Assessment report on vulnerability and the long-term ecosystem impacts of climate change and air pollution at the remote pristine Natura 2000 site in Eastern Finland using long-term ecosystem data

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Table of contents

1. Introduction

1.1 Long-range transported air pollutants and climate change

Increased emissions of air pollutants and greenhouse gases into the atmosphere since the 1950s have escalated environmental problems from the local to the global scale. The long-range transport of sulphur (SO_2) and nitrogen compounds (NO_x, NH_x) has caused widespread acidification of acid-sensitive aquatic and terrestrial ecosystems in Europe and North America (e.g. Wright et al. 2005). A sustained accumulation of deposited inorganic N in forest soil and vegetation also poses a threat to ecosystems through nutrient enrichment and nutrient imbalance (e.g. Bergström and Jansson 2006) and deteriorated tree mineral nutrition (Jonard et al. 2014). It also poses a threat to biodiversity, as a consequence of the eutrophication of sensitive ecosystems (e.g. Bobbink et al. 2010, Dirnböck et al. 2014).

At the same time, emissions of greenhouse gases into the atmosphere are causing global warming, and consequent climate change affects freshwater and terrestrial ecosystems. There is growing evidence that, for example, lakes throughout the world, particularly in northern Europe and North America have been subject to climate change-driven warming (e.g. Schneider and Hook 2010). A substantial body of research demonstrates the sensitivity of lakes to the climate and shows that physical, chemical and biological lake properties respond rapidly to climate-related changes (e.g. Adrian et al. 2009). Many of the retention and release processes for sulphate and inorganic N in catchment soil are sensitive to climatic variables, and would, therefore, be affected by climate change (e.g. Moore et al. 2010, Mitchell et al. 2013). Inter-annual variations in water chemistry related to variations in the deposition of air pollutants and climate are greater than the expected improvement in water chemical status in 2020. The effects of climate variability and change are expected to offset and delay chemical and biological recovery of acid-sensitive waters, for example (de Wit et al. 2015).

Long-term observations are crucial for ecosystem monitoring in general and for forested ecosystem in particular, because such systems have high capacities to store atmospheric inputs and feedback loops may be slow. Forest vegetation is an effective receptor of airborne material delivered in both wet and dry forms because of the reactivity and large surface area of the canopy. The forest floor, including the organic layer is also effective retaining deposition inputs. The soil solution also reflects the atmospheric inputs, but the influence is weaker due to various processes in the soil including weathering, ion exchange, adsorbtion/desorption, decomposition and immobilisation. Furthermore, understorey vegetation, which consists of a remarkable part of the total biodiversity of boreal forests, has a great indicative value when impacts of atmospheric deposition and other environmental changes such as climate change in forest ecosystems are studied.

1.2 Aim of the assessment

Natura 2000 is a network of nature protection areas and is made up of Special Areas of Conservation and Special Protection Areas designated respectively under the Habitats Directive and Birds Directive. Approximately 97% of Finland´s Natura 2000 areas are established in previously protected areas, wilderness areas and areas taking part in nature conservation programmes.

Priority Actions Framework (PAF) is mainly designed "to maintain and restore, at a favourable conservation status, natural habitats and species of EU importance, whilst taking account of economic, social and cultural requirements and regional and local characteristics". Assessment of ecosystem services (ES) is a prioritized action in the Finnish Priority Actions Framework (PAF) for Natura 2000, where a key vision is that the favourable status of biodiversity and ES will be ensured by 2050.

Due to the widespread global pressures, even remote pristine ecosystems with no direct human impact, such as protected Natura areas, are susceptible to harmful anthropogenic environmental changes. In this report, we demonstrate the vulnerability and ecosystem impacts of global change drivers at a selected sensitive Natura 2000 site in Eastern Finland using long-term ecosystem data. We assess the effect of long-term changes in air pollution and climate on small boreal lake, evaluate the change of DOC concentration from atmosphere through the terrestrial area to streams and lakes, and assess the changes connected to the extreme or altered weather events using long-term ecosystem monitoring data between 1987 and 2019. Extreme weather events were identified from the modelled weather data. The concept of critical loads (CLs) is the basis for air pollution control policies in Europe (Amann et al. 2011), and critical loads were determined for the demonstration site with respect to the acidification of surface waters, and eutrophication of the habitat. The exceedances of critical loads of acidification and eutrophication were determined using observed S and N deposition.
Furthermore, we studied long-term changes in understorey vegetation.

2. Material and Methods

2.1 Data

2.1.1 Demonstration site

Impacts of global change drivers are demonstrated in an intensively monitored reference site Lake Iso Hietajärvi and its catchment (Fig. 1) (locating in the catchment area of the FRESHABIT Koitajoki target area), which is located in protected Natura 2000 area (Patvinsuo national park) and represents key habitat of this region.

The site belongs to ICP IM (International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems, www.syke.fi/nature/icpim), ICP Waters (International Cooperative Programme for assessment and monitoring of the effects of air pollution on rivers and lakes, http://www.icp-waters.no/), ICP Forests (International Cooperative Programme on Assessment and Monitoring of Air Pollution Effects on Forests, http://icp-forests.net/) and National Emission Ceilings Directive (NECD) networks aimed at monitoring and detecting long-term impacts of air pollutants and climate change. Data from different compartments in terrestrial and aquatic ecosystems have been collected since the late 1980s, providing unique data sets for assessing the long-term impacts of climate change and air pollutants (www.syke.fi/nature/icpim, ICP IM network).

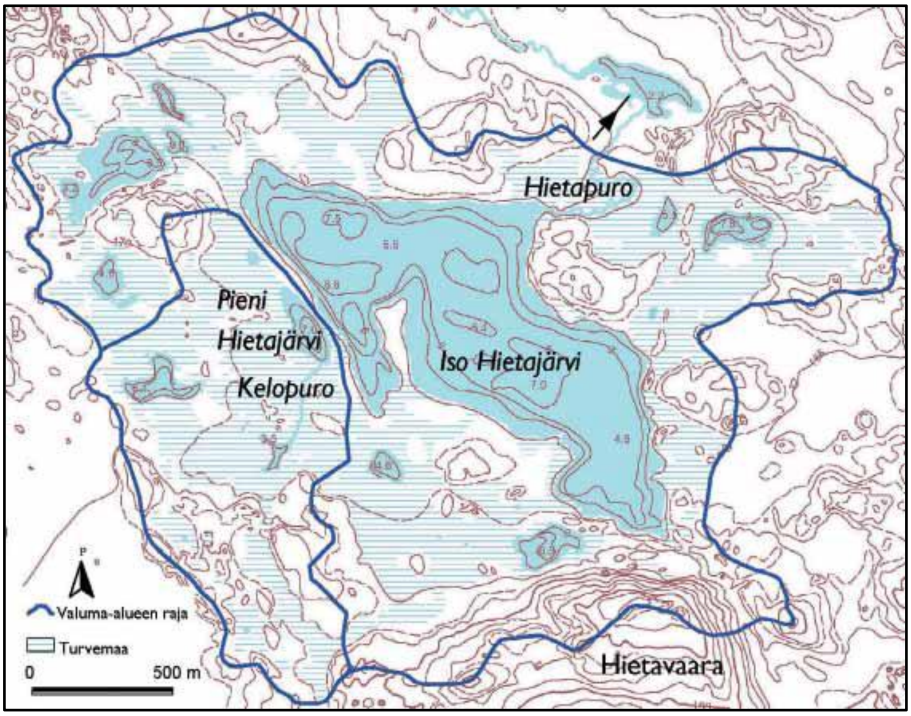

Figure 1. The demonstration site. Blue line indicates the catchment area of the Lake Iso Hietajärvi. Figure: Anita Rämö.

The demonstration lake, L. Iso Hietajärvi, is a boreal, small (A=0.83 km^2) headwater lake with a small pristine forested catchment (A=4.64 km^2) in Eastern Finland (Figs. 1 and 2). Small headwater lakes can be considered as "early warning indicators", since those type of lakes are susceptible to air pollutants and effects of variations in climate and reflect sensitively the changes of the global change drivers. The forest dominantly consists of the Scots pine (*Pinus sylvestris*), Norway spruce (*Picea abies*), with the aspen (*Populus tremula*) and the birch (*Betula* spp.). The forest, peatland and lakes cover 55%, 35% and 23% of the total catchment area, respectively. The bedrock is acid-sensitive, dominated by slow-weathering granitoids and gneisses. The catchment is located inside a conservation area and has been intact for over 50 years, and therefore long-range transported air pollutants and climate change are the only external disturbances.

Figure 2. Lake Iso Hietajärvi.

2.1.2 Deposition, soil water, litterfall and needles monitoring

Samples for bulk deposition (largely wet deposition but also including some dry deposition), including the precipitation amount and chemistry of bulk precipitation, were collected in an open area within the catchment, using continuously open HDPE (high-density polyethylene) plastic funnel collectors (Fig. 3). During the winter conditions (snow cover), cylindrical HDPE collectors were used to collect the bulk deposition samples in winter snowfall. In addition, Luke has set own bulk deposition collectors to the same open area, which are similar type as used for throughfall sampling (see later in connection of throughfall).

Figure 3. Collection of bulk deposition samples. White funnels are official collectors of Finnish Meteorological Institute (right) and Finnish Environment Institute (left), orange funnels are set by Natural Resources Institute Finland (Luke).

Precipitation which passes through the canopy to the forest floor (throughfall) was also sampled (Fig. 4). It is well known that precipitation under the forest canopy differs in quality and quantity from that of precipitation collected in an open area due to the wash-off of dry deposition and strong canopy interactions, such as e.g. leachates produced by the canopy, and uptake of N by plant tissue and through stomata (e.g. Draaijers and Erisman 1995). Throughfall (TF) samples were collected from the intensive observation plot, which is locating in the forest part of the catchment.

Figure 4. Throughfall (orange funnel) and litterfall collectors (green funnels) in L. Iso Hietajärvi catchment.

The observation plot consists of three subplots (size 30 × 40 m or 40 × 40 m) and a surrounding mantle. At one of the subplots are locating throughall, litterfall and soil water collectors, at the other one vegetation related studies are carried out, third one is for soil sampling and whole observation plot for tree measurements (see more Merilä et al. 2007). Throughfall samples were collected using funnel-shaped collectors, which were placed systematically around the plot or in a grid under the canopy (Fig. 4). During the winter, purpose-made snow collectors (a plastic ring and attached plastic bag) were used to collect snowfall under the canopy.

Precipitation is furthermore modified as it infiltrates and percolates through the soil. Therefore, soil solution chemistry is a valuable indicator of soil-mediated effects of stress factors on both forests and the surrounding water ecosystems. The soil solution (SW) was collected using either zero-tension lysimeters at depth of 5 cm (under organic layer) or suction cup lysimeters at depth of 20 cm (upper part of the mineral soil) (Fig. 5). There were 3–6 lysimeters at both depths.

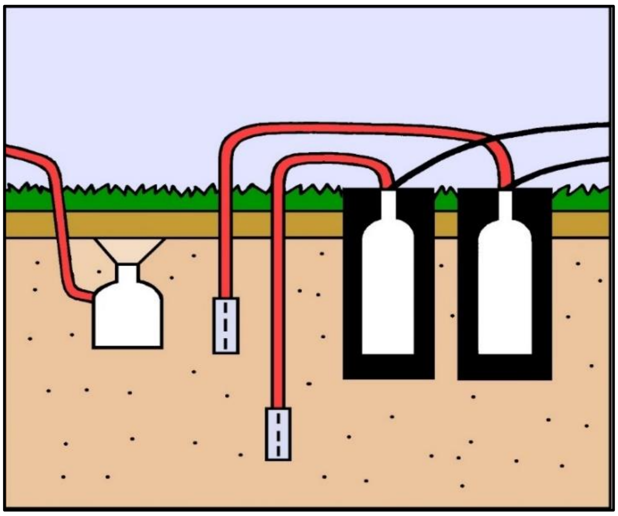

Figure 5. Schematic figure of soil water collectors. On the left zero-tension collector and on the right suction cup lysimeter. Figure: Liisa Ukonmaanaho.

The bulk deposition/precipitation (BP) samples were collected weekly and analysed as a monthly composite sample. Throughfall samples were collected in general at 2-week intervals during the snow free period and at 4-week intervals in winter (winter collection since 1995). In the study area, snowfall usually occurred from November to April. However, in some of the years, the sample collection was performed at 4-week intervals throughout the year. Throughfall samples from 12 (1990–1998) to 20 (1999–2018) collectors were pooled to a composite sample representative for a certain stand. Weekly samples can be analysed or mixed with monthly samples before analyses. The soil water (SW) was collected same interval as throughfall during snow-free period. The samples from zero-tension lysimeters are removed by means of a plastic tube leading down into the collection acid washed bottle, while suction cup lysimeters soil water were drawn using a vacuum suction of ca. 60 kPa and collected into acid-washed glass bottles placed in a covered bucket dug into the soil to keep the samples cool and in the dark (Fig. 5).

In order to study whether there is changes in litterfall (LF) chemical composition or amount, we collected also litterfall from the study area using 12 litterfall traps, which located systematically on a 10 × 10 m grid on the plot. The top of the funnel-shaped traps, with a collecting area of 0.5 m^2, stood at a height of 1.5 m above the forest floor (Fig. 4). The LF was collected in a replaceable cotton bag attached to the bottom of the LF trap. The LF was sampled at two- or four-week intervals during the snow-free period (May to November), and once at the end of winter. The LF production (dry mass per unit area) was calculated by dividing the total LF mass by surface area of the traps. In addition, tree-specific sample branches with current (C) and previous-year (C+1) needles were collected from the uppermost third of predominant or dominant trees (n=10) on each study plot once during October–November, uneven years, starting in 2005.

As Iso Hietajärvi demonstration site is dominantly forested catchment, dry deposition (gases and particles filtered by the canopy) highly contributes to the total deposition. The total deposition of non-marine, anthropogenic SO_4 to the catchment including wet and dry deposition fractions was estimated from the bulk deposition and throughfall measurements by calculating the annual deposition, both to the defined open area (bulk) and the forest area (throughfall), and then summing up the area-weighted open area and throughfall deposition. To distinguish changes in anthropogenic SO_4 from climate-related variations in sea salt, trends for SO_4 deposition was calculated using non-marine fractions. The sea salt-corrected fractions were calculated by subtracting the marine contribution estimated from the ratio of the ion to Cl in seawater (Lyman and Fleming 1940). Because of a strong canopy interaction for reduced N (NH_4) and oxidized N (NO_3), bulk deposition measurements were used for the total deposition of inorganic nitrogen.

After sampling, samples have been stored in dark and cool conditions. Chemical analyses from liquid and solid samples were determined using standardized methods (e.g. Ukonmaanaho et al. 2008, Ruoho-Airola et al. 2014, Ukonmaanaho et al. 2014).

2.1.3 Surface water chemistry and hydrometeorological monitoring

Physico-chemical monitoring of the lake was carried out by the Environmental Administration (Finnish Environment Institute SYKE and Centre for Economic Development, Transport and the Environment for Pohjois-Karjala). Water sampling, altogether 7–8 times, has been carried out monthly in March, April-August, and once in October and December from the depths of 1, 3 and 5

m. All samples were taken at the site of the maximum depth, in the middle of the lake. For SO_4 and sum of base cations ($BC=Ca^{2+}+Mg^{2+}+Na^{+}+K^{+}$) in lake chemistry, the non-marine fractions were used, and are later denoted as xSO_4 and xBC.

Samples for runoff water chemistry were collected biweekly during the high-flow seasons in spring (April–May) and autumn (September–October), and monthly during the base-flow period in winter and summer at the catchment outlet stream. Continuous runoff monitoring (based on water level recording calibrated against discharge) was performed at the overflow-measuring weir in the outlet stream, 140 m downstream from the lake. Monthly runoff (mm $month^{-1}$) was calculated as a sum of mean daily runoff values.

Mean monthly air temperatures were calculated for the period 1990–2019. Air temperatures for the region of L. Iso Hietajärvi were collected from Finnish Meteorological Institute's observation station Lieksa Lampela (WMOn 2796) situating 37 km north-west from the study area.

2.1.4 Trend analysis

The Seasonal Kendall test (SKT, Hirsch et al. 1982) was used for detecting long-term monotonic trends. For air temperature, runoff volume, water chemistry and temperature monthly values were used. SKT is one of the most popular trend analyses for detecting monotonic trends in water chemistry records because it is not particularly sensitive to missing data or outliers and is robust with respect to non-normality and serial character (e.g. seasonal changes). The long-term trends for lake chemistry were calculated only for samples taken from the depth of 1 m for the period 1990–2019. On an annual scale, the samples represented almost the entire year (March–December). The gradient of the trend (annual change), i.e. the slope of the linear trend, was calculated according to Sen's slope estimation method (Sen 1968). A statistical significance threshold of $p < 0.05$ was applied to the trend analysis.

2.1.5 Sulphur and nitrogen deposition and critical load exceedances

There is an increased risk for ecosystems to become acidified, or eutrophied, if the deposition of sulphur (S) and nitrogen (N) are persistently higher than the critical loads of acidification, or eutrophication (Posch et al. 2015). The difference between the acidifying, or eutrophying, deposition and the critical load of acidification, or eutrophication, at a certain site is called the exceedance of acidifying or eutrophying CL (Ex_{aci} or Ex_{eut}). Critical loads of acidification and eutrophication have been determined for L. Iso Hietajärvi (Holmberg et al. 2013, Forsius et al. 2021).

The critical load of acidification was determined on the basis of a critical runoff ANC of 20 µeq l^{-1}, to avoid harmful effects of fish in case of lower ANC in runoff. Both S and N deposition contribute to the acidity of soil and surface water, and the acidity critical load of acidity is not a single value, but a function characterized by values such as $CL_{max}S$ and $CL_{max}N$. The acidity critical load for L. Iso Hietajärvi was determined using the FAB model (Henriksen and Posch 2001, Posch et al. 2012).

For eutrophication, the critical load CL_{eut} was determined as the minimum of the empirical and the mass balance critical load (Posch et al. 2015). Bobbink and Hettelingh (2011) reviewed observational and experimental studies on the impact of N deposition on ecosystems in Europe. Following their review, they provided a comprehensive set of habitat specific empirical critical loads for N that represent the thresholds for N deposition below which eutrophication effects do not occur according to present knowledge (Bobbink and Hettelingh 2011). The mass balance critical load was determined on the basis of an acceptable concentration of N in runoff of 1.3 mg l^{-1}, in order to avoid harmful effects on vegetation such as nutrient imbalances, or sensitivity to fungal disease or frost (Mapping Manual, Table V.5, 2017).

For each year in the period 1990–2017, Ex_{aci} was calculated from the observed and S and N depositions and the acidity critical load function, and Ex_{eut} was obtained as the difference between the observed N deposition and CL_{eut} (Forsius et al. 2021). Here we report the year-to-year variation in runoff ANC and TIN as a function of the exceedances of critical loads of acidification (Ex_{aci}) and eutrophication (Ex_{eut}).

2.1.6 Forest

BP, TF, SW, needle and LF concentration values were screened for gross outliers and error were corrected. The remaining gross outliers were replaced by regression estimates based on the relationships with other determinants. Monthly values were used, excluding green needles, which were taken every second year. Acid neutralizing capacity (ANC) was calculated as the difference between the sum of dissolved base cations (Ca^{2+}, K^{+}, Mg^{2+}, Na^{+}) and the sum of dissolved 'strong acid anions' (SO_4^{2-}, NO_3^{-}, Cl^{-}) on an equivalent basis. It indicates the acidification potential of water and is associated with the deposition of strong acids, taking the buffering capacity effect on base cations into account (Neal et al. 2001). The Seasonal Kendall test (SKT, Hirsch et al. 1982) was used for detecting long-term monotonic trends. In 'terrestrial observation' chapter 3.7, yearly values were used for BP, TF, SW, LF, green needles and stream water trend analysis.

Annual mean air temperatures and precipitation sums were derived for each site using a model by Venäläinen et al. (2005), which is based on daily mean temperature and precipitation data interpolated onto a 10 ×10 km grid provided by the Finnish Meteorological Institute weather stations.

2.1.7 Understorey vegetation

2.1.7.1 Sampling design

The Finnish Environment Institute (SYKE) was responsible for the inventory of understorey vegetation in the Hietajärvi forest habitat in 1990–1998 as a part of ICP Integrated Monitoring Programme. During this period there were altogether 24 vegetation quadrats sized 1 m^2 and 36 quadrats sized 0.25 m^2 for monitoring of the cover % of plant species and their population dynamics in the two plots (FI03/01 and FI03/04) located in the forest area (Kokko et al. 2002). The sampling design for vegetation survey changed after 1996, when the Finnish Forest Research Institute (nowadays Natural Resources Institute Finland, Luke) started monitoring of the forest ecosystem processes including understorey vegetation as a part of the ICP Forests Level II programme (Merilä et al. 2007). Then three sub-plots sized 30 m × 30 m were established inside the Level II plot (total area with a buffer zone 0.5 ha).

In the new design a 30 m × 30 m (sub-plot no 1) subplot was marked inside the plot FI03/04. Altogether 16 sampling quadrats for vegetation assessment were established systematically (4 × 4 design) on the sub-plot (Fig. 6). The size of the quadrat was 2 m^2 (1.41 m × 1.41 m). The distance of the upper left corner of the first quadrat from the side of the plot was 304 cm, and the distance between the sides of two adjacent quadrats was 608 cm. The location of the quadrats was marked permanently with white plastic stakes at two diagonal corners. The code of the quadrat (1–16) was signed on the upper part of the stakes in black paint. In addition, four 10 m × 10 m quadrats (A - D) were marked out with orange plastic stakes to give four areas for smaller vegetation quadrats (Fig. 6). These areas form the monitoring area comparable with the ICP Forests Level II monitoring programme (Common Sample Area is 400 m^2). The plant species growing outside the small quadrats were recorded within the areas of 4 × 100 m^2 (A - D) and 900 m^2.

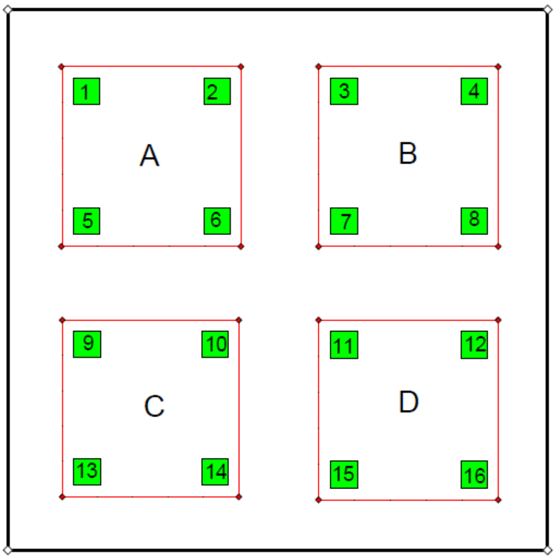

Figure 6. The sub-plot 30 m × 30 m in size used for the inventory of understorey vegetation. The cover % of plant species was assessed on the small (green) sample quadrats (sized 1.4 m × 1.4 m). Species found outside the small quadrats were recorded within red areas (each 10 m × 10 m) and the whole plot.

2.1.7.2 Measurements of plant cover and structure of tree stand

Visual coverage of the plant species was assessed using the following scale: 0.01, 0.1, 0.2, 0.5, 1, 2, …99, 100 %. The bottom layer (mosses and lichens), the field layer (vascular plants, height < 50 cm: trees and shrubs, dwarf shrubs, herbs, and grasses and sedges) and the shrub layer (height 50–200 cm: trees and shrubs) were investigated. A 2 m^2 frame divided into 100 small quadrats by a net of elastic strings was used in assessing of the percentage covers (Fig. 7). An open frame without a net was placed on the sites where a tree, shrubs or high vegetation were growing (Fig. 7). Plants on stones or decayed wood were excluded. The coverage of leaf and needle litter, dead plant material, small diameter decaying wood, dead branches, fallen tree stems, stumps, bare mineral soil and stones were also assessed, each in own group. Species occurring on the vegetation plot, but not found on the sample quadrat, were recorded with 0.01% cover. More information of the sampling strategy (e.g. required number and size of sample quadrats) is given in Salemaa et al. 2000.

Three or four botanists worked in the plot simultaneously, so that each assessed vegetation on their own quadrats within a 10 m × 10 m area. Field tests were carried out to check that the assessment level stayed similar between the observers. Samples of unknown plant species were later identified by specialists. All the surveys were carried out in the end of July (June in 2003) during maximum biomass stage of understorey vegetation.

Stand measurements were carried out in the area of the whole plot, but here we present the stand data from the same sub-plot (no 1) as the vegetation survey in 2009. Stem number, volume, basal area, diameter and tree height were measured from living and dead trees with a breast height diameter of at least 4.5 cm. In addition, the height of lower crown limit was measured from living trees. Lying dead trees were not included in the measurements.

Figure 7. Hannu Nousiainen assessing the cover % of plant species using a 2 m^2 frame divided into 100 small quadrats with a net (year 2009) (left), and Juha-Pekka Hotanen assessing the cover % of plant species using a frame having an open corner and without a net (right). Photo: Maija Salemaa.

2.1.7.3 Climatic variables, nitrogen deposition and nitrogen content of plant species

The effective temperature sum, cumulative precipitation and average minimum temperature of days were determined for each year during 1998–2019 from the data base of the Finnish Meteorological Institute (FMI) at a daily resolution for a grid scale of 1 × 1 km^2 (Venäläinen et al. 2005). We used annual precipitation sums, since beside summer precipitation water volume of snow have importance for soil moisture (Ilvesniemi et al. 2010). The effective temperature sum (growing degree days GDD °C) was defined as the sum of the positive differences between diurnal mean temperatures and 5 °C yr^{-1}.

Throughfall precipitation and nitrogen deposition were collected within the forest stand with 20 rainfall collectors (funnels, diameter 20 cm) resulting the area-based estimate for the forest floor (Fig. 4). Bulk deposition of nitrogen was collected in the nearby open area with 3 rainfall collectors (Fig. 3). The data of N deposition in Hietajärvi in 2009 and average during 2007–2009 for other ICP Forests Level II plots in Finland has been published in the supplementary data of Salemaa et al. (2020).

The plant material for the analysis of nitrogen (N) and carbon (C) concentrations and contents was collected in an EU Life+ project FutMon in 2009. We took systematically biomass samples (total n = 28) along transects in the buffer zone of the vegetation monitoring plot. Biomass was divided into functional plant groups as follows: deciduous and evergreen dwarf shrubs (leaves and stems separately), herbs, grasses and bryophytes. Bryophyte biomass included only the upper living part (with 2.5–3 annual growths). The N and C concentration was determined using CHN analyser (LECO) (details of sampling and chemical analysis of bryophytes in Salemaa et al. 2020).

3. Results and discussion

3.1 Runoff and air and water temperature

In 1990–2019, the mean annual runoff was 384 mm yr^{-1} without any significant trend neither on annual nor monthly basis. Only a weak decreasing trend in June was detected (Table 1). Given the interest in leaching of constituents from the catchment, high annual and/or summer/autumn runoff during the study period occurred in 1992–1993, 1998, 2004, 2007–2008, 2013 and 2017 (Fig. 8).

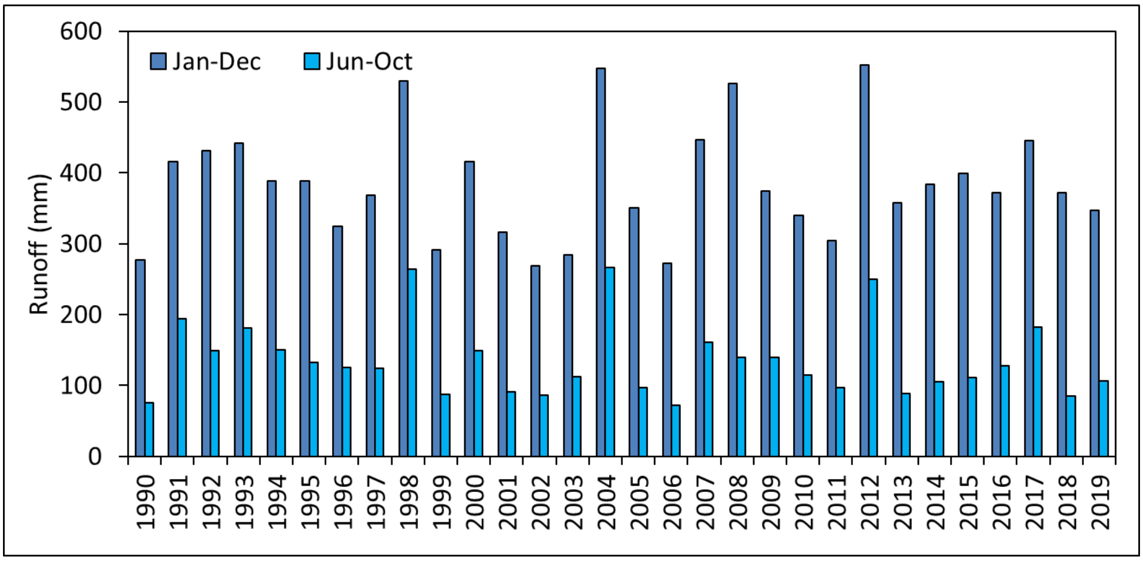

Figure 8. Annual and seasonal (June-October) runoff at the measurement station of the Iso Hietajärvi catchment in 1990–2019.

Table 1. Long-term temporal monthly and annual trends (SKT, Sen's slope) for runoff in the L. Iso Hietajärvi catchment in 1990–2019. For the annual change (mm yr^{-1}), a statistically significant trend ($p < 0.05$) is denoted with asterisk. A weak trend ($p < 0.1$) is indicated with +.

Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan-Dec
0.32	0.18	0.09	0.21	-0.08	-0.40^{+}	-0.28	-0.08	-0.1	-0.19	0	0.24	0.01

There was a significant increase in annual air temperature in L. Iso Hietajärvi area from 1990 to 2019 (Table 2). Seasonally, the increase was most pronounced in spring (May, *p*=0.009), but significant increase was detected also in late summer (August, *p*=0.04) and autumn (September, *p*=0.04 and November, *p*=0.01). This is in line with the trend on air temperatures observed for the whole of Finland (Tietäväinen et al. 2010).

Table 2. Long-term temporal monthly and annual trends (SKT, Sen's slope) for air temperatures in the Iso Hietajärvi area in 1990–2019. For the annual change (slope, °C yr^{-1}), a statistically significant trend ($p < 0.05$) is denoted with asterisk.

Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan-Dec
-0.02	0.05	0.01	0.04	0.11*	0.004	0.04	0.05*	0.07*	0.01	0.19*	0.11	0.05*

At the same period 1990–2019, the annual water temperature of the surface layer (0–1 m) has significantly increased in L. Iso Hietajärvi (Table 3). The trend was most pronounced in September during the autumn overturn, when water temperature was significantly increased in all water layers. Epilimnetic water temperatures are shown to reflect warming trends in air temperature in North America (Arhonditsis et al. 2004, Coats et al. 2006, Stainsby et al. 2011), Europe and Eurasia (Livingstone 2003, Hampton et al. 2008), and Africa (O'Reilly et al. 2003). Significant warming of lakes has taken place throughout the world, but lakes in northern Europe and North America are warming most rapidly (Schneider and Hook 2010, Hook et al. 2012). As water temperature is closely linked to change in air temperature, the predicted increase in air temperature due to climate change will result in increased surface water temperatures in the future. Physical modeling studies of medium-sized lakes in the temperate zone predict that temperatures will increase more in the upper regions of the water column than in the lower regions, resulting in generally steeper vertical temperature gradients and enhanced thermal stability (Hondzo and Stefan 1993, Stefan et al. 1998, Peeters et al. 2002).

Table 3. Long-term temporal monthly trends (SKT, Sen's slope) for water temperatures in the L. Iso Hietajärvi in 1990–2019. For the annual change (°C yr^{-1}), a statistically significant trend ($p < 0.05$) is denoted with asterisk. A weak trend ($p < 0.1$) is indicated with +.

Depth (m)	Mar	May	Jun	Jul	Aug	Sep	Oct	Mar-Oct
1	0	0.09	0.06	0.1	-0.03	0.10*	0.1	0.04*
3	-0.02	0.07	0.07	0.04	-0.02	0.11*	0.1	0.04^{+}
5	-0.01	0.05	0	-0.02	-0.03	0.11*	0.1	0.01

3.2 Sulphur and nitrogen deposition

Successful emission reduction measures in Europe over the past 30–40 years have led to a declining deposition of air pollutants in Europe (Colette et al. 2016), as shown at Iso Hietajärvi demonstration site (Fig. 9). The emission control programmes have been particularly successful for sulphur (S), and the total deposition of anthropogenic (non-marine fraction) sulphate (xSO_4) decreased in the Iso Hietajärvi area by 70–80% between 1990 and 2017.

Nitrogen (N) emissions have also decreased and have resulted in a decrease of total inorganic nitrogen ($TIN=NO_3+NH_4$) deposition, but the decrease of TIN deposition has been generally smaller than that of xSO_4. European N emissions have decreased less than those of S, and the bulk deposition of TIN has generally exceeded xSO_4 deposition on an equivalent basis since the late 1990s.

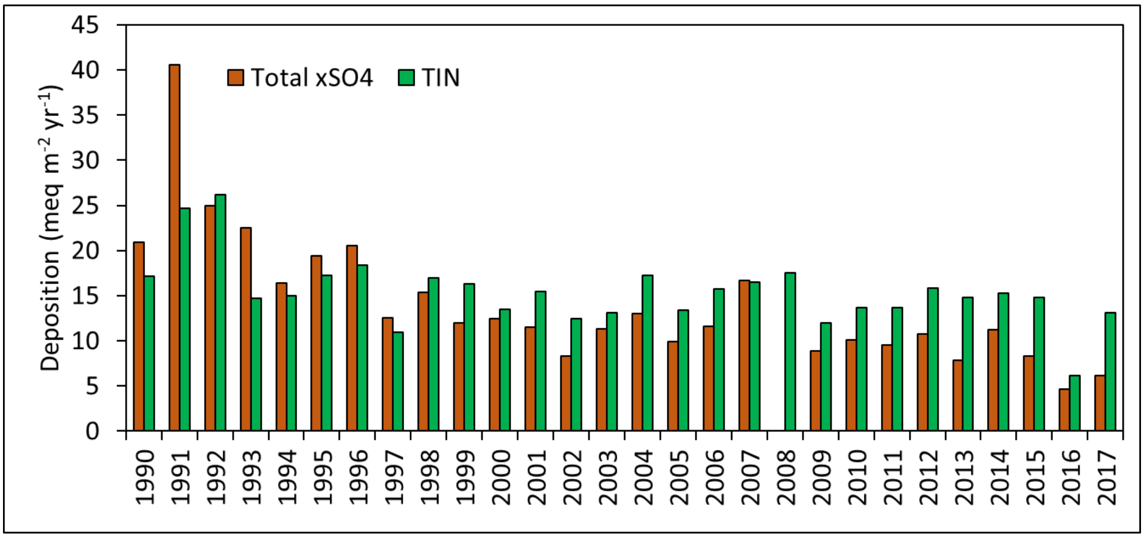

Figure 9. The annual total deposition of non-marine xSO_4 (x denotes non-marine, anthropogenic fraction) and TIN (TIN=NO_3+NH_4) in the Iso Hietajärvi region in 1990–2017. (Data source for bulk deposition of xSO_4 and TIN: Finnish Meteorological Institute).

3.3 Acidification parameters and trace (heavy) metals

Lake Iso Hietajärvi exhibits neutral or only weak acidic conditions with the mean pH 6.7 (range 5.9–7.3) between 1990 and 2019. L. Iso Hietajärvi is located in low SO_4 deposition area, and airborne acidification has not markedly taken place in the lake, although the lake can be considered sensitive to acidification with low buffering capacity (mean alkalinity 88 µeq l^{-1} in 1990–2019) and has been affected to some extent to acid deposition. Due to the decreased acid S deposition, sulphate concentration has significantly decreased by about 60% during the study period 1990–2019, and the acidification reversal was also recorded in L. Iso Hietajärvi (Fig. 10, Table 4).

Table 4. Long-term temporal trends (SKT, Sen's slope) for pH, conductivity, alkalinity, Acid Neutralizing Capacity (ANC = (Ca+Mg+Na+K) – (SO_4+Cl+NO_3)), sum of non-marine (x denotes non-marine fraction) base cations (xBC), non-marine sulphate (xSO_4), chloride (Cl), total organic carbon (TOC), water colour and silicon dioxide in the L. Iso Hietajärvi in 1990–2019. For the annual change, a statistically significant trend ($p < 0.05$) is denoted with asterisk. A weak trend ($p < 0.1$) is indicated with +.

Variable	Unit	Mar	May	Jun	Jul	Aug	Oct	Dec	Mar-Dec
pH	pH-unit	0.01*	0.02*	0.00	0.00	-0.00	0.00	0.00	0.00^{+}
Conductivity	mS m^{-1} yr^{-1}	-0.01^{+}	-0.01*	-0.00^{+}	-0.01*	-0.01*	0.00	-0.01^{+}	-0.01*
Alkalinity	µeq l^{-1} yr^{-1}	0.56*	0.44*	0.66*	0.50*	0.57*	0.81*	0.50	0.57*
ANC	µeq l^{-1} yr^{-1}	0.86*	0.44*	0.72*	0.77*	0.81*	1.20*	0.06	0.72*
xBC	µeq l^{-1} yr^{-1}	0.11	-0.25	0.03	0.05	0.00	0.32	-0.48^{+}	0.00
xSO_4	µeq l^{-1} yr^{-1}	-0.70*	-0.65*	-0.54*	-0.60*	-0.63*	-0.73*	-0.80*	-0.64*
Cl	µeq l^{-1} yr^{-1}	-0.12*	0.00	0.00*	0.00^{+}	0.00	0.00	-0.00*	0.00*
TOC	mg l^{-1} yr^{-1}	0.06*	0.07*	0.07*	0.07*	0.06*	0.05*	0.05	0.06*
Colour	mg Pt l^{-1} yr^{-1}	0.31*	0.44*	0.67*	0.50*	0.44*	0.63*	0.71*	0.50*
SiO_2	mg l^{-1} yr^{-1}	0.03	0.03*	0.03^{+}	0.03^{+}	0.02	0.04*	0.05^{+}	0.03^{+}

Base cation concentrations exhibit no decreasing trend, while sulphate concentration was decreased, indicating the improved acid-base status of soils, and led to significant increase in buffering capacity (alkalinity, measured and ANC, calculated) in the lake water. A weak increasing trend on an annual basis was detected for pH-value. Seasonally, the increase of pH was significant in winter and spring.

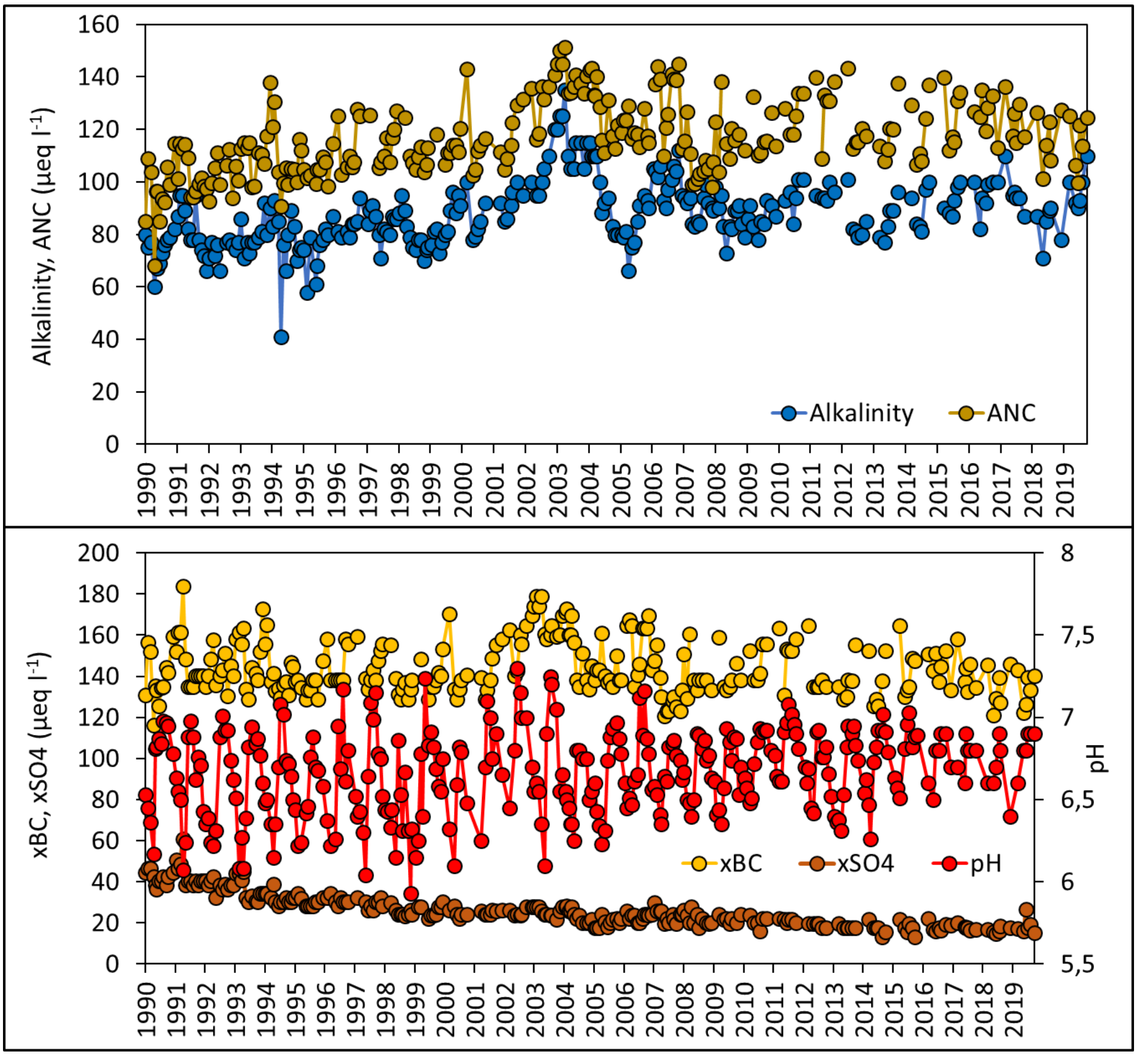

Figure 10. Time series for alkalinity and Acid Neutralising Capacity (ANC= [Ca+Mg+Na+K]-$[SO_4+Cl+NO_3]$ (up) and non-marine base cations (xBC) and sulphate (xSO_4), and pH (down) at the depth of 1 m in L. Iso Hietajärvi in 1990–2019.

Many studies have reported that accompanying a decreasing SO_4 deposition, a net release (output > input) of SO_4 from internal soil S sources has taken place in many forested catchments in Europe and North America (e.g. De Vries et al. 2001, 2003, Forsius et al. 2005, Mitchell et al. 2013, Vuorenmaa et al. 2017). This process may be considered as a time-lagged recovery of terrestrial systems fuelled by the mobilization of legacy S pools accumulated during times of a high atmospheric SO_4 deposition. The recovery of forested catchments from SO_4 deposition – in terms of SO_4 net release – appeared to be most pronounced in catchments with the highest SO_4 deposition level and also having the strongest decrease in the SO_4 deposition load, but this process has taken place also in low SO_4 deposition areas (Vuorenmaa et al. 2017). In the low SO_4 deposition area at the site L. Iso Hietajärvi, SO_4 was mainly retained in the catchment (input > output), but the retention rate declined over the study period, and at the end of the study period, the catchment shifted towards a net release (output > input) (Fig. 11).

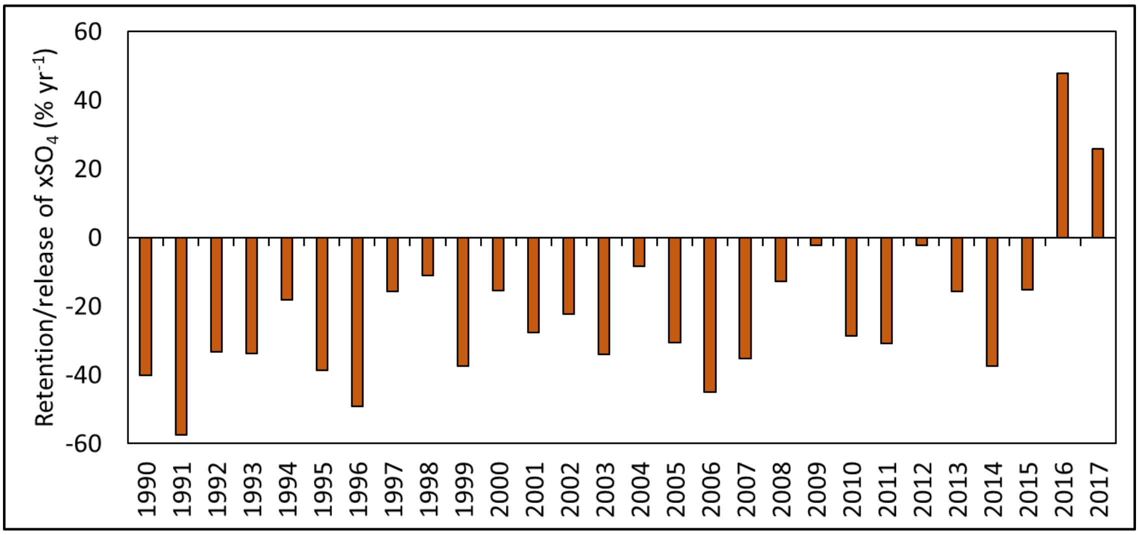

Figure 11. Retention/release of xSO_4 (% yr $^{-1}$) in the L. Iso Hietajärvi catchment in 1990–2017.

Due to the net release of sulphate, it is suggested that SO_4 remains the dominant source of actual soil acidification despite the generally lower input of sulphur than nitrogen in European forested ecosystems (De Vries et al. 2003). This is important for assessing the effects of emission reductions on acidification recovery. Several processes, including desorption and excess mineralisation, and re-oxidation of reduced sulphur regulate the long-term response of soil S, and a differentiation is necessary for predictions of future responses (e.g. Prechtel et al. 2001).

Besides changes in deposition, the variation in runoff contributes to the net release of mobilized SO_4 in forested catchments (Vuorenmaa et al. 2017). Because many of the S retention and release processes are also sensitive to changes in climatic variables, further analysis of processes regulating mobilization and the release of SO_4 in terrestrial ecosystems are needed to allow an evaluation of the effects, not only of emission reduction policies, but also of the changing climate.

Along with decreased acidifying emissions, emissions of trace (heavy) metals, particularly of Hg, Cd and Pb, substantially decreased in Europe (Travnikov et al. 2012), and in line with this, the deposition of the trace (heavy) metals in South Finland clearly declined over the past decades (Ruoho-Airola et al. 2014). It is obvious that decrease of long-range transported trace metals deposition has also taken place in remote Iso Hietajärvi region. The general decrease of trace metals deposition was reflected to some extent in lake water concentrations (Table 5). A significant decreasing trend was detected for arsenic (As), chrome (Cr), lead (Pb) and nickel (Ni) between 1994 and 2019. For copper (Cu) the median value and maximum concentrations, indicated by 90% percentiles, decreased between the 1990s and 2010s.

Concentration of Cd was mostly below the detection limits (< 0.03 µg l^{-1} in 1994–2004 and < 0.01 µg l^{-1} in 2005–2019). Total Hg determinations from the lake water started in December 2002, and analytical detection limit has changed over time, therefore water chemistry data is insufficient to reveal long-term pattern in concentration.

Table 5. Percentiles (10%, median and 90%) of the concentrations for the three periods (1994–1999, 2000–2009 and 2010–2019) and long-term temporal trends (SKT, Sen's slope) in 1994–2019 for trace metals Al tot, As, Cd, Cr, Cu, Pb, Ni, Zn and Hg in the L. Iso Hietajärvi. For the annual change, a statistically significant trend ($p < 0.05$) is denoted with asterisk. n.d.=no data.

Variable	Percentiles 1994-1999 ($\mu g\ l^{-1}$)			Percentiles 2000-2009 ($\mu g\ l^{-1}$)			Percentiles 2010-2019 ($\mu g\ l^{-1}$)			Trend 1994-2019
	10%	50%	90%	10%	50%	90%	10%	50%	90%	($\mu g\ l^{-1}\ yr^{-1}$)
Al tot	7.00	10	19	4.52	10	17	5.00	10	14	-0.000
As	0.08	0.11	0.14	0.10	0.12	0.13	0.08	0.10	0.12	-0.001*
Cd	<0.03	<0.03	<0.03	<0.01	<0.03	<0.03	<0.003	0.004	0.005	n.d.
Cr	<0.10	0.16	0.74	0.06	0.10	0.37	0.04	0.08	0.10	-0.004*
Cu	0.08	0.15	0.40	0.10	0.14	0.30	0.004	0.11	0.31	-0.001
Pb	0.05	0.08	0.31	0.04	0.07	0.14	0.04	0.06	0.11	-0.001*
Ni	0.08	0.13	0.21	0.09	0.10	0.24	0.06	0.10	0.13	-0.002*
Zn	0.15	0.50	1.66	0.50	0.50	1.08	0.35	0.80	1.40	0.000
Hg	n.d.	n.d.	n.d.	<0.002	<0.002	0.002	0.001	0.001	0.003	n.d.

The decrease of trace metal deposition was not fully reflected in lake water concentrations, because the response to atmospheric deposition is difficult to differentiate from other factors, such as natural catchment acidity due to organic soils and delays in the hydrological transport of the trace metals from the catchment due to their strong retention in the soil (e.g. Ukonmaanaho et al. 2001). Acidity particularly controls the levels of As, Cd and Zn, while organic matter controls the levels of Cr, Fe, Cu and Ni in headwater lakes, and Pb, Mn and Al are affected by both factors. Humic substances act as carriers of trace metals from catchment soils to surface waters, irrespective of the source of trace metals.

3.4 Total organic carbon and water colour

Lake Iso Hietajärvi can be considered as a clear water lake (mean water colour and total organic carbon concentrations (TOC) 29 mg Pt l^{-1} and 4.7 mg l^{-1}, respectively, in 1990–2019), but during the past 30 years, L. Iso Hietajärvi has shown proceeding brownification with a significant increase in both TOC concentration and water colour (Table 4, Fig. 12).

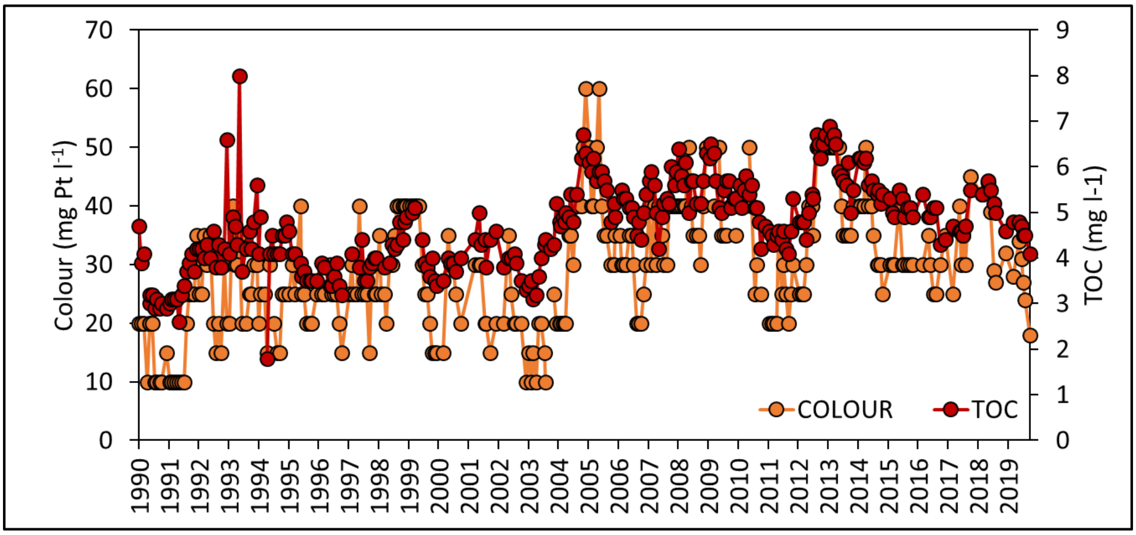

Figure 12. Time series for water colour and total organic carbon (TOC) concentrations at the depth of 1 m in the L. Iso Hietajärvi in 1990–2019.

From 1990 to 2019, TOC concentration has increased approximately by 2 mg l^{-1} and water colour 15 mg Pt l^{-1}.

The increases in TOC and water colour in L. Iso Hietajärvi were in agreement with earlier observations from Finland and elsewhere in Europe and North America (e.g. Vuorenmaa et al. 2006, Monteith et al. 2007, de Wit et al. 2016). Browning in the 1990s and early 2000s has been attributed dominantly to improved air chemistry i.e. substantially decreased acid sulphate deposition and variations in sea-salt deposition, acting through chemically-controlled organic matter solubility in catchment soils (e.g. Monteith et al. 2007). Recently, changes in climatic conditions, such as increased precipitation and discharge, are exerting greater influence on variation and increasing TOC concentrations in surface waters (e.g. de Wit et al. 2016).

Increased TOC concentration and water colour, and the consequent decrease in light penetration into the lake may have large ecological impacts on small boreal lakes, such as decreasing primary production (Arvola et al. 2014) and decreasing feeding efficiency and growth of perch (Rask et al. 2014). Increased TOC and water colour may lead to heat absorption in shallower water layers and is reported to strengthen the thermal stratification after ice-off in spring, causing incomplete spring overturn and deteriorated oxygen conditions in the lower part of the water column (Vuorenmaa et al. 2014). In anoxic conditions, it is likely that phosphorus stored in the sediment will be released into the water, causing eutrophication.

Variation in runoff and leaching of humic-derived organic acids may also have affected alkalinity in L. Iso Hietajärvi, because in the pH range 4–7 a significant fraction of organic acids can be considered strong and may have a large influence on pH and alkalinity (e.g. Munson and Gherini 1993). Runoff-induced surges of organic acids can be an important factor suppressing recovery of pH and alkalinity in acid sensitive Finnish lakes (e.g. Vuorenmaa and Forsius 2008).

3.5 Nitrogen, phosphorus and oxygen

During the period 1990–2019, there was a significant decreasing trend in total inorganic nitrogen concentrations (TIN=NO_3-N+NH_4-N) on an annual basis. Seasonally, the decrease in nitrate (NO_3-N) concentration was pronounced in summer (Table 6). The trend slopes for NO_3-N and NH_4-N were generally decreasing rather than increasing, which is in agreement with declined TIN deposition in the region, and annual deposition amounts. The trend slopes of TIN concentrations in surface waters have been generally decreasing rather than increasing also in other undisturbed forested catchments elsewhere in Europe (Vuorenmaa et al. 2018).

Studies from European forested ecosystems have shown that nitrate leaching mainly occurs when the inorganic N deposition input is above a critical deposition threshold of ca. 10 kg ha^{-1} yr^{-1} (e.g. Dise and Wright 1995). During the period 2010–2017, the mean annual TIN deposition in the region was 13 meq m^{-2} yr^{-1} (1.8 kg ha^{-1} yr^{-1}), which was clearly below the critical deposition threshold, which should mean low deposition-driven risk of N leaching. Moreover, the input-output budgets of inorganic nitrogen for the Iso Hietajärvi catchment showed high net retention (95%) of inorganic nitrogen (Vuorenmaa et al. 2017). Total nitrogen concentration did not exhibit any long-term trend on an annual basis in 1990–2019.

Table 6. Long-term temporal trends (SKT, Sen's slope) for nitrate (NO_3-N), ammonium (NH_4-N), total inorganic nitrogen (TIN=NO_3-N+NH_4-N), total nitrogen (tot N), organic nitrogen (Org N), total phosphorus (tot P), N:P-ratio and hypolimnion oxygen (O_2) (bottom-layer) in L. Iso Hietajärvi in 1990–2019. For the annual change, a statistically significant trend ($p < 0.05$) is denoted with asterisks. A weak trend ($p < 0.1$) is indicated with +.

Variable	Unit	Mar	May	Jun	Jul	Aug	Oct	Dec	Mar-Dec
NO_3-N	$\mu g\ l^{-1}\ yr^{-1}$	0.27	-0.19	-0.00*	-0.00*	-0.00*	-0.00	0.19	-0.00*
NH_4-N	$\mu g\ l^{-1}\ yr^{-1}$	-0.33	-0.06^{+}	-0.00	0.00	0.00	-0.05	-0.33*	-0.00*
TIN	$\mu g\ l^{-1}\ yr^{-1}$	-0.44	-0.42	-0.00	-0.00	-0.00	-0.17^{+}	-0.13	-0.03*
tot N	$\mu g\ l^{-1}\ yr^{-1}$	-0.00	-0.67	-0.16	-0.91*	0.00	-0.00	0.53	-0.00
Org N	$\mu g\ l^{-1}\ yr^{-1}$	0.18	-0.41	-0.48	-0.93*	0.10	0.11	0.79	-0.00
tot P	$\mu g\ l^{-1}\ yr^{-1}$	-0.00*	-0.08*	-0.00	-0.00	-0.07*	-0.00	-0.00	-0.01*
N:P-ratio		0.42^{+}	0.11	0.07	0.00	0.30^{+}	0.14	0.28	0.17^{+}
O_2, hypolimnion	$mg\ l^{-1}\ yr^{-1}$	0.04	-0.03	-0.06*	-0.05	-0.03	-0.03^{+}	0.07^{+}	-0.02*

Lake Iso Hietajärvi is a nutrient poor, oligotrophic lake with the mean total phosphorus (tot P) concentration 5.7 $\mu g\ l^{-1}$ (range 2–14 $\mu g\ l^{-1}$) in 1990–2019 (Fig. 13). During the same period, tot P concentration showed significant decreasing trend (Table 6). Seasonally, the decreasing trend was pronounced in early summer (June). In the same period, N:P-ratio exhibited weak increasing trend, indicating the increasing importance of phosphorus as a limiting nutrient, which has been detected in other small boreal lakes in Finland (Arvola et al. 2014). L. Iso Hietajärvi has not been suffered anoxic conditions in the hypolimnion (bottom layer), although long-term records show gradual decrease in the bottom layer oxygen concentrations (Table 6). This may be due to increased organic matter in the lake and consequent accelerated decomposition in the bottom layer. Increased leaching of dissolved organic matter accompanied with organic phosphorus has not increased tot P concentrations in L. Iso Hietajärvi.

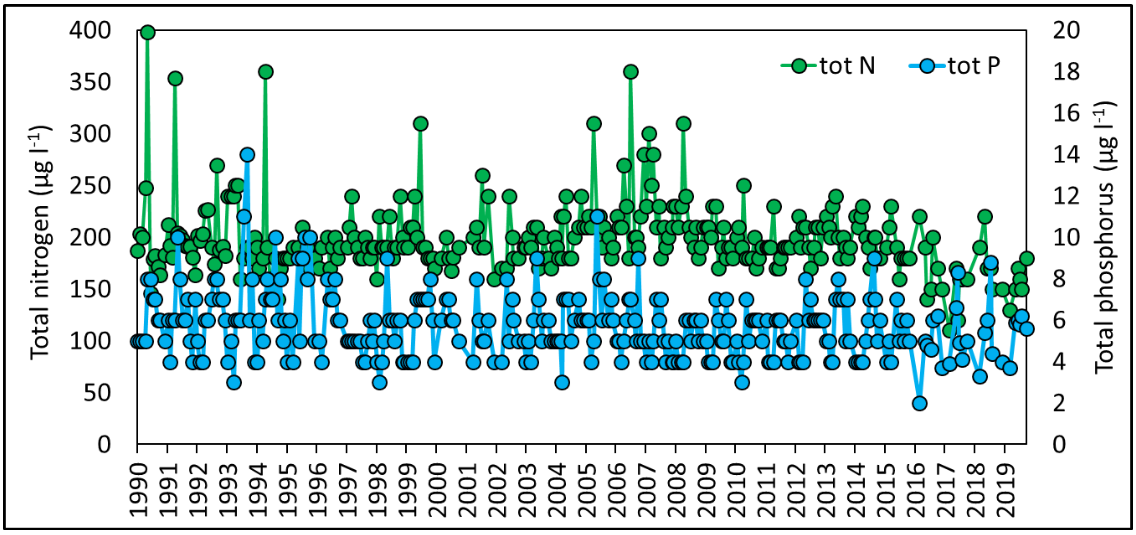

Figure 13. Time series for total nitrogen (tot N) and total phosphorus (tot P) concentrations at the depth of 1 m in the L. Iso Hietajärvi in 1990–2019.

3.6 Sulphur and nitrogen deposition and exceedances of critical loads

The acidity critical load function at Iso Hietajärvi site is determined by the values $CL_{max}N$=2960 eq ha^{-1} yr^{-1}, and $CL_{max}S$=653 eq ha^{-1} yr^{-1}, using the critical ANC concentration of 20 µeq l^{-1} (Forsius et al. 2021). The annual values of the observed ANC concentrations were plotted on the y-axis in a scatter diagram as a function of EX_{aci} on the x-axis (Fig. 14). As can be seen in the graph, the acidity critical load was not exceeded during the observation period, and the critical ANC concentration of 20 µeq l^{-1} was not, either, violated during the observation period.

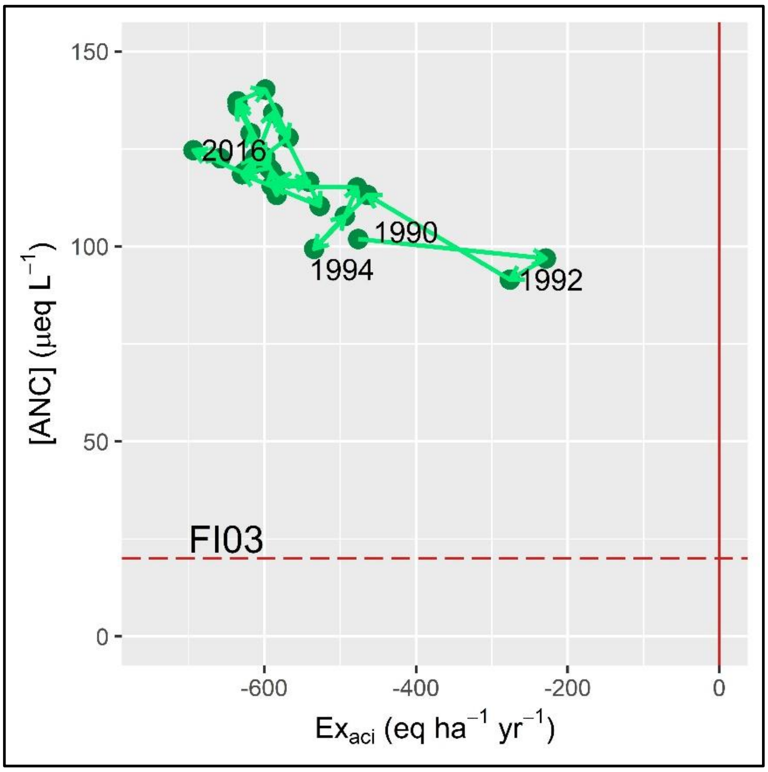

Figure 14. Hietajärvi (FI03) year-to-year variation in runoff ANC (µeq l^{-1}) plotted against EX_{aci}, the exceedance of critical loads of acidification (eq ha^{-1} yr^{-1}). The vertical solid line (Ex_{aci}=0) shows the division between non-exceedance (negative values of Ex_{aci}), and exceedance (positive values of Ex_{aci}). The horizontal dashed line represents the critical ANC concentration 20 µeq l^{-1}.

The empirical critical load of eutrophication at Hietajärvi was estimated to 5–8(10) kg N ha^{-1} yr^{-1}, using the range for *Picea* taiga woodland (G3.A) and mixed taiga woodland with *Betula* (G4.2) suggested by Bobbink et al. (2010) for habitats classified according to the EUNIS (European Nature Information System) habitat system for Europe (Davies et al. 2004). The mass balance critical load of eutrophication at Hietajärvi was determined as 5 kg N ha^{-1} yr^{-1}, equaling the minimum value of the empirical critical load, thus the CL of eutrophication at Hietajärvi is 5 kg N ha^{-1} yr^{-1}. The annual values of the observed TIN concentrations were plotted on the y-axis in a scatter diagram as a function of EX_{eut} on the x-axis (Fig. 15). The graph shows that the critical loads were exceeded in the beginning of the period 1990–2017. The observed TIN concentrations in runoff were, however, lower than the acceptable concentration of N 1.3 mg l^{-1} during the whole period.

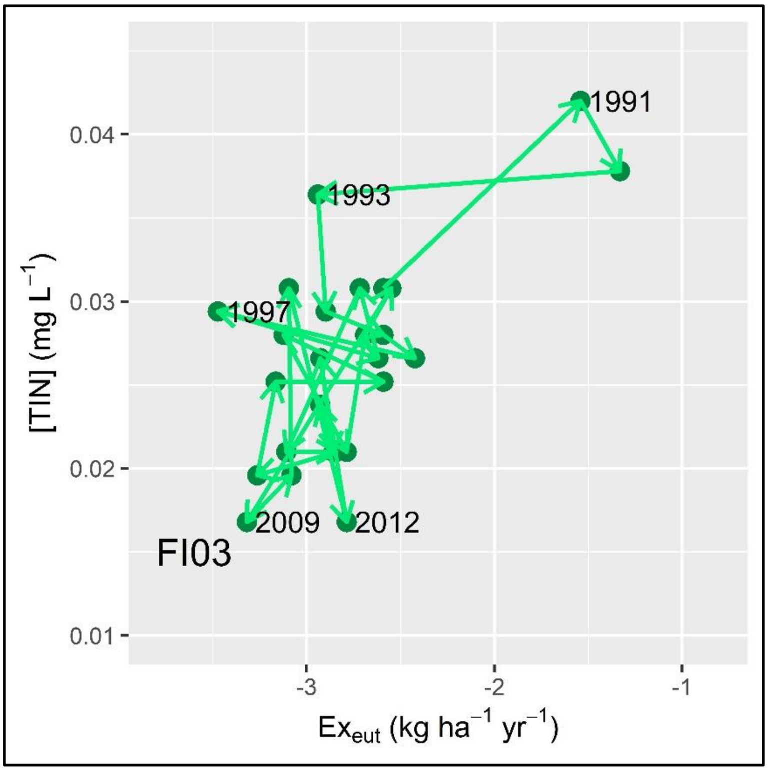

Figure 15. Hietajärvi (FI03) year-to-year variation in runoff TIN (mg l^{-1}) plotted against EX_{eut}, the exceedance of critical loads of eutrophication (kg ha^{-1} yr^{-1}). No exceedance occurred, and the vertical solid line (Ex_{eut}=0), representing the division between non-exceedance (negative values of Ex_{eut}), and exceedance (positive values of Ex_{eut}), falls outside the plot.

The critical concentrations (ANC 20 µeq l^{-1}, TIN 1.3 mg l^{-1}) were not violated any time during the observation period, and the ANC concentrations have increased (Table 4, Fig. 10) and the TIN concentrations decreased over the observation period (Table 6). For eutrophication at Hietajärvi site, there is a pattern that low TIN concentrations are coupled to low exceedance values (more negative Ex_{eut}).

3.7 Long-term changes in terrestrial part of the catchment

3.7.1 Element concentrations in bulk precipitation, throughfall, soil water, foliar, litterfall and stream water

In addition of atmospheric deposition, the terrestrial part of the ecosystem is important source of nutrients and DOC to recipient water bodies. Forest vegetation is an effective receptor of airborne material delivered in both wet and dry forms because of the reactivity and large surface area of the canopy (e.g. Kimmins 1987). In addition, forest floor, including the organic layer retains effectively deposition inputs. The soil solution also reflects the atmospheric inputs, but the influence is weaker due to various processes in the soil including weathering, ion exchange, adsorbtion/desorption, decomposition and immobilisation. Despite retention of atmospheric inputs to the biomass and soil, there is a leaching of elements from soil to the lake.

We studied dissolved organic carbon (DOC), inorganic nitrogen (NO_3-N and NH_4-N), total nitrogen (total N), sulphate sulphur (SO_4-S), base cations (Ca+Mg+K+Na), strong acid anions (SO_4, NO_3-N, Cl) and ANC (Acid Neutralizing Capacity: base cations–strong acid anions) concentrations from aquatic (BP=bulk precipitation, TF=throughfall, SW=soil water) and foliage (green needles and litterfall) samples from Hietajärvi forest area and compared the values to the stream water (output of L. Iso Hietajärvi). The mean annual element concentrations from 1989 to 2018 for BP and stream, 1990–2018 for TF and SW 20 cm and from 2001 SW 5 cm are shown in Table 7. In general element concentrations were lowest in BP and highest in SW either in depth of 5 or 20 cm. For example, the SO_4-S concentration in BP was lowest as expected, but its concentration increased when precipitation passed through forest canopy as TF, and peaked in SW at depth of 5 cm, in a deeper soil layer concentration again decreased. In stream water SO_4-S concentration was only little higher than in BP. Source of SO_4-S in stream water is from surrounding terrestrial area, especially wetland area. In Table 8 is presented nutrient concentrations of living pine needles (Scots pine, *Pinus Sylvestris*, 1995–2017) and litterfall (Scots pine, *Pinus sylvestris* and other fraction for 1999–2017, for birch, silver birch, *Betula pendula*, 2008–2017).

The average DOC concentration in the BP was lower than other aquatic samples, only 2.5 mg l^{-1}, but the concentration of DOC increased as it passed through the tree canopy, the extent of these changes is related to the composition and characteristics of the forest stand (Starr and Ukonmaanaho 2004). In boreal forests, the DOC concentration in TF varies typically between 2 and 24 mg l^{-1} depending on the season and on the forest site type (Starr and Ukonmaanaho 2004, Fröberg et al 2006, Wu et al. 2010, Ukonmaanaho et al. 2014). In Hietajärvi the DOC concentration in TF was on average 9.0 mg l^{-1}. The rainfall reaching the forest floor is further modified as it infiltrates and percolates through the soil. The greatest changes occur in the upper soil, reflecting the distribution and characteristics of the litter and soil organic matter (Aber and Melillo 1991, Starr and Ukonmaanaho 2004). The DOC concentration in soil water under organic layer was on average 41.5 mg l^{-1} being almost twice higher than in soil solution in depth of 20 cm, and over four times higher than DOC concentration in other solutions. Typically, in boreal forest soil solution DOC concentration lies in a range of 5 to 70 mg l^{-1} (Fröberg et al 2006, Lindroos et al. 2008, Ukonmaanaho et al. 2014). In the soils the main production of DOC takes place in the upper soil layers and it is controlled mainly by biological processes (decomposition of litter, humus, root exudates), implying that the DOC production is sensitive to changes in soil temperature and moisture (Michalzik and Matzner 1999, Kalbitz et al. 2000).

The highest concentrations of inorganic nitrogen (NO_3-N and NH_4-N) were in BP. The concentration of inorganic nitrogen decreased when passing through the canopy and soil due to the active uptake by the tree foliage and epiphytes and further in the soil due to the uptake by roots and microbial processes. In boreal forests nitrogen (N) limits the growth of trees and therefore N is taken up actively by vegetation as reported earlier (e.g. Mälkönen 1974, Mustajärvi et al. 2008). In stream water NO_3-N concentration was similar level as in SW, NH_4-N was little higher in SW than in stream water. The total nitrogen concentration was highest in SW and lowest in stream water, however, share of organic nitrogen was over 80 % in above-mentioned samples, but in TF 31% and in BP less than 15 %. In Finland typical organic nitrogen concentration in deposition has been on average 24% (Vuorenmaa et al. 2001). Organic nitrogen is important part of the soil organic matter; thus, it is probable that leached organic nitrogen from soil explains higher share of it in soil and stream water.

Sum of base cation concentrations were highest in stream water, however, when observing individual base cations, potassium (K) and calcium (Ca) concentrations were higher in SW 5 cm than stream water, excluding K, which concentration was highest in SW. Potassium is a very mobile nutrient, in

TF it is origin almost entirely from foliage (green needles), where K has mainly leached through cation exchange. In general, the enrichment of nutrients in TF is related to the washing off of dry deposition and leachates produced by the canopy, while in SW, the enrichment is related to ionic exchange reactions in the organic layer depending mainly on the quality of litter and humus, and in deeper layers dissolution and weathering processes (e.g Johnson 1992). The decrease in water volume, which takes place when water passes through the canopy and percolates through the soil layers, has an increasing effect on the concentrations. In addition to evaporation, root uptake and microbial immobilization have an influence on SW quality; the greatest impact occurs in the humus layer where most of the plant roots are growing (Gosz 1981).

Table 7. Mean solute concentrations and standard deviations (*sd*) in 1990–2018 in different aquatic media in forest (BP=bulk precipitation, TF=throughfall, SW=soil water, Stream=stream water from the outlet of L. Iso Hietajärvi). BP and stream water results since 1989, and SW 5 cm since 2000.

		BD	TF	SW 5 cm	SW 20 cm	Stream
mm (sum)		631	567			386
n		263-371	252-359	212-229	358-494	596-665
pH*		5.1	4.9	4.30	5.00	6.30
		0.26	*0.23*	*0.430*	*0.710*	*0.260*
DOC/TOC, mg l^{-1}	mean	2.5$^{\#}$	9.00	41.5$^{\#}$	25.0$^{\#}$	5.6$^{\#}$
	sd	*3.530*	*6.040*	*25.680*	*23.670*	*2.030*
Ca, mg l^{-1}	mean	0.15	0.35	1.37	0.88	1.31
	sd	*0.112*	*0.263*	*1.177*	*0.725*	*0.203*
Mg, mg l^{-1}	mean	0.04	0.11	0.27	0.21	0.34
	sd	*0.061*	*0.082*	*0.184*	*0.168*	*0.059*
K, mg l^{-1}	mean	0.11	0.60	1.05	0.76	0.43
	sd	*0.164*	*0.527*	*1.387*	*0.790*	*0.088*
Na, mg l^{-1}	mean	0.14	0.24	0.62	0.86	1.11
	sd	*0.083*	*0.124*	*0.303*	*0.425*	*0.153*
NO$_3$N, mg l^{-1}	mean	0.24	0.21	0.02$^{¤}$	0.02$^{¤}$	0.02
	sd	*0.129*	*0.176*	*0.006*	*0.003*	*0.029*
NH$_4$N, mg l^{-1}	mean	0.21	0.17	0.14	0.10	0.01
	sd	*0.230*	*0.238*	*0.283*	*0.197*	*0.013*
N$^{\$}$, mg l^{-1}	mean	0.52	0.55	0.90	0.78	0.21
	sd	*0.409*	*0.473*	*0.592*	*0.666*	*0.048*
SO$_4$S, mg l^{-1}	mean	0.38	0.50	3.35	1.17	0.45
	sd	*0.267*	*0.397*	*1.078*	*1.087*	*0.158*
Base cations$^{\%}$, µeq l^{-1}	mean	33.27	52.20	144.22	112.95	152.12
	sd	*24.792*	*31.862*	*86.684*	*62.821*	*22.275*
Strong acid anions$^{\%}$, µeq l^{-1}	mean	47.20	58.87	86.21	89.48	39.24
	sd	*30.108*	*42.245*	*70.934*	*71.437*	*12.443*
ANC$^{\%}$, µeq l^{-1}	mean	-13.05	-6.48	58.01	59.01	112.93
	sd	*22.593*	*36.014*	*77.243*	*78.966*	*23.187*

*only 2018, #BP 1998-2018, SW5 cm 2000-2018, SW20 cm 1991-2018, stream 1989-2018, $^{¤}$most of the values under detection limit

$^{\$}$measurements started in BP 1998, TF 1999, SW 5cm 2000, SW20 1994, stream 1989

$^{\%}$Base cations = (Ca+K+Mg+Na), strong acid anions = (NO$_3$+SO$_4$+ Cl), ANC = base cations-strong acid anions

We studied also nutrients of green pine needles, litterfall (LF) pine needles, LF birch leaves and other LF fractions over the study period (Table 8). In general, on average all nutrients, excluding calcium (Ca), were higher in current year needles than previous year needles and litterfall. However, LF birch leaves deviated from this pattern. When needles are senescing most mobile nutrients are transformed back to trunk or younger needles, therefore concentrations are usually lower in LF needles than in living needles. However, Ca was an exception, its concentration was higher in all LF fractions than green pine needles. Calcium continues to accumulate in foliage up to the time of senescence, and thus Ca concentration in litterfall usually exceeds foliar Ca content. This is because Ca is a major component of permanent plant tissues, such as pectates in cell wall. While, for example, K, which concentration was significantly higher in green needles than LF needles, is very mobile nutrient and it stays primarily in ionic form within plant, playing a major role in osmoregulation of stomatal opening and closing and therefore it is leaching easily from the living needles and due that its content is lower in LF needles (e.g. Johnson 1992), but high in TF as was mentioned in a previous chapter. There was also difference in concentrations between different LF fractions, usually nutrient concentrations of birch LF leaves were highest and pine LF needles lowest, which is in line with earlier studies (e.g. Johansson 1995). The other LF fraction is composed various components such as seeds, flowers, bark, small branches and animal faeces, which nutrient content may vary a lot, for example, in flowers and seeds nutrient concentrations are usually much higher than, in bark, which explains higher concentration in other LF fraction than LF pine needles fraction (Ukonmaanaho et al. 2020).

Table 8. Nutrient concentration of green needles (c=current year needles, c+1, one-year old needles) and litterfall (pine needles, birch leaves and other litterfall).

	Green pine needles		Litterfall		
	c	c+1	pine needles	birch leaves	other litterfall
n	25	25	128	18-22	128
Years	1995−2017	1995−2017	1999−2017	2008−2017	1999−2017
g m^{-2} year^{-1}			114.97	7.31	90.86
sd			*27.658*	*3.840*	*12.959*
C%	53.79	54.40	54.85	54.33	53.35
sd	*0.905*	*0.483*	*0.828*	*1.041*	*0.707*
N mg g^{-1}	11.43	11.51	6.61	8.85	7.97
sd	*1.070*	*0.841*	*1.816*	*2.701*	*1.609*
Ca mg g^{-1}	1.78	3.07	3.75	9.07	3.76
sd	*0.263*	*0.278*	*0.520*	*2.331*	*0.736*
Mg mg g^{-1}	0.99	0.87	0.59	2.16	0.42
sd	*0.100*	*0.091*	*0.099*	*1.068*	*0.176*
K mg g^{-1}	5.12	4.32	1.39	2.95	1.07
sd	*0.249*	*0.246*	*0.718*	*1.273*	*0.386*
S mg g^{-1}	0.79	0.81	0.53	0.59	0.59
sd	*0.060*	*0.060*	*0.088*	*0.190*	*0.078*
P mg g^{-1}	1.32	1.18	0.60	1.93	0.73
sd	*0.090*	*0.071*	*0.213*	*0.373*	*0.210*

3.7.2 Long-term temporal trends in bulk precipitation, throughfall, soil water, stream water, foliage and litterfall

We studied also long-term temporal trends in forest ecosystem at Hietajärvi. As a result of air quality controls, the SO_4-S deposition has considerably decreased throughout Europe and it can be seen also in Hietajärvi (Table 9, Figs. 9 and 16). The trend analyses showed a significant decrease in BP and TF SO_4-S concentrations and sulphur concentration in green needles and LF (Table 10, Fig 17) during the past decades. In addition, SW at depth of 5 cm and stream water had a decreasing trend, although not significant. Therefore, the reduced SO_2 emissions have not resulted in only reduced deposition loads to the forest canopy and litterfall, but the reduction has also taken place in the soil and in the stream water. Decreasing trend in SO_4-S concentrations obviously reflected also to strong acid anions, which had a significant decreasing trend in all aquatic samples, excluding soil water in mineral soil.

Table 9. Long-term temporal trends (SKT, Sen's slope) for bulk precipitation (BP), throughfall (TF), soil solution at the depth of 5 cm (SW 5 cm) and at depth of 20 cm (SW 20 cm) and stream water from outlet of L. Iso Hietajärvi (stream). Values in bold indicate statistically significant trend ($p < 0.05$).

	BP	TF	SW 5cm	SW 20cm	Stream
Period	1989-2018	1990-2018	2000-2018	1990-2018	1989-2018
Precipitation/runoff mm yr^{-1}	1.276	1.84	-	-	0.407
DOC/TOC mg l^{-1} yr^{-1}	-0.044	0.015	0.563	**0.818**	**0.08**
Ca, mg l^{-1} yr^{-1}	0.001	-0.001	0.028	0.005	<0.001
Mg, mg l^{-1} yr^{-1}	<0.001	<0.001	0.005	<0.001	<0.001
K, mg l^{-1} yr^{-1}	0.001	0.004	-0.041	-0.014	-0.001
Na, mg l^{-1} yr^{-1}	-0.001	-0.002	-0.003	-0.006	-<0.001
NO_3-N, mg l^{-1} yr^{-1}	**-0.004**	-0.002	$0^{\#}$	$0^{\#}$	-<0.001
NH_4-N, mg l^{-1} yr^{-1}	-0.002	**-0.004**	0.004	**0.007**	**-<0.001**
Total N, mg l^{-1} yr^{-1}	**-0.012**	**-0.013**	0.014	0.019	<0.001
SO_4-S, mg l^{-1} yr^{-1}	**-0.015**	**-0.022**	**-0.04**	0.005	**-0.012**
Base cations, µeq l^{-1} yr^{-1}	-0.103	-0.011	-0.116	-0.453	0.02
Strong acids, µeq l^{-1} yr^{-1}	**-1.344**	**-2.279**	-3.257	0.144	**-0.816**
ANC, µeq l^{-1} yr^{-1}	**1.231**	**2.286**	1.773	-0.413	**0.790**

$^{\#}$ Results have been mainly under instrument detection limit → no trend seen.

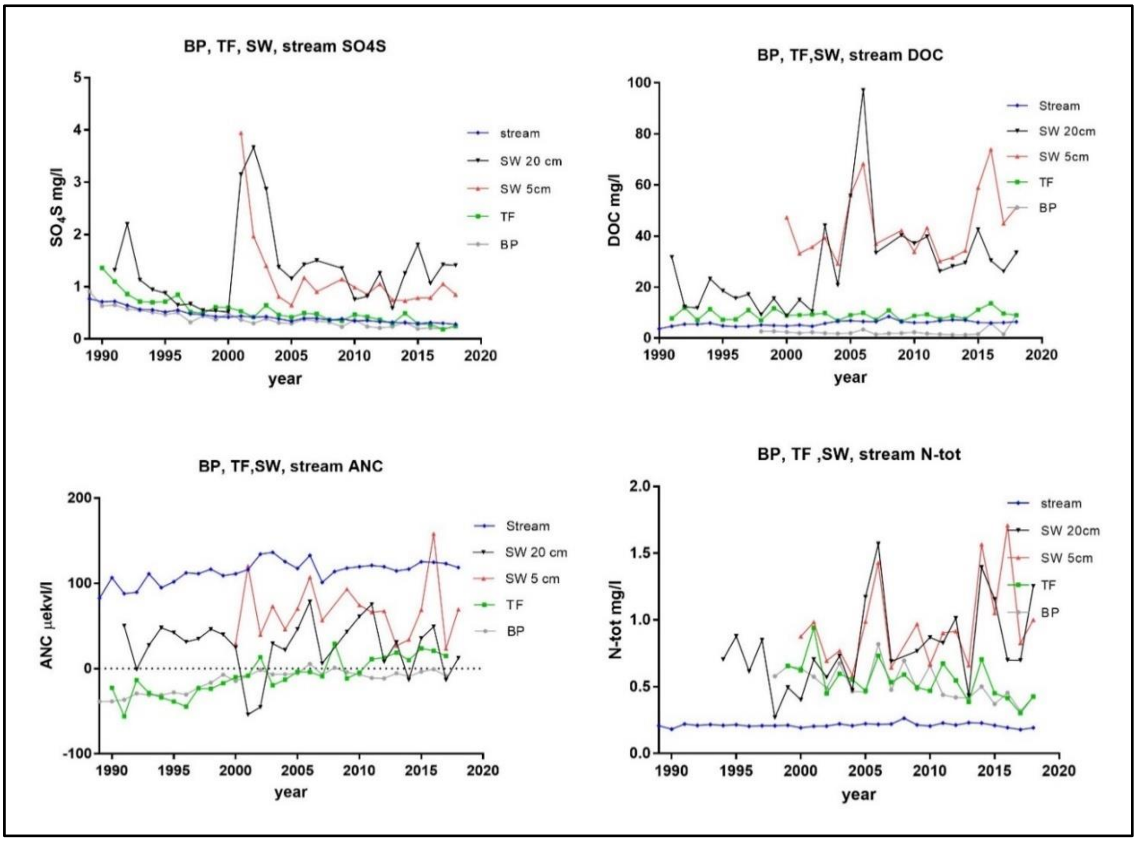

Fig 16. Time series for SO_4-S, DOC, ANC and total N in bulk precipitation (BP), throughfall (TF), soil water (SW) and stream water (at the outlet of L. Iso Hietajärvi) in 1990–2018.

Sum of base cation concentrations in BP were slightly decreasing, but not significantly. The main emission sources of base cations are soil dust, energy production, industrial processes, traffic and seawater. In Finland, main single anthropogenic sources are combustion of wood and dust raised by traffic (Ruoho-Airola et al. 2003). The slightly decreasing trend in BP is probably due that in 1990s emissions from the oil shale power plants in Estonia increased the base cation deposition in the South and Southeastern Finland earlier, but due to the application of dust removal technology, the recent base cation emissions of the region have decreased (Ruoho-Airola et al. 2003). All other aquatic solutions, except stream water also showed slightly decreasing trend in base cations, which probably in TF is reflect of decreased base cations concentration in BP, while in soil water, the decline may be related to the decline of SO_4-S concentrations. According to Singh et al. (1980), a reduction in the amount of SO_4 anions as required to accompany cations could lead to a reduction in base cation concentrations in the soil solution. In green needles and litterfall, there was no significant trends in base cations (Ca, Mg, K), except in Mg, which significantly decreased in pine LF needles. Slightly decreasing trend in Mg concentrations was also seen in green needles and other LF fractions, but in aquatic samples, for example, in BP and TF, there was no change. Probably there has been less uptake of Mg from soil, which is seen as decreased green needle and LF concentrations. Traditionally nutrient content of needles is used as an indicator of the tree vitality and nutrition level in soil, in addition indicator of air pollution. However, despite decreased Mg concentration in green needles, nutrient concentrations are in balance in living needles.

The combined effects of changes in SO_4-S, strong acids and base cation concentrations reflected as a significantly increasing trend in ANC value in the studied solutions, excluding SW at depth of 20 cm which had slightly decreasing trend. In view of the general decrease in BP, TF, SW 5 cm and stream water SO_4-S concentrations, the decrease in ANC in SW at the depth of 20 cm is somewhat contradictory. However, the slight decrease in base cation concentrations can be the main cause for the slightly decreasing trend in ANC in mineral soil layer and may be related, for example, to a natural succession of the forest ecosystem. When trees and other vegetation take up base cations from the soil, protons are released into the soil. In a mature natural forest ecosystem, the increase in acidity is neutralized by nutrients released through decomposition and mineralization of litter. However, it is possible that in an old forest, there can be an imbalance between base cation uptake and release from litter mineralization (Bérden et al. 1987).

In all water samples DOC concentration, showed significant or slightly increasing trend, excluding BP. Increased DOC concentration in SW indicates that decomposition of organic material has increased in soil. Probably, the increased temperature and precipitation due to climate change has reflected as a longer growing season, which in turn has led to a higher net primary production and a higher foliar litter production rate. Since the senescing foliar biomass is the primary source of DOC-producing substrate, a higher input rate of litter may reflect as a higher DOC production in soils (Fröberg et al. 2006). However, there was no significant increase in litterfall amount during the study period, only amount of pine LF needle fraction had a slightly positive trend. Presumably the biomass of ground vegetation has increased, and decaying material from there has increased decomposition and release of DOC. Actually, ground litter estimations indicated that, for example, in year 2003, when SW both in depth 5 cm and 20 cm started to increase, share of litter leaves was two to five times more than in other observed years (estimations have made every 5th year) (Table 13, in chapter 3.7.4.2).

In Hietajärvi region, inorganic nitrogen bulk deposition is about 2.1 kg ha^{-1} yr^{-1} and it is decreasing (unpublished results from Hietajärvi by Luke 2011-2017). We found a decreasing inorganic nitrogen trend in forest, excluding SW under the mineral soil (20 cm) (Table 9). Trend is obviously related to the slightly decreased inorganic nitrogen deposition trend. In Finland inorganic nitrogen deposition has not been critical high during the study period, although study period started already in beginning of the 1990s, when elevated nitrogen concentrations were common and still are, for example, in central Europe. Similar decreasing trend in N concentration was also seen in litterfall (both foliar and other fraction) but not in green needles (Table 10). In contrast green needles had a significant positive trend. Plants take up part of the nitrogen directly through precipitation, therefore decreased concentration in green needles would have been more expected due to the decreased deposition. However, it is possible that earlier higher N deposition is still in internal N flux of forest ecosystem and can be expressed as elevated concentration of needles. In N limited boreal forests, biological fixation of N_2 gas by bryophyte- and lichen-associated cyanobacteria on forest floor is also an important source of plant-available N (DeLuca et al. 2002, Salemaa et al. 2019). Further, warm growing seasons may have accelerated mineralization of nutrients from the organic matter resulting in increased plant availability of N and consequently, in higher N concentration of needles. The same phenomenon has also been observed on other ICP Forests Level II plots in Finland (unpubl.results). In soil solution inorganic nitrogen concentrations were often under instrument detection limit, thus results are only indicative. Similarly decreasing trend was also seen in total nitrogen concentration in different solutions, but significant only in BP and TF which is a sign that N emission have decreased during study period.

Table 10. Long-term temporal trends (SKT, Sen’s slope) for green needles (C and c+1 years) and litterfall (needles and other litterfall fraction). Values in bold indicate statistically significant trends ($p < 0.05$). Green needles from period 1995–2017, litterfall from 1999–2017, except birch litterfall 2008–2017.

	Green needles		Litterfall		
	c	c+1	pine litter	birch litter	other LF
Sum of dry weight g m^{-2} yr^{-1}			0.750	-0.027	-0.264
N, mg g^{-1} yr^{-1}	**0.246**	**0.244**	-0.006	**-0.763**	-0.021
S, mg g^{-1} yr^{-1}	-0.001	-0.003	0.000	-0.033	0.001
P, mg g^{-1} yr^{-1}	0.006	0.002	0.006	-0.007	0.009
Ca, mg g^{-1} yr^{1}	0.003	-0.003	0.016	0.285	0.022
Mg, mg g^{-1} yr^{-1}	-0.003	-0.005	**-0.009**	0.139	-0.004
K, mg g^{-1} yr^{-1}	0.005	-0.010	-0.033	0.093	**-0.023**
C, % dw. yr^{-1}	-0.125	-0.133	0.033	-0.094	**0.002**

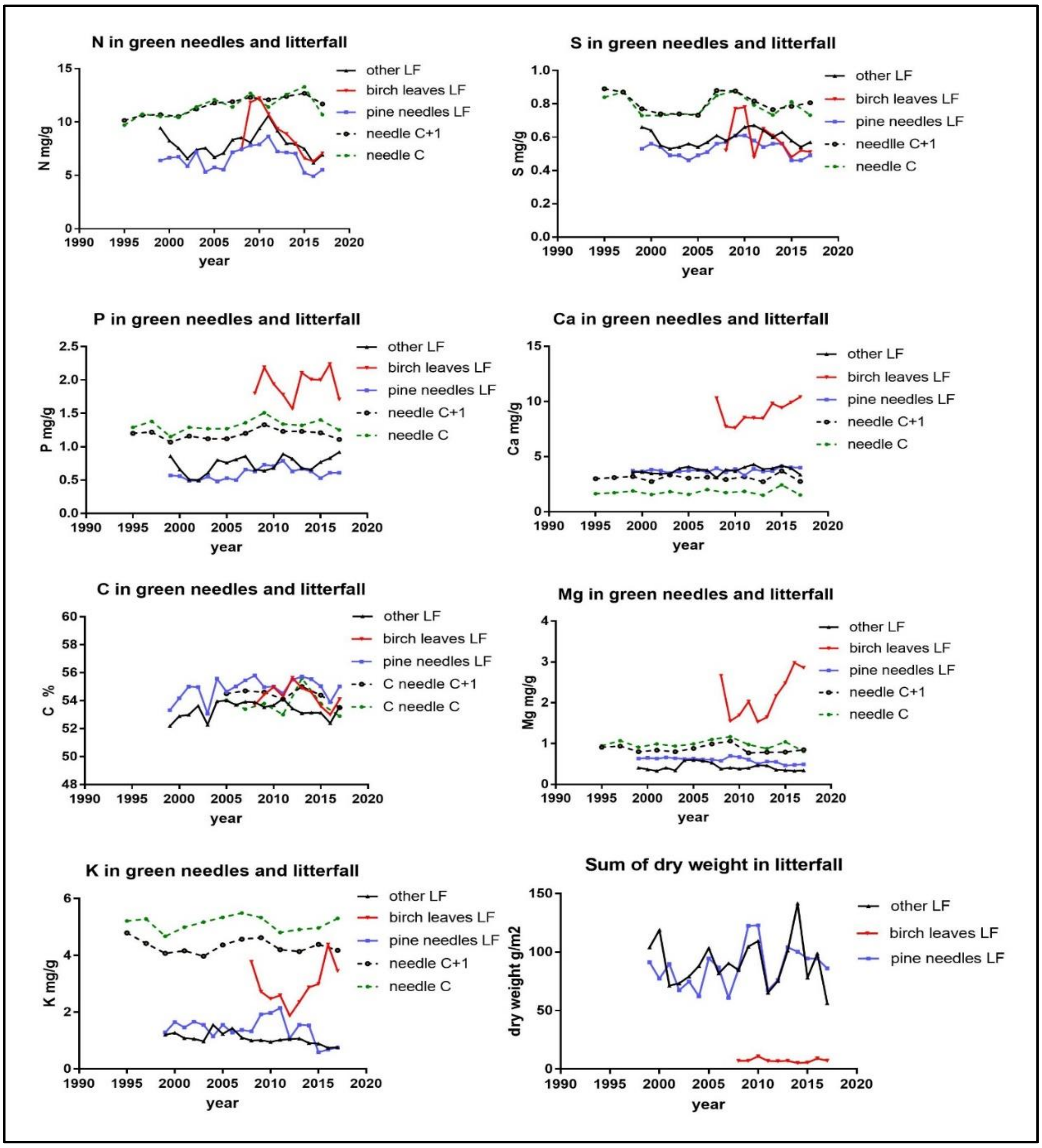

Figure 17. Time series for N, S, P, Ca, C%, Mg and K in green needles (c and c+1, 1995–2017) and litterfall (pine needles and other LF fraction, 1999–2017 and birch leaves 2008–2017). Dry weight trend only in litterfall.

3.7.3 Do extreme weather events change DOC export from terrestrial to aquatic ecosystems?

In addition to general increase of air temperature and precipitation and consequent increase of temperature in fresh waters, climate change has predicted to increase extreme weather events in northern countries. Extreme events are defined as episodes or occurrences of a statistically rare and/or unusual weather pattern that alters ecosystem beyond the limits of typical of normal variability (Smith 2011). Due to the extreme weather events key processes that regulate production and transport of the solutions, such as dissolved organic carbon (DOC) from terrestrial area to streams and lakes can be potentially altered, as have extreme rain events shown to increase DOC export from peatland and forest soils (Dinsmore et al. 2013, Intergovernmental Panel on Climate Change (IPCC) 2014).

In Hietajärvi area the annual precipitation sum was an average 617 mm and mean temperature +2.7 °C (Table 11). Annual mean air temperatures and precipitation sums were derived using a model by Venäläinen et al. (2005). We identified the years from the study period 1990–2018, when annual temperature was over or less than average as well as years when annual precipitation sum was lower or greater than average. Three most warm, cold, dry and wet years where chosen (Table 11).

Table 11. Identified cold, warm, dry and wet years at Hietajärvi region. Precipitation and temperature have calculated using a model by Venäläinen et al. (2005).

	Year	Annual average temp.	Annual precipitation sum
		°C	mm
1990–2018		2.7	617
Cold years	1998	1.27	
	2010	1.36	
	1993	1.36	
Warm years	2015	4.21	
	2014	3.89	
	2013	3.82	
Dry years	1999		368
	2001		419
	2000		439
Wet years	2012		846
	2008		844
	2004		842

In addition, we chose three years when DOC concentrations in TF and SW 20cm and TOC concentration in stream water was highest (D/TOC+) or lowest (T/DOC-). TF DOC concentration in DOC+ years was 30 to 51% more than on average and 22 to 27 % less than on average in DOC- years, correspondingly at stream water in TOC+ years was 25 to 51% higher than on average and in TOC- years 20 to 34% lower than on average. In SW differences were greater, in SW 20 cm DOC+ concentration was nearly 300% more in 2008, which was also one of the years 2008, when precipitation sum peaked (Fig.18). Although we found that some max/min temp/precipitation years coincided with max/min aquatic DOC concentrations, we did not identify any clear pattern between DOC concentrations and temperature/precipitation. In addition, there was no strong correlation between temperature or precipitation and DOC concentrations in different water samples. We also looked single extreme weather events such as ’Unto storm 5.7.2002’, ‘Asta storm 30.7.2010’ and ‘Elviira storm 27.6.2013’, but we did not see clear ‘storm’ effect on concentrations, neither after very rainy July in 2004 (153 mm) and in 2012 (161 mm). Therefore, it looks like that so far, the extreme weather events, which are expected to be more frequent in a future, have not had very strong impact on the study area. However, we should keep in mind that study area is a pristine, protected area, where forestry has not been practiced for decades, therefore the forest structure is diverse compared to managed forest and biodiversity in vegetation and other biota has remained, all these characteristics are supposed to increase resilience in fight against the effects of extreme weather events.

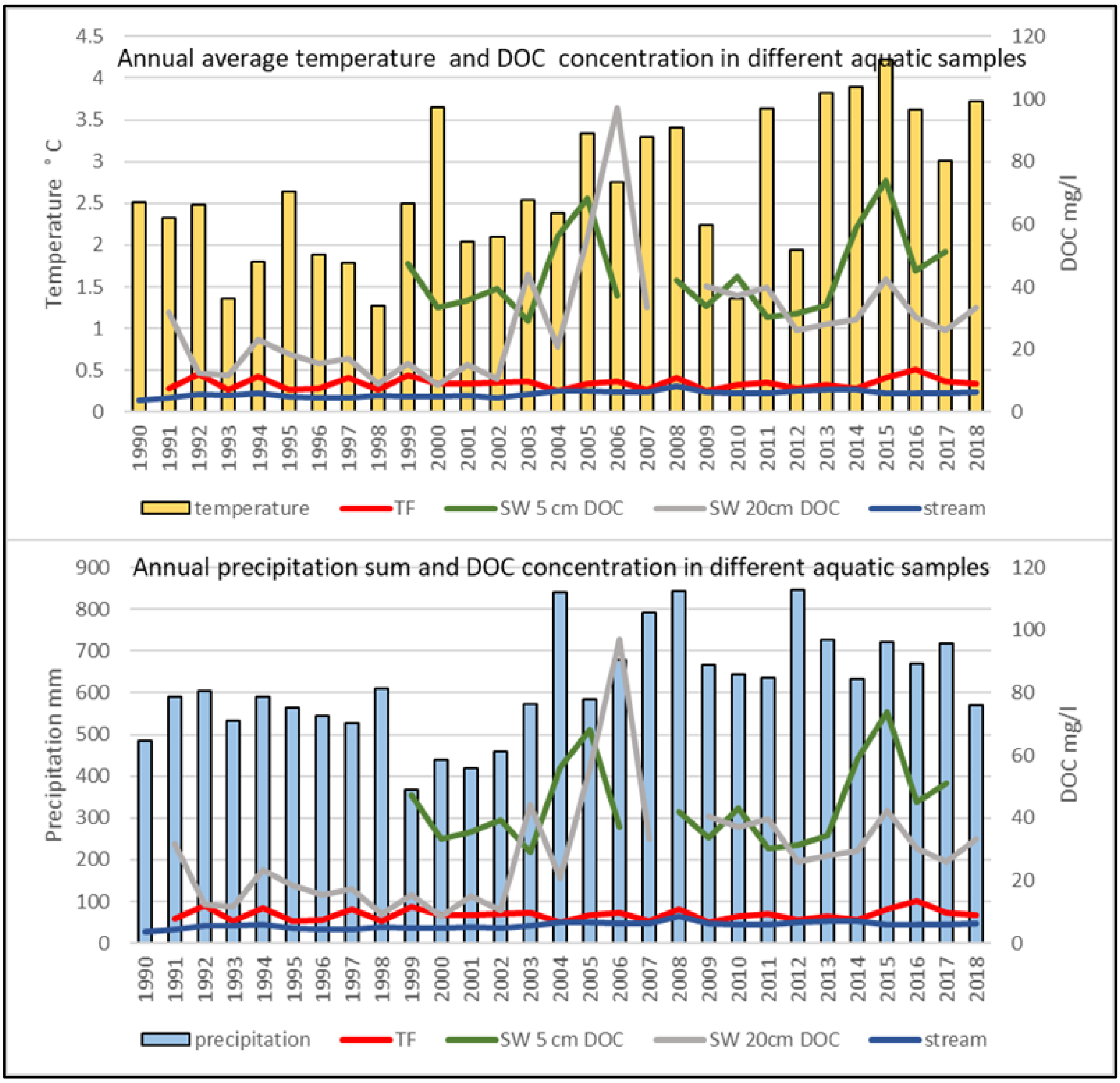

Fig. 18. Annual average temperature sum and average DOC concentrations in water samples (up), and annual precipitation sum and average DOC concentrations in different water samples (down).

3.7.4 Long-term changes in vegetation

3.7.4.1 Stand characteristics

The Hietajärvi forest monitoring site characterizes an old-growth pine (*Pinus sylvestris*) dominated *Empetrum-Vaccinium* (EVT) forest type of middle boreal zone. In addition to pine (93% from the total living volume) some birches (*Betula pendula* and *B. pubescens*, 7%) grew as mixture in the subplot 1 (Table 12). In the other subplots, some spruces were present, but in general, the whole study site represents very homogenous dryish pine heath forest. Although the study site located at a protected area, the amount of standing dead wood was very low (< 0.1 – 5.8 m^3 ha^{-1} in different subplots) in 2009. There were some lying dead stems and stumps on the ground (Fig. 19), but their volume has not been measured.

The eldest pines were over 130 years-old (calculated from annual rings of stem discs of cut sample trees), but birches represented younger age-class. As a habitat for understorey vegetation, the stand was sparse with high light level on the forest floor. According to visual estimation of crowns carried out by botanists, the canopy cover was 27% in 2014 and 21% in 2019. This indicates that the light level has slightly increased probably due to defoliation of crowns of old pines (Figs. 4 and 19).

Table 12. Stand parameters in the sub-plots no 1–3 at Hietajärvi study site in 2009. Basal area and volume have been measured with bark.

Parameter Subplot 1	Number of stems, ha^{-1}	Basal area, m^{-2} ha^{-1}	Volume, m^{-3} ha^{-1}	Mean diameter, cm	Mean height, m	Height of lower crown limit, m
Living stock, tot	450	22.3	244.1	32.7	24.1	10.8
Pine	381	20.3	226.7	33.9	24.6	11.0
Birch	69	2.0	17.3	22.0	19.1	9.7
Dead pines	6		0.01	3.1	3.7	
Subplot 2						
Living stock, tot	468	27.1	292.8	34.3	23.7	11.7
Pine	425	26.3	285.5	34.9	23.9	11.6
Spruce	12	0.1	0.3	10.0	7.9	12.3
Birch	31	0.8	7.1	19.5	18.8	0.9
Dead pines	6	0.3	3.1	24.5	22.2	
Subplot 3						
Living stock, tot	352	23.6	249.3	33.2	23.2	13.3
Pine	279	21.8	233.8	34.4	23.6	14.6
Birch	73	1.9	15.4	19.7	18.2	8.0
Dead pines	8	0.5	5.2	28.5	22.7	
Dead birches	4	0.1	0.6	15.7	15.6	

Figure 19. An overview of the Hietajärvi monitoring plot with some dead tree trunks and stumps in 2009. Leena Hamberg doing vegetation assessment of a sample quadrat in front (right). Cover of tree crowns in the Hietajärvi monitoring plot in 2014 (left). Photo: Maija Salemaa.

3.7.4.2 Number and cover of plant species

A total of 27 plant species (including 8 lichen species) was found in the field and bottom layers in the Hietajärvi subplot no 1 during the whole monitoring period 1998–2019. (Table 13). The shrub layer was scanty (cover 0.6%), and it consisted of only two species (pine and aspen - *Populus tremula*). The total number of vascular plant species in the field layer was 10 including 4 dwarf shrub, 4 tree and 2 herb species. No grass species grew in the subplot. The species number was 9 in bryophytes and 8 in lichens.

The most abundant species in the field layer was cowberry (*Vaccinium vitis-idaea*, its cover varied from 26 to 37%), followed by bilberry (*Vaccinium myrtillus*) with an average cover of 23%, heather (*Calluna vulgaris*, 6%) and black crowberry (*Empetrum nigrum*, 1 %). The herb group was formed only by *Melampyrum pratense*. Its cover varied from 0.1 to 4.5% in different years. *Pyrola chlorantha* was sporadic and found once.

The bottom layer was formed mainly by tall feather mosses (*Pleurozium schrebens*, *Hylocomium splendens* and *Ptilium crista-castrensis*). They covered almost completely (90%) the forest floor. Bryophytes in *Dicranum* family and lichens in *Cladina* (reindeer lichens), *Cladonia* (cup lichens), *Cetraria* and *Peltigera* families were present only in small patches (cover < 1%).

Table 13. The mean percentage cover (%) and number of plant and lichen species and ground litter in five different inventories (years 1998, 2003, 2009, 2014 and 2019) and the species occurrence over the years. *=species outside the sample quadrats but inside the area of 900 m^2. The mean cover values lower than 0.1 have been transformed to 0.1.

Species groups	**1998**	**2003**	**2009**	**2014**	**2019**	**Present**
Woody species, height < 50 cm						
Picea abies	0.00	<0.01	0.00	0.00	0.00	1
Pinus sylvestris	0.01	0.04	0.01	0.00	<0.01	1
Populus tremula	0.19	0.12	0.14	0.00	0.00	1
Salix caprea			*			1
Calluna vulgaris	4.85	6.05	6.44	6.87	5.06	1
Empetrum nigrum	0.61	0.86	0.44	0.56	1.03	1
Vaccinium myrtillus	22.56	24.44	23.62	24.06	23.69	1
Vaccinium vitis-idaea	25.62	30.75	34.94	36.87	32.37	1
Total cover	53.84	62.26	65.59	68.37	62.15	
Number of species	6	7	6	4	5	8
Herbs						
Melampyrum pratense	3.26	0.07	4.50	0.99	4.44	1
Pyrola chlorantha	<0.01	0.00	0.00	0.00	0.00	1
Total cover	3.26	0.07	4.50	0.99	4.44	
Number of species	2	1	1	1	1	2
Bryophytes						
Dicranum drummondii	0.00	0.03	0.00	0.00	0.00	1.
Dicranum fuscencens	1.23	1.32	0.33	0.05	0.66	1
Dicranum majus	0.00	0.14	0.31	0.58	0.88	1
Dicranum polysetum	2.9	2.42	3.16	3.18	3.5	1
Dicranum scoparium	0.01	0.27	0.07	0.03	0.24	1
Hylocomium splendens	17.51	19.03	21.94	25.84	15.94	1
Pleurozium schrebens	58.37	57.06	44.00	44.50	47.69	1
Pohlia nutans		*	*	*		1
Ptilium crista-castrensis	16.25	12.87	23.31	21.56	19.37	1
Total cover	96.28	93.16	93.12	95.74	88.31	
Number of species	6	8	7	7	7	9
Lichens						
Cetraria islandica	0.00	0.02	0.00	0.00	0.01	1
Cladina arbuscula	0.14	0.02	0.12	0.03	0.04	1
Cladina rangiferina	1.04	0.42	0.11	0.04	0.11	1
Cladina stellaris	0.01	0.00	0.00	0.00	0.00	1
Cladonia fimbriata					*	1
Cladonia sulphurina					*	1
Cladonia gracilis ssp. turbinata					*	1
Peltigera aphthosa	0.09	0.16	0.37	0.37	0.44	1
Total cover	1.28	0.63	0.61	0.44	0.59	
Number of species	4	4	3	3	4	8
All species						
Number of vasculars	8	8	7	5	6	10
Number of cryptogams	10	12	10	12	13	17

Ground litter	1998	2003	2009	2014	2019
Bark	-	1.9	2.4	1.8	2.3
Needles	1.5	2.5	1.1	1.4	2.6
Leaves	4.4	10.8	1.5	1.7	2.9
Dead plants	1.2	1.5	1.1	1.1	0.7
Dead wood diam.> 2cm	0.5	0.4	0.2	0.1	0.3
Dead wood diam. < 2 cm	4.6	3.6	1.8	1.1	2.4
Cones	0.1	0.1	0.1	0.1	0.1
Laying stems	0.0	0.1	1.2	2.5	1.9

3.7.4.3 Changes in species number and cover

The total percentage cover of the understorey vegetation stayed very stable in the Hietajärvi monitoring plot during the period 1998–2019 (Figs. 20a and 21). The temporal species turnover was very low in all species groups, which is typical for old succession stages of boreal forests. The cover of cowberry (*Vaccinium vitis-idaea*) increased approximately 8–10% during 2003–2019 (Fig. 20 b,c, Table 13). The cover of *V. myrtillus,* instead, did not change. The herb *Melampyrum pratense* showed yearly variation in occurrence.

Although the time series was short (21 years) and it consisted of only 5 study points, it seems that climatic factors regulated the abundance relationships of the plant species. There was a slight covariation in the cover% of bilberry (Pearson r=0.85, *p*=0.08) and cowberry (r=0.83, *p*=0.08) with the annual temperature sum (Fig. 22 a,b). When the temperature sum was high, the cover of dwarf shrubs increased. In the years with high precipitation sum, the temperature sum was low, and this kind of weather correlated with lower cover of bilberry. Furthermore, the lower the annual average minimum temperature was, the lower was the cover of bilberry (r=0.78, *p*=0.12) and cowberry (r= 0.84, *p*=0.07). On the other hand, *Melampyrum pratense* had the highest cover in the rainy years with low temperature sum.

The distribution areas of many boreal plant species shift northwards in the future due to increasing temperature according to modelling scenarios (Villèn-Perèz et al. 2020). Cowberry was among the dwarf shrub species, which have gained more cover right above the tree line on a northern fjäll (Vuorinen et al. 2017). It is possible, that cowberry benefits of the warmer climate in the future, because it resists draught. However, as a fast-growing competitor, cowberry can suppress the spreading of other plant species. For instance, the cover of a bryophyte species *Pleurozium schreberi* correlated negatively with the cover of cowberry in this data (r =–0.90, *p*=0.04). The cover of *Ptilium crista-castrensis* decreased with increase of the leaf litter of birches laying on the forest floor (r = –0.92, *p*=0.02). So, many natural factors, which are intermingled with the climatic factors, regulate the cover changes of plant species in boreal forests.

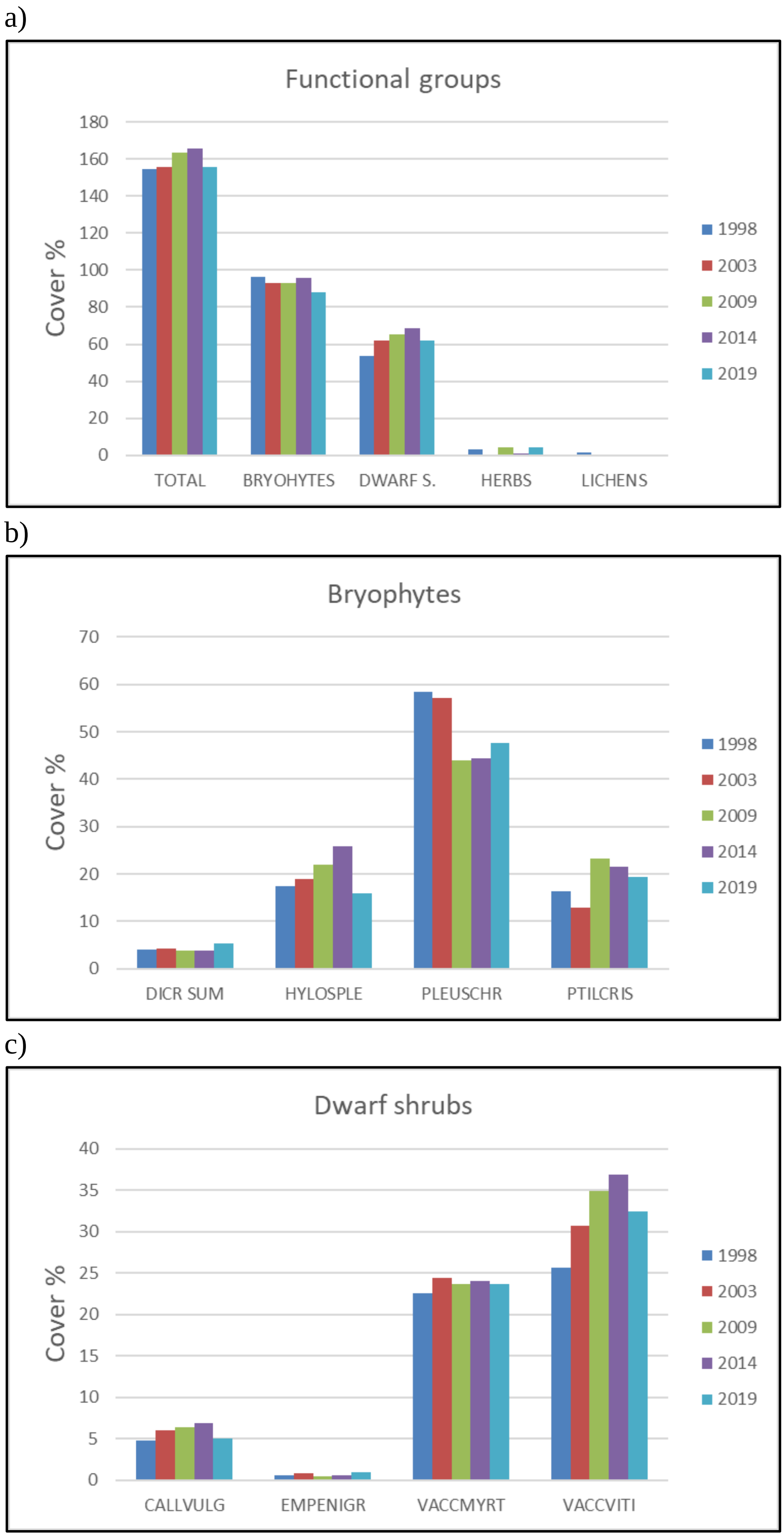

Figure 20. The cover (%) of understorey vegetation and its species groups in the Hietajärvi plot during 1998–2019. a) sum of all species (TOTAL), and separately bryophytes, dwarf shrubs, herbs and lichens, b) bryophyte species *Dicranum* spp. (DICR SUM) (mostly *D. fuscescens* and *D. polysetum*, Table 13), *Hylocomium splendens* (HYLOSPLE), *Pleurozium schreberi* (PLEUSCHR) and and *Ptilium crista-castrensis* (PTILCRIS), c) dwarf shrubs *Calluna vulgaris* (CALLVULG), *Empetrum nigrum* (EMPENIGR), *Vaccinium myrtillus* (VACCMYRT) and *V. vitis-idaea* (VACCVITI).

Figure 21. Characteristic understorey vegetation in the Hietajärvi forest monitoring plot: cowberry, bilberry, black crowberry in the field layer and *Ptilium crista-castrensis* in the bottom layer (left), and cowberry, bilberry and *Melampyrum pratense* in the field layer and *Pleurozium schreberi* and *Dicranum* spp. in the bottom layer (right).

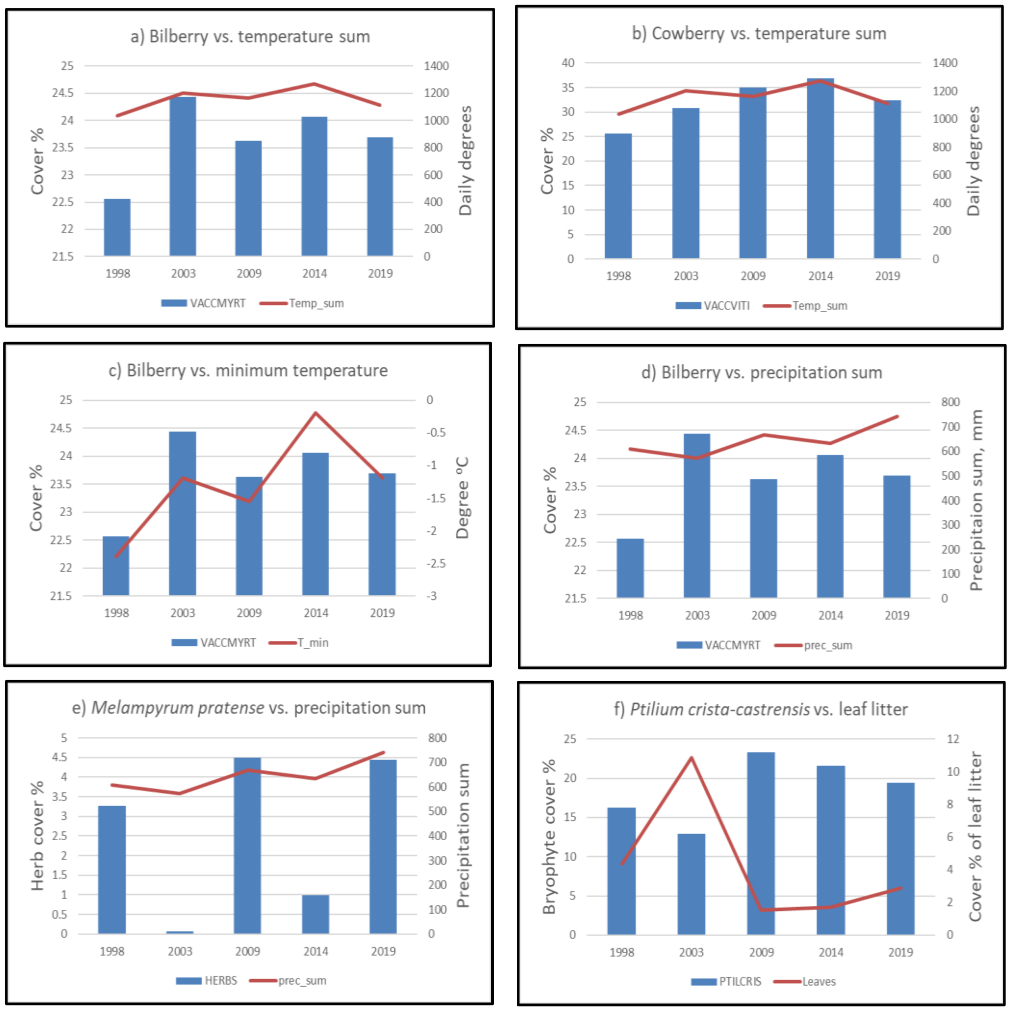

Figure 22. The relationship between the cover (%) of a) bilberry and b) cowberry with the annual temperature sum (daily degrees). The relationship between the cover (%) of bilberry and c) the average lowest temperature of months and d) annual precipitation sum. The relationship of e) the cover of *Melampyrum pratense* and precipitation sum and f) the cover of *Ptilium crista-castrensis* and the leaf litter of birches.

3.7.4.4 Nitrogen deposition and nitrogen concentration of plants

The total nitrogen (N) deposition was at the same level in bulk precipitation (BP) and throughfall (TF) in the Hietajärvi study plot in the year 2009 (Table 14). The total inorganic N deposition (TIN=NH_4-N+NO_3-N) was slightly higher in BP than in TF indicating tree crowns (or their epiphytes) uptake N when it passes through the canopy. On the other hand, dissolved organic N (DON) deposition was higher in TF than in BP indicating that DON was leached from needles or epiphytic organisms.

Table 14. Annual average precipitation (mm yr^{-1}) and different forms of nitrogen (N) deposition (kg ha^{-1} yr^{-1}) in the open site and in the forest of the Hietajärvi plot in the year 2009.

Habitat	Collection	Precipitation	NH_4-N	NO_3-N	DON	TIN	N_{tot}
Open site	BP	567	0.99	0.99	0.55	1.98	2.53
Forest	TF	627	0.72	0.99	0.69	1.71	2.40

In general, the N concentration of all bryophyte species as well as evergreen dwarf shrubs were relatively low (Table 15). Bryophytes uptake N mainly from precipitation through their surfaces, which makes them competent bioindicators of N deposition, although part of the N in bryophyte tissues probably comes from N leachate of litter in forest habitat (Salemaa et al. 2020). The total N concentration of the upper part of the bryophyte species in 2009 were as follows: *Hylocomium splendens* 0.87%, *Pleurozium schreberi* 1.02%, *Dicranum* spp. 1.13% and other bryophyte species 1.04%. In the lichen species (*Cetraria islandica* and *Cladina* spp.) the average N concentration was 0.4%. These N concentrations were so low, that it can be concluded that bryophytes had not reached the N saturated stage in their tissues. In fact, according to earlier studies TF deposition level should be over 3.3 kg ha^{-1} and bryophyte N concentration over 1.4% to cause inhibition of N_2 fixation by cyanobacteria living on *H. splendens* and *P. schreberi* (Salemaa et al. 2019). The corresponding values of these variables were lower in the Hietajärvi data, indicating that bryophyte growth was here N limited. Similarly, the N concentration of dwarf shrubs was low (mostly < 1 %). Only leaves of bilberry and *Melampyrum pratense* had little higher concentration, 1.4 and 1.9%, respectively. All these biological indicators suggest that the N deposition level in the Hietajärvi forest area is under the critical N load.

Table 15. Biomass weighted averages of N and C concentrations (%) in different plant groups and plant parts. Data is based on 28 samples of aboveground biomass on 30 cm × 30 cm quadrats. Evergreen dwarf shrubs were *Calluna vulgaris*, *Empetrum nigrum* and *V. vitis-idaea*. Bryophyte concentrations from the upper living part of thalli.

Hietajärvi forest	N	C
	%	%
Bryophytes	1.0	48.5
Lichens	0.4	45.3
Herbs	1.9	48.7
V. myrtillus leaves	1.4	52.9
V. myrtillus stems	0.8	52.4
Evergreen dwarf shrubs leaves	0.8	53.4
Evergreen dwarf shrubs stems	0.6	52.1

4. Conclusions

Lake Iso Hietajärvi demonstration site is a pristine, sensitive Natura 2000 area in Eastern Finland. The catchment is located inside a conservation area, and therefore is not affected by direct human disturbance. Our results verify that even remote pristine ecosystems, such as protected Natura areas, are susceptible to harmful environmental changes due to global pressures. On the other hand, the ecosystems have resilience to recover, if impacts of global change drivers are decreasing.

The international emission abatement actions for air pollutants have led to a recovery from acidification, and to a lesser extent, a decrease in trace metal loadings. However, processes regulating sulphur retention and release in the catchment are still not fully understood. The increase in dissolved organic matter and organic carbon concentration and consequent brownification of lake waters may have large ecological impacts on lake ecosystems and changing carbon dynamics in the lakes is one the key challenges in the future. These processes – net release of sulphate and browning of lakes – were driven by changed acid deposition, but climate-driven changes in hydrological conditions are becoming increasingly important, as atmospheric SO_4 input has declined. Another challenge is the enrichment in nutrients due to changing in-lake processes and climate-driven conditions, which may play an important role in affecting the processes in pristine lakes.

The critical loads of acidity and eutrophication were not exceeded at the catchment area of Lake Iso Hietajärvi during the observation period. In concert with decreasing eutrophication critical loads, also the inorganic nitrogen concentrations have decreased.

Precipitation was strongly modified before it entered from the terrestrial part of the L. Iso Hietajärvi catchment to the surface water. For example, when precipitation passed through the canopy to the soil, inorganic nitrogen concentration decreased due to the uptake of tree canopy, while base cation concentrations increased due to leaching and wash-off nutrients from the canopy. Long-term nutrient concentration trends confirmed that observed decrease in SO_4-S deposition was seen also in different parts of the forest ecosystem. Instead, increase in DOC concentration as has been observed in surface waters throughout Europe and North America, was not noticeable in the forest part of the Hietajärvi region, only DOC concentration in SW 20 cm increased significantly over studied period. We could not find very clear effects of extreme weather events to the forest ecosystem, which probably indicates the resilience of the pristine Hietajärvi region.

Over 20 years monitoring has produced new information of the succession of undisturbed old-growth forest in the Hietajärvi area. The cover % of cowberry (*Vaccinium vitis-idaea*) showed an increasing trend during the study period 1998–2019, and there was a slight covariation in the cover % and the annual temperature sum. On the other hand, the cover % of bilberry has changed only little. Because many natural factors are intermingled with the climatic factors regulating the cover changes of plant species in boreal forests, it is very difficult to draw conclusions whether climate change already has affected the understorey vegetation. However, if cowberry intensify its biomass production due to increased temperature, this may cause changes in the other plant community and affect even nutrient fluxes in the forest ecosystem. The low N deposition and low N concentrations of bryophytes and vascular species indicated that biomass production of terrestrial plants was N limited and the deposition level was under the critical N load in the studied forest ecosystem.

Our assessment strongly emphasizes the importance of the integrated aquatic and terrestrial long-term monitoring on the effects of air pollution, climate change and their interactions, due to the complex processes involved. Ecological monitoring under international agreements and legislation, such as United Nations Economic Commission for Europe Convention on Long-Range Transboundary Air Pollution (UNECE CLRTAP) and National Emission Ceilings Directive (NECD), are key activities set up to evaluate the effects, not only of emission reduction policies, but also of the changing climate.

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