peninsula have been selected for the determination of the forms of metals. 2 selected lakes near the settlement Umba, 1 near Epish and 1 on the way to Kovdor.

Tuble 1. Gharacteristics of the faites								
Parameters	Umba	Umba	Epish	Kovdor				
pН	6.86	4.55	7.03	5.53				
Cond20	45	20	81	19				
Color	42	127	10	128				
COD Mn	7.39	20.57	1.90	24.92				
AIK	323	0	431	39				

Table 1. Characteristics of the lakes

The results of the distribution of iron and aluminum (Fig. 3) forms showed an increase in the degree of binding to organic materials in water under the pH below 6 and color than 100. This is, firstly reduction and hydrolysis processes and secondly, different ratios of concentrations of iron and aluminum in system. Iron concentration excess over the concentration of aluminum is more than 2 times the characteristic lakes with a pH below 6. The concentrations of aluminum and iron are approximately the same (near 300 μ g/l) to lakes with pH above 6.



Figure 3. The distribution of iron and aluminum by forms

Behavior of heavy metals such as copper, lead, cadmium and zinc depends on their content in natural waters (rule the material balance), and the physic-chemical nature (conditional, concentration stability constants of complexes).

According to the obtained data, lead and zinc are characterized a high degree of binding and copper and cadmium opposite are characterized by the sediment and dissolved forms of a finding. The behavior of groups of elements that are associated with leaching of trace elements (such as heavy metals, iron, aluminum) such elements scandium subgroup shows a high affinity to hard centers of Pearson by humus substances. This fact is directly related to the physical and chemical nature of the elements. Especially indicative the fact of increasing the degree of binding of scandium with organic components with increasing anthropogenic load (details will be discussed later), which is associated with an increase in the content of accompanying elements in natural waters.

The behavior of nickel (human primary indicator of contamination) and cobalt varies with pH and amount of organic substances. Naturel water with a pH of less than 6 and a color of more than 100 are characterized by high degree of complexation. In addition, the constant hydrolyzability of nickel in 2 times higher than cobalt so alkaline conditions (above pH 7) cobalt complexes with humic substance largely.

In unpolluted natural waters lanthanides and actinides influence on the complexation is extremely small due to trace amounts in the system, as well as elements of the subgroup of chromium.

The data have allowed constructing the following series of activity of metal at various pH

pH< 6 Fe>=Al>Y>Pb>Zn>Cu>Sc>Ni>Co>Cd pH>6 Cu>Fe>Al>Sc>Co>Ni>Zn>Pb>Y>Cd

Lakes with direct source of pollution

Samples of the 4 lakes near industrial sites were selected for the study of finding forms of metals. 2 samples were taken near the Arctic village, near the plant, 1 sample was selected near the mining processing complex in Kovdor, 1 sample from Lake Monche (Lovozero) near the copper-nickel manufacture.

Table 2. Characteristics of the lakes								
	Zap	olyar						
	y		Kovdor,	Apatity,				
Parameters	(Ni	ckel)	(Smelter)	(Lovozero)				
рН	7.01	6.50	8.21	6.62				
Cond20								
	99	56	273	28				
.								
Color	21	58	43	15				
COD Mp								
	4.80	10.54	6.86	3.12				
A 11Z								
AIK	299	316	1156	169				

Table 2. Characteristics of the lakes

Natural waters characterized by high alkalinity and pH about 7. The iron ions at such pH values are more form hydroxy compounds and sorption aggregation compared with aluminum ions. Therefore, iron is characterized by three forms of being in natural waters. Under conditions of high load anthropogenic, chromium ions characterized by sufficient complexing capacity (pH 6.5), which may be due to an increase in the concentration of the metal is several times as compared with lakes without direct sources. Manganese in such conditions has a high capacity to form suspensions. The complexation of heavy metals is modified as follows: as well as for lakes without a direct source of pollution, zinc is complexed by more than 50%, copper also forms complexes with organic matter actively due to a significant increase in concentration. Depending on the type of copper coming from the wastewater, copper may form sorption unit and the low-molecular inorganic compound. Forms of a finding of nickel in natural waters with a direct source of pollution range from units to sorption complexes with organic matter. A significant

increase in metal concentration shifts the equilibrium in the system towards formation of high-molecular compounds.

An interesting feature of the distribution of elements on the forms such natural waters is increasing the complexation with organic matter for the elements of the lanthanide series. Lanthanide elements are associated elements of many rocks of the Kola Peninsula, which explains the increase in their concentration in the areas near the plant.

The affinity of these elements to an organic substance as follows: Fe>Al>Zn>Ni>Cu>Pb>La>Ce>Co

Changing forms of finding metals with increasing distance from the source of pollution – Cu-Ni smelter (% of complex form). Samples of the 8 lakes removal of copper-nickel plant were selected for the study of the metals forms. pH of all lakes were less than 7. In some lakes Alk were more than 1000 μ eq/l.

						•		
Parameter	1	2	3	4	5	6	7	8
рН	6.50	6.55	6.25	6.18	6.64	5.99	6.16	6.48
Cond20	56	24	22	29	27	38	27	30
Color	58	18	30	51	28	55	67	62
COD Mn	10.54	3.76	6.87	9.29	5.58	13.74	11.00	10.67
AlK	316	85	73	140	151	67	116	156

Table 3. Characteristics of the lakes

Aluminum ions and iron is characterized by decrease of percentage complexation in the distance from the source of pollution, due to the change in concentration ratios of these elements. An interesting feature here is this type of gradual decline in the subgroup elements of tin (in particular, the Itria) to remove contamination. It is a measure of the degree of water pollution, as it itry accompanying element in the enrichment water. Zinc ions to remove the source of pollution are characterized by a sharp decrease to 0%. While lead ions irregularly varying degrees of complexation with organic matter. Very significant reduction and complex forms of nickel and copper, as well as their contents are different variations indicate a decrease in aerobic of pollution away from the source. Reduced nickel chelation with respect to point out a copper to copper ions greater affinity to organic ligands and also to reduce the influence of the factor of the material balance (Fig. 4).



Figur. 4 The distribution of some elements by forms. The distribution of metal affinity for organic substances should be Lake1 - Lake 4 Ni>Cu>Fe>=Al>Y and Lake 4 – Lake 8 Fe>Al>Cu>Ni>Y

In addition, experiments were performed to calculations different forms of metals (a technique Dinu Moiseenko. 2015) based chemical composition of the lake water. Results showed high similarity - more than 80% for ions were identified.

4. Conclusions

1. The lake near the source of pollution is typical aluminum content greater than the natural acidification of lakes. This affects the mass balance and distribution of other elements by forms.

2. In lakes near pollution dominated the content of zinc and complex forms. For the acidification of lakes with a natural characteristic of the effect of ions of copper and lead in the overall forming.

3. With increasing distance from the source of pollution - gradual decline in the elements of tin (in particular, the Itria) was used to remove contamination.

5. Air pollution impacts on Debed River waters in different parts of the watershed

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1. Introduction

The river Kura-Araks basin is the region, where the entrance of various chemicals of transboundary and trans-regional origin is one of the most intensive on the planet, especially on the territory of Armenia (where , e.g. 50% of total flow of the river Araks is formed, and deposit of chemicals from precipitations makes 30-40g/km2/year), south Azerbaijan, Iran. Especially negative effect on water quality is produced by acid rains, which wash the topsoil, change the acid conditions of the river waters and discharge heavy metals into river waters.

In Armenia negative effects of the mining industry activities is most strongly felt in the north and in the south of the country, where the environment has been continuously affected by both wastewater, and air pollution discharges. In particular, intensive development of mining activity within the Debed River basin in north part of Armenia causing heavy metal pollution of river water and water acidulation, which has a local character. Despite the fact that some of the actions being taken, however the issue of water pollution of the river is vital.

2. Study area

2.1. General characteristics of the Debed River and its main tributaries

The Debed River – one of the main tributaries of Kura River - has the highest water level among all rivers in the Northern Armenia. The river is 178 km long, and its watershed covers an area of 4,050 km² (Fig.1). It heads from an altitude of 870m, from the point of cross-section of rivers Pambak and Dzoraget, it then flows into Khrami River, in the territory of Georgia. River Debed separates the mountain ridges of Somkheti and Gugarats. Until the point of Ayrum, the river flows through a deep gorge (300-500m), then passes by a lowlands. The remaining, more low water rivers of the area are mainly the tributaries to Debed, and area characterized by high speed flows and abrupt deviations of the river bed. The Valley of Debed River has a form of a narrow and deep valley, which relatively expands in the area of Alaverdi. Tributary Lalvar passes through the city of Alaverdi.



Fig.1.Hydrological Map of Debed River and monitoring sites.

River feeding is of combined character, the ground feeding coefficient varying from 34 to 56%. Mean annual water discharge is 35.8 m³/sec. Twenty-one settlements are located along the river banks, including the cities of Spitak, Vanadzor, and Alaverdi. The Akhtala Mining and Dressing Plant and the Alaverdi Mining and Smelting Enterprise operate within the limits of the basin.

Our investigations based on the data of 9 monitoring sites on the river, which is located in different parts of the watershed (Fig.1).

2.2. Historical assessment of urbanization

Alaverdi is an area with rich history, which is reflected in a number of historical-cultural monuments located within the territory. The copper mines of the area have always been in the center of attention of many foreign companies. In the 19th century already, the French constructed a small factory there, which was later reconstructed by the Greeks. The Tbilisi-Alexandropol railroad, which passes by the left bank gorge of Debed River, has been constructed in 1899. After that, the final processing facilities for copper were constructed in Debed valley, where Alaverdi Mining and Smelting Enterprise is currently located. The village located in the gorge of Debed River, was named "Manes". In 1912 that village, as well as the upper residence thereof (by Alaverdi and the factory "Piritis"), had a permanent population of 3235 people, except the workers of the plant. In the years of the World War I, the number of residents had decreased to 2711 (in 1915).

The mining activity and industrial development in Alaverdi started in 1960s, when very large investments were made by the Soviets for the purposes of reconstruction and operationalization of the plant.

2.3. Description of economic activities

Till the 90s Alaverdi was developing as a center of mining industry. The earthquake and the transit to a new socio-economic reality had its impacts on the industries of Alaverdi. The copper plant, the apparel plant, the beer factory and the diary production stopped. The production of construction material decreased dramatically.

After the closure of the mining plant in 1988-1989 years, the territory thereof was used by various industrial facilities, with regularly changing profiles and capacities. The mining plant, reopened in 1997(Table 1) with trend of increasing, today comprises the largest portion of the GDP for the Alaverdi Community and the whole Marz of Lori. The copper processing branch of the mining facility employs 75% of the total number of labor of the city, occupies 10% of construction industry, 10% of food industry and 5% of light industry[3].

f copper , tonnes	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008
Volume o produced	285,98	2789,3	535	6840,42	4955,2	4669,9	6258,7	9475,7	9882,49	8817,55	6954,05	6480,46

Table 1. Alaverdi Copper Molybdenum Plant production dynamics for the 1997-2008 period.

Source: Data provided by ACP company, 2009.

2.4. Summary of local socio-economic factors

The economic sector of Alaverdi is very weak, despite the fact that the region has a number of significant factors that could contribute to its economic development: favorable environment (mild climate, forests, etc.), human resources (specialists, who are experienced in mining, agricultural and other production) and natural resources. Taking into account the rich forested areas around Alaverdi and the existence of a large number of historical monuments, tourism has been identified as one of the priority sectors of development for the region. That objective can be performed through improvement of community infrastructure and development of services, which require additional training and capacity building. The demographic indicators of Alaverdi are also concerning. Within a period of 20 years the number of the

population has decreased by 40%. Most of the young families and the labor force have left the city because of the socio-economic conditions. Besides, the seasonal migration in the region, mainly to the Russian Federation, for job search has become traditional.

2.5. Description of the river pollution sources and conditions of the ambient air in Alaverdi

The pollution of the river within the area of the city is mainly because of the discharges of domestic wastewaters into the river, as well as by direct release of solid waste and industrial wastes into the river, and indirect spreading of air pollution. Flow of the river, saturated with industrial wastewaters, used for irrigational purposes, seriously pollutes soil (basically with copper) along the coastal territories. According to the data by the year 1990 Alaverdi Mining and Smelting Enterprise annually discharges 1.1 mln m3 polluted wastewaters, containing significant amount of sulphur trioxide, sulphur acid and heavy metals.

The inflow of pollution through the Lalvar tributary is significant. The water in Lalvar would seem clean from the first sight, however, flowing through iron and copper mines and passing closely by ACP factory those waters accumulate significant concentrations of copper, molybdenum and vanadium therein, which directly flows into Debed. That is clearly visible in the point of confluence of Lalvar and Debed rivers, where the colors of the water and the stones in it differs. It might be mentioned also that Debed is a high-speed and a high-assimilatory capacity river; and the pollution from Alaverdi wastewaters has significant impact only in separate areas, and has not a total impact.

2.5.1. Emissions from a permanent source.

In the end of 80s, the maximum annual quantity of emissions to the ambient air in Alaverdi was over 41 thousand tones, or the 26% of the emissions from the all stationary sources in the city. In 2007 there were 13 stationary pollution sources, two of which had defined permissible records of pollution quantity; and the total number of emissions from the static pollution sources was 25,2 thousand tones that were completely released into the atmosphere. 99,4% of the emissions into the atmosphere are of sulfur anhydride (25021,4 tones); carbon oxide comprised 0,1% (27,4 tones). The total quantity of the heavy metals in the ambient air was 38,6 tones. The amount of dust released into the atmosphere was 196,8 tonnes, 4% of which (7t) was the organic dust. It should be mentioned that there is no pollution prevention systems enacted hence all the substances released from the pollution sources appear in the environment [3].

The average calculation of ambient air pollution spread in the territory of Alaverdi and the vicinities thereof is clearly presented below (Table 2), where the 4 pollution zones are shown[3].

MAC exceedances	The calculated densities within	Lack of winds /dead calm/	Priority directions of V= 5m/sec		wind,	
	the borders of the zones, mg/c.b. m		East	Sout h- East	South -West	Wes t
1-st zone: At a distance of 90 m from the source – 80,6 at a distance of 180 m – 30,5, at the border – 1,2 times	0,6	386	454	363	386	499
2-nd zone– up to 0,44 times	0,22	613	840	704	749	885
3-d zone– up to 0,3 times	0,15	885	1226	931	1067	1294
4-th zone– up to 0,2 times	0,1	1175	1859	1680	1703	1930

Table 2. Average circumference of spread for sulfur dioxide

3. Methods

The **KNS** index was applied to assess the role of the contribution of sulfates and nitrates in acidulation processes; the index was estimated as the ratio of $NO_3^{-}/(SO_4^{2-}+NO_3^{-})$, along with the **Alk/SO4 index**,

reflecting the ratio of water alkalinity versus the amount of sulfates. Regions where sulfates are the determining factor of acidulation have **KNS** values approaching 0, increasing as the contribution of nitrates in the anionic composition of the waters builds up.

Samples were collected, conserved, transported and stored following the Standard Operational Procedures (SOPs) developed based on the methods of ISO [1]. Field measurements were done on a monthly basis. In-situ measurements included hydrogen index (pH), conductivity, turbidity, dissolved oxygen, temperature and salinity and were done using portable analyzer.

Common SO₄, HCO₃, NO₂ and NO₃ ions in water were studied on a quarterly basis and were determined in non-acidified and filtered samples. Element and compound concentrations were measured following the developed ISO-based SOPs [2].

4. Results and discussion

To assess water quality by the acidification criteria, we carried out certain studies and calculations. According to the measurements carried out at the Alaverdi-Akhtala and Alaverdi -Lalvar sites, it was revealed that pH varies in certain range depending on the season and the year and can change to very low values (5.8-6.8, which is the upper limit of the acidulation hazard).

According to the data [4], the amount of sulfur dioxide in the air within the Debed river basin, and, in particular, in the region of Alaverdi, has been 6 to 7 times higher than the MPC. (Fig.2).

The analysis of the condition of the ambient air in Alaverdi shows that the stationary source of air pollution – the copper processing plant. It does not have air cleaning modern equipment, which can provide full treatment of the polluted gases. Although I would like to note, that a few last years the whole volume of emissions are released into the open environment after the partial filtration through the new chimney. Anyway today we have again the problem with pollution.



Fig.2. Sulfur dioxide in the air within Debed River basin during 2013-2014

As concerns sulfate contents in the river water, it should be observed that in the Akhtala and Lalvar rivers (tributaries of Debed River), these contents exceeded the MPC from 5 to 6 times during 2014 (Fig.3).



Fig.3. The sulfate contents in the water of the river Debed in different monitoring sites.

In the meantime, the studies of Alk/SO4 indicated that in the mentioned study regions the ratio varied in the limits of 2.08-11.9 (Fig. 4). According to the standards, the rates of the studied indices attest to onset of water acidulation process in the Akhtala and Lalvar Rivers region, which poses potential hazard for the water ecosystem.



Fig.4. Alk/SO4 for the river water in studied watershed.

The index of KNS, calculated for the all sites in the basin, bears evidence of an increased contribution and share of sulfates in the water acidulation process. For instance, the index varied in the limits of 0- 0.17 in the region of the Pambak River (main tributary of the river Debed) near the Vanadzor city, in the area of the Lalvar and Aghtala tributaries it was in the range of 0.07-0.09, while the range established near Shnogh and Ayrum settlements was about 0.012-0.15 (Fig.5).



Fig.5. Index of KNS, calculated for the all monitoring sites in the basin.

5. Conclusion

As a summary, it is stated that the water acidulation process is continued now in the Alaverdi-Akhtala and another Alaverdi - Lalvar River regions, which indicate on progressing trend of acidification in the basin of the river Debed.

And though today water acidification processes are merely local nature, lack of attention to the observed pollution and concrete actions for the suspension of the effects can lead to the spread of negative processes along a river flow and the creation of conditions for the expansion of the area of its impact.

From the point of view of the river ecosystem the research of the river flora and fauna is needed for the assessment of the possible changes in its composition and strength in connection with the influence of the new factor.

Regretfully, lack of information about the quality of atmospheric precipitation in the considered region does not allow us to achieve better accuracy in estimating the influx of air sulfates and the critical load imposed on the river waters.

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6. Mercury in fish in a TOC-changing world: A model to improve the general dietary regulations and focus on individual lakes versus large scale catchment advices

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1. Introduction

Fish is the number one source of methyl mercury (MeHg) to humans (Campbell *et al.* 2003). As nearly 100 % of the MeHg in fish muscle is absorbed during digestion (Carrier et al. 2001), trade levels and recommendations for consume is an important environmental management tool to protect people in general, and especially "groups at risk": pregnant and breastfeeding women, and children.

The European Water Framework Directive (EWFD) recommends avoiding consumption of fish filet with concentrations as low as 0.02 mg Hg/kg wet weight (w.w.) (EWFD 2011), but still use a trade level of 0.5 mg Hg/kg w.w. (0.5 ppm). US-EPA (2014) recommendations for "Groups at risk" are to avoid consumption of fish containing maximum 0.2 mg Hg/kg w.w. Norway has the same trade level as EU, but has at present no dietary advice for Hg content, except a general recommendation to the public not to eat pike (*Esox lucidus*) at all, European perch (*Perca fluviatilis* – perch from here on) longer than 25 cm, and brown trout (*Salmo trutta*) and Arctic charr (*Salvelinus alpinus*) larger than 1 kg. Groups at risk should not eat any freshwater fish (Matportalen 2016).

Monitoring data on mercury in Norwegian freshwater fish have demonstrated increased Hg (measured as total (THg)) load in populations of European perch over a 20-year period (Fjeld and Rognerud 2009). One theory for such an increase links the observed Hg increase to an increase in total organic carbon (TOC) in freshwater, imposed by reduced acidification (Monteith et al. 2007) and reduction in aluminium (Al) concentration that could have precipitated the organic from the water. The theory is supported by observations of higher concentrations of MeHg is found in fish inhabiting TOC-rich freshwater habitats (Braaten et al. 2014).

Providing fish Hg data from every lake in Norway, in order to provide safe food recommendations, is an impossible task. A model-based approach may prove more rational, provided that the models are based on representative datasets. The Norwegian University of Life Sciences, NMBU, have for many years used master students as a mean to uncover the Hg situation in important freshwater lakes in Norway (Rosseland and Borgstrøm 2014).

2. Methods

In the 2012 -2014 period, four lakes in the Degernes and Visterflo area in the County of Østfold, SE Norway, all located within the catchment of River Glomma, were investigated through gillnetfishing surveys (Myreng 2013, Hartmann 2014, Våge 2014), (Figure 1). The gillnet surveys and fish organ sampling followed the ICP Waters manual (ICP Waters 2010). Water samples were analyzed at NIVA

laboratory. Sediment samples were analyzed for trace metals, including selenium and radionuclides, and trace metals and Hg in fish muscle tissue were analyzed at NMBU. Stable isotopes of N and C in fish muscle and individuals of species at all trophic levels in the food web were analyzed at NMBU (Øvre Sandvann) and Institute of Energy Technology (IFE), (Lake Holmetjenn, Lake Djupetjenn and Visterflo). All individuals were aged using species-specific structures (otoliths in perch, minnows and brown trout, operculum in roach and metapterygoid bone in pike).

3. Results and discussion

The water quality at the Degernes area (Lake Sandvann, Lake Holmetjenn and Lake Djupetjenn) were typical for the region with low calcium levels (1.3-2.0 mg Ca/L), low selenium (<1-1.1 μ g Se/L), and moderate TOC (7-9 mg C/L) and pH (5.4-6.2). The Visterflo area, however, where influenced by some brackish water, Table 1.

Lake Sandvann had four species of fish: brown trout, perch, roach (*Rutilus rutilus*) and minnow (*Phoxinus*) *phoxinus*), all being at the same trophic level in the food web, Figure 2. Lake Djupetjenn and Lake Holmetjenn contained mainly perch and pike, while Lake Visterflo had a mixture of species. In the model presented here, we use perch only from the four lakes.



Figure 1. Study areas in Østfold County, SW Norway, with lakes used in the model marked with red rings.

Table 1. Some of the water	chemistry	variables	from th	e four	lakes	used i	in the	model.	The lake	upstream
Lake Sandvann is limed.										

	Visterflo	Øvre Sandv.	Djupetj.	Holmetj.
pН	6.98	6.2	5.45	5.74
Ca mg/l	5.41	2.0	1.32	1.48
TOC mgC/l	6.4	8.9	8.7	7.6
Se µg/l	<1	1.1	<1	<1



Figure 2. Stable isotopes of N (${}^{15}N/{}^{14}N = \delta^{15}N$) and C (${}^{13}C/{}^{12}C = \delta^{13}C$) in fish muscle, invertebrates, macrophytes and periphyton in Lake Sandvann. Shaded area represent "the same" level in the food web. After Myreng (2013).



Figure 3. Total Hg(mg/kg ww) in fish muscle from the five fish species in Lake Sandvann. After: Myreng (2013). Red lines indicate trade level (upper) and recommended level for consumption.



Figure 4. Total Hg in muscle of perch from Lake Sandvann (Myreng 2013, left), Lake Holmetjenn and Djupetjenn (pooled samples, Våge 2014, middle), and Lake Visterflo (Våge 2014, right). The 25 cm length recommendation in Norway, and the 0.2 and 0.5 mg THg/kg w.w. levels are marked with red lines.

All four fish species in Lake Sandvann exceeded the 0.2 ppm Hg level at sizes below 10 g (Figure 3), and the largest perch had a muscle Hg concentration of 2.5 mg Hg/kg w.w., (Figure 4 left). Lake Holmetjenn and Lake Djupetjenn exceeded the 0.2 and 0.5 ppm Hg levels in perch at 10 and 12 cm, respectively, (Figure 4 middle). In Lake Visterflo, where there are several non-piscivore prey fish for perch, the 0.2 and 0.5 ppm Hg levels were exceeded around 25 cm (Figure 4, right).

A perch THg model was fitted based on data from these four lakes. A linear mixed effects modelling (LME) approach was used to allow for lake-level processes being modeled as random effects and specific well-documented THg drivers included as fixed effects. The candidate fixed effects were [variable (process)]: Age (bioaccumulation); $\delta^{15}N$ (biomagnification); individual growth rate (biodilution); Se

(modifier); δ^{13} C (habitat use), and sex (life-history differences). A maximum of three-way interactions was allowed among the fixed effect components. Model selection was based on AICc and back-wards selection. The most supported LME model had the following fixed effects structure:

$$\ln(THg) = -2.2 + 0.15age - 0.06\tilde{L} + 0.12\delta^{15}N$$

where \tilde{L} =standardized age-specific length [(X-mean)/SD] – a proxy for growth rate. In addition, lake was included as random effect. The fixed part of the LME model explained 56 % of the THg variation and by adding the random among-lake variance component 88 % was explained. Hence, among-lake differences accounted for 32 % of the THg variation. The fixed model structure showed that THg could most effectively (parsimoniously) be predicted from the positive effect from bioaccumulation (age), the negative effect from individual growth rate and the positive effect from biomagnification (trophic level). Model predictions are illustrated in Figure 5.

There was no indication of a modifying effect from selenium (Se) on muscle THg for the measured Selevels in this study (max Se= 0.84).



Figure 5. Model predictions, as function of age and trophic level ($\delta^{15}N$) from the most supported perch THg mixed effects model under three individual growth rates (slow, medium and fast growth). The contour lines represent THg muscle concentrations (mg Hg/kg). The 0.2 mg/kg and 0.5 mg/kg isoclines are provided in red to ease comparisons among growth rate effects. Predictions are centered according to an average lake effect.

4. Conclusions

- Apart from Lake Visterflo, all lakes had perch < 25 cm exceeding dietary advice levels and trade levels of THg
- Local Lake Owner Unions sell fishing cards to families without any warning about high THg
- A general regional or national dietary advice based on fish species, weight or length, CAN NOT be used in the future there is a **need of local advices!**
- Trends of TOC linked to THg_{fish} must be monitored in selected lakes
- The LME approach used in this study yielded promising and reasonable parameter estimates for predicting THg in perch based on age, size and trophic-level data.
- Since a large proportion of perch THg could be attributed to lake-level factors, future analyses should include more lakes in order to quantify catchment/lake-level processes affecting THg in perch.
- Future fish studies should include Se in water and fish tissue for a more thorough assessment of the modifying potential of this chemical.

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7. Minutes of the 31st Task Force meeting of the ICP Waters programme held in Monte Verità, Switzerland, October 6-8 2015

CONVENTION ON LONG-RANGE TRANSBOUNDARY AIR POLLUTION

Working Group on Effects

International Cooperative Programme on Assessment and Monitoring of the Effects of Air Pollution on Rivers and Lakes

MINUTES

of the 31st meeting of the Programme Task Force held in Monte Verità, Switzerland, October 6-8, 2015

Key messages from the Task Force meeting

Acidification of surface waters is still a concern in acid-sensitive ecosystems Substantial decreases in sulphur deposition have occurred in large parts of world as a result of international cooperation on emission reductions. The reduction of sulphur deposition has led to significant chemical recovery of surface waters, but not yet to a return to pre-industrial conditions. Associated biological recovery proceeds more slowly than chemical recovery. In some regions, the deposition of nitrogen has become more important than sulphur for surface water acidification, and delays chemical and biological recovery. Further reduction of deposition of sulphur and nitrogen are important to further enhance protection of acid-sensitive aquatic ecosystems.

Mercury In many lakes in Sweden, mercury concentrations in fish are above limits that are advised for human consumption, and even further above environmental quality standards proposed by the EU Water Framework Directive to protect ecosystem health. The mercury levels in Swedish fish are in the range that is typical for fish in boreal regions in Europa and North America. International cooperation to reduce heavy metal emissions has been effective in particular for lead and cadmium, but to a lesser extent for mercury. Continued monitoring is needed to document the fate of mercury in the terrestrial and aquatic environment, and more research is needed to understand drivers of temporal trends in mercury in aquatic ecosystems.

Biodiversity and biological recovery A large European survey of aquatic insects, more specifically benthic (bottom-dwelling) macro-invertebrates, in streams and lakes was used to document changes in aquatic biodiversity. The dominating three insect groups (EPT: Ephemeroptera (mayflies), Plecoptera (stoneflies), and Trichoptera (caddisflies)) show an increase in EPT diversity in many sites, correlated with decreases in sulphate concentrations in surface waters. This suggests a strong link between chemical recovery and increased biodiversity of acidified surface waters. Further reductions in atmospheric deposition of sulphur are expected to further promote aquatic biodiversity.

Afforestation may re-acidify surface waters in the Czech Republic Afforestation of acidified catchments in the Czech Republic may lead to a substantial re-acidification of currently recovering surface waters. A modelling study using scenarios of future forest cover and a dynamic model indicates that increased forest growth (associated with higher base cation uptake) and increased dry deposition will lead to future acidification, even under future reductions in sulphur deposition. The results of this study are relevant for other regions where substantial changes in forest cover and forest management are expected, for instance where afforestation is used to mitigate climate change.

Continued browning of surface waters A large water chemistry database for Europe and North America shows that concentrations of dissolved organic carbon (DOC) continue to rise and do not show any indication of levelling out. Changes in atmospheric deposition appear to be the dominant driver of surface water browning, and currently the relative importance of climate drivers are being investigated. More DOC in surface waters makes drinking water treatment more costly and may lower freshwater productivity, while mercury uptake in aquatic food webs is also affected.

Eastern Europe, the Caucasus and Central Asia Aquatic ecosystems in the Russian Federation and in Armenia are strongly impacted by atmospheric deposition. In lakes in the Kola Peninsula in the Russian Federation, the forms and complexes in which metals are present in lakes is affected by anthropogenic deposition. In Armenia, hardly any policy instruments to abate atmospheric pollutions have been implemented. Mining and release of household and industrial waste into rivers are other major sources of surface water pollution in Armenia. The meeting of the International Cooperative Programme on Assessment and Monitoring of the Effects of Air Pollution on Rivers and Lakes (ICP Waters) was attended by 30 experts from the following Parties to the Convention on Long-range Transboundary Air Pollution (CLRTAP): Armenia, the Czech Republic, Finland, Italy, Netherlands, Norway, Russian Federation, Sweden, Switzerland, the United Kingdom of Great Britain and Northern Ireland and the United States of America. In addition a representative for ICP Integrated Monitoring and ICP Modelling and Mapping participated. The list of participants is attached as Annex I.

Introductions

- 2. Mr. Lorenzo Sonognini, Director Monte Verità, Switzerland, welcomed all participants to the ICP Waters meeting in Monte Verità. Mr. Sonognini gave an introduction to the history, geography, economic development of the region and Mr. Sonognini showed a short film about the history of Monte Verità.
- 3. Mr. Claudio Zali, Head of the Department of the Territory, Government councillor of the Canton of Ticino, Switzerland opened the meeting by thanking the community of Monte Verita, and the Swiss environmental federation. Mr Zali recognised the importance of long term monitoring and presented the history of Swiss surface water issues. Mr. Zali described the unique geography of the alpine region and introduced the anthropogenic acidification problems and the swiss Federal Office for the Environment.
- 4. Mr. Gaston Theis, Swiss Federal Office for the Environment, welcomed all participants to the ICP Waters meeting in Monte Verità and thanked the Ms. Sandra Steingruber, the local organizer of the meeting. Mr. Theis acknowledged the importance of the cooperation and learning process between scientists and policy makers, and presented future strategies of the interface between science and policy. He also pointed out that this is a key in future environmental management. He stated that simple tools to communicate scientific results to stakeholders and policy makers are needed. Mr. Theis also stated that the acidification problem is not solved and presented atmospheric nitrogen deposition as an important pollutant in the future, as it has effects on acidification, eutrophication and future climatic problems. He expressed his thanks to ICP Waters for the important work done to support the LTRAP Convention.
- 5. Mr. Gunnar Skotte (Norway), Chair of the Programme Task Force, thanked Ms. Steingruber, the local organizer, and the Swiss Federal Office for the Environment, for hosting the meeting and the introductory speakers for their presentations and warm welcoming words. Then Mr. Skotte welcomed all participants to the 31st Task Force Meeting of ICP Waters in Monte Verità, especially the representatives from ICP Modelling and mapping and ICP Integrated Monitoring.
- 6. The Task Force adopted the agenda of the meeting (Annex II).
- 7. Ms. Steingruber (Switzerland, local host) welcomed all participants to Monte Verità, and gave general information on the meeting and the excursion.
- 8. Ms. Steingruber (Switzerland) then gave a presentation on deposition of acidifying pollutants in Southern Switzerland and their effects on high altitude lake chemistry: 30 years of environmental monitoring. Nitrogen, instead of sulphur, has become the largest driver of surface water acidification in southern Switzerland. This is particularly true in particular in high altitude lakes, because of the high amount of precipitation in this region. Ms. Steingruber presented the monitoring programs and gave examples of trends in wet deposition, showing that sulphate concentrations have declined. Nitrate and ammonia in surface waters had no significant declines in southern Switzerland. Ms. Steingruber demonstrated that the high altitude lakes are extremely acid-sensitive because of their bedrock and low buffering capacity from surrounding soils.
- 9. The presentation from Ms Steingruber raised questions from the audience with regard to biological recovery in the lakes, and presence of high-frequency monitoring. She answered that biological monitoring records started after 2000 and are yet too short to document significant biological recovery. Also, an ICP IM site has started monitoring in one of the high alpine lakes, including some high-frequency measurements.

- 10. Mr. Skotte presented news and emerging issues from the joint session of EMEP Steering Body and WGE. Mr. Per Inge Grennfelt has been re-elected as the Chair of WGE. The Netherlands have announced a severe reduction in budget in the funding to the CCE, which is worrying because of the synthesizing role that CCE has under the WGE. Mr. Skotte gave an overview of the relevant work plan items in the Conventions work plan for 2016-2017. A common data portal for all bodies under the WGE was discussed at the joint EMEP/WGE meeting. Mr. Skotte presented some highlights from the WGE trends report. This report received contributions from all ICPs, EMEP and AMAP and will be finalized in November 2015. Mr. Skotte gave an overview of the LRTAP assessment report, which is planned to be finalized at end of February 2016. He gave and overview of some results from the different ICPs under the WGE, with relevance for the work under ICP Waters. (I) A new grid and finer resolution in EMEP deposition might affect the critical loads calculations of exceedances, highlighting a need for a better regional assessment of acidified lakes. (II) Health effects give higher momentum on air pollution policy and it is a challenge for us to increase visibility of ecosystem effects of air pollution. (III) Increased focus on biodiversity. He highlighted the value of the ICP waters data base on invertebrates, which could be relevant for further development of critical loads for acidification of surface waters.
- 11. The presentation by Mr Skotte raised a discussion on the role of science for the choice of which organisms to protect, for instance fish, or more general, biodiversity. There was a discussion on the interaction between science and policy, where it was suggested that the role of policy was to decide what organisms to protect, whereas science would supply relevant data. Other viewpoints were that monitoring networks would need to focus on species that are important to society.
- 12. Mr. Jussi Vuorenmaa (Finland) reported from the ICP Integrated Monitoring Programme (ICP IM). He described the purpose, extent and priorities of the programme. He then presented ongoing work and highlights from reports and scientific papers, and planned cooperation with other ICPs.
- 13. Mr. Max Posch (CCE) reported from the ICP Mapping and Modelling Programme. He described the purpose and gave information on the updated Critical Loads maps and the different Critical Loads calculations. He then presented on-going work and highlights and planned cooperation with other ICPs.

Reports from the ICP Waters Programme activities 2014/2015

- 14. Ms. Heleen de Wit (Norway) from the Programme Centre reported on the status of the ICP Waters programme and on common work under the WGE for effect-based approach. She emphasised that ICP Waters should produce policy-relevant results. She further presented the ICP Waters programme aims, main area of activities and recent reports. The state of the water chemistry and biology data base and number of participants at the TF meeting was presented (Annex III). Current work includes chemical trend assessment by Mr. Garmo. Biological trend analysis by Mr. Velle. Ms de Wit is responsible for the coordination of the WGE trend assessment report. She mentioned the ICP Waters contribution to the LRTAP Assessment report. Ms. de Wit presented discussion points for Thursday, including the joint meeting with ICP IM, and the work plan for 2016-2017.
- 15. Ms. Heleen de Wit (Norway) reported on representation of ICP Waters in other bodies/meetings under the Convention. ICP Waters was represented at the following meetings:
 - Oct 14: JEG-meeting, Sitges, Spain Ms. de Wit
 - Jan 15: Assessment Report Workshop Mr. Wright, Ms. de Wit
 - March 15: WGE, Extended Bureau, Geneva Mr. Skotte, Ms. de Wit
 - April 15: TF Modelling and Mapping, CCE WS, Zagreb Ms. de Wit
 - April 15: TF Materials, Oslo Mr. Skotte
 - May 15: ICP IM, Minsk Ms. de Wit
 - Sept 15: Joint session of EMEP Steering Body & WGE, Geneva Mr. Skotte, Ms. de Wit

- 16. Ms. Heleen de Wit (Norway) presented new reports and papers and from ICP Waters. These are:
 - ICP Waters report 119/2015 Trend report Chemical and biological recovery in acidsensitive waters: trends and prognosis. Garmo Ø.A., de Wit, H.A. and Fjellheim A.
 - ICP Waters report 121/2015 Biological intercomparison 1814. Fjellheim, Arne; Johannessen, Arne; Landås, Torunn Svanevik
 - ICP Waters report 122/2015 Proceedings Task Force meeting 2014, Proceedings of the 30th Task Force meeting of the ICP Waters Programme in Grimstad, Norway 14th –16th October, 2014. Heleen de Wit and Bente Wathne
 - ICP Waters report 123/2015 Chemical intercomparison 1529
 - Intercomparison 1529: pH, Conductivity, Alkalinity, NO3-N, Cl, SO4, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. Carlos Escudero
 - Master thesis, Gea van der Lee, 2015, "Invertebrate community change in acidified rivers and lakes in Southern Norway", Master thesis Utrecht University. Supervision K.T. Rebell (Utrecht University), H.A. de Wit, G. Velle, A. Fjellheim (ICP Waters).
- 17. Ms. Heleen de Wit (Norway) outlined current work. This can be summarised as follows:
 - The trend assessment of biodiversity and climate
 - The WGE trend assessment common report from all ICPs
 - The CLTRAP EMEP/WGE Assessment Report

Presentation of ICP Waters reports in progress

- 18. Mr. Velle (Programme sub-centre) presented preliminary results for the 2015 ICP waters report. The aim of the analysis was to tease out effects climate change on biodiversity in acidified lakes, i.e. the effects of increased temperature and precipitation on biodiversity. Mr. Velle stressed that there are multiple stressors that affect biodiversity. Mr. Velle presented the climate data. There were significant changes in temperature and in precipitation for different sites. Benthic invertebrate data was available for the period 1981 to 2014. A re-analysis of the 2013 report was made with improvements and the methodology improved with including EPT taxa. The results with the new EPT taxa showed increases in diversity in lake and river systems with decreases in sulphur as the most important driver for diversity changes. The new analysis included testing multiple stressors including climate and water chemical parameters, and characterization of the biological community was characterized with several indices, such as a 'stability index'.
- 19. The presentation inspired a long discussion on methodology and interpretation of the preliminary results. Communities in river and lakes appeared to respond differently to drivers of change, especially sulphur and precipitation. The large spatial and temporal variation in the database was commented upon, which may be relevant to understand differences between rivers and lakes. Also, differences in dispersal and ecological inertia were suggested as possible causes for the differences between lakes and rivers. Stability indices in relation to stability or variation in precipitations. It was suggested to classify sites according to water chemistry, rather than according to country.
- 20. Mr. Velle continued to present more results using statistical Bayesian modelling, on lag time responses to changes in chemistry and climate. He concluded that maximum diversity has not been reached in the monitoring sites in the database, expecting a further increase in biodiversity given the chances in water chemistry. However, climate warming may slow down these increases.
- 21. Mr. Velle also presented a recent MSc thesis based on a subset of the data in the ICP Waters database, which had similar focus as the ICP Waters biodiversity and climate report. Here, changes in biodiversity were mainly linked to water chemistry, while temperature affected variation rather than trends in biodiversity.
- 22. The Task Force expressed its satisfaction with the progress made by Mr. Velle. Three reviewers for the report volunteered, i.e. Ms. Heleen de Wit (Programme centre), Mr. Jens Fölster (Sweden) and Mr. Don Monteith (UK).

23. Mr Garmo (Programme centre) presented the 2014 ICP Waters report: Chemical and biological recovery in acid-sensitive waters: trends and prognosis of water chemistry for 2020. The report was published in 2015 (119/2015). The report has two main chapters, i.e. on trends and modelling of water chemistry and on trends in biological recovery. The main conclusion of the water chemistry trends was that sulphur concentrations have declined, pH is increasing, alkalinity and ANC patterns are showing general increase but in some regions no change was identified. Biological recovery from acidification was found where long time series were available, but not everywhere. The modelled improvements in water chemistry in 2020 were marginal compared with measured water chemistry in 2012, despite a the reduction in deposition.

Water chemistry

- 24. Ms. Michela Rogora (Italy) presented recent trends in major ions in sub-alpine lakes and rivers in Northern Italy. Sodium and chloride concentrations have increased substantially for the large deep sub-alpine lakes. Also, chloride during winter has increased in rivers that drain into lake Maggiore. Road salts appear to be the driver of the identified increases of chloride. Ms. Rogora invited the participants of the Task Force meeting to the SIL conference in Torino in 2016.
- 25. Ms. Marine Nalbandyan (Armenia) reported on Air Pollution Impacts on Debed River Waters in Different Parts of the watershed. Ms. Nalbandyan provided a rich history of the region and the industrial development and its effects on the water quality. The anthropogenic pressures are based on waste water and industrial pollution. Remediation measures have shown to be effective in the region. Ms. Nalbandyan discussed the role of economic wealth and environmental objectives.
- 26. Mr. Jens Fölster (Sweden) reported on high quality long-term monitoring: Swedish experiences with assessments of surface water acidification and recovery. Mr. Fölster gave a detailed description of the history of Swedish environmental monitoring programs and the link between science and policy. Mr. Fölster gave examples of how to maintain a successful monitoring program and one key points was the cooperation between scientist and society. He also gave examples on how monitoring can be adapted to different environmental stressors and emerging pollutants. Mr Fölster stated that open access of data is an important aspect to keep as the data goes beyond the university and becomes socially relevant.

Water chemistry - Dissolved organic carbon

- 27. The presentation of Mr. John Gunn (Canada) on Input and the Recovery of Carbon-Starved Ecosystems was given by Mr. Rosseland. The Sudbury area received extremely high deposition of sulphur up to 1960s, resulting in large damage to forest and aquatic biota. Emissions from Sudbury were reduced greatly as a result of pollution control policy, and after this forest health improved as demonstrated by satellite images showing greening. Also, there was strong chemical and biological recovery in lakes. He showed relationships between catchment greenness (NDVI) and particulate organic carbon (POC) in streams. In addition, links were shown between forest cover and DOC, and between DOC and lake productivity. It was suggested that a non-linear relationship between DOC and fish production can be found.
- 28. Ms. Heleen de Wit (Progamme Center) gave a presentation the on-going work with the DOC trend analysis. Ms. de Wit presented the current status of the trend report and gave a description of the different data sources. The results shows that most lakes still have increasing DOC concentrations, and that these still correlate strongly with sulphate concentrations. Climatic factors were also tested with the DOC trends and are becoming increasingly important. Atmospheric chemistry is the strongest driver of changes in DOC. The data analysis is under progress and more data will be added to the database and further investigations will be done.

Biological responses to air pollution

29. Mr. Andreas Bruder (Switzerland) presented a study on Lago Nero - a high alpine system to study the consequences of environmental change for lake ecosystems. He gave a rich history of the Lago Nero catchment and on the monitoring program of the area. The deposition pattern shows high inter-annual variability and the relation to biota should be examined in more detail for

remote systems. Mr. Bruder stressed the importance of high frequency sampling. It is important to include these remote lakes in larger programs as they provide important threshold for acid deposition systems susceptible to changes in high altitude areas. Mr. Vuorenmaa (ICP IM) welcomed Mr. Bruder and the alpine lakes to be part of ICP IM.

30. Mr. Salar Valinia (Programme Centre) presented a comparison between two independent estimates of reference conditions with the use of biological indicators species and geochemical modelling. The two independent estimates of acidification gave the same results in 78 of 85 cases. The results showed that biological recovery is lagging behind chemical recovery, particularly for fish. Mr. Bruder asked how biological integrity for several species, not just roach, would relate to the conceptual model. Mr Valinia stated that selection of the most acid sensitive species in the catchment is meaningful for acidification responses, but that the conceptual model will work with multiple species and different thresholds.

Heavy metals and POPs

- 31. Mr. Bjørn Olav Rosseland (Programme Centre) gave a presentation on mercury in fish in a TOCchanging world. A model was presented to improve the general dietary regulations and focus on individual lakes versus large-scale catchment advices. Mr Rosseland showed the relationship between TOC and different forms of mercury. He stated that the exceedance of mercury thresholds, set to protect human health, depends on the number of fish species in lakes, because this affects the length of the food web. Mr. Rosseland concluded that fish in most lakes in his study exceeded the critical limit for mercury. National advisories are not specific enough for local conditions. Mr Åkerblom asked what drivers besides TOC can affect mercury trends and what relationship there was to selenium. Ms. Skjelkvåle asked if the selenium was available in the monitoring program and if it was concentrations in the biota or water column. Mr Vuorenmaa gave an example of perch studies in Finland and connected high mercury content with recovering lakes.
- 32. Ms. Dinu (Russian Federation) gave a presentation on metals forms distribution in the surface waters of the Kola North under anthropogenic deposition. Ms. Dinu presented results on different forms of metals in lakes around Kola research centre. The results showed different experimental and modelling exercises. In areas far from point source of pollution Al and Fe form complexes with organic matter when pH is below 7. Ms. Dinu concluded that metal formation was dependent on what anthropogenic driver is available.
- 33. Mr. Åkerblom (Sweden) gave a presentation on the trends in mercury in the environment, including soils and fish. There is an accumulation of mercury in forest soils, because circa 90% of deposited Hg is retained in soils. Samples of mercury in fish were available from the beginning of 1965. However, the data sources were various monitoring programmes with different aims and designs, focusing on different fish species and regions. The data were standardized with regard to fish species using a transfer function, but differences in representation of regions was not accounted for. The general pattern shows a decline for mercury in fish in Swedish lakes. The causes of this decline are not understood, but it is likely that some explaining factors are removal of point sources and decreased mercury deposition. However, some of the demonstrated temporal patterns may in fact reflect underlying regional patterns, changes in sampled fish species and/or fish size, and lack of even distribution of sampled lakes. This requires further study, also with regard to investigating the role of climate change. Mr Åkerblom concluded that extending the database to wider regional representation in Europe would a valuable way to move forward.
- 34. Ms. de Wit (Programme centre) gave a presentation of 2016 ICP Waters mercury report and the new project on mercury in Norway. Ms. de Wit mentioned that the ICP Waters report will be done in cooperation with the Norwegian mercury project, which is in fact collaboration between Norway, Sweden and Canada. Ms. de Wit opened the possibility for including more data than from Norway, Sweden and Canada. Mr. Monteith answered that there are some data from the UK. Mr. Posch said that ICP forests are performing a moss vegetation survey so data might be available. Mr. Åkerblom expressed the wish that the ICP Waters report should be more than a Nordic report. Mr. Vuorenmaa mentioned that Mr. Rask (an expert on mercury in fish in Finland) may be able to supply data. Mr. Rosseland said that relevant data could be obtained from master

students, and he was willing to assist to help obtain these data. Mr. Stoddard suggested that it may be interesting to study links between DOC and mercury. Ms de Wit suggested that Mr. Bruder (Switzerland) could send fish samples from the new Swiss ICP IM site to NIVA for analysis, to investigate if mercury might be an environmental problem here. Mr Skotte enquired if the Task Force meeting was satisfied with the suggested focus on mercury, and the Task Force expressed its support for the choice for mercury as a thematic report for 2016.

Critical loads

35. Mr Hruska (Czech Republic) gave a presentation on changes in stream water chemistry under future scenarios of acid deposition and forestry practices. Mr. Hruska showed how the catchment has recovered from 1980 as it was deforested and highly affected by acid deposition. Today, the catchment receives about 10 kg/ha sulphur deposition. Mr. Hruska argued that there might be a decline in stream water pH due to forestry effects in the future scenarios. Mr. Hruska concluded the catchment might be re-acidified in the future, i.e. increased forest uptake and less base cations transported to surface waters. The presentation inspired a discussion on relationships between scenarios of deposition, forestry and stream water chemistry, an issue which is becoming increasingly relevant as there is a tendency to more intense forest management to mitigate climate change.

Intercalibration/intercomparison

- 36. Mr. Garmo (Programme centre) presented the results from the chemical intercomparison. 39 laboratories from 19 countries participated, which is higher than last year but lower than previously. The reduction in number of participating laboratories compared with previously is related to the introduction of a participation fee for laboratories which do not contribute with analyses for environmental monitoring programmes, and to a more even geographic distribution of participating laboratories. The chemical comparison included all the usual variables were included, aluminium and TOC. On average, 76% of results were within the accepted error margin and laboratory performance was slightly better than last year. Performance for trace metals and major ions was high, in contrast to pH and TOC. For pH, there is a systematic error in the analysis and it was noted that the recommendation of the Programme Centre to report pH method (stirring or no stirring) was not followed by all participating laboratories. It was noted that the sample had a pH of circa 7, somewhat high when considering that the focus of ICP Waters is acidified waters. For TOC, many systematic errors were found in the analysis, which might be related to methodological differences and possibly the somewhat low TOC concentration of the sample. Mr. Garmo enquired if the concentration for nitrate was within the distribution of the lakes assessed within ICP Waters. Mr Garmo asked if the Task Force has the wish to include more parameters. Mr. Hruska informed that the laboratory at his institute was content with higher concentrations, as these are more representative of the water chemistry in the monitored surface waters in the Czech Republic. Ms. de Wit questioned the spiking as the concentrations are much higher than those identified in Norway. Mr. Rosseland asked for labile inorganic aluminium to be added. Ms. de Wit asked the Task Force for reacting with regard to concentration levels and pH. Mr. Fölster suggested that pH and alkalinity should be lower. Mr. Fölster asked if it was possible to send out two samples with contrasting water chemistry instead of one. Ms de Wit replied that this good suggestion, but unfortunately not possible within the budget, as using two samples will increase the work load significantly. However, contrasting samples in terms of concentration levels, pH and TOC between years allow for potentially interesting comparisons of performance between years. It was concluded that the chemical comparison showed a satisfying performance for the majority of participating laboratories, and that the Programme Centre will aim for contrasting concentrations in the water sample between years, but that pH should preferably reflect acidified waters, thus lay between 5 and 6, possibly up to 6.5.
- 37. Mr. Arne Fjellheim (Programme subcentre) presented the results from the 19th biological intercomparison. Test samples normally contain about 50 different species of macroinvertebrates from different countries. Six laboratories participated in the biological intercalibration 2015, which is an increase from the previous year. The laboratories identified a high portion of the individuals in the test samples. The quality of the identification of the different groups was above the level of

acceptance for all laboratories. The taxonomic quality was sufficient for stating the acidity index. The average quality assurance index ranged between 91.9 and 98.5, well above the accepted criteria of 80 % - indicating good taxonomic work. Ms. de Wit thanked Mr. Fjellheim, who will retire in 2015, for his successful work on intercomparison and his contributions to the ICP Waters programme.

38. Mr. Wright (Programme centre) presented the draft CLRTAP Assessment Report 2015, which will be finalized in 2016. The report is available on the CLRTAP webpage (www.unece.org/fileadmin/DAM/env/documents/2015/AIR/EMEP/CLRTAP_Assessment_R eport_2016_draft_3_sept_2015.pdf). Mr. Wright gave examples from different parts of the CLRTAP assessment report. One important message from the report is that acidification remains an issue in surface waters, and is not yet solved. However, acidification of surface waters is also used as a success story to demonstrate what emission policy can achieve for improvement of the environment. It was suggested to include some text on mercury in biota in the report. Ms de Wit will coordinate the ICP Waters response to the CLRTAP Assessment Report 2015. The Swiss delegates will send their comments to ICP Waters.

Current issues and workplan

- 39. Ms. de Wit (Programme Centre) presented the WGE trend report. The work with the report started in October 2014 and will be finished in November 2015, and will be an input to the CLTRAP assessment report. The focus was on presenting trends and the presented results are based on existing work. All ICPs contributed to the report and Ms. de Wit was responsible for the coordination of the report. Ms. de Wit underlined that the report was written in excellent cooperation between all groups under the Working Group on Effects and also with contributions from outside (EMEP and AMAP). Mr. Skotte thanked Ms. de Wit for her hard work on the report. The content and key messages of the report were discussed. Suggestions for a way to reach out to a wider audience included use of symbols from traffic (speedometer, traffic lights) to convey summarized results, and a youtube video. It was suggested to print the report on paper rather than delivering the report only in digital format.
- 40. Ms. de Wit presented the ICP Waters work plan for 2016 and the recommendations from the ICP Review done under the WGE, specific for the ICP program. She underlined that ICP Waters is among the best ICPs with regard to the follow-up of these recommendations. Mr. Posch stated that nitrogen is also an acidifying compound and should be highlighted in the acidification assessment. Suggestions were made on trend assessments on Hg and possibilities for cooperation with ICP IM and ICP vegetation. Ms. de Wit mentioned that the next ICP Waters meeting will be organized together with ICP IM, where possibilities for more joint work on trend assessments will be discussed. It was also pointed out by the Task Force meeting that acidification and recovery remains an important issue. The new report on mercury is in line with recommendations from the ICP review given to ICP waters.
- 41. Ms. de Wit showed a short review of the different ICP's webpages, presented earlier at the joint EMEP-WGE meeting in September, which pointed to possible improvements of the ICP waters webpage. An example would be to highlight better links to other ICPs, and to national data portals. The Programme Centre will improve the ICP Waters homepage keeping these recommendations in mind.
- 42. Ms. de Wit proposed two suggestions for future ICP Waters thematic reports, i.e. a regional acidification assessment of lakes and rivers and a report focusing on dose-response relationships with regard to aquatic biodiversity. A regional assessment will be highly policy relevant, as one of its aims would be to quantify how many lakes remain under threat of acidification. Mr. Stoddard pointed towards the need for an extension of the ICP Waters database, as the current database is not sufficient for a regional assessment. Mr. Posch (CCE) stated that the number of lakes under threat of acidification is likely to be a very useful complement to critical loads maps. Mr. Stoddard wondered if it was possible to include rivers in the assessment, which was acknowledged by the Task Force as an important suggestion, because rivers also have strong relevance to policy makers. The Task Force agreed that focal centres may have an important role in such a regional assessment, and was positive to the suggested theme.

- 43. The second suggestion for a thematic report was to develop aquatic biodiversity dose-response relationships. A similar approach has been developed under the Task Force of Modelling and Mapping for a few years already with regard to terrestrial biodiversity, using the so-called habitat suitability index. Mr. Velle mentioned that there are challenges with regard to quality assurance of the ICP Waters database for such an effort. Mr. Posch gave a presentation on the work on biodiversity critical loads and the habitat suitability index from ICP Modelling and Mapping, and explained how biodiversity may be linked to exceedance of critical loads. Ms. de Wit thanked Mr Posch for his background presentation, and suggested that this approach may be useful for developing critical loads to protect surface waters from losses of biodiversity. Ms. Skjelvåle suggested that monitoring and work on critical limits under the EU-Water Framework Directive may be relevant to this topic. Mr. Monteith suggested that the current work and database on invertebrate biodiversity done by Mr Velle is highly relevant for this topic. Mr. Stoddard agreed and suggested that relationships between macro-invertebrates and environmental variables may be developed, also based on work done outside of ICP Waters. Mr Velle saw several challenges for the development of a habitat suitability index for macroinvertebrates. It was concluded that the topic is highly relevant for work under the Convention. Ms. de Wit proposed that the Programme Centre will consider asking an expert to present challenges and possibilities related to this topic, to be discussed at a future Task Force meeting.
- 44. Ms. de Wit concluded from the previous discussion that it appeared that the Task Force members' preference was the thematic report on the regional assessment of surface water acidification. The Task Force approved of this conclusion.

Other business

- 45. Ms. de Wit (Programme centre) announced that the TF meeting 2016 will be held in or near to Oslo, Norway, jointly with the Task Force meeting of ICP IM, May 24-26.
- 46. Ms. de Wit reminded the Task Force meeting that from now on, ICP Waters Task Force meetings will be organized in the spring instead of the fall. Such timing fits better with the schedule for the other meetings under the Convention.
- 47. Mr. Gunnar Skotte (Norway) thanked all participants for a constructive meeting and thanked the host Ms. Steingruber and the Swiss Federal Offices for an excellent hosting of the meeting, including an interesting excursion and very pleasant Task Force dinner. He thanked the Programme Centre for the organization of the meeting.
- 48. The Task Force expressed its appreciation to the Programme Centre for its scientific and coordinating work and acknowledged its important contribution to the programme's successful implementation. Finally, the Task Force recognizes that the contributions of the National Focal Centres and cooperating institutes are essential for the high quality output from the programme.

Adoption of the minutes

49. The decisions in the ICP Waters meeting as written in the minutes were adopted by the Task Force.

8. Annex I: Participants at the ICP Waters 31st Task Force meeting

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9. Annex II: Agenda for the 31st Task Force ICP Waters October 6-8, 2015

Introductions

- Meeting welcome, Lorenzo Sonognini, director Monte Verità, Switzerland
- Meeting welcome: Claudio Zali, Head of the Department of the Territory, Government councillor of the Canton of Ticino, Switzerland
- Opening words, Gaston Theis, Swiss Federal Office for the Environment, Switzerland
- Adoption of the agenda, Gunnar Skotte, ICP Waters Chairperson
- General information about the meeting and excursion, Sandra Steingruber, local host
- Deposition of acidifying pollutants in Southern Switzerland and its effects on high altitude lake chemistry: 30 years of environmental monitoring, *Sandra Steingruber, Switzerland*
- Report from the Working Group on Effects, Gunnar Skotte, ICP Waters Chairperson
- Report from ICP Integrated Monitoring, Jussi Vuorenmaa, ICP IM
- Report from ICP Modelling and Mapping, Max Posch, CCE
- Reports from other ICPs TBA

Reports from the ICP Waters Programme activities 2014/2015

• Status of the ICP Waters programme, and report on common work for effect-oriented programmes under the Working Group on Effects, *Heleen de Wit, Programme centre*

Presentation of ICP Waters reports

- The 2015 ICP Waters report: An attempt to tweeze out the effect of climate change as driver of biodiversity in acidified waters, *Gaute Velle, Programme subcentre*
- The 2014 ICP Waters report: Chemical and biological recovery in acid-sensitive waters: trends and prognosis, *Øyvind Garmo, Programme centre*

Water chemistry

- Recent trends in major ions in subalpine lakes and rivers in Northern Italy, Michela Rogora, Italy
- Air Pollution Impacts on Debed River Waters in Different Parts of the Watershed, *Marine Nalbandyan, Armenia*
- High quality long-term monitoring: Swedish experiences with assessments of surface water acidification and recovery, *Jens Fölster, Sweden*

Water chemistry - dissolved organic carbon

- Terrestrial DOC Input and the Recovery of Carbon-Starved Ecosystems, John Gunn, Canada
- DOC trend analysis, Heleen de Wit, Programme Centre

Biological responses to air pollution

- Lago Nero a high alpine system to study the consequences of environmental change for lake ecosystems, *Andreas Bruder, Switzerland*
- Comparing biological indicators and geochemical modelling from the beginning of the 20th century, *Salar Valinia, Programme centre*

Heavy metals and POPs

- Mercury in fish in a TOC-changing world: A model to improve the general dietary regulations and focus on individual lakes versus large-scale catchment advices, *Bjørn Olav Rosseland, Programme centre*
- Metals forms distribution in the surface waters of the Kola North under anthropogenic loads, *Marina Dinu*, Russian Federation
- Trends in mercury in fish, Staffan Åkerblom, Sweden
- The 2016 Mercury report, Heleen de Wit, Programme centre

Critical loads/Dynamic modelling

• Changes in stremwater chemistry under future scenarios of acid deposition and forestry practices (Uhlířská chatchment), *Jakub Hruška, Czech Republic*

Intercalibration/intercomparison

- Chemical intercomparison, Øyvind Garmo, Programme centre
- Biological intercalibration, Arne Fjellheim, Programme subcentre

Common work for all ICPs

- The WGE trend report, Heleen de Wit, Programme centre
- The Assessment Report, Dick Wright, Programme centre

Current issues and workplan

- Current issues that have emerged from WGE: joint meetings, data portal, open data access, *Heleen de Wit, Programme centre*
- Draft 2015-2016 Workplan, Heleen de Wit, Programme centre

Other Business

• TF meeting 2016

Adoption of the minutes

	Chemical	Biological	Participation	Participating in	Participating in
	data	data	in TF	chemical	biological
			2013-2015	2013-2015	2013-2015
A	2012		-		
Armenia	2013		•		
Austria	2015			•	
Belarus	2015				
Canada	2015R		•		
Croatia					
Czech Rep.	2015	2011	•	•	
Estonia	2015		•	•	•
Finland	2015		•	•	
France				•	
Germany	2015	2014		•	•
Ireland	2014			•	
Italy	2015		•	•	
Latvia	2015	2014	•	•	•
Moldova	2015?				
Montenegro	2015R				
Netherlands				•	
Norway	2015	2014	•	•	•
Poland	2013		•	•	
Russia			•	•	
Spain	2014			•	
Sweden	2015	2013	•	•	•
Switzerland	2015	2014	•	•	•
UK	2015R	2013	•	•	•
USA	2015R		•		
Total	20	7	14	17	7
Belgium				•	
Iceland					
Lithuania					
Portugal					
Romania					
Serbia			1	•	
Slovenia			1		
Total			1	19	

10. Annex III: Status participation in the ICP Waters programme as of October 2015

11. Reports and publications from the ICP Waters programme

All reports from the ICP Waters programme from 2000 up to present are listed below. Reports before year 2000 can be listed on request. All reports are available from the Programme Centre. Reports and recent publications are also accessible through the ICP Waters website; http://www.icp-waters.no/

De Wit, H., Valinia, S. and Steingruber, S. Proceedings of the 31st Task Force meeting of the ICP Waters Programme in Monte Verità, Switzerland 6th –8th October, 2015. ICP Waters report 126/2015

De Wit, H., Hettelingh, J.P. and Harmens, H. 2015. Trends in ecosystem and health responses to long-range transported atmospheric pollutants. **ICP Waters report 125/2015**

- Fjellheim, A., Johannessen, A. and Landås, T.S. 2015. Biological intercalibration: Invertebrates 1915. ICP Waters report 124/2015
- Escudero-Oñate, C. 2015 Intercomparison 1529: pH, Conductivity, Alkalinity, NO3-N, Cl, SO4, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. **ICP Waters report 123/2015**
- de Wit, H., Wathne, B. M. (eds) 2015. Proceedings of the 30th Task Force meeting of the ICP Waters Programme in Grimstad, Norway 14th-16th October, 2014. **ICP Waters report 122/2015**
- Fjellheim, A., Johannessen, A. and Landås, T.S. 2014. Biological intercalibration: Invertebrates 1814. ICP Waters Report 121/2014
- Escuedero-Oñate. 2014. Intercom¬parison 1428: pH, Conductivity, Alkalinity, NO3-N, Cl, SO4, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. ICP Waters Report 120/2014
- De Wit, H. A., Garmo Ø. A. and Fjellheim A. Chemical and biological recovery in acid-sensitive waters: trends and prognosis. ICP Waters Report 119/2014
- Fjellheim, A., Johannessen, A. and Landås, T.S. 2013. Biological intercalibration: Invertebrates 1713. ICP Waters Report 118/2014
- de Wit, H., Bente M. Wathne, B. M. and Hruśka, J. (eds) 2014. Proceedings of the 29th Task Force meeting of the ICP Waters Programme in Český Krumlov, Czech Republic 1st –3rd October, 2013. **ICP Waters report 117/2014**
- Escuedero-Oñate, C. Intercomparison 1327: pH, Conductivity, Alkalinity, NO3-N, Cl, SO4, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni and Zn. **ICP Waters Report 116/2013**
- Holen, S., R.F. Wright, I. Seifert. 2013. Effects of long-range transported air pollution (LTRAP) on freshwater ecosystem services. **ICP Waters Report 115/2013**
- Velle, G., Telford, R.J., Curtis, C., Eriksson, L., Fjellheim, A., Frolova, M., Fölster J., Grudule N., Halvorsen G.A., Hildrew A., Hoffmann A., Indriksone I., Kamasová L., Kopáček J., Orton S., Krám P., Monteith D.T., Senoo T., Shilland E.M., Stuchlík E., Wiklund M.L., de Wit, H., Skjelkvaale B.L. 2013. Biodiversity in freshwaters. Temporal trends and response to water chemistry. ICP Waters Report 114/2013
- Fjellheim, A., Johannessen, A. and Landås, T.S. 2013. Biological intercalibration: Invertebrates 1612. ICP Waters Report 113/2013
- Skjelkvåle, B.L., Wathne, B.M., de Wit, H. and Michela Rogora (eds.) 2013. Proceedings of the 28th Task Force meeting of the ICP Waters Programme in Verbania Pallanza, Italy, October 8 10, 2012. **ICP Waters Report 112/2013**
- Dahl, I. 2012. Intercomparison 1226: pH, Conductivity, Alkalinity, NO3-N, Cl, SO4, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni and Zn. **ICP Waters report 111/2012**
- Skjelkvåle, B.L., Wathne B. M. and Moiseenko, T. (eds.) 2010. Proceedings of the 27th meeting of the ICP Waters Programme Task Force in Sochi, Russia, October 19 21, 2011. **ICP Waters report 110/2012**
- Fjellheim, A., Johannessen, A., Svanevik Landås, T. 2011. Biological intercalibration: Invertebrates 1511. NIVA-report SNO 6264-2011, **ICP Waters report 109/2011**.
- Wright, R.F., Helliwell, R., Hruska, J., Larssen, T., Rogora, M., Rzychoń, D., Skjelkvåle, B.L. and Worsztynowicz, A. 2011. Impacts of Air Pollution on Freshwater Acidification under Future Emission Reduction Scenarios; ICP Waters contribution to WGE report. NIVA-report SNO 6243-2011. ICP Waters report 108/2011.
- Dahl, I and Hagebø, E. 2011. Intercomparison 1125: pH, Cond, HCO3, NO3-N, Cl, SO4, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA-report SNO 6222-2011. ICP Waters report 107/2011.
- Skjelkvåle B.L. and de Wit, H. (Eds). 2011. Trends in precipitation chemistry, surface water chemistry and aquatic biota in acidified areas in Europe and North America from 1990 to 2008. NIVA-report SNO 6218-2011 **ICP Waters report** 106/2011.
- ICP Waters Programme Centre 2010. ICP Waters Programme manual. NIVA SNO 6074-2010. ICP Waters report 105/2010. 91 s. ISBN 978-82-577-5953-7,

- Skjelkvåle, B.L., Wathne B. M. and Vuorenmaa J. (eds.) 2010. Proceedings of the 26th meeting of the ICP Waters Programme Task Force in Helsinki, Finland, October 4 6, 2010. **ICP Waters report 104/2010**
- Fjellheim, A. 2010. Biological intercalibration: Invertebrates 1410. NIVA-report SNO 6087-2010, ICP Waters report 103/2010.
- Hovind, H. 2010. Intercomparison 1024: pH, Cond, HCO₃, NO₃-N, Cl, SO₄, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA-report SNO 6029-2010. **ICP Waters report 102/2010**.
- De Wit, H. A. and Lindholm M., 2010. Nutrient enrichment effects of atmospheric N deposition on biology in oligotrophic surface waters a review. NIVA-report SNO 6007 2010. ICP Waters report 101/2010
- Skjelkvåle, B.L., De Wit, H and and Jeffries, D. (eds.) 2010. Proceedings of presentations of national activities to the 25th meeting of the ICP Waters Programme Task Force in Burlington, Canada, October 19-21 2009. NIVA-report SNO 5995 - 2010. ICP Waters report 100/2010.
- Fjellheim, A. 2009. Biological intercalibration: Invertebrates 1309. NIVA-report SNO 5883-2009, ICP Waters report 99/2009.
- Hovind, H. 2009. Intercomparison 0923: pH, Cond, HCO₃, NO₃-N, Cl, SO₄, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA-report SNO 5845-2009. **ICP Waters report 98/2009**.
- Ranneklev, S.B., De Wit, H., Jenssen, M. T. S. and Skjelkvåle, B.L., 2009. An assessment of Hg in the freshwater aquatic environment related to long-range transported air pollution in Europe and North America. NIVA-report SNO 5844-2009. ICP Waters report 97/2009.
- Skjelkvåle, B.L., Jenssen, M. T. S. and De Wit, H (eds.) 2009. Proceedings of the 24th meeting of the ICP Waters Programme Task Force in Budapest, Hungary, October 6 – 8, 2008. NIVA-report SNO 5770-2009. **ICP Waters report 96/2008.**
- Fjellheim, A and Raddum, G.G. 2008. Biological intercalibration: Invertebrates 1208. NIVA-report SNO 5706-2008, ICP Waters report 95/2008
- Skjelkvåle, B.L., and De Wit, H. (eds.) 2008. ICP Waters 20 year with monitoring effects of long-range transboundary air pollution on surface waters in Europe and North-America. NIVA-report SNO 5684-2008. ICP Waters report 94/2008.
- Hovind, H. 2008. Intercomparison 0822: pH, Cond, HCO3, NO3-N, Cl, SO4, Ca, Mg, Na, K, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA-report SNO 5660-2008. **ICP Waters report 93/2008**.
- De Wit, H. Jenssen, M. T. S. and Skjelkvåle, B.L. (eds.) 2008. Proceedings of the 23rd meeting of the ICP Waters Programme Task Force in Nancy, France, October 8 – 10, 2007. NIVA-report SNO 5567-2008. **ICP Waters report 92/2008**.
- Fjellheim, A and Raddum, G.G. 2008. Biological intercalibration: Invertebrates 1107. NIVA-report SNO 5551 2008, ICP Waters report 91/2008
- Hovind, H. 2007. Intercomparison 0721: pH, Cond, HCO3, NO3-N, Cl, SO4, Ca, Mg, Na, K, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA-report SNO 5486-2007. ICP Waters report 90/2007
- Wright, R.F., Posch, M., Cosby, B. J., Forsius, M., and Skjelkvåle, B. L. 2007. Review of the Gothenburg Protocol: Chemical and biological responses in surface waters and soils. NIVA-report SNO 5475-2007. ICP Waters report 89/2007
- Skjelkvåle, B.L., Forsius, M., Wright, R.F., de Wit, H., Raddum, G.G., and Sjøeng, A.S.M. 2006. Joint Workshop on Confounding Factors in Recovery from Acid Deposition in Surface Waters, 9-10 October 2006, Bergen, Norway; Summary and Abstracts. NIVA-report SNO 5310-2006. ICP Waters report 88/2006.
- De Wit, H. and Skjelkvåle, B.L. (eds). 2007. Trends in surface water chemistry and biota; The importance of confounding factors. NIVA-report SNO 5385-2007. **ICP Waters report 87/2007.**
- Hovind, H. 2006. Intercomparison 0620. pH, K25, HCO3, NO₃ + NO₂, Cl, SO₄, Ca, Mg, Na, K, total aluminium, aluminium reactive and nonlabile, TOC, COD-Mn. Fe, Mn, Cd, Pb, Cu, Ni and Zn. NIVA-report SNO 5285-2006. ICP Waters report 86/2006.
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