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

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# 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), and 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 assess the vulnerability and ecosystem impacts of global change drivers at selected sensitive Natura 2000 site in South 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 Valkea-Kotinen 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 intensively monitored reference site Lake Valkea-Kotinen and its catchment (Fig. 1) (located in the catchment area of the FRESHABIT Vanajavesi target area), which are located in protected Natura 2000 area (Evo nature reserve) 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) and ICP Forests (International Cooperative Programme on Assessment and Monitoring of Air Pollution Effects on Forests, http://icp-forests.net/ ) networks aimed at monitoring and detecting long-term impacts of air pollutants and climate change. Intensive 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).

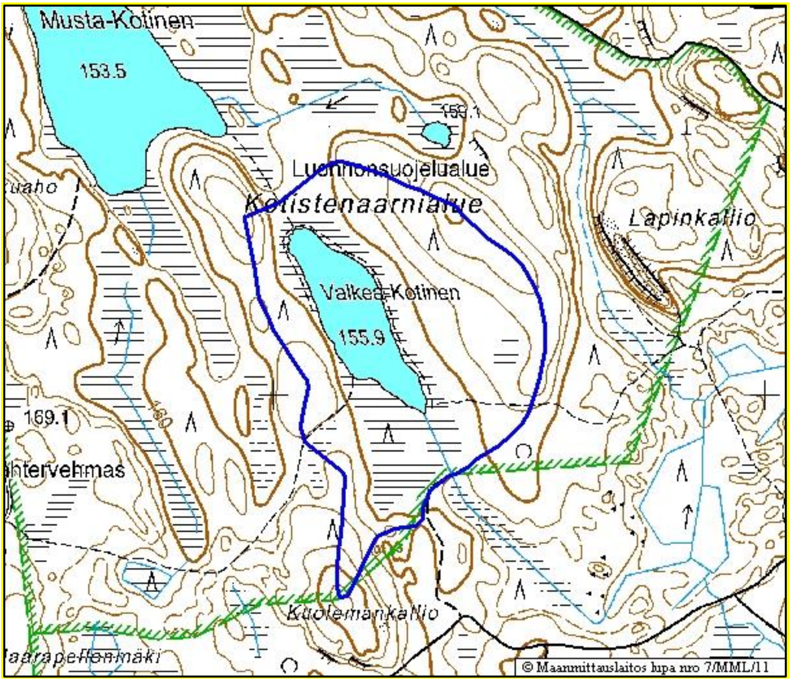

Figure 1. The demonstration site. Blue line indicates the catchment area of the L. Valkea-Kotinen.

The demonstration lake, L. Valkea-Kotinen, is a boreal, small (A=0.042 km$^2$) headwater lake with a small pristine forested catchment (A=0.22 km$^2$) in South 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 old-growth Norway spruce (*Picea abies*), with the Scots pine (*Pinus sylvestris*), the aspen (*Populus tremula*) and the birch (*Betula* spp.). The forest and peatland cover 86% and 19% 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 100 years, and therefore long-range transported air pollutants and climate change are the only external disturbances.

Figure 2. Lake Valkea-Kotinen. Photo: Erkki Oksanen

### 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, orange funnels are set by Natural Resources Institute Finland (Luke).
Photo: Jussi Vuorenmaa

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 located in the forest part of the catchment. The observation plot consists of three subplots (size 30 x 40 m or 40 x 40 m) and a surrounding mantle. On one of the subplots located throughall, litterfall and soil water collectors, on the other one vegetation related studies were carried out, third one was 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.

Figure 4. Throughfall (orange funnel) and litterfall collectors (green funnels) in Valkea-Kotinen. Photo: Liisa Ukonmaanaho.

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.

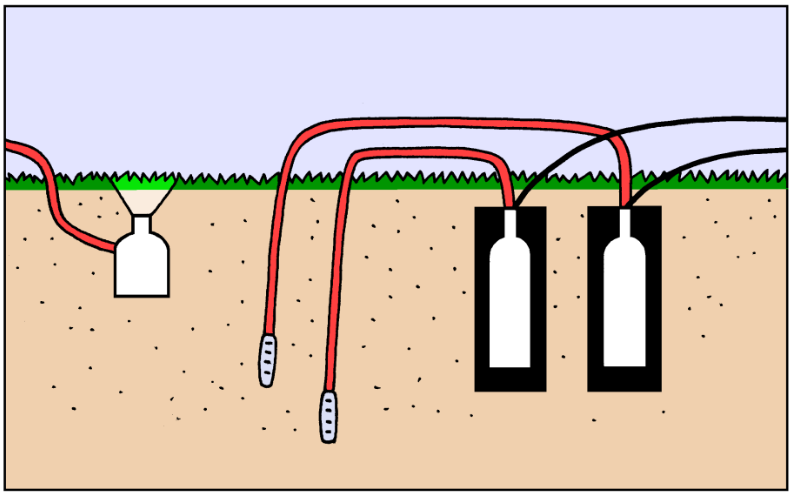

Figure 5. Schematic figure of soil water collectors. On the left zero-tension collector and on the right suction cup lysimeter. Figure: Liisa Ukonmaanaho.

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 is 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 c. 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 20 x 20 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 Valkea-Kotinen demonstration site is almost totally 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 Häme. Water sampling, altogether seven times, has been carried out monthly in March–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. Continuous runoff monitoring (based on water level recording calibrated against discharge) was performed at the overflow-measuring weir in the outflow 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 during 1990–2019. Air temperatures for the region of L. Valkea-Kotinen were collected from Finnish Meteorological Institute's observation station Lammi Pappila (WMOn 02767) situating 20 km south from the study areas.

### 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 S and 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 Valkea-Kotinen (Holmberg et al. 2013, Forsius et al. 2020).

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 Valkea-Kotinen 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. 2020). 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, 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 organic ‘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.

### 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 Valkea-Kotinen forest habitat in 1988–1998 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 (VG103 and VG108) located in the forest area (Kokko et al. 2002). The sampling design for vegetation survey changed in 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 two sub-plots sized 30 m x 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 x 30 m sub-plot was marked inside the plot VG103 (50 m x 50 m) for vegetation monitoring. Altogether 16 sampling quadrats for vegetation assessment were established systematically (4 x 4 design) on the sub-plot (Fig. 6). The size of the quadrat was 2 $m^2$ (1.41 m x 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 x 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 x 100 m$^2$ (A - D) and 900 m$^2$.

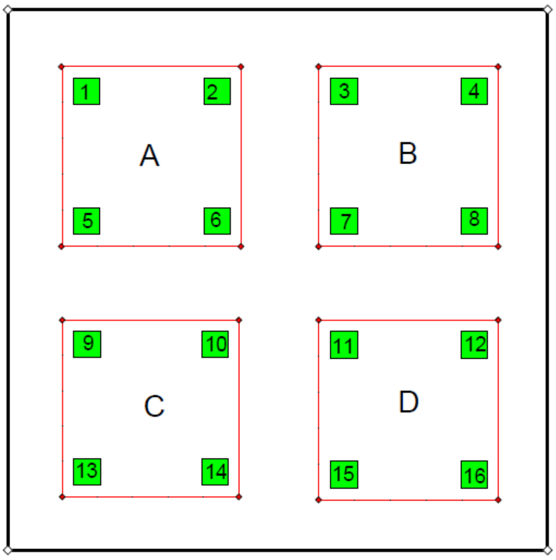

Figure 6. The sub-plot 30 m x 30 m in size used for the inventory of understorey vegetation. The cover % of plant species was assessed on the small (green) sample quadrats (sized 1.4 m x 1.4 m). Species found outside the small quadrats were recorded within red areas (each 10 m x 10 m) and the whole plot.

### 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. 7a). An open frame without a net was placed on the sites where a tree, shrubs or high vegetation were growing (Fig. 7b). 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 x 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 as the vegetation survey in 2009. Stem number, volume, basal area, diameter, tree height and height of lower crown limit were measured from living and dead trees with a breast height diameter of at least 4.5 cm. Lying dead trees were not included to the measurements. A demonstration study of the tree crown dimensions was done in 2002–2003. These values were used in calculations of crown projections to ground surface, which measure the shading area of the canopy.

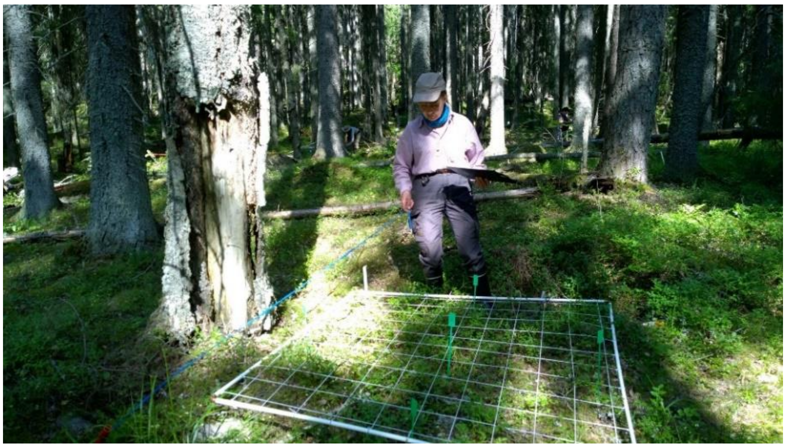

Figure 7a. Anneli Viherä-Aarnio studying understorey vegetation in the Valkea-Kotinen monitoring plot in 2009. Sample frame (1.41 m x 1.41 m) with a net was used in assessing plant species cover %. Photo: Maija Salemaa

Figure 7b. Leena Hamberg assessing plant covers with an open frame around a tree stem.
Photo: Maija Salemaa

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

The effective temperature sums and cumulative precipitation 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 average N deposition in Valkea-Kotinen and other ICP Forests Level II plots in Finland during 2007– 2009 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 191 mm $yr^{-1}$ without any significant trend neither on annual nor monthly basis (Table 1). Given the interest in leaching of constituents from the catchment, high summer/autumn runoff during the study period occurred in 1998, 2004, 2012, 2013 and 2017 (Fig. 8).

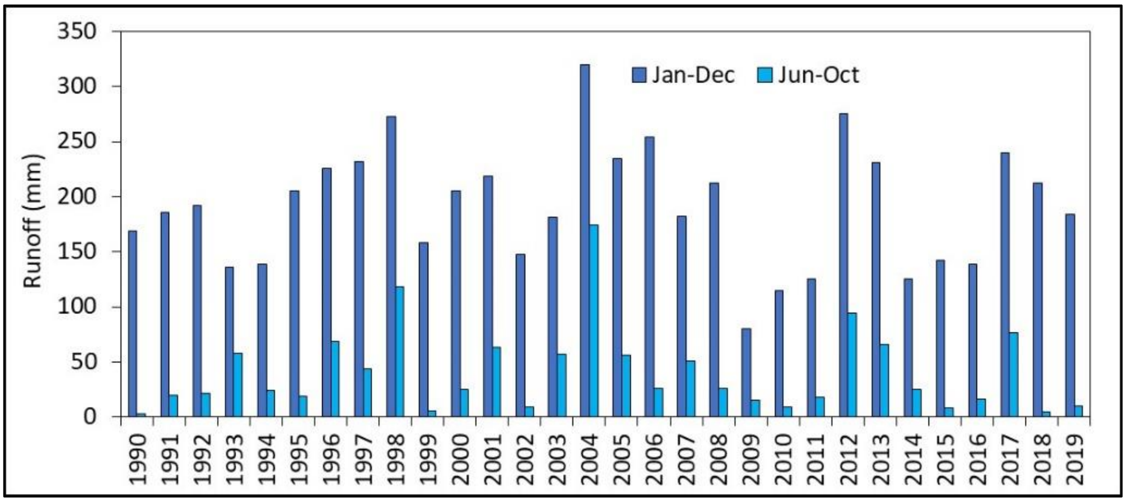

Figure 8. Annual and seasonal (June–October) runoff at the measurement station of the Valkea-Kotinen catchment in 1990–2019.

Table 1. Long-term temporal monthly and annual trends (SKT, Sen's slope) for runoff in Valkea-Kotinen catchment in 1990–2019. For the annual change (mm $yr^{-1}$), 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 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 0.05 | 0.07 | -0.12 | -0.47 | -0.53 | -0.18 | 0 | 0 | -0.002 | -0.11 | 0.15 | 0.49 | -0.02 |

There was a significant increase ($p < 0.05$) in air temperature in January–December in L. Valkea-Kotinen area from 1990 to 2019 (Table 2). The increase was pronounced in autumn, because September and November exhibited the significant increase (0.08 °C $yr^{-1}$ and 0.12 °C $yr^{-1}$, respectively) on a monthly basis. Air temperature exhibited weak increasing trend ($p < 0.1$) in spring (May).

During the last 50 years, the air temperature in the Valkea-Kotinen region has risen by about 0.4 °C per decade (Jylhä et al. 2014). This is in good agreement with the trend on air temperatures observed for the whole of Finland (Tietäväinen et al. 2010). One indicator for rising air temperatures has been the earlier ice-off in spring and later freezing in autumn on the L. Valkea-Kotinen, which has resulted in the shortening of ice-cover period in the course of last 20 years, by approximately 1.5 days per year as an average (Jylhä et al. 2014). Climate change projections have suggested that warming is expected to continue in the region throughout the 21st century by 0.3–0.4 °C per decade (Jylhä et al. 2014).

Table 2. Long-term temporal monthly and annual trends (SKT, Sen's slope) for air temperatures in the study area in 1990–2019. For the annual change (slope, °C $yr^{-1}$), 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 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| -0.08 | 0.01 | -0.01 | 0.04 | $0.08^{+}$ | 0.01 | 0.03 | 0.03 | 0.08* | 0.002 | 0.12* | 0.07 | 0.04* |

While air temperature has increased, water temperature in L. Valkea-Kotinen has not correspondingly increased in 1990–2019. However, significant increases in epilimnetic (1 m) temperatures in March, and hypolimnetic (5 m) temperatures in May were detected (Table 3).

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

| Depth (m) | Mar | May | Jun | Jul | Aug | Sep | Oct | Mar-Oct |
|---|---|---|---|---|---|---|---|---|
| 1 | 0.03* | 0.09 | 0.01 | -0.01 | 0.03 | -0.14 | 0 | 0.02 |
| 3 | 0.01 | 0.03 | 0.02 | 0 | -0.03 | -0.02 | 0.01 | 0.01 |
| 5 | -0.01 | 0.03* | 0 | -0.02 | -0.01 | -0.01 | 0.005 | 0 |

### 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 Valkea-Kotinen demonstration site (Fig. 9). The emission control programmes have been particularly successful for sulphur (S), and the deposition of anthropogenic (non-marine fraction) sulphate ($xSO_4$) decreased in Valkea-Kotinen area by 80–90% 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.

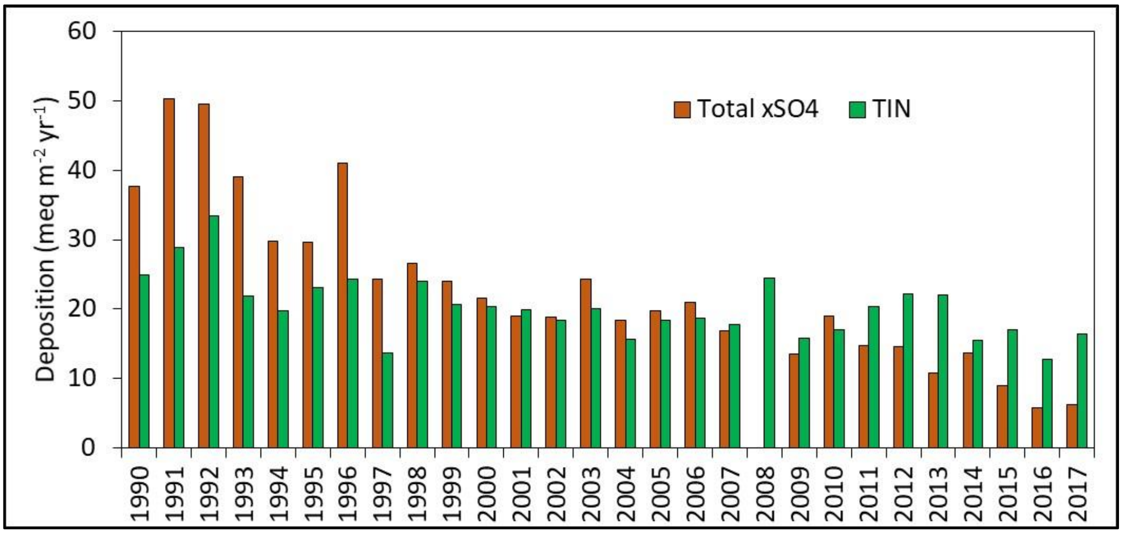

Figure 9. The annual total deposition of non-marine $xSO_4$ (x denotes non-marine, anthropogenic fraction) and TIN ($TIN=NO_3+NH_4$) in Valkea-Kotinen region in 1990–2017. (Data source for bulk deposition of $xSO_4$ and TIN: Finnish Meteorological Institute).

### 3.3 Acidification parameters and trace (heavy) metals

In 1990s, L. Valkea-Kotinen exhibited acidic conditions, and alkalinity and pH values were commonly $< 0$ µeq $l^{-1}$ and $\leq 5$, respectively (Fig. 10). Due to the decreased acid deposition, sulphate concentration has significantly ($p < 0.05$) decreased by about 50% during the study period 1990–2019, and the acidification reversal was also recorded in L. Valkea-Kotinen. Base cations also declined in the lake, but to a lesser extent than sulphate (Fig. 10, Table 4), indicating the improved acid-base status of soils, and led to significant increase in buffering capacity (alkalinity and ANC) and pH in lake water. Increase of alkalinity has been pronounced since the early 2000s.

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 L. Valkea-Kotinen in 1990–2019. For the annual change, a statistically significant trend ($p < 0.05$) is denoted with asterisk.

| Variable | Unit | Mar | May | Jun | Jul | Aug | Oct | Dec | Mar-Dec |
|---|---|---|---|---|---|---|---|---|---|
| pH | pH-unit | 0.01* | 0.01* | 0.01* | 0.01* | 0.01 | 0.01* | 0.01* | 0.01* |
| Conductivity | mS m$^{-1}$ yr$^{-1}$ | -0.04* | -0.03* | -0.04* | -0.04* | -0.03* | -0.03* | -0.03* | -0.03* |
| Alkalinity | µeq l$^{-1}$ yr$^{-1}$ | 1.26* | 1.20* | 1.20* | 1.00* | 1.22* | 1.28* | 1.50* | 1.22* |
| ANC | µeq l$^{-1}$ yr$^{-1}$ | 1.52* | 0.96* | 0.98* | 0.75 | 0.60 | 1.09* | 1.53* | 1.00* |
| xBC | µeq l$^{-1}$ yr$^{-1}$ | -1.89* | -1.87* | -2.16* | -2.03* | -2.02* | -1.63* | -1.53* | -1.90* |
| $xSO_4$ | µeq l$^{-1}$ yr$^{-1}$ | -3.05* | -2.56* | -3.11* | -2.80* | -2.69* | -2.92* | -3.16* | -2.89* |
| Cl | µeq l$^{-1}$ yr$^{-1}$ | -0.23* | -0.13* | -0.10* | -0.19* | -0.14* | -0.14* | -0.38* | -0.18* |
| TOC | mg l$^{-1}$ yr$^{-1}$ | 0.21* | 0.14* | 0.14* | 0.12* | 0.11* | 0.15* | 0.11 | 0.14* |
| Colour | mg Pt l$^{-1}$ yr$^{-1}$ | 2.50* | 2.00* | 1.70* | 1.43* | 1.43* | 2.00* | 3.16* | 1.88* |
| $SiO_2$ | mg l$^{-1}$ yr$^{-1}$ | 0.06* | 0.06* | 0.03 | 0.06* | 0.04* | 0.06* | 0.04 | 0.05* |

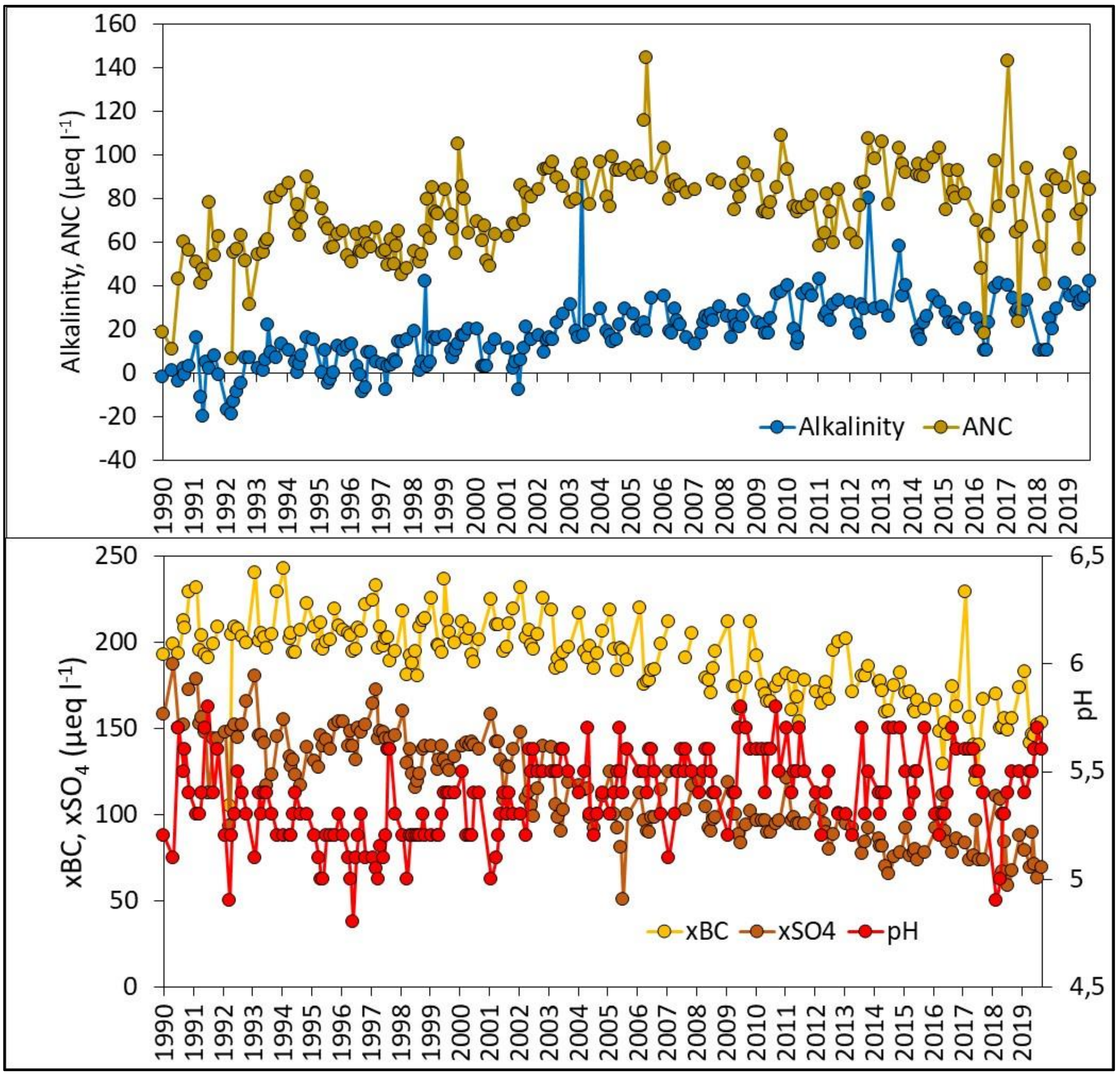

Figure 10. Time series for alkalinity and ANC$_{CB}$ (up) and non-marine base cations (xBC) and sulphate ($xSO_4$), and pH (down) at the depth of 1 m in L. Valkea-Kotinen in 1990–2019.

In L. Valkea-Kotinen, the decrease in $xSO_4$ concentration was slower than expected, on the basis of the clear decrease of total $xSO_4$ deposition (80–90%). This indicates a delayed response in $SO_4$ output in the catchment to decreased deposition. At the beginning of the 1990s, the L. Valkea-Kotinen catchment retained 30% of sulphate deposition due to strong retention of $SO_4$ in peatlands and in peaty soils, but after the mid-1990s the catchment shifted from retention to net release (output > input) of sulphate (Fig. 11). This shows that forest soils are now recovering from acid deposition by releasing of stored airborne sulphur that had accumulated in the past (e.g. De Vries et al. 2001).

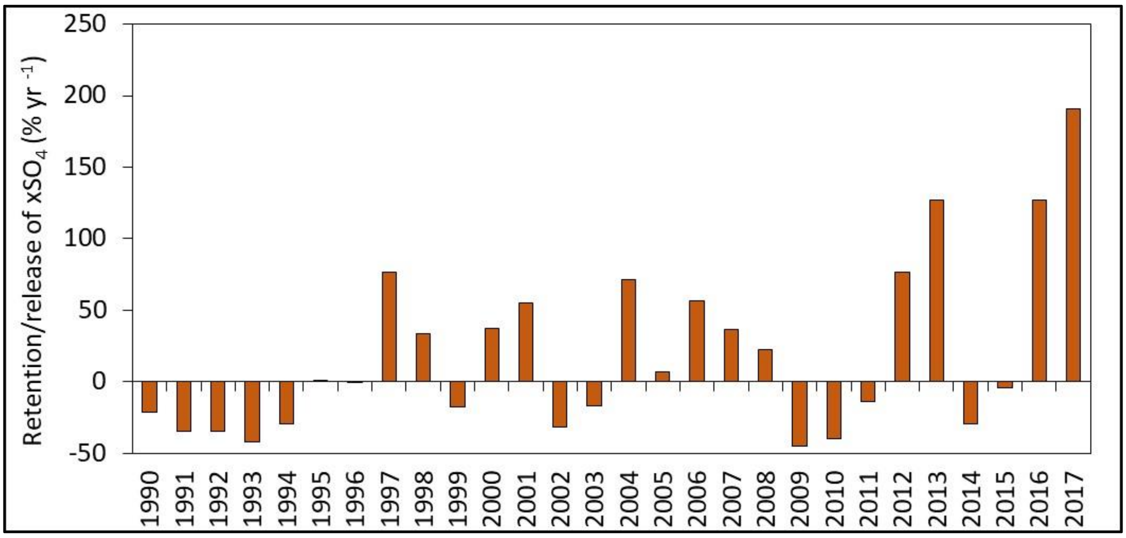

Figure 11. Retention/release of $xSO_4$ (% yr $^{-1}$) in Valkea-Kotinen catchment in 1990–2017.

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 the Valkea-Kotinen catchment clearly declined over the past decades (Ruoho-Airola et al. 2014). The general decrease of trace metals deposition was reflected to some extent in lake water concentrations (Table 5). Total aluminium (Al tot) increased significantly, but a significant decreasing trend was detected for arsenic (As), lead (Pb) and nickel

(Ni) between 1994 and 2019. For zinc (Zn), copper (Cu) and chrome (Cr) the 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, V and Hg in L. Valkea-Kotinen. For the annual change, a statistically significant trend ($p < 0.05$) is denoted with asterisk. n.d.=no data.

| Variable | Percentiles 1994-1999 (µg l$^{-1}$) | | | Percentiles 2000-2009 (µg l$^{-1}$) | | | Percentiles 2010-2019 (µg l$^{-1}$) | | | Trend 1994-2019 |
|---|---|---|---|---|---|---|---|---|---|---|
| | 10% | 50% | 90% | 10% | 50% | 90% | 10% | 50% | 90% | (µg l$^{-1}$ yr$^{-1}$) |
| Al tot | 112 | 130 | 151 | 130 | 150 | 200 | 130 | 160 | 217 | 1.814* |
| As | 0.22 | 0.27 | 0.32 | 0.24 | 0.27 | 0.30 | 0.22 | 0.24 | 0.29 | -0.001* |
| Cd | <0.03 | <0.03 | 0.03 | <0.01 | <0.03 | <0.03 | <0.01 | <0.01 | 0.015 | n.d. |
| Cr | 0.16 | 0.27 | 0.45 | 0.20 | 0.30 | 0.42 | 0.15 | 0.24 | 0.33 | -0.001 |
| Cu | 0.14 | 0.22 | 0.73 | 0.17 | 0.24 | 0.32 | 0.13 | 0.26 | 0.5 | 0.002 |
| Pb | 0.30 | 0.42 | 0.65 | 0.25 | 0.35 | 0.52 | 0.21 | 0.35 | 0.54 | -0.003* |
| Ni | 0.39 | 0.48 | 1.11 | 0.36 | 0.43 | 0.51 | 0.30 | 0.39 | 0.5 | -0.004* |
| Zn | 2.9 | 4.5 | 33 | 3.0 | 6.8 | 25 | 2.4 | 3.65 | 5.6 | -0.039 |
| V | 0.26 | 0.29 | 0.36 | 0.24 | 0.29 | 0.40 | 0.21 | 0.28 | 0.38 | -0.001 |
| Hg | n.d. | n.d. | n.d. | 0.003 | 0.004 | 0.006 | 0.002 | 0.003 | 0.01 | n.d. |

The clear 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 head water 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.

In contrast to other trace metals, Al tot significantly increased in 1994–2019. L. Valkea-Kotinen is undergoing a recovery from acidification, and pH-value and buffering capacity are increasing. Therefore, processes other than acidification may have controlled Al tot leaching. It is known that aluminium in soil percolation water is largely controlled by the concentration of organic complexes (DOC) (e.g. Lindroos et al. 2011), and an increase in Al tot concentration in L. Valkea-Kotinen may have a link to the increased export of DOC from the catchment. Weathering of minerals in forest soil (like aluminium) is, inter alia, temperature-dependent, and it has also been suggested that an increased DOC concentration in soil solution increases the weathering release of Al (Lindroos et al. 2003). Air temperature significantly increased and snow cover thickness decreased in the Valkea-Kotinen region, and the leaching of DOC to surface water significantly increased in L. Valkea-Kotinen, all of which suggests an enhancement of the weathering rate of Al. Observed significant increase in silicon dioxide ($SiO_2$) (Table 4) may indicate general increase of weathering of minerals due to increased temperature. The results altogether suggest that climate may be an important driver behind the enhanced weathering and observed increase in Al tot.

### 3.4 Total organic carbon and water colour

During the past 30 years, L. Valkea-Kotinen showed further brownification with a significant increase ($p < 0.05$) in both total organic carbon (TOC) concentration and water colour (Table 4). TOC and water colour were elevated particularly during the 2000s (Fig. 12). From 1990 to 2019, TOC concentration has increased approximately by 4 mg $l^{-1}$ and water colour 50 mg Pt $l^{-1}$ (Table 4).

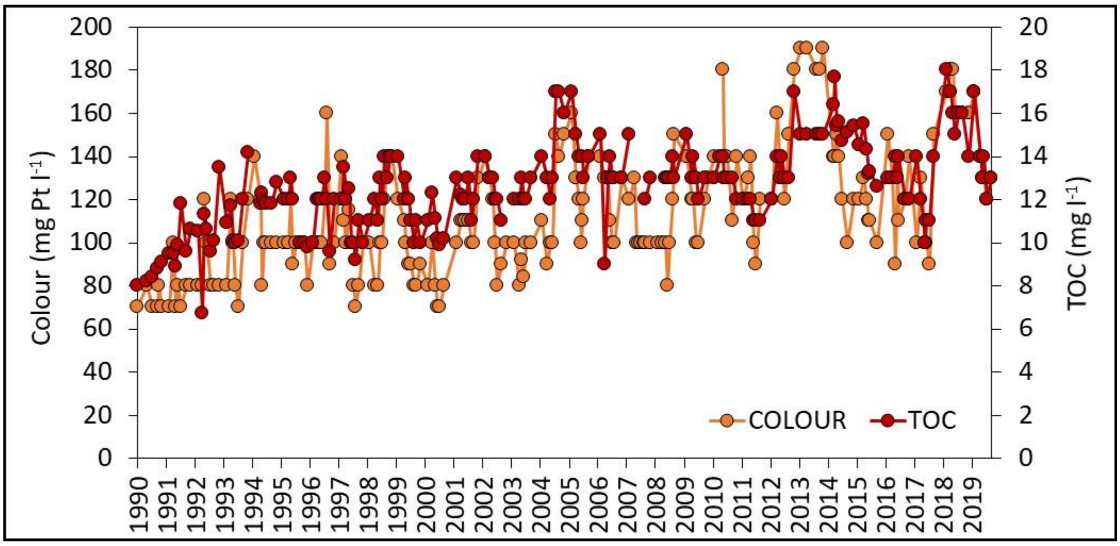

Figure 12. Time series for water colour and total organic carbon (TOC) concentrations at the depth of 1 m in L. Valkea-Kotinen in 1990–2019.

The increase in TOC and water colour in L. Valkea-Kotinen was in agreement with earlier observations from Finland and elsewhere in Europe and North America (e.g. Vuorenmaa et al. 2006, Monteith et al. 2007). 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 L. Valkea-Kotinen, 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 within couple of day after ice-off in L. Valkea-Kotinen, 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. Oxygen deficiency in the hypolimnion of L. Valkea-Kotinen have also caused production of methyl-Hg (Verta et al. 2010) and its accumulation in fish (Rask et al. 2010).

Variation in runoff and leaching of humic-derived organic acids may also have affected alkalinity in L. Valkea-Kotinen, 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 and phosphorus

During the period 1990–2019, there was no significant long-term trend in total inorganic nitrogen concentrations (TIN=$NO_3$-N+$NH_4$-N) on an annual basis. However, a seasonal pattern was detected: TIN concentration decreased significantly in the epilimnion in winter (Table 6). The trend slopes 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 18 meq m$^{-2}$ yr$^{-1}$ (2.5 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 Valkea-Kotinen catchment showed high net retention (> 95%) of inorganic nitrogen (Vuorenmaa et al. 2017). Total nitrogen concentration did not exhibit any long-term trend 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) and N:P-ratio in L. Valkea-Kotinen 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 | µg l$^{-1}$ yr$^{-1}$ | -0.42 | -0.14 | 0.00 | 0.00 | 0.00 | -0.16 | -0.29 | 0.00 |
| $NH_4$-N | µg l$^{-1}$ yr$^{-1}$ | -1.36* | -0.05 | 0.00 | 0.00 | 0.00 | -0.48 | 0.13 | 0.00 |
| TIN | µg l$^{-1}$ yr$^{-1}$ | -1.69* | -0.37 | -0.01 | 0.12 | 0.00 | -0.47 | -0.05 | -0.10 |
| tot N | µg l$^{-1}$ yr$^{-1}$ | 0.00 | -2.50 | 1.43 | 1.13 | -1.67 | 0.74 | 2.73 | 0.00 |
| Org N | µg l$^{-1}$ yr$^{-1}$ | 2.60* | -0.36 | 0.72 | 1.13 | -1.87 | 2.00 | 3.38 | 1.00 |
| tot P | µg l$^{-1}$ yr$^{-1}$ | 0.00 | 0.10 | 0.25* | 0.14* | 0.00 | 0.09 | 0.05 | 0.09 |
| N:P-ratio | | -0.09 | -0.17 | -0.23* | -0.15 | -0.12 | -0.19 | 0.03 | -0.16+ |

No consistent trend was found in total phosphorus (tot P) concentration in the epilimnion in 1990–2019 (Table 6), but short-term patterns were evident. Concentration of tot P in 1990–1999 decreased (-0.62 µg l$^{-1}$ yr$^{-1}$, $p < 0.01$), and in 2000–2019 it increased (0.31 µg l$^{-1}$ yr$^{-1}$, $p < 0.01$) (Fig. 13).

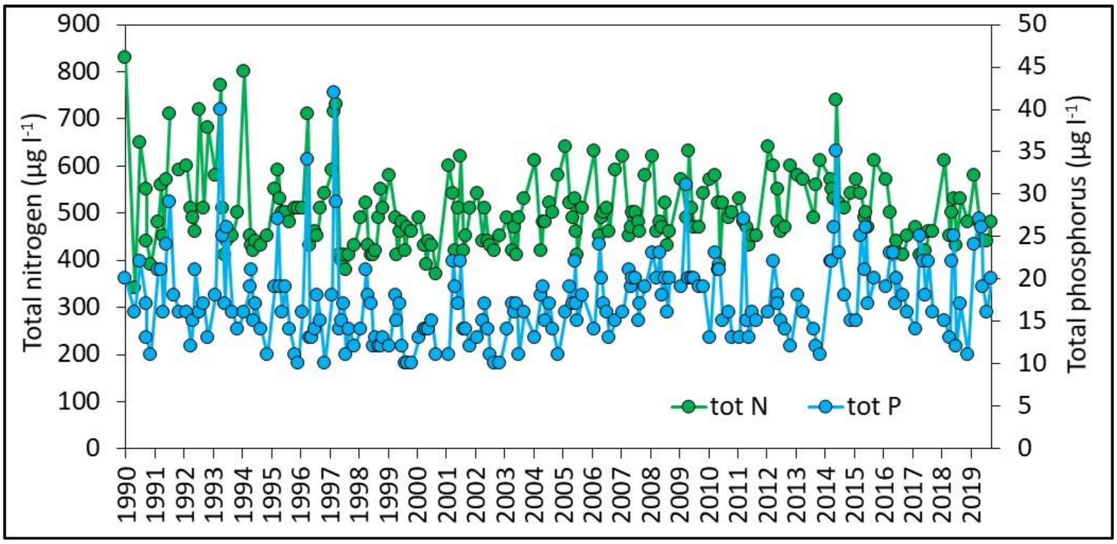

Figure 13. Time series for total nitrogen (tot N) and total phosphorus (tot P) concentrations at the depth of 1 m in L. Valkea-Kotinen in 1990–2019.

One possible reason for the variability in tot P concentration might be deteriorated oxygen conditions in the hypolimnion, and enhanced release of phosphorus from the sediment in the 2000s. The incomplete spring overturn seems to be a significant factor influencing tot P concentration in the L. Valkea-Kotinen (Arvola et al. 2014, Vuorenmaa et al. 2014).

Hydrological conditions, such as wet summer/autumn in 2004, 2012, 2013 and 2017 (Fig. 2) may also have increased leaching of phosphorus, since dissolved organic matter (DOM) is considered to be a major carrier of organic P (e.g. Kortelainen et al. 2006).

### 3.6 Sulphur and nitrogen deposition and exceedances of critical loads

The acidity critical load function at Valkea-Kotinen is determined by the values $CL_{max}N=2060$ eq ha$^{-1}$ yr$^{-1}$, and $CL_{max}S=373$ eq ha$^{-1}$ yr$^{-1}$, using the critical ANC concentration of 20 µeq l$^{-1}$. 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). The graph shows that for some years in the beginning of the period 1990–2017, the acidity critical load was exceeded. The critical ANC concentration of 20 µeq l$^{-1}$ was not, however, violated during the observation period.

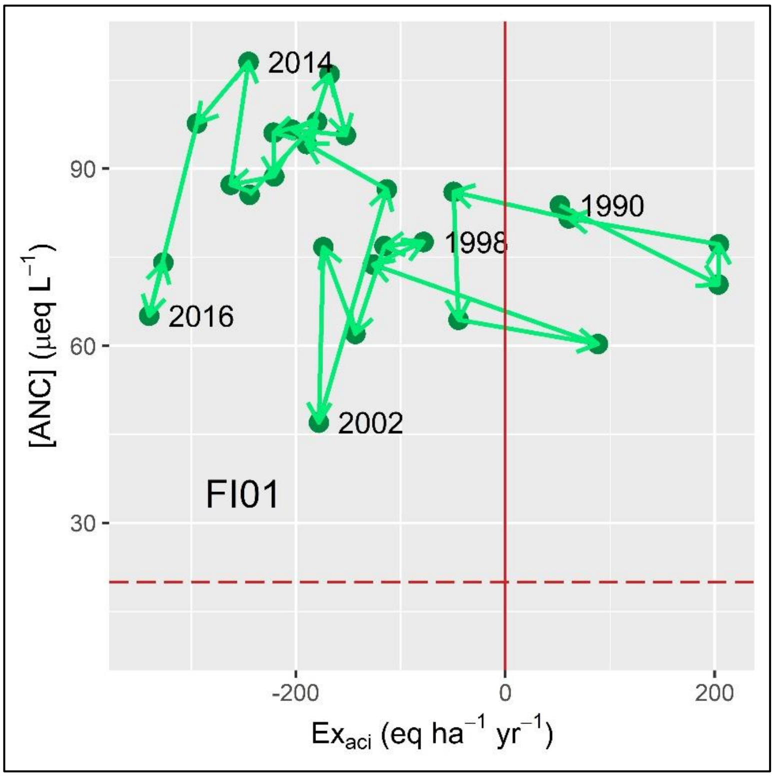

Figure 14. Valkea-Kotinen (FI01) year-to-year variation in runoff ANC (µeq $l^{-1}$) plotted against $EX_{aci}$, the exceedance of critical loads of acidification (eq $ha^{-1}$ $yr^{-1}$). The vertical solid line ($Ex_{aci}$=0) shows the division between non-exceedance (negative values of $Ex_{aci}$), 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 Valkea-Kotinen 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 Valkea-Kotinen was determined as 3.3 kg N $ha^{-1}$ $yr^{-1}$. As this value is lower than the empirical CL, the CL of eutrophication at Valkea-Kotinen is 3.3 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.

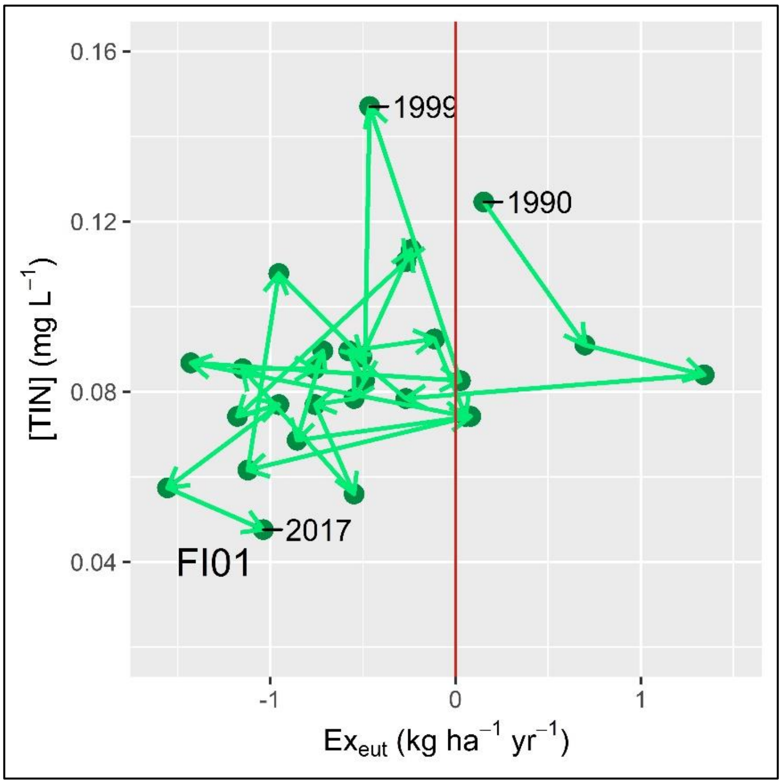

Figure 15. Valkea-Kotinen (FI01) year-to-year variation in runoff TIN (mg $l^{-1}$) plotted against $Ex_{eut}$, the exceedance of critical loads of eutrophication (kg $ha^{-1}$ $yr^{-1}$). The vertical solid line ($Ex_{eut}$=0) shows the division between non-exceedance (negative values of $Ex_{eut}$), and exceedance (positive values of $Ex_{eut}$).

The critical concentrations (ANC 20 µeq $l^{-1}$, TIN 1.3 mg $l^{-1}$) were not violated during the observation period, the exceedances have decreased over the observation period (Figs. 14, 15). For eutrophication at Valkea-Kotinen, 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) and 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 Valkea-Kotinen forest area and compared the values to the stream water (output of L. Valkea-Kotinen). The mean annual element concentrations during 1990–2018 are shown in Tables 7 and 8. In general element concentrations were lowest in BP and highest in SW either in depth of 5 or 20 cm. However, the concentrations of inorganic N and K were exceptions, where BP had highest inorganic N concentration and TF highest K concentration.

The $SO_4$-S concentration in BP was lowest as expected, but it was surprising that $SO_4$-S concentration in soil water at depth 20 cm was higher than under the organic layer, this is probably due to desorption of previously retained $SO_4$ from the deposition or the mineralization of organic pools (Mitchell 1992, Ukonmaanaho 2001). The $SO_4$-S concentration in stream water was similar level as SW at depth of 20 cm. Source of high $SO_4$-S concentration in stream water is from surrounding terrestrial area, especially wetland area, as mentioned in a previous chapter.

The DOC concentration in the BP was lower than other aquatic samples, only 2 mg $l^{-1}$, however, the concentration of DOC increases as it passes through the tree canopy and is related to the composition of tree species 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 Valkea-Kotinen the DOC concentration in TF was on average 17.6 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). DOC concentrations in soil water under organic layer was on average 63.5 mg $l^{-1}$ being almost three times higher than DOC concentrations in other solutes. At the depth of 20 cm, DOC concentration was less, on average 21.3 mg $l^{-1}$. Typically, in boreal forest soil solution DOC concentration lie 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 concentrations 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 inorganic N concentration was similar level as in SW. The total nitrogen concentration was highest in SW and lowest in BP, showing almost similar pattern as DOC. The share of organic nitrogen is great in total nitrogen. At soil solution share of organic nitrogen was > 99% of the total nitrogen, but in BP only 30%.

Base cation concentrations were highest in SW, excluding potassium (K), which concentration was highest in TF. Potassium concentration in green needles was relatively high compared litterfall (LF) needles or other LF fractions. It is known that K is mobile cation, therefore the K in the TF is likely leached almost entirely from the foliage (green needles). In general, the enrichment of 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, as well. 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. Valkea-Kotinen.

| Media | BP | TF | SW 5 cm | SW 20 cm | Stream |
|---|---:|---:|---:|---:|---:|
| mm (sum) | 645 | 461 | | | 191 |
| n | 243–351 | 284–375 | 128–145 | 493–541 | 532–537 |
| DOC/TOC, mg $l^{-1}$ | 2.0 | 17.6 | 63.5 | 21.3 | 22.2 |
| sd | 1.90 | 4.86 | 38.60 | 15.43 | 9.62 |
| Ca, mg $l^{-1}$ | 0.20 | 0.90 | 2.51 | 2.14 | 2.44 |
| sd | 0.196 | 0.355 | 0.51 | 1.76 | 0.713 |
| Mg, mg $l^{-1}$ | 0.05 | 0.35 | 0.83 | 0.92 | 0.73 |
| sd | 0.058 | 0.132 | 0.510 | 0.444 | 0.179 |
| K, mg $l^{-1}$ | 0.10 | 3.26 | 2.81 | 0.91 | 0.32 |
| sd | 0.146 | 1.101 | 2.370 | 1.106 | 0.110 |
| Na, mg $l^{-1}$ | 0.17 | 0.63 | 0.78 | 1.92 | 1.41 |
| sd | 0.114 | 1.101 | 0.410 | 0.881 | 0.166 |
| $NO_3$-N mg $l^{-1}$ | 0.30 | 0.27 | 0.02 | 0.03 | 0.03 |
| sd | 0.204 | 0.208 | 0.020 | 0.097 | 0.030 |
| $NH_4$-N, mg $l^{-1}$ | 0.25 | 0.22 | 0.10 | 0.04 | 0.05 |
| sd | 0.266 | 0.187 | 0.130 | 0.100 | 0.044 |
| N, mg $l^{-1}$¤ | 0.59 | 0.79 | 2.47 | 1.21 | 0.64 |
| sd | 0.446 | 0.308 | 9.490 | 1.247 | 0.157 |
| $SO_4$-S mg $l^{-1}$ | 0.41 | 1.12 | 0.53 | 3.29 | 2.16 |
| sd | 0.354 | 0.560 | 0.420 | 1.857 | 0.639 |
| Base cations*, µeq $l^{-1}$ | 40.5 | 184.4 | 298.3 | 289.1 | 250.7 |
| sd | 31.49 | 56.25 | 159.07 | 106.69 | 55.49 |
| Strong acid anions# µeq $l^{-1}$ | 53.5 | 123.8 | 72.0 | 258.8 | 173.4 |
| sd | 35.78 | 55.05 | 58.48 | 134.39 | 67.49 |
| ANC, µeq $l^{-1}$ | -12.99 | 60.65 | 226.3 | 23.98 | 76.7 |
| sd | 22.27 | 28.90 | 139.65 | 80.70 | 58.23 |

¤ 2002-2018, *(Ca+K+Mg+Na), # ($NO_3$+ $NH_4$+ $SO_4$+ Cl)

We studied also nutrients in green and LF needles and other LF fractions over 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. When needles are senescing most mobile nutrients are transformed back to trunk or younger needles, therefore concentrations are usually lower in LF needles than living needles. However, Ca was an exception, its concentration was highest in LF 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 tissue, playing a major role in osmoregulation of stomatal opening and closing and therefore it is leaching easily from the living needles and therefore its content is lower in LF needles (e.g. Johnson 2002). There were also difference in needle LF and other LF fraction, usually nutrient concentrations of other LF fraction were higher than needle LF. The other LF fraction may compose 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 (Ukonmaanaho et al. 2020).

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

| | | Green needles | | Litterfall | |
|---|---|---|---|---|---|
| | | c | c+1 | needles | other litterfall |
| n | | 25 | 25 | 148-160 | 148-160 |
| years | | 1995–2017 | 1995–2017 | 1999–2017 | 1999–2017 |
| g $m^{-2}$ $yr^{-1}$ | sum | | | 209 | 147 |
| | sd | | | 43.8 | 39.6 |
| C % | mean | 51.9 | 51.8 | 52.6 | 53.2 |
| | sd | 0.62 | 0.36 | 2.04 | 1.49 |
| N mg $g^{-1}$ | mean | 11.9 | 10.9 | 8.6 | 13.4 |
| | sd | 1.24 | 0.94 | 2.89 | 4.81 |
| Ca $mg^{-1}g$ | mean | 4.00 | 6.38 | 8.67 | 5.34 |
| | sd | 0.563 | 0.662 | 3.278 | 1.655 |
| Mg mg $^{-1}$ | mean | 1.04 | 0.96 | 1.08 | 1.10 |
| | sd | 0.087 | 0.084 | 0.778 | 0.709 |
| K mg $^{-1}$ | mean | 6.87 | 4.84 | 2.12 | 3.01 |
| | sd | 0.571 | 0.580 | 1.133 | 2.244 |
| S mg $^{-1}$ | mean | 0.82 | 0.78 | 0.68 | 1.04 |
| | sd | 0.055 | 0.054 | 0.171 | 0.236 |
| P mg $^{-1}$ | mean | 1.35 | 1.00 | 0.62 | 1.01 |
| | sd | 0.125 | 0.070 | 0.247 | 0.490 |

### 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 Valkea-Kotinen. As a result of air quality controls, the $SO_4$-S deposition has considerably decreased throughout Europe and it can be seen also in Valkea-Kotinen (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 20 cm and stream water had a decreasing trend  Therefore, the reduced $SO_2$ emissions have resulted not only in 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 liquid samples, excluding soil water under organic layer.

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. Valkea-Kotinen (stream). Values in bold indicate statistically significant trend ($p < 0.05$).

| | BP | TF | SW 5 cm | SW 20 cm | Stream |
|---|---|---|---|---|---|
| n | 243–351 | 284–375 | 128–145 | 414–541 | 532–537 |
| Period | 1990–2018 | 1990–2018 | 2002–2017 | 1990–2018 | 1990–2018 |
| Precipitation/runoff, mm $yr^{-1}$ | 0.040 | -0.37 | | | 0.446 |
| DOC/TOC, mg $l^{-1}$ $yr^{-1}$ | **-0.056** | **0.19** | 0.119 | -0.289 | **0.183** |
| Ca, mg $l^{-1}$ $yr^{-1}$ | 0.001 | 0.004 | 0.040 | **-0.113** | **-0.025** |
| Mg, mg $l^{-1}$ $yr^{-1}$ | 0.000 | 0.001 | 0.026 | **-0.031** | **-0.009** |
| K, mg $l^{-1}$ $yr^{-1}$ | 0.001 | **0.048** | -0.046 | **-0.042** | **-0.002** |
| Na, mg $l^{-1}$ $yr^{-1}$ | **-0.001** | **0.022** | 0.020 | **0.021** | -0.003 |
| $NO_3$-N, mg $l^{-1}$ $yr^{-1}$ | **-0.004** | **-0.002** | -0.001# | 0.00# | -0.172 |
| $NH_4$-N, mg $l^{-1}$ $yr^{-1}$ | **-0.003** | **-0.005** | **-0.005** | 0.002 | -0.460 |
| Total N, mg $l^{-1}$ $yr^{-1}$ | **-0.013** | -0.006 | 0.039 | -0.056 | **2.553** |
| $SO_4$-S, mg $l^{-1}$ $yr^{-1}$ | **-0.016** | **-0.044** | -0.018 | **-0.164** | **-0.042** |
| Base cations, µeq $l^{-1}$ $yr^{-1}$ | -0.30 | **1.93** | 2.601 | **-9.791** | **-2.231** |
| Strong acid anions, µeq $l^{-1}$ $yr^{-1}$ | **-1.32** | **-2.18** | -2.67 | **-11.09** | **-2.764** |
| ANC, µeq $l^{-1}$ $yr^{-1}$ | **1.17** | **4.51** | 2.48 | -1.90 | 0.375 |

# Results have been mainly under instrument detection limit → no trend seen.

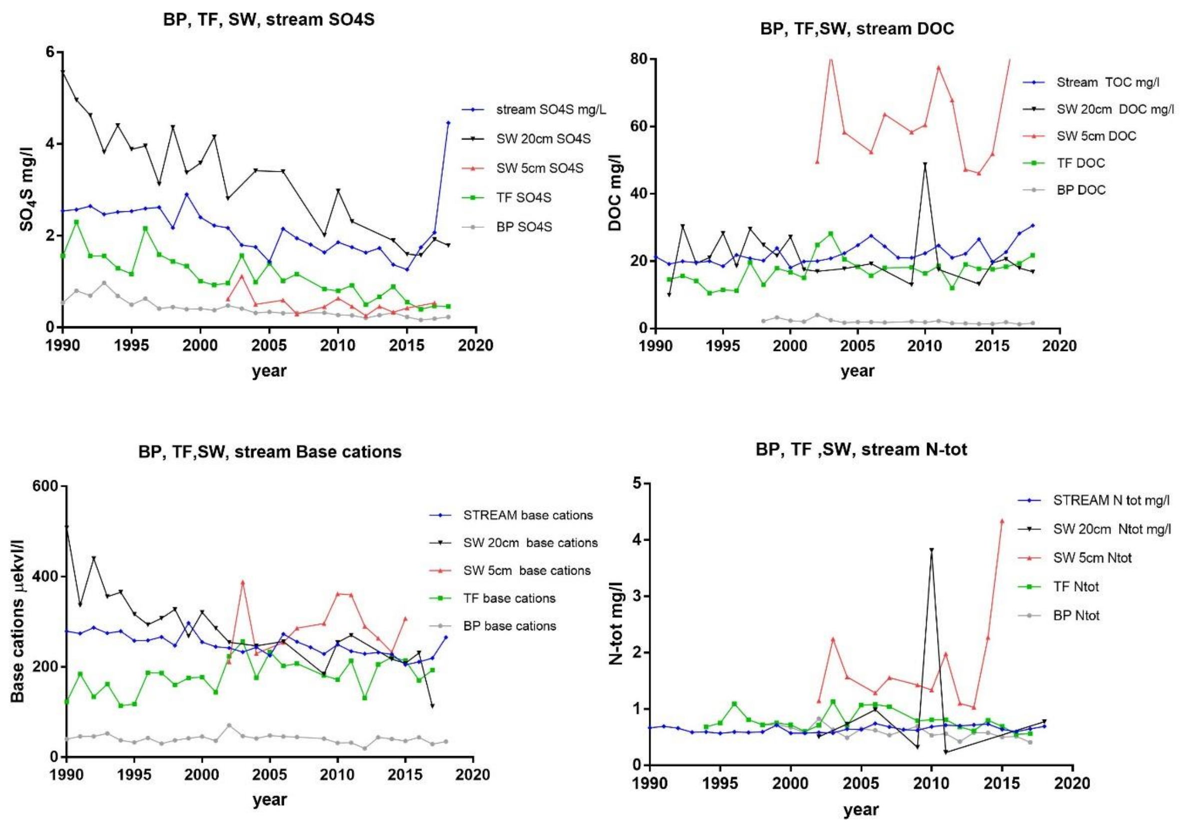

Fig 16. Time series for $SO_4$-S, DOC, base cations and total N in bulk precipitation (BP), throughfall (TF), soil water (SW) and stream water (at the outlet of L. Valkea-Kotinen) in 1990–2018.

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, but due to the application of dust removal technology, the recent base cation emissions of the region have decreased (Ruoho-Airola et al. 2003). However, in TF there was a significant increasing trend, while in soil solution at depth of 20 cm and in stream water a significant decreasing trend was detected. Because of a decreasing trend of base cations in BP, it is obvious that the detected significant increase in base cation concentrations in TF is related to the intercepted dry deposition on the tree canopy, and the consequent wash-off by precipitation. The share of dry deposition of Ca, K and Mg is usually less than 20 % of the total deposition in Finland (Ruoho-Airola et al. 2003). In addition to dry deposition, wash-off and leaching from the above ground biomass and tree litter are the most relevant sources, which increase base cation concentrations in TF. Single base cations Ca, Mg, K and Na concentrations had a positive trend in TF, but trend was significant only for K. In litterfall (needles and other litterfall fraction) and green needles there was a decreasing trend in Mg and K concentrations. In general base cation concentrations are usually high in needles, however, due to cation exchange reaction hydrogen ions in TF can replace adsorbed cations, especially K, which is seen also in our results. The base cation concentrations decreased significantly ($p<0.05$) in SW at the depth of 20 cm, but had increasing trend at 5 cm. Increasing trends in TF base cations, may have had effect also on soil solution under organic soil layer (humus), but a decline in soil water at 20 cm is probably due that roots have taken available cations efficiently. On the other hand, the decline may be related to the decline in $SO_4$-S concentrations. According to Singh et al. (1980) a reduction in the amount of $SO_4$ anions as required to accompany cations would lead to a reduction in base cation concentrations in the soil solution. The clear decline in $SO_4$-S concentration in stream water explains significant decreasing trend of base cations in stream water.

The combined effects of changes in $SO_4$-S, strong acids and base cation concentrations reflected as an increasing trend in ANC value in the studied solutions, excluding SW at the depth of 20 cm. In view of the general decrease in BP, TF, SW and stream water $SO_4$-S concentrations, the decrease in ANC in SW at the depth of 20 cm is somewhat contradictory. However, the decrease in base cation concentrations can be the main cause for the 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).

DOC in TF was positively correlated with air temperature, which is in agreement with other studies (Solinger 2001, Kalbitz et al. 2000, Ukonmaanaho et al. 2014), and this was expected because in the growing season DOC concentrations were usually higher compared to winter conditions. Obviously the slightly increased temperature in our study site has reflected as longer growing seasons, which in turn leads to a higher net primary production and a higher foliar litter production rate. Since the senescing needle mass is the primary source of DOC-producing substrate, a higher input rate of litter may reflect a higher DOC production in the canopy and subsequently an increased DOC concentration in TF (as has been observed in organic soils) (Fröberg et al. 2006). However, there was no significant increase in litterfall amount during the study period, although amount of needle LF fraction had a slightly positive trend. It is obvious, however, that release of organic compounds from decaying tissues is enhanced in old growth forests as on Valkea-Kotinen area, where for example, a wide set of biota (e.g. *Usnea*) live on the canopy surface. Pollen can also be source of DOC, particularly in spring and beginning of the summer, when pollen concentrations are high in atmosphere, as preliminary results of Verstraeten et al. (2020) have shown. Animal exudates may also be important DOC sources, especially during insect outbreaks such as Buckley 1987 and Domisch (2009) have indicated. However, there was no sign of insect attacks at our site during the study period. In case of SW there were also significant decreasing DOC concentration under mineral soil layer (20 cm) and slightly positive trend under organic soil layer (5 cm). Lush vegetation in old growth forest and lot of decaying material, explain slightly increasing DOC trend under organic soil layer, however, this effect did not last to the 20 cm depth, where lack of decaying material has led slightly decreasing DOC trend. Other option is that with declining $SO_4$-S inputs the DOC competes more efficiently with $SO_4$-S for adsorption sites in the soil and therefore a decrease in DOC concentration in mineral soil horizons can be expected with decreasing concentration of $SO_4$-S (Gobran and Nilsson 1988). In stream water DOC has a significant increasing trend as has indicated in one of the previous chapters.

In Valkea-Kotinen region, inorganic nitrogen bulk deposition is about 2.5 kg ha$^{-1}$ yr$^{-1}$ and is decreasing. We found also 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 the 1990s, when elevated nitrogen concentrations were common and still are, for example, in central Europe. Similar decreasing trend in N concentration was seen also in litterfall (both foliar and other fraction) but not in green needles. In contrast they had a positive trend, in older needles the trend was significant, which can be explained by more efficient nitrogen retranslocation from senescing needles to the younger needles. 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, which is sign that N emission have decreased during study period. In stream water total N had increasing trend, probably because lot of organic nitrogen has leached from the surrounding terrestrial area to the lake and stream. Large part of the total nitrogen is in organic fraction.

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.

| | Green needles | | Litterfall | |
|---|---|---|---|---|
| | current year | current + 1 year | needle LF | other LF |
| dry weight g m$^{-2}$ yr$^{-1}$ | | | 2.362 | -0.698 |
| N, mg g$^{-1}$ yr$^{-1}$ | 0.229 | **0.184** | **-0.024** | **-0.304** |
| S, mg g$^{-1}$ yr$^{-1}$ | -0.001 | 0.000 | -0.001 | **-0.016** |
| P mg g$^{-1}$ yr$^{-1}$ | -0.009 | -0.005 | 0.001 | **-0.018** |
| Ca, mg g$^{-1}$ yr$^{-1}$ | 0.083 | 0.052 | 0.040 | 0.017 |
| Mg, mg g$^{-1}$ yr$^{-1}$ | -0.012 | -0.002 | -0.008 | -0.042 |
| K, mg g$^{-1}$ yr$^{-1}$ | -0.074 | **-0.079** | **-0.044** | **-0.142** |
| C, % yr$^{-1}$ | 0.000 | -0.095 | **0.084** | 0.044 |

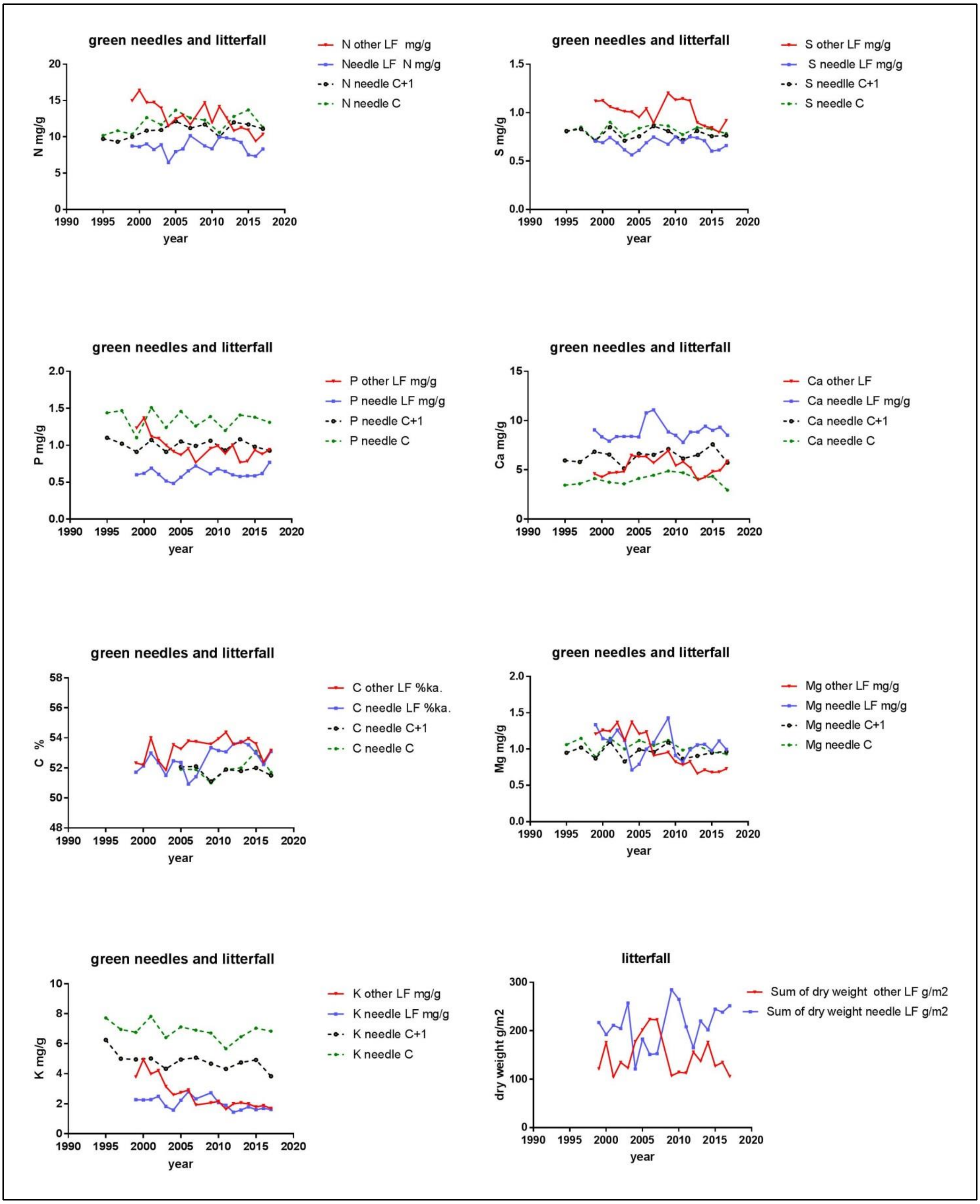

Figure 17. Time series for N, S, P, Ca, C%, Mg and K in green needles (c and c+1) and litterfall (needles and other LF fraction) in 1990–2018. Dry weight trend only in litterfall.

### 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 Valkea-Kotinen area the annual precipitation sum was an average 588 mm and mean temperature 4.3 °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 Valkea-Kotinen region.

| | Year | Annual average temp. | Annual precipitation sum |
|---|---|---|---|
| | | °C | mm |
| 1990–2018 | | 4.3 | 588 |
| Cold years | 2010 | 2.66 | |
| | 1996 | 2.97 | |
| | 1998 | 2.34 | |
| Warm years | 2015 | 5.40 | |
| | 2000 | 5.34 | |
| | 2014 | 5.02 | |
| Dry years | 2002 | | 422 |
| | 2003 | | 472 |
| | 2018 | | 478 |
| Wet years | 1998 | | 678 |
| | 2017 | | 706 |
| | 2012 | | 791 |

In addition, we chose three years when DOC concentrations in TF and TOC concentration in stream water was highest (D/TOC+) or lowest (T/DOC-). TF DOC concentration in DOC+ years was 27 to 58% more than on average and 36 to 38 % less than on average in DOC- years, correspondingly at stream water in TOC+ years was 22 to 27% higher than on average and in TOC- years 14 to 17% lower than on average. Results indicated that during the three driest years, the DOC concentration was highest in TF, while this was not so clear in stream water, only in 2018 there was high TOC concentration in stream water (Fig. 18). In all, there was an indication that both dry and wet periods have effect on DOC concentrations, although we could not see effect of wet season. In 2018 (dry year), $SO_4$-S concentration also increased suddenly in stream water, which was probably due to decreased water table level in surrounding peatland area during dry and hot summer, which resulted in oxidization of sulphur to sulphate in soil, which after rainfall leached to the surface water. 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 1996, 2004 and 2005. 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 conservation area, with old growth forest, high amount of biomass, high biodiversity in vegetation and other biota and the forest structure is diverse compared to managed forest, all these characteristics perhaps protect against the effects of extreme weather events.

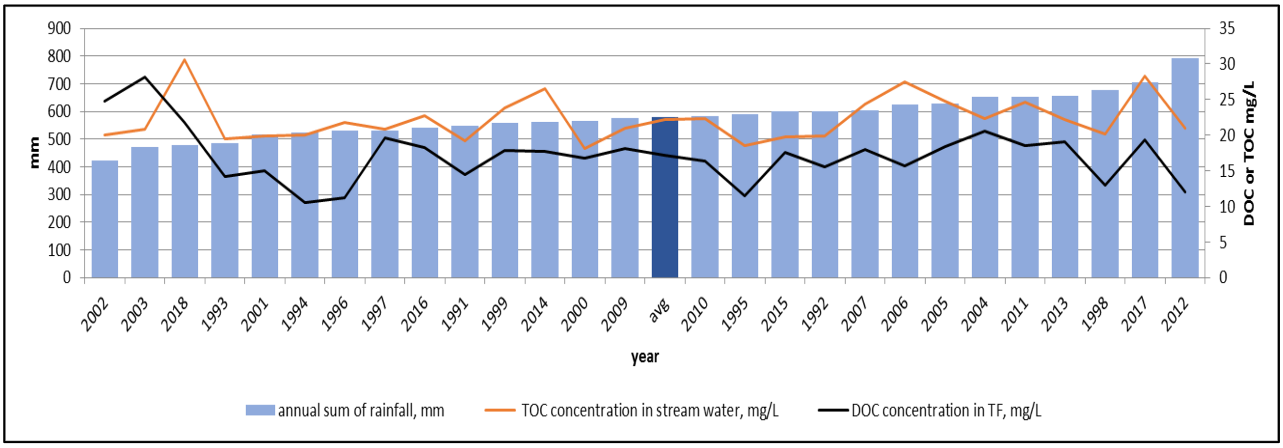

Figure 18. Study years from driest to wettest including DOC concentration in TF and TOC in stream water, dark blue column in the middle is average precipitation amount during study period (1990-2018).

### 3.7.4 Long-term changes in vegetation

#### 3.7.4.1 Stand characteristics

The Valkea-Kotinen forest monitoring site represents an old-growth spruce dominated *Oxalis-Myrtillus* forest type. In addition to spruce (*Picea abies*, 63 % from the total living volume), birch (*Betula* spp., 32 %) and pine (*Pinus sylvestris*, 5 %) grew as mixture (Table 12). The forest is located at a protected area, and there have not been carried out forest management practices. The amount of dead and decaying wood was very high in 2009. The volume of standing dead trees was almost 100 $m^3$ $ha^{-1}$ (Table 1). Birches represented the highest proportion of the volume of dead standing trees. The size of dead spruces was smaller (average stem diameter 8 cm) than that of dead birches (22 cm). There were also lots of lying dead stems on the ground (Fig. 19a), but their volume has not been measured.

The stand was uneven-aged and the eldest trees (which probably are pines) were over 200 years-old (calculated from annual rings of stem discs of cut sample trees). As a habitat for understorey vegetation, the stand was very shady, although there were some canopy openings where the light level was higher. According to the measured dimensions of the crown surface of trees, the total projection cover of crowns was 88 % in 2004–2005. This value is rather high because it does not take radiation filtered through the branches into account. According to visual estimation of crowns carried out by botanists, the canopy cover was 74 % in 2014 and 64 % in 2019. This indicates that the light level has increased probably due to defoliation of tree crowns or falling of dead trees (Figs. 4 and 19b). There was a younger and sparser stand compartment neighbouring the study plot, which may affect the growth conditions of understorey vegetation, too.

Table 12. Stand parameters in the sub-plot no. 1 at Valkea-Kotinen study site in 2009. Basal area and volume have been measured with bark.

| Parameter | Number of stems, ha$^{-1}$ | Basal area, m$^{-2}$ ha$^{-1}$ | Volume, m$^{-3}$ ha$^{-1}$ | Average diameter, cm | Average height, m | Height of lower crown limit, m |
|---|---:|---:|---:|---:|---:|---:|
| Living stock, tot | 992.4 | 49.7 | 596.7 | 22.3 | 20.4 | 11.1 |
| Pine | 16.8 | 2.1 | 27.6 | 40.1 | 29.3 | 21.7 |
| Spruce | 756.9 | 31.8 | 374.6 | 20.0 | 18.1 | 8.4 |
| Birch | 218.7 | 15.8 | 194.5 | 29.9 | 27.9 | 19.8 |
| Dead trees, tot | 420.5 | 8.6 | 98.0 | 13.6 | 15.2 | 10.6 |
| Dead spruces | 252.3 | 2.2 | 19.6 | 8.3 | 8.1 | 4.8 |
| Dead birches | 168.2 | 6.4 | 78.3 | 21.6 | 25.9 | 19.4 |

Figure 19a. Standing and lying dead trees. Anneli Viherä-Aarnio marks the corners of a sample quadrat (2019). Photo: Maija Salemaa.

Figure 19b. Cover of tree crowns in 2019. Photo: Maija Salemaa

### 3.7.4.2 Number and cover of plant species

A total of 71 plant species was found in the field and bottom layers in the Valkea-Kotinen plot during the whole monitoring period 1998–2019, 10 of which were found outside the sample quadrats (Table 13). The shrub layer was scanty, and it consisted of only three species (spruce, birch spp. and rowan - *Sorbus aucuparia*). The total number of vascular plant species in the field layer was 32. The number of herb species was the highest (20), but the number of grass and sedge species was low (4). Species with woody stem included four dwarf shrub and four tree species (8 species). The species number was relatively high in bryophytes (24 in Musci and 10 in Hepaticae). Some cup lichen species (5) were found near tree trunks, too. Although species growing on lying dead wood or on stumps were not recorded, it is possible that there were much small pieces of dead wood inside humus layer offering a substrate for liverworts and cup lichens.

The most abundant species in the field layer was bilberry (*Vaccinium myrtillus*, cover 10–20%), which likes semi-shade conditions. On the other hand, cowberry (*Vaccinium vitis-idaea*) had low abundance (1 %), but it was consistently present on the sample quadrats. Interestingly, we found some “giant shoots” of cowberry in 2009 when they grew near lying decayed stems. The cover of herbs and graminoids was all the time relatively low, 5–6 % and 1 %, respectively. The most abundant herbs *Maianthemum bifolium* and *Melampyrum sylvaticum* are tolerant to shade, but it seems that besides tree canopy also abundant bilberry canopy (at height 10–30 cm) restricted their growth by shading. From grasses the most abundant species was *Calamagrostis arundinacea.*

Table 13. The mean percentage cover (%) and number of plant and lichen species in five different inventories (years 1998, 2003, 2009, 2014 and 2019) and the species occurrence over the years. x = species outside the sample quadrats but inside the area of 400 $m^2$. * = species outside sample quadrats, but inside area of 900 $m^2$. The mean cover values lower than 0.1 have been transformed to 0.1.

| Species group | **1998** | **2003** | **2009** | **2014** | **2019** | **Present** |
|---|---|---|---|---|---|---|
| **Woody species** | | | | | | |
| *Acer platanoides* | | x | | | | x |
| *Frangula alnus* | | * | | | | * |
| *Picea abies* | 0.1 | 0.1 | 0.1 | 0.2 | 0.1 | 1 |
| *Sorbus aucuparia* | 0.6 | 0.1 | 0.3 | 0.0 | 0.1 | 1 |
| *Linnaea borealis* | 0.4 | 0.2 | 0.8 | 0.6 | 0.6 | 1 |
| *Lycopodium annotinum* | | x | | | | x |
| *Vaccinium myrtillus* | 10.3 | 12.2 | 21.3 | 11.6 | 23.3 | 1 |
| *Vaccinium vitis-idaea* | 0.8 | 0.8 | 1.0 | 1.9 | 2.8 | 1 |
| Total cover % | 12.1 | 13.3 | 23.4 | 14.4 | 26.8 | |
| Number of species | 5 | 8 | 5 | 5 | 5 | 8 |
| **Herbs** | | | | | | |
| *Athyrium filix-femina* | | * | | | | * |
| *Convallaria majalis* | 0.2 | 0.2 | 0.1 | 0.1 | 0.1 | 1 |
| *Dryopteris carthusiana* | | * | | | | * |
| *Goodyera repens* | 0.0 | 0.1 | 0.1 | 0.3 | 0.3 | 1 |
| *Gymnocarpium dryopteris* | 0.6 | 0.0 | 0.0 | 0.0 | 0.0 | 1 |
| *Hieracium sp.* | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 1 |
| *Lathyrus vernus* | | * | * | | | * |
| *Maianthemum bifolium* | 2.3 | 2.9 | 2.5 | 1.8 | 1.4 | 1 |
| *Melampyrum pratense* | 0.2 | 0.1 | 0.0 | 0.0 | 1.6 | 1 |
| *Melampyrum sylvaticum* | 0.4 | 0.4 | 0.7 | 3.2 | 2.2 | 1 |
| *Monotropa hypopitys* | | x | | | | x |
| *Orthilia secunda* | 0.1 | 0.1 | 0.0 | 0.0 | 0.1 | 1 |
| *Oxalis acetosella* | 1.1 | 1.2 | 1.0 | 0.9 | 0.2 | 1 |
| *Pyrola minor* | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 1 |
| *Pyrola rotundifolia* | | * | x | x | | * |
| *Rubus saxatilis* | 0.3 | 0.0 | x | 0.0 | 0.0 | 1 |
| *Solidago virgaurea* | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 1 |
| *Trientalis europaea* | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 1 |
| *Veronica chamaedrys* | 0.0 | 0.0 | 0.1 | 0.0 | 0.0 | 1 |
| *Veronica officinalis* | 0.1 | 0.1 | 0.0 | 0.1 | 0.2 | 1 |
| Total cover % | 5.3 | 5.0 | 4.7 | 6.5 | 6.0 | |
| Number of species | 14 | 14 | 11 | 9 | 11 | 20 |
| **Grasses and sedges** | | | | | | |
| *Calamagrostis arundinacea* | 0.6 | 0.4 | 0.3 | 0.5 | 0.2 | 1 |
| *Carex digitata* | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 1 |
| *Deschampsia flexuosa* | 0.1 | 0.1 | 0.1 | 0.6 | 0.3 | 1 |
| *Luzula pilosa* | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 1 |
| Total cover % | 0.9 | 0.6 | 0.5 | 1.2 | 0.8 | |
| Number of species | 4 | 4 | 4 | 4 | 4 | 4 |

Table 13 continues.

| Species group | **1998** | **2003** | **2009** | **2014** | **2019** | **Present** |
|---|---|---|---|---|---|---|
| **Bryophytes** | | | | | | |
| **Musci** | | | | | | |
| *Brachythecium mildeanum* | 0.00 | 0.00 | 0.02 | 0.00 | 0.00 | 1 |
| *Sciuro-hypnum curtum* | 0.01 | 0.06 | 0.01 | 0.10 | 0.01 | 1 |
| *Sciuro-hypnum reflexum* | 0.00 | 0.21 | 0.01 | 0.01 | 0.00 | 1 |
| *Brachythecium salebrosum* | 0.00 | 0.18 | 0.00 | 0.00 | 0.00 | 1 |
| *Sciuro-hypnum starkei* | 0.00 | 0.30 | 0.09 | 0.02 | 0.00 | 1 |
| *Ceratodon purpureus* | | x | | | | x |
| *Dicranum flexicaule* | 0.00 | 0.01 | 0.00 | 0.00 | 0.00 | 1 |
| *Dicranum fuscescens* | 0.06 | 0.01 | 0.01 | 0.01 | 0.08 | 1 |
| *Dicranum majus* | 0.36 | 0.81 | 0.25 | 0.21 | 0.35 | 1 |
| *Dicranum montanum* | 0.00 | 0.01 | 0.00 | 0.00 | 0.01 | 1 |
| *Dicranum polysetum* | 0.10 | 0.10 | 0.01 | 0.18 | 0.39 | 1 |
| *Dicranum scoparium* | 0.74 | 2.41 | 0.73 | 1.12 | 0.26 | 1 |
| *Hylocomium splendens* | 15.24 | 31.44 | 35.54 | 44.88 | 44.31 | 1 |
| *Hypnum pallescens* | 0.00 | 0.01 | 0.01 | 0.00 | 0.00 | 1 |
| *Hypnum recurvatum* | 0.00 | 0.01 | 0.00 | 0.00 | 0.00 | 1 |
| *Plagiothecium denticulatum* | 0.00 | 0.02 | 0.00 | 0.00 | 0.00 | 1 |
| *Plagiothecium laetum* | 0.06 | 0.15 | 0.11 | 0.13 | 0.01 | 1 |
| *Plagiothecium curvifolium* | 0.00 | 0.01 | 0.00 | 0.00 | 0.00 | 1 |
| *Platygyrium repens* | 0.00 | 0.01 | 0.01 | 0.00 | 0.00 | 1 |
| *Pleurozium schreberi* | 1.76 | 5.00 | 6.16 | 5.27 | 7.25 | 1 |
| *Pohlia nutans* | 0.00 | 0.01 | 0.00 | 0.00 | 0.00 | 1 |
| *Rhytidiadelphus triquetrus* | 0.16 | 0.46 | 0.19 | 0.27 | 0.56 | 1 |
| *Sanionia uncinata* | 0.00 | 0.11 | 0.13 | 0.00 | 0.00 | 1 |
| *Schistostega pennata* | | x | x | | | x |
| Total cover % | 18.47 | 41.25 | 43.25 | 52.18 | 53.21 | |
| Number of species | 9 | 22 | 15 | 11 | 10 | 24 |
| | | | | | | |
| **Hepaticae** | | | | | | |
| *Blepharostoma trichophyllum* | 0.00 | 0.02 | 0.01 | 0.01 | 0.00 | 1 |
| *Cephalozia bicuspidata* | 0.00 | 0.00 | 0.01 | 0.00 | 0.00 | 1 |
| *Chiloscyphus latifolius* | 0.00 | 0.01 | 0.01 | 0.00 | 0.00 | 1 |
| *Chiloscyphus polyanthos* | 0.00 | 0.01 | 0.01 | 0.00 | 0.00 | 1 |
| *Chiloscyphus profundus* | 0.00 | 0.02 | 0.01 | 0.02 | 0.00 | 1 |
| *Jamesionella autumnalis* | 0.00 | 0.01 | 0.00 | 0.00 | 0.00 | 1 |
| *Lepidozia reptans* | 0.00 | 0.01 | 0.06 | 0.03 | 0.00 | 1 |
| *Lophozia ventricosa* | 0.00 | 0.01 | 0.00 | 0.00 | 0.00 | 1 |
| *Ptilidium ciliare* | 0.00 | 0.01 | 0.00 | 0.00 | 0.00 | 1 |
| *Ptilidium pulcherrinum* | 0.00 | 0.01 | 0.00 | 0.08 | 0.00 | 1 |
| Total cover % | 0.00 | 0.06 | 0.07 | 0.04 | 0.00 | |
| Number of species | 0 | 9 | 6 | 4 | 0 | 10 |

Table 13 continues.

| Species group | 1998 | 2003 | 2009 | 2014 | 2019 | Present |
|---|---|---|---|---|---|---|
| **Lichens** | 0.00 | 0.10 | 0.0 | 0.00 | 0.00 | 1 |
| *Cladonia coniocraea* | | | | | | |
| *Cladonia cornuta* | 0.00 | 0.01 | 0.00 | 0.06 | 0.00 | 1 |
| *Cladonia digitata* | 0.00 | 0.01 | 0.00 | 0.00 | 0.00 | 1 |
| *Cladonia fimbriata* | 0.00 | 0.01 | 0.00 | 0.00 | 0.00 | 1 |
| *Cladonia squamosa* | 0.00 | 0.03 | 0.00 | 0.00 | 0.00 | 1 |
| Total cover % | 0.000 | 0.153 | 0.001 | 0.063 | 0.000 | |
| Number of species | 0 | 5 | 1 | 1 | 0 | 5 |
| **All species** | | | | | | |
| Number of vasculars | 23 | 20 | 18 | 18 | 20 | 32 |
| Number of cryptogams | 9 | 35 | 20 | 16 | 10 | 39 |

### 3.7.4.3 Changes in species number and cover

The total percentage cover of the understorey vegetation has increased in the Valkea-Kotinen monitoring plot during the period 1998–2019 (Fig. 20a). The sampling design changed after 1998, and therefore results concerning comparisons between 1998 and 2003–2019 should be interpreted with caution. Anyway, both the cover of bryophytes (mainly *Hylocomium splendens*) and dwarf shrubs (mainly *Vaccinium myrtillus*) have increased at least 10 % and the total cover more than 20 % during 2003–2019 (Fig. 20 b,c, Table 13). If we include the year 1998 to the comparison, the increasing trend is much higher – 30 % for bryophytes and 40 % for the total cover. Herbs showed a slight increase during 2009–2019, too. The number of vascular species in different years remained relatively stable, the highest variation was found in the number of herbs. The species identification of bryophytes is demanding in field conditions and often samples for microscopic analysis are needed. The number of bryophyte species was highest in the year 2003 (Table 13) when we took many samples which were identified by a professional taxonomist (M.Sc. Nijole Kalinauskaite).

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. The cover of bilberry was slightly lower in the years with high temperature sum and low precipitation sum (years 2003 and 2014) which indicated dry growing conditions (Fig. 21a). On the other hand, the cover of bilberry increased in 2019 when precipitation sum was high (Fig. 21b) and the previous year (2018, not shown in the figure) was warm. In addition to nutrient and water availability, also the amount of light regulates the abundance of plant species in forests habitats (Verhayen et al. 2012, Tonteri et al. 2016). It is possible that particularly bilberry and herbs have benefited from the increase of light level due to decrease in canopy cover in otherwise shady forest.

According to modelling scenarios (Villèn-Perèz et al. 2020), the distribution areas of many boreal plant species shift northwards in the future due to increasing temperature. In addition, the areas of the highest abundance of cowberry, many herbs, grasses and bryophytes move to more suitable climatic areas. Bilberry increased in the Valkeajärvi-Kotinen area during 1998–2019, but according to a simulation model (Frolov et al. 2020) bilberry suffers more than cowberry from the effects of climate change. Thin leaves of bilberry are more sensitive to drought than thicker wax-covered leaves of cowberry. On the other hand, the rhizomes and roots of bilberry can grow to deep soil layers which may protect them from drying. Although dry periods will restrict the growth of bilberry, its shoots have good regrowth ability after damage (Tolvanen et al. 1994), which helps bilberry to survive over stressful periods.

a)

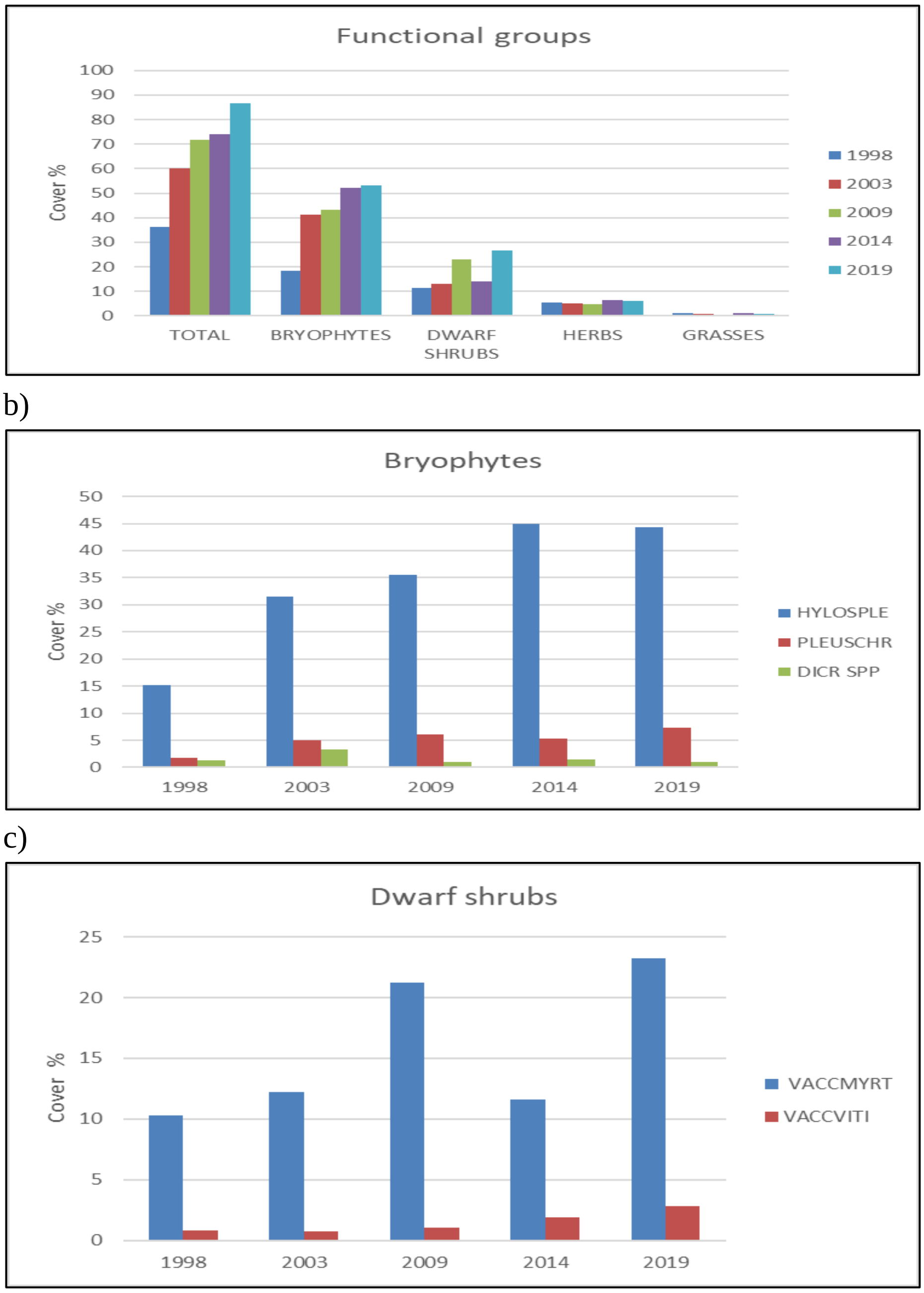

Figure 20. The cover (%) of understorey vegetation and its species groups during 1998–2019. a) sum of all species (TOTAL), and separately bryophytes, dwarf shrubs, herbs and grasses, b) bryophyte species *Hylocomium splendens* (HYLOSPLE), *Pleurozium schreberi* (PLEUSCHR) and *Dicranum* spp. (DICR SPP) (mostly *D. majus*, *D. polysetum* and *D. scoparium*, Table 2) and c) dwarf shrubs *Vaccinium myrtillus* (VACCMYRT) and *V. vitis-idaea* (VACCVITI).

The total cover of bryophytes correlated negatively with the cover of dead birch leaves laying on the forest floor. Thus, the cover of bryophytes increased when the cover of leaf litter (Fig. 21c) or total litter (Fig. 21d) decreased. We suggest that the amount of leaf litter on the ground has decreased because many birch trees have decayed and dead. We found quite recently fallen birch stems in the monitoring plot in 2019 (Fig. 7a). The cover of *Hylocomium splendens* increased in the old-growth spruce forests during the monitoring period. It has an ability to spread wide shoot segments over the other bryophyte species, which probably gives competition advantage to it in shady environment.

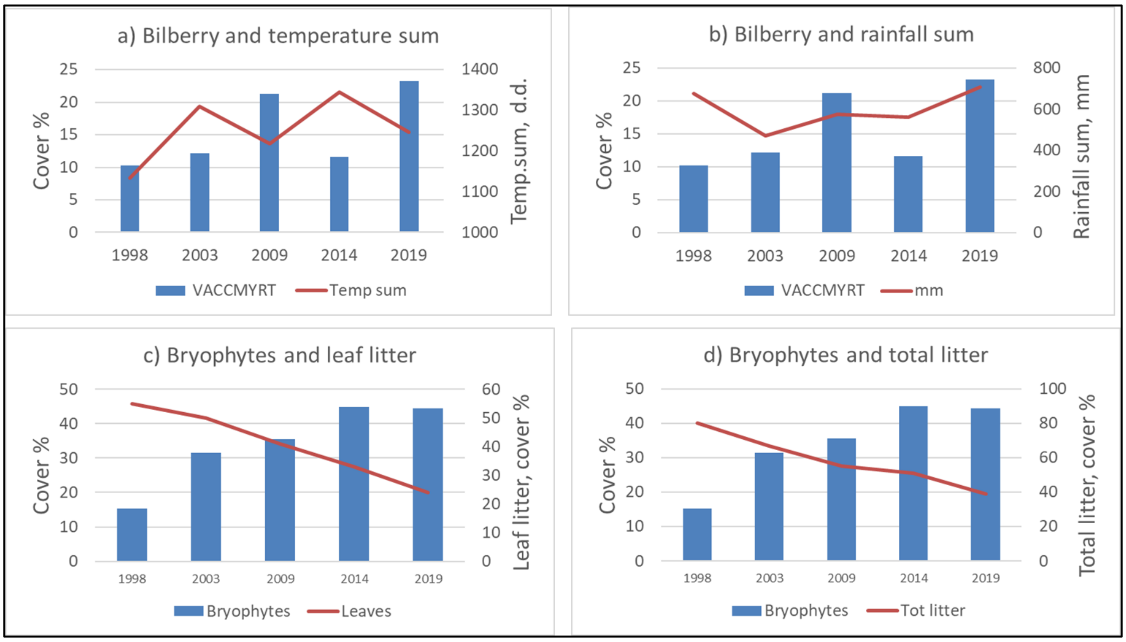

Figure 21. The relationship between the cover (%) of bilberry and a) annual temperature sum (daily degrees) and b) annual rainfall sum (mm). The relationship between the total cover (%) of bryophytes and cover of c) dead leaves and d) total litter (leaves and other dead material dropped from tree crowns) on the ground.

### 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 Valkea-Kotinen study plot (Table 7). The total inorganic N deposition (TIN=$NH_4$-N + $NO_3$-N) was 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. Thus, the amount of DON and TIN deposition were at the same level in the forest habitat. Bryophytes uptake N mainly from precipitation through their surfaces, which makes them competent bioindicators of N deposition. In experimental exposures (Forsum et al. 2009) and correlative analyses (Salemaa et al. 2020) DON has been shown to serve as an important source of N for boreal bryophytes, which is probably true also in the Valkea-Kotinen forest. The total N concentration of the upper part of the bryophyte species in 2009 were as follows: *Hylocomium splendens* 1.70%, *Pleurozium schreberi* 1.58%, *Dicranum* spp. 2.11% and other species 1.71%. The occurrence of nitrophilic bryophytes

(*Sciuro-hypnum*, *Brachythecium* and *Plagiothecium* genera) indicates relatively high nitrogen level of the site.

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 Valkea-Kotinen plot across the years 2006, 2007 and 2009.

| Habitat | Collection | Precipitation | $NH_4$-N | $NO_3$-N | DON | TIN | $N_{tot}$ |
|---|---|---|---|---|---|---|---|
| Open site | BP | 597 | 1.13 | 1.50 | 0.49 | 2.63 | 3.12 |
| Forest | TF | 410 | 0.57 | 1.00 | 1.77 | 1.57 | 3.34 |

We applied a model equation (Salemaa et al. 2020) using the N concentration of the most abundant moss species (*Hylocomium splendens*) as a predictor for total annual N ($N_{tot}$) deposition in Valkea-Kotinen study area. The used equation is based on the data from 15 Finnish ICP Forests Level II plots, including Valkea-Kotinen plot. The model produced higher estimate for $N_{tot}$ in bulk deposition (4.7 kg $ha^{-1}yr^{-1}$) and lower estimate for TF (2.5 kg $ha^{-1}yr^{-1}$) than the measured $N_{tot}$ deposition values. Their average 3.6 kg N $ha^{-1}yr^{-1}$ was quite near the measured values (Table 14). Applying the modelled open site N concentration for *H. splendens* (1 %) in the equation (method in Salemaa et al. 2020), the estimate for $N_{tot}$ in bulk deposition was 3.9 kg N $ha^{-1}$. The slightly higher values for predicted $N_{tot}$ deposition may be caused by the effect of N leached from litterfall (Ukonmaanaho and Starr 2001) and absorbed by bryophytes as seen as higher N concentration in their tissue.

N concentration of bryophytes, herbs, grasses and bilberry leaves were at the same level (1.7–2.0 %) (Table 15). In bilberry stems and cowberry stems and leaves the N concentration varied from 0.7% to 1.0%. The bryophyte layer (only living upper parts of the shoots included) consisted of almost the same amount of biomass (319 kg $ha^{-1}$) as bilberry and cowberry layer (318 kg $ha^{-1}$). Their C contents were quite similar, too. However, the bryophyte layer consisted more N than the bilberry and cowberry layer, because the stems of dwarf shrubs had relatively low N concentration (0.7–0.9%).

The TF deposition level of 3.3 kg $ha^{-1}$ and bryophyte N concentration of 1.48% have been found to cause inhibition of biological $N_2$ fixation by cyanobacteria living on *H. splendens* (Salemaa et al. 2019). The Valkea-Kotinen monitoring plot showed corresponding N deposition level and higher N concentration of bryophytes (1.7 %) in 2009 than mentioned above. It is possible that bryophytes were approaching N saturated stage already at this N level in forest. DON passing through tree canopies (throughfall) and N leachate from litterfall have a big role as N supply for forest bryophytes (Salemaa et al. 2020). Although TIN in bulk deposition has showed a decreasing trend in Valkea-Kotinen study area during 1990–2017 (Table 9), the earlier higher N deposition might have been accumulated in the forest ecosystem, and now it appeared as high N concentration in bryophytes. Our results emphasize the importance of bryophyte layer in accumulating different N forms and affecting internal N flux of the forest ecosystem. Already N deposition level < 5 kg $ha^{-1}$ $yr^{-1}$ may cause slow changes in the boreal forest ecosystem, e.g. in the structure of bryophyte communities (cf. Nordin et al. 2005). This should be considered when evaluating the critical N loads for the most sensitive organisms of boreal forests.

Table 15. N and C concentrations (%) and quantities (kg dry weight $ha^{-1}$) in different plant groups and plant parts. Data is based on 28 samples of aboveground biomass on 30 cm x 30 cm quadrats.

| | N | C | Mean mass | N | C |
|---|---|---|---|---|---|
| | % | % | kg d.w. $ha^{-1}$ | kg $ha^{-1}$ | kg $ha^{-1}$ |
| Bryophytes | 1.7 | 48.8 | 316.4 | 5.3 | 154.6 |
| Herbs | 1.7 | 45.5 | 17.6 | 0.3 | 8.0 |
| Grasses | 2.0 | 45.6 | 36.3 | 0.7 | 16.6 |
| *V. myrtillus* leaves | 2.0 | 51.2 | 60.9 | 1.2 | 31.2 |
| *V. myrtillus* stems | 0.9 | 51.9 | 232.1 | 2.0 | 120.5 |
| *V. vitis-idaea* leaves | 1.1 | 53.3 | 16.2 | 0.2 | 8.6 |
| *V. vitis-idaea* stems | 0.7 | 51.6 | 8.5 | 0.1 | 4.4 |

## 4. Conclusions

Lake Valkea-Kotinen demonstration site is a pristine, sensitive Natura 2000 area in South 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 are no longer exceeded at the catchment area of Valkea-Kotinen. In concert with decreasing eutrophication critical loads, also the inorganic nitrogen concentrations have decreased. However, in the forest area of the catchment the N concentration of bryophytes is still relatively high (1.7 %). This may indicate that earlier higher N deposition has been accumulated in the forest ecosystem, and it now appears as high N concentration in bryophytes. Therefore, it is possible that slow changes in the forest ecosystem, e.g. in the structure of bryophyte communities take place already at N deposition level < 5 kg $ha^{-1}$ $yr^{-1}$. This is an important point when evaluating the critical N loads for the most sensitive organisms of boreal forests.

Precipitation is strongly modified before it enters from the terrestrial part of the Valkea-Kotinen catchment to the surface water. Concentrations of inorganic nitrogen decreased due to the uptake of tree canopy and other vegetation, while for example base cation concentrations mainly increased. Long-term trends in concentrations confirmed that observed decrease in $SO_4$-S deposition was seen also in different parts of the forest ecosystem. Instead, increase in DOC concentration in L. Valkea-

Kotinen, as has been observed in surface waters throughout Europe and North America, was not noticeable in the forest part of the Valkea-Kotinen region, only DOC concentration in throughfall increased significantly over studied period. Nutrition status of green needles was stable, only K concentration decreased significantly in one-year old needles, similar decreasing trend was found also in litterfall. We could not find very clear effects of extreme weather events to the forest ecosystem, which probably indicates the resilience of the old growth forest of Valkea-Kotinen region.

Over 20 years monitoring has produced valuable knowledge of the succession of undisturbed old-growth forest in Valkea-Kotinen, but it is still difficult to draw conclusions how climate change affects understorey vegetation. The cover of bilberry showed an increasing trend during the monitoring period 1998 - 2019, but its cover decreased in dry years. It is probable that bilberry benefits from a slight increase of temperature, but it suffers if extreme dry periods in summers are yearly repeated and stay long time. If bilberry sheds dry leaves repeatedly, it can be detrimental to its carbon assimilation and ability to recover after damage. On the other hand, snowless winters may cause freezing of shoots. Shading trees act against the effects of climate change, because canopy prevents heating of the forest floor. The canopy cover of the study plot decreased during the monitoring period possibly due to crown defoliation and falling of old and decayed trees. The cover of bryophytes showed interaction with the amount of leaf and needle litter on the ground. The nitrogen concentration of bryophytes was relatively high (1.7 %) compared to other spruce plots in southern Finland. It could be possible that bryophytes were approaching nitrogen saturation stage at the measured nitrogen concentration of tissues. If the excess of nitrogen in TF deposition is available for other organisms and plants, and amount of light increases because tree falling, eutrophication of plant communities is possible (Verheyen et al. 2012, Dirnböck et al. 2014). Therefore, nitrogen economy of bryophytes has many effects on the function of boreal 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.

## 5. References

Aber, J.D. & Melillo, J.M. (Eds.) 1991.Terrestrial Ecosystems. Saunders College Publishing. ISBN 0-03-047443-4

Adrian, R., O'Reilly, C.M., Zagarese, H., Baines, S.B., Hessen, D.O., Keller, W., Livingstone, D.M., Sommaruga, R., Straile, D., Van Donk, E., Weyhenmeyer, G.A. & Winder, M. 2009. Lakes as sentinels of climate change. Limnol. Oceanogr. 54: 2283–2297.

Amann, M., Bertok, I., Borken-Kleefeld, J., Cofala, J., Heyes, C., Höglund-Isaksson, L., Klimont, Z., Nguyen, B., Posch, M., Rafaj, P., Sandler, R., Schöpp, W., Wagner, F. & Winiwarter, W. 2011. Cost-effective control of air quality and greenhouse gases in Europe: Modeling and policy applications. Environmental Modelling & Software 26 (12): 1489−1501.

Andersson, S., Nilsson, S.I., & Saetre, P. 2000. Leaching of dissolved organic carbon (DOC) and dissolved organic nitrogen (DON) in mor humus as affected by temperature and pH. Soil Biology and Biochemistry 32: 1−10.

Arvola, L., Salonen, K., Keskitalo, J., Tulonen, T., Järvinen, M. & Huotari, J. 2014. Plankton metabolism and sedimentation in a small boreal lake — a long-term perspective. Boreal Env. Res. 19 (suppl. A): 83–96.

Berdén, M., Nilsson, S., Rosèn, K., & Tyler, G. 1987. Soil acidification – extent, causes and consequences. An evaluation of literature information and current research. National Swedish environment Protection Board. Report 3292. 164 p.

Bergstedt, J. & Milberg, P. 2001. The impact of logging intensity on field-layer vegetation in Swedish boreal forest. Forest Ecology and Management 154: 105–115.

Bergström, A. & Jansson, M. 2006. Atmospheric nitrogen deposition has caused nitrogen enrichment and eutrophication of lakes in the northern hemisphere. Global Change Biology 12: 635–643.

Bobbink, R., Hicks, K., Galloway, J., Spranger, T., Alkemade, R., Ashmore, M., Bustamante, M., Cinderby, S., Davidson, E., Dentener, F., Emmett, B., Erisman, J.W., Fenn, M., Gilliam, F., Nordin, A., Pardo, L. & de Vries, W. 2010. Global assessment of nitrogen deposition effects on terrestrial plant diversity: a synthesis. Ecol. Appl. 20: 30–59.

Bobbink, R. & Hettelingh, J.-P. 2011. Review and revision of empirical critical loads and dose-response relationships. Proceedings of an expert workshop, Noordwijkerhout 23-24 June 2010. Report 680359002, RIVM, Bilthoven, the Netherlands.

Buckley, R. (1987). Ant-plant–Homopteran interactions. Advances in Ecological Research 16: 53−85.

Colette, A., Aas, W., Banin, L., Braban, C. F., Ferm, M., González Ortiz, A., Ilyin, I., Mar, K., Pandolfi, M., Putaud, J.-P., Shatalov, V., Solberg, S., Spindler, G., Tarasova, O., Vana, M., Adani, M., Almodovar, P., Berton, E., Bessagnet, B., Bohlin-Nizzetto, P., Boruvkova, J., Breivik, K., Briganti, G., Cappelletti, A., Cuvelier, K., Derwent, R., D'Isidoro, M., Fagerli, H., Funk, C., Garcia Vivanco, M., Haeuber, R., Hueglin, C., Jenkins, S., Kerr, J., de Leeuw, F., Lynch, J., Manders, A., Mircea, M., Pay, M. T., Pritula, D., Querol, X., Raffort, V., Reiss, I., Roustan, Y., Sauvage, S., Scavo, K., Simpson, D., Smith, R. I., Tang, Y. S., Theobald, M., Tørseth, K., Tsyro, S., van Pul, A., Vidic, S., Wallasch, M. & Wind, P. 2012. Air pollution trends in the EMEP region between 1990 and 2012. Joint Report of the EMEP Task Force on Measurements and Modelling (TFMM), Chemical Co-ordinating Centre (CCC), Meteorological Synthesizing Centre-East (MSC-E), Meteorological Synthesizing Centre-West (MSC-W). Norwegian Institute for Air Research, Kjeller, Norway.

Davies, C.E., Moss, D. & Hill, M.O. 2004. 'EUNIS habitat classification revised 2004. Report to: European Environment Agency-European Topic Centre on Nature Protection and Biodiversity' (http://www.eea.europa.eu/themes/biodiversity/eunis/eunis-habitat-classification).

De Vries, W., Reinds, G.J., van der Salm, C., Draaijers, G.P.J., Bleeker, A., Erisman, J.W., Auee, J., Gundersen, P., Kristensen, H.L., van Doben, H., de Zwart, D., Derome, J., Voogd, J.C.H., Vel, E., 2001. Intensive Monitoring of Forest Ecosystems in Europe. Technical Report 2001. Forest Intensive Monitoring Coordinating Institute, Heerenveen, The Netherlands.

De Vries, W., van der Salm, C., Reinds, G.J., Dise, N.B., Gundersen, P., Erisman, J.W. & Posch, M. 2003. Assessment of the dynamics in nitrogen and carbon sequestration of European forest soils. Alterra-Report 818. Alterra, Wageningen, the Netherlands.

de Wit, H., Hettelingh, J.-P. & Harmens, H. (Eds.) 2015. Trends in ecosystem and health responses to long-range transported atmospheric pollutants. ICP Waters report 125/2015, No. 6946-2015, Norwegian Institute for Water Research, Oslo.

de Wit, H.A., Valinia, S., Weyhenmeyer, G.A., Futter, M.N., Kortelainen, P., Austnes, K., Hessen, D.O., Raike, A., Laudon, H. & Vuorenmaa, J. 2016. Current Browning of Surface Waters Will Be Further Promoted by Wetter Climate. Environmental Science and Technology Letters 12 (3): 430–435.

Dinsmore, K.J., M.F Billett, and K.E Dyson. 2013. Temperature and precipitation drive temporal variability in aquatic carbon and GHG concentrations and fluxes in a peatland catchment. Global Change Biology 19 (7): 2133–2148.

Dirnböck, T., Grandin, U., Bernhardt-Römermann, M., Beudert, B., Canullo, R., Forsius, M., Grabner, M-T.,Holmberg, M., Kleemola, S., Lundin, L., Mirtl, M., Neumann, M., Pompei, E., Salemaa, M., Starlinger, F., Staszewski, T. & Uzięblo, A. K. 2014. Forest floor vegetation response to nitrogen deposition in Europe. Global Change Biology 20: 429–440.

Dise, N.B. & Wright, R.F. 1995. Nitrogen leaching from European forests in relation to nitrogen deposition. For. Ecol. Manage. 71: 153–162.

Domisch, T., Finér, L., Neuvonen, S., Niemelä, P., Risch, A.C., Kilpeläinen, J., Ohashi, M., & Jurgensen, M.F. 2009. Foraging activity and dietary spectrum of wood ants (Formica rufa group) and their role in nutrient fluxes in boreal forests. Ecological Entomology 34(3): 369−377.

Draaijers, G.P.J. & Erisman, J.W. 1995. A canopy budget model to assess atmospheric deposition from throughfall measurements. Water Air Soil Pollut. 85: 2253–2258.

Forsius, M., Posch, M., Holmberg, M., Vuorenmaa, J., Kleemola, S., Augustaitis, A., Beudert, B., Bochenek, W., Clarke, N., de Wit, H.A., Dirnböck, T., Frey, J., Grandin, U., Hakola, H., Kobler, J., Krám, P., Lindroos, A.-J., Löfgren, S., Pecka, T., Rönnback, P., Skotak, K., Szpikowski, J., Ukonmaanaho, L., Valinia, S., Váňa, M.. Assessing critical load exceedances and ecosystem impacts of anthropogenic nitrogen and sulphur deposition at unmanaged forested catchments in Europe. In review 2020 with Science of the Total Environment.

Forsum, Å., Dahlman, L., Näsholm, T. & Nordin, A. 2006. Nitrogen utilization by Hylocomium splendens in a boreal forest fertilization experiment. Functional Ecology 20: 421–426.

Fröberg, M., Berggren, D., Bergkvist, B., Bryant, C., & Mulder, J. 2006. Concentration and fluxes of dissolved organic carbon (DOC) in three Norway spruce stands along a climatic gradient in Sweden. Biogeochemistry 77(1): 1−23.

Frolov, P., Zubkova, E., Shanin, V., Bykhovets, S., (Data Curation), Makipaa, R. & Salemaa, M. 2020. CAMPUS-S – The model of ground layer vegetation populations in forestecosystems and their contribution to the dynamics of carbon and nitrogen.II. Parameterization, validation and simulation experiments. Ecological Modelling 431, 109183.

Gobran, G. R., & Nilsson, S.I. 1988. Effects of forest floor leachate on sulfate retention in a spodosol soil. Journal of Environmental Quality 17(2): 235−239

Gosz, J.R. 1981. Nitrogen cycling in coniferous ecosystems. In: Clark, F.E. & Roswell, T. (eds.), Terrestrial nitrogen cycles. Ecological Bulletins, Stockholm 33: 405−426.

Grennfelt, P., Engleryd, A., Forsius, M., Hov, Ø., Rodhe, H. & Cowling. E. 2020. Acid rain and air pollution– 50 years of progress in environmental science and policy. Ambio 49: 849–864.

Henriksen, A., Posch, M., 2001. Steady-state models for calculating critical loads of acidity for surface waters. Water Air Soil Pollut. Focus 1: 375-398.

Hirsch, R.M., Slack, J.R., Smith, R.A., 1982. Nonparametric tests for trend in water quality. Water Resour. Res. 18: 107–121.

Holmberg, M., Vuorenmaa, J., Posch, M., Forsius, M., Lundin, L., Kleemola, S., Augustaitis, A., Beudert, B., de Wit, H.A., Dirnböck, T., Evans, C.D., Frey. J., Grandin, U., Indriksone. I., Krám, P., Pompei, E., Schulte-Bisping, H., Srybnyn, A., Vána, M., 2013. Relationship between critical load exceedances and empirical impact indicators at Integrated Monitoring sites across Europe. Ecol. Ind. 24: 256–265.

Ilvesniemi, H., Pumpanen, J., Duursma, R., Hari, P., Keronen, P., Kolari, P., Kulmala, M., Mammarella, I., Nikinmaa, E., Rannik, Ü., Pohja, T., Siivola, E. & Vesala, T. 2010: Water balance of a boreal Scots pine forest. Boreal Env. Res. 15: 375–396.

IPPC 2014. Climate change (2014). Synthesis Report. Contribution of working groups I,, II and III to 5th assessment Report of the Intergovernmental Panel on Climate Change (core writing team, R:K. Pachauri and L.A. Meyrer (eds.)). IPCC, Geneva, Switzerland, 1-104.

Johnson, D.W.1992. Base cations, introduction. In: Johnson,D.W. & Lindberg, S.E. (editors). Atmospheric deposition and forest nutrient cycling. Ecological studies 91: 232–235

Jonard, M., Fürst, A., Verstraeten, A., Thimonier, A., Timmermann, V., Potočić, N., Waldner, P., Benham, S., Hansen, K., Merilä, P., Ponette, Q., De La Cruz, A.C., Roskams, P., Nicolas, M., Croisé, L., Ingerslev, M., Matteucci, G., Decinti, B., Bascietto, M. & Rautio, P. 2014. Tree mineral nutrition is deteriorating in Europe. Global Change Biology 21: 418–430.

Jylhä, K., Laapas, M., Ruosteenoja, K., Arvola, L., Drebs, A., Kersalo, J., Saku, S., Gregow, H., Hannula, H.-R. and Pirinen, P. 2014. Climate variability and trends in the Valkea-Kotinen region, southern Finland: comparisons between the past, current and projected climate. Boreal Environ. Res. 19 (suppl. A): 4–30.

Kalbitz, K., Solinger S., Park, J-H., Michalzik, B., & Matzner E. 2000. Controls on the dynamics of dissolved organic matter in soils: A review. Soil Science 165: 728−736.

Kimmins, J.P. 1987. Forest Ecology. MacMillan p. 531

Kokko, A., Mäkelä,K, Tuominen S. 2002. Aluskasvillisuuden seuranta Suomen ympäristön yhdennetyn seurannan alueella 1988-1998. Suomen ympäristö 544, 97 http://www.vyh.fi/palvelut/julkaisu/elektro/sy544/sy544.htm

Kortelainen, P., Mattsson, T., Finér, L., Ahtiainen, M., Saukkonen, S. & Sallantaus, T. 2006. Controls on the export of C, N, P and Fe from undisturbed boreal catchments, Finland. Aquat. Sci. 68: 453–468.

Lindroos A.-J., Derome J., Derome K. & Smolander A. 2011. The effect of Scots pine, Norway spruce and silver birch on the chemical composition of stand throughfall and upper soil percolation water in northern Finland. Boreal Env. Res. 16: 240–250.

Lindroos, A.-J., Derome, J., Mustajärvi, K., Nöjd, P., Beuker, E., & Helmisaari, H.-S. 2008. Fluxes of dissolved organic carbon in stand throughfall and percolation water in 12 boreal coniferous stands on mineral soils in Finland. Boreal Environment Research, 13 (suppl. B): 22−34.

Lyman, J. & Fleming, R.H. 1940. Composition of seawater. J. Mar. Res. 3: 134–146.

Merilä, P., Kilponen T. & Derome J. (eds.) 2007. Forest condition monitoring in Finland – National Report 2002–2005.Working papers of the Finnish Forest Research Institute 45, 166 p.

Mälkönen, E. 1974. Annual primary production and nutrient cycle in some Scots pine stands. Comm.Inst.For.Fenn. 84(5):1–87.

Mapping Manual 2017. German Environment Agency, Coordination Centre for Effects. www.icpmapping.org, Accessed date: 3 April 2020.

Michalzik, B., Kalbitz, K., Park, J. H., Solinger, S., & Matzner, E. (2001). Fluxes and concentrations of dissolved organic carbon and nitrogen-a synthesis for temperate forests. Biogeochemistry 52(2): 173−205.

Mitchell, M.J., Driscoll, C.T., McHale, P.J., Roy, K.M. & Dong, Z. 2013. Lake/watershed sulfur budgets and their response to decreases in atmospheric sulfur deposition: watershed and climate controls. Hydrological Processes 27: 710–720.

Mitchell, M.J. 1992. Retention of loss of sulfur for IFS sites and evaluation of relative importance of processes. In: Johnson, D.W. & Lindberg, S.E. (editors). Atmosphgeric deposition and forest nutrient cycling. Ecological studies 91: 129–133.

Monteith D.T., Stoddard J.L., Evans C.D., de Wit H.A., Forsius M., Høgåsen T., Wilander A., Skjelkvåle B.L., Jeffries D.S., Vuorenmaa J., Keller, B., Kopáček J. & Vesely J. 2007. Dissolved organic carbon trends resulting from changes in atmospheric deposition chemistry. Nature 450: 537–540.

Moore, K., Jennings, E., Allott, N., May, L., Järvinen, M., Arvola, L., Tamm, T., Järvet, A., Nöges, T., Pierson, D. & Schneiderman, E. 2010. Modelling the effects of climate change on inorganic nitrogen transport from catchments to lakes, in: George, D.G. (Ed.), The impact of climate change on European lakes, Aquatic Ecol. Series 4, Springer, Amsterdam, pp. 179–197.

Munson, R.K. & Gherini, S.A. 1993. Influence of organic acids on the pH and acid-neutralizing capacity of Adirondack lakes. Water Resour. Res. 29: 891–899.

Mustajärvi, K., Merilä, P., Derome, J., Lindroos, A.-J., Helmisaari, H.-S., Nöjd, P., & Ukonmaanaho, L. 2008. Fluxes of dissolved organic and inorganic nitrogen in relation to stand characteristics and latitude in Scots pine and Norway spruce stands in Finland. Boreal Envronment Research 13 (supp.B): 3−21.

Nordin, A., Strengbom, J., Witzell, J., Näsholm, T. & Ericson, L. 2005. Ambio 34(1): 20–24.

Neal, C., Reynolds, B., Neal, M., Pugh, B., Hill, L. & Wickham, H. 2001. Long term changes in the water quality of rainfall, cloud water and stream water for moorland, forested and clearfelled catchments at Plynlimon, mid-Wales. Hydr Earth Syst Sci 5(3): 459 –476.

Posch M., Aherne J., Forsius M., Rask M., 2012. Past, present and future exceedance of critical loads of acidity for surface waters in Finland. Environ. Sci. Technol. 46, 4507-4514. https://pubs.acs.org/doi/10.1021/es300332r.

Posch, M., De Vries, W. & Sverdrup, H.U. 2015. Mass balance models to derive critical loads of nitrogen and acidity for terrestrial and aquatic ecosystems. In: Critical Loads and Dynamic Risk Assessments: Nitrogen, Acidity and Metals in Terrestrial and Aquatic Ecosystems; De Vries, W., Hettelingh, J.-P., Posch, M. (Eds.); Springer: Dordrecht, 2015; pp. 171–205.

Prechtel, A., Alewell, C., Armbruster, M., Bittersohhl, J., Cullen, J.M., Evans, C.D., Helliwell, R.C., Kopáček, J., Marchetto, A., Matzner, E., Messenburg, H., Moldan, F., Moritz, F., Vesely, J. & Wright, R.F. 2001. Response of sulphur dynamics in European catchments to decreasing sulphate deposition. Hydrol. Earth Syst. Sci. 5: 311–326.

Priha, O. 1999. Microbial activities in soils under Scots pine, Norway spruce and silver birch. Finnish Forest Research Institute, research papers 731. 50 p.

Rask M., Verta M., Korhonen M., Salo S., Forsius M., Arvola L., Jones R.I. & Kiljunen M. 2010. Does lake thermocline depth affect methyl mercury concentrations in fish? Biogeochemistry 101: 311–322.

Rask, M., Sairanen, S., Vesala, S., Arvola, L., Estlander, S. & Olin, M. 2014. Population dynamics and growth of perch in a small, humic lake over a twenty-year period — importance of abiotic and biotic factors. Boreal Env. Res. 19 (suppl. A): 112–123.

Ruoho-Airola T., Hatakka T., Kyllönen K., Makkonen U. & Porvari P. 2014. Temporal trends in the bulk deposition and atmospheric concentration of acidifying compounds and trace elements in the Finnish Integrated Monitoring catchment Valkea-Kotinen during 1988–2011. Boreal Env. Res. 19 (suppl. A): 31–46.

Ruoho-Airola, T. 2004. Temporal and regional patterns of atmospheric components affecting acidification in Finland. Academic dissertation. Finnish Meteorological Institute Contributions No. 44, 45 p + 5 attachments.

Ruoho-Airola, T., Alaviippola, B., Salminen, K., & Varjoranta, R. 2003. An investigation of base cation deposition in Finland. Boreal Environment Research 8: 83−95.

Salemaa, M., Derome, J. & Nöjd, P. 2008. Response of boreal forest vegetation to the fertility status of the organic layer along a climatic gradient. Boreal Environment Research 13 (suppl. B): 48–66.

Salemaa, M., Kieloaho, A.-J., Lindroos, A.-J., Merilä, P., Poikolainen, J. & Manninen, S. 2020. Forest mosses sensitively indicate nitrogen deposition in boreal background areas. Environmental Pollution 261, 114054.

Salemaa, M., Lindroos, A.-J., Merilä, P., Mäkipää, R. & Smolander, A. 2019. $N_2$ fixation associated with the bryophyte layer is suppressed by low levels of nitrogen deposition in boreal forests. Science of the Total Environment 653: 995–1004.

Salemaa, M., Monni, S., Royo Peris, F. & Uhlig, C. 2000. Sampling strategy for the assessment of temporal changes in ground vegetation in boreal forests. Finnish Forest Research Institute, Research papers 743: 117–127.

Schneider, P. & Hook, S.J. 2010. Space observations of inland water bodies show rapid surface warming since 1985. Geophys. Res. Lett. 37: L22405.

Seidling, W., Hamberg, L., Máliš, F., Salemaa, M., Kutnar, L., Czerepko, J., Kompa, T., Buriánek, V., Dupouey, J.-L., Vodálová, A. & Canullo, R. 2020. Comparing observer performance in vegetation records by efficiency graphs derived from rarefaction curves. Ecological Indicators 109: 11 p.

Singh, B.R., Abrahamsen, G., & Stuannes, A. 1980. Effect of simulated acid rain on sulphate movement in acid forest soils. Soil Science Society of America Journal 44: 75−80.

Solinger, S., Kalbitz, K., & Matzner, R. 2001. Controls on the dynamics of dissolved organic carbon and nitrogen in a Central European deciduous forest. Biogeochemistry 55: 327−349.

Starr, M. & Ukonmaanaho, L. 2004. Levels and characteristics of TOC in throughfall, forest floor and soil solution in undisturbed boreal forest ecosystems. Water Air and Soil Pollution, Focus 4: 715−729.

Tietäväinen, H., Tuomenvirta, H. & Venäläinen, A. 2010. Annual and seasonal mean temperatures in Finland during the last 160 years based on gridded temperature data. Int. J. Climatol. 30 (15): 2247–2256.

Tolvanen, A., Laine, K., Pakonen, T., Saari, E., Havas, P., 1994. Responses to harvesting intensity in a clonal dwarf shrub, the bilberry (Vaccinium myrtillus L.). Vegetatio. 110(2): 163–169.

Tonteri, T., Salemaa, M., Rautio, P., Hallikainen, V., Korpela, L., and Merilä, P. 2016. Forest management regulates temporal change in the cover of boreal plant species. Forest Ecology and Management 381: 115–124.

Travnikov, O., Ilyin, I., Rozovskaya, O., Varygina, M., Aas, W., Uggerud, H.T., Mareckova, K. & Wankmueller, R. 2012. Long-term changes of heavy metal transboundary pollution of the environment (1990–2010). EMEP contribution to the revision of the Heavy Metal Protocol. EMEP MSC-E & CCC & CEIP Status Report 2/2012.

Ukonmaanaho, L., Pitman, R, Bastrup-Birk, A, Breda, N. & Rautio, P. 2020: Part XIII: Sampling and Analysis of Litterfall. In: UNECE ICP Forests Programme Co-ordinating Centre (ed.): Manual on methods and criteria for harmonized sampling, assessment, monitoring and analysis of the effects of air pollution on forests. Thünen Institute for Forests Ecosystems, Eberswalde, Germany, 14 p. + Annex.

Ukonmaanaho, L., & Starr, M. 2002. Major nutrients and acidity: budgets and trends at four remote boreal stands in Finland during the 1990s. The Science of Total Environment 297: 21−41.

Ukonmaanaho, L., Merilä, P., Nöjd, P., & Nieminen, T.M. 2008. Litterfall production and nutrient return to the forest floor in Scots pine and Norway spruce stands in Finland. Boreal Environment Research, 13(supp.B): 67−91.

Ukonmaanaho, L., Starr, M., Mannio, J. & Ruoho-Airola, T. 2001. Heavy metal budgets for two headwater forested catchments in background areas of Finland. Environ. Pollut. 114: 63–75.

Ukonmaanaho, L., Starr,M., Lindroos, A.-J., Nieminen T.M. 2014. Long-term changes in acidity and DOC in throughfall and soil water in Finnish forests. Environ. Monit. Assess. 186: 7733–7752 (+ erratum).

Vanha-Majamaa, I., Shorohova, E., Kushnevskaya, H., Jalonen, J. 2017. Resilience of understorey vegetation after variable retention felling in boreal Norway spruce forest – A ten-year-perspective. Forest ecology and management 393: 12−28.

Venäläinen, A., Tuomenvirta, H., Pirinen, P. & Drebs, A. 2005. A basic climate data set 1961–2000-description and illustrations. – Finnish Meteorol. Inst. Rep. 5: 27.

Verheyen, K., Baeten, L., De Frenne, P., Bernhardt-Romermann, M., Brunet, J., Cornelis, J., Decocq, G., Dierschke, H., Eriksson, O., Hedl, R., Heinken, T., Hermy, M., Hommel, P., Kirby, K., Naaf, T., Peterken, G., Petrik, P., Pfadenhauer, J., Van Calster, H., Walther, G.R., Wulf, M., & Verstraeten,

G. (2012) Driving factors behind the eutrophication signal in understorey plant communities of deciduous temperate forests. Journal of Ecology 100: 352–365.

Verstraeten, Arne et al. + 41 co-authors 2020. Impact of pollen on throughfall biochemistry in European temperate and boral forests. Poster, EGU, Wien 2020.

Verta M., Salo S. Porvari P., Korhonen M., Paloheimo A. & Munthe J. 2010. Climate induced thermocline change has an effect on the methyl mercury cycle in small boreal lakes. Sci. Total Environ. 408: 3639–3647.

Villén-Peréz, S., Heikkinen, J., Salemaa, M., Mäkipää, R. 2020. Global warming will affect the maximum potential abundance of boreal plant species. Ecography 43: 1−11.

Vuorenmaa J., Forsius M. & Mannio J. 2006. Increasing trends of total organic carbon concentrations in 659 small forest lakes in Finland from 1987 to 2003. Sci. Total Environ. 365: 47–65.

Vuorenmaa, J. & Forsius, M. 2008. Recovery of acidified Finnish lakes: trends, patterns and dependence of catchment characteristics. Hydrol. Earth Syst. Sci. 12: 465–478.

Vuorenmaa, J., Augustaitis, A., Beudert, B., Clarke, N., de Wit, H.A., Dirnböck, T., Frey, J., Forsius, M., Indriksone, I., Kleemola, S., Kobler, J., Krám, P., Lindroos, A.-J., Lundin, L., Ruoho-Airola, T., Ukonmaanaho, L. & Váňa, M. 2017. Long-term sulphate and inorganic nitrogen mass balance budgets in European ICP Integrated Monitoring catchments (1990–2012). Ecological Indicators 76: 15–29.

Vuorenmaa, J. Augustaitis, A., Beudert, B., Bochenek, W., Clarke, N., de Wit, H., Dirnböck, T., Frey, J., Hakola, H., Kleemola, S., Kobler, J., Krám, P., Lindroos, A.-J., Lundin, L., Löfgren, S., Marchetto, A., Pecka, T., Schulte-Bisping, H., Skotak, K., Srybny, A., Szpikowski, J., Ukonmaanaho, L., Váňa, M., Åkerblom, S. & Forsius, M. 2018. Long-term changes (1990–2015) in the atmospheric deposition and runoff water chemistry of sulphate, inorganic nitrogen and acidity for forested catchments in Europe in relation to changes in emissions and hydrometeorological conditions. Science of the Total Environment 625: 1129–1145.

Vuorenmaa, J., Salonen, K., Arvola, L., Mannio, J., Rask, M. & Horppila, P. 2014: Water quality of a small headwater lake reflects long-term variations in deposition, climate and in-lake processes. Boreal Env. Res. 19 (suppl. A): 47–65.

Wright, R.F., Camarero, L., Cosby, B.J., Ferrier, R.C., Forsius, M., Helliwell, R., Jenkins, A., Kopáček, J., Larssen, T., Majer, V., Moldan, F., Posch, M., Rogora, M. & Schöpp, W. 2005. Recovery of acidified European surface waters. Environ. Sci. Technol. 39: 64A–72A.

Wu, Y., Clarke, N., & Mulder, N. 2010. Dissolved organic carbon concentrations in throughfall and soil waters at Level II monitoring plots in Norway: short- and long-term variations. Water Air and Soil pollution 205: 273−288.

Zellweger et al., 2020. Forest microclimate dynamics drive plant responses to warming. Science 368: 772–775.