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Duration of forest fertilization effects on streamwater chemistry in a catchment in central Sweden

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ABSTRACT

Demands for forest biomass production for energy, construction and carbon storage purposes are increasing, and therefore measures to increase tree growth are required. One potential measure is nitrogen (N) fertilization, as N is usually the most growth-limiting nutrient in boreal forests and partly due to decreasing atmospheric N deposition in northern Europe in recent decades. However, N fertilization can have adverse effects, such as soil acidification and N leaching, particularly nitrate leaching via streamwater flow. To mitigate the acidification risk, dolomite ($CaMg(CO_3)^2$) is added to N fertilizer boron (B) as increased tree growth hamper tree uptake of this essential micronutrient.

This study examined the effects of forest fertilization on streamwater chemistry in the 45 ha Swedish catchment Risfallet (RF), around 80% of which was treated with fertilizer. That was rather exceptional, as most previous catchments studied have had <50% treated area, which may give weaker treatment signals. A paired catchment method combined with the control area and calibration period technique was applied to evaluate leaching effects from forest fertilization.

Effects over 7.5 years were compared with previously reported initial effects in the first year, in order to assess the duration of fertilization effects on surface water. High excess outflow of N over five years was detected, with 20% of the applied amount leached and with nitrate dominating total nitrogen. Excess outflow of Ca and Mg was highest in the first year. Effects on pH were limited, with calculated untreated pH on 5.9 being on average 0.4 units lower during the first six months and then remaining at 0.2 units lower.

Recommended could be to mainly fertilize well-drained soil, avoid wet areas and open streams. Consider the hydrological conditions while weather would be more hazardous to foresee.

1. Introduction

There is great interest in increasing forest productivity in terms of conventional products such as timber and paper, but also for the purposes of renewable energy supply, environmental services, and mitigation of climate change by carbon sequestration (Garcia-Fernandez et al., 2008; Favero et al., 2020). In Sweden, the forest industry needs to maintain production of primary forest products, while at the same time adjusting to societal needs and environmental concerns. To meet these challenges, a circular production chain is suggested (Kumar et al., 2020). This would include all stages in biomass utilization from forest management, environmental concerns, production of gods available for various sectors, waste treatment and the machinery involved.

Potential measures to increase forest growth include improved forest management, use of alternative tree species, tree breeding, fertilization, and further afforestation (Rytter et al., 2016). Boreal forests are considered growth-limited because of insufficient soil nitrogen (N) availability (Tamm, 1991; Hyvönen et al., 2008). Experiments examining the benefits of N addition have shown aboveground biomass increment for especially coniferous forests (Nohrstedt, 2001; Albaugh et al., 2016; Högberg et al., 2017; Lee et al., 2020). Around 40–50 years ago, forest fertilization was common practice in Sweden. Later, with high atmospheric N deposition caused by airborne industrial pollution, no forest N fertilization was needed in regions where high deposition occurred, such as in the southern parts of the Nordic countries. However, N deposition (and subsequent leaching) in Sweden have declined in the past 20 years (Forsius et al., 2020). This is mainly because of cessation of high anthropogenic atmospheric N deposition and as a result of growing

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environmental awareness (Lindkvist et al., 2011). Deposition in central Sweden is currently less than 5 kg N ha^{-1} yr^{-1} (Löfgren, 2020), so interest in the benefits of applying N fertilizer to forests is returning. There was some limited forest fertilization in the period 1980–2000, but in recent years fertilization activities have increased. The area of fertilized forest in Sweden increased to 24 000 ha in 2010 (SCB, 2014) and to 25 000 ha in 2017 (SKS, 2019).

However, applying high N input results in increased leaching of N to watercourses (Grip, 1982; Binkley et al., 1999; Davis et al., 2012). A deposition of 10 kg N ha^{-1} yr^{-1} elevates the risk of N leaching, while 25 kg N ha^{-1} yr^{-1} is often cited as the threshold for significantly increased N leaching (Dise and Wright, 1995; Gundersen et al., 2006). Conventional Nordic forest fertilization involves addition of 150 kg N ha^{-1} on 1–3 occasions during a forest rotation, with the effects of each fertilization event expected to last at least seven years (Nohrstedt, 2001; Saarsalmi and Mälkönen, 2001). This has positive effects in increasing forest biomass production and in contributing to carbon sequestration in biomass and soil (Sathre et al., 2010; Maaroufi et al., 2015; Bowden et al., 2019). However, it can also have negative effects, as high N application rate increases the risk of leaching and the potential for N storage in the ecosystem. High nitrogen storage could mean concentrations close to the threshold were enhanced leaching occur. Elevated leaching could occur in particular from sites with high deposition and low soil carbon: nitrogen (C/N) ratio (Binkley et al., 1999; Gundersen et al., 2006). Application of N fertilizer can also cause soil acidification due to nitrification and soil exchange acidity (Högbom and Nohrstedt, 2000; Saarsalmi et al., 2014).

Most existing information on the effects of forest N fertilization derives mainly from studies carried out more than 40 years ago, when airborne practice was common. To achieve more site-adapted addition, and possibly avoid unnecessary N fertilization of wet soils, land-based tractor spreading could be used instead of the earlier usual aerial application method. In a peatland fertilization, the peaks in concentration the first period after application were explained by direct input of fertilizer from the aerial application (Lundin and Bergquist, 1985). Fertilized sites should mainly be well-drained, with groundwater levels below one metre.

In 2012, the Swedish company Sveaskog fertilized a 35-year-old pine (*Pinus silvestris*) forest on a well-drained area in east-central Sweden. Part of the fertilized area was the well-defined catchment Risfallet (RF), which has been studied in soil, water, and acidification research since the mid-1980s (Knutsson et al., 1995). This monitoring of RF catchment for a long period before fertilization provided very good background information for the present study, as did similar monitoring data from ongoing measurements in an unfertilized control area nearby, Gusseltjärn (GT) catchment. This furnished the possibility to compare the catchments and to assess differences arising as effects of N fertilization in RT catchment. Initial effects of large-scale forest fertilization with N, Ca, Mg, and B on surface water chemistry and leaching from RF catchment in the first year after fertilization have been reported previously (Lundin and Nilsson, 2014). Over seven years have now elapsed since fertilization, so the longer-term effects (2012–2019) were evaluated in this study. We hypothesis that leaching of fertilization compounds continues longer than one year dependent on soil water turnover time. Further the fertilization compounds influence leaching of other chemical elements related to soil and forest processes. In particular, the duration of effects of fertilization on concentrations of chemical elements in surface water and leaching in outflow from the catchment were assessed.

2. Material and methods

2.1. Site characteristics

This investigation had the advantage of good availability of long-term data collected over a 25 year period previous to forest fertilization and in an unfertilized control catchment for the same period. Other advantages were that Risfallet is a rather homogeneous catchment and that around 80% of the entire catchment was fertilized in 2012, avoiding wet soils and mires. This provided the opportunity to monitor effects of forest fertilization on water and element turnover at catchment scale. Previous studies of forestry operations have often been based on catchments with less than 50% treated area, which means that the signal from fertilization in the RF case could be measured more appropriate. In cases with smaller parts of catchments are related to dilution effects. Further benefits were good availability of data for the control GT catchment, only 120 km away (Fig. 1), which has similar geophysical conditions and hydrology as RF catchment (Lundin, 1995) (Table 1). The GT catchment being part of the experimental forest Siljansfors in was equipped with instruments for forest monitoring in 1973, as part of research at the Swedish University of Agricultural Sciences (SLU). Coniferous forests, mainly >100-year-old pine, dominate GT catchment, with very limited forestry operations carried out (Bergqvist and Grip, 1975; pers. Comm. with head of park admin.). The pine forest in RF catchment was younger (35 years) at the time of fertilization. Both catchments are largely dominated by well-drained, rather coarse-grained, glacial till soils, with mesic soil water conditions and the groundwater level often found below 1–3 m (Maxe, 1995). In the first months, following fertilization occasion elevated water levels occurred being above one meters depth and partly close to ground surface. The moraine shows influences from the underlying bedrock being granite in RF catchment and porphyry in GT catchment, showing somewhat poorer soil nutrient conditions in GT catchment (Lundin and Nilsson, 2014).

2.2. Forest fertilization

Forest fertilization in Sweden mainly involves application of nitrogen, which is normally the limiting nutrient for high growth. The content of ammonium nitrate (NH_4NO_3) in the commonly used forest fertilizer in Sweden has an acidifying effect on the soil. Calcium (Ca) and magnesium (Mg) are now added to N fertilizer in the form of dolomite ($CaMg(CO_3)^2$) to mitigate the acidification risk, and increased forest growth influence boron (B) availability (Wikner, 1983), so B is also added. This specialist forest fertilizer, called SkogCAN, supplying 150 kg N, 22 kg Ca, 12 kg Mg and ~1 kg B per hectare, was distributed in RT catchment by tractor in 2012, in a total amount of 550 kg/ha. In Swedish forestry this is a recommended dose (Bergh et al., 2014). Tractor was used for avoiding fertilizer spraying to watercourses.

2.3. Paired catchment method

Based on the data available, the most suitable approach for investigating fertilization effects was considered to be a catchment concept using the control area and calibration period technique. The long-term calibration period (1987–2011 regarding meteorology and hydrology) showed good agreement between the two catchments, making it possible to use the control area (GT) to calculate hydrological and water chemistry conditions for the period after treatment (fertilization) for RF catchment as in untreated state. Discharges were calculated from measured water levels registration by chart recorders and calculated on daily basis while water chemistry was based on monthly samples and outflow from interpolated daily concentrations and multiplied with daily discharges. Statistical values used monthly values. Comparisons were then made with the actual measured values in RF catchment after treatment, with differences showing the impact of treatment (Lundin and Nilsson, 2014).

The sub-period 1997–2011 in the calibration data was used for assessing water chemistry. Fertilization was carried out over around one week in the end of June 2012 and initial effects of fertilization (to July 2013) have been reported (Lundin and Nilsson, 2014). The present study examined the longer-term effects of fertilization, covering the period 2012–2019.

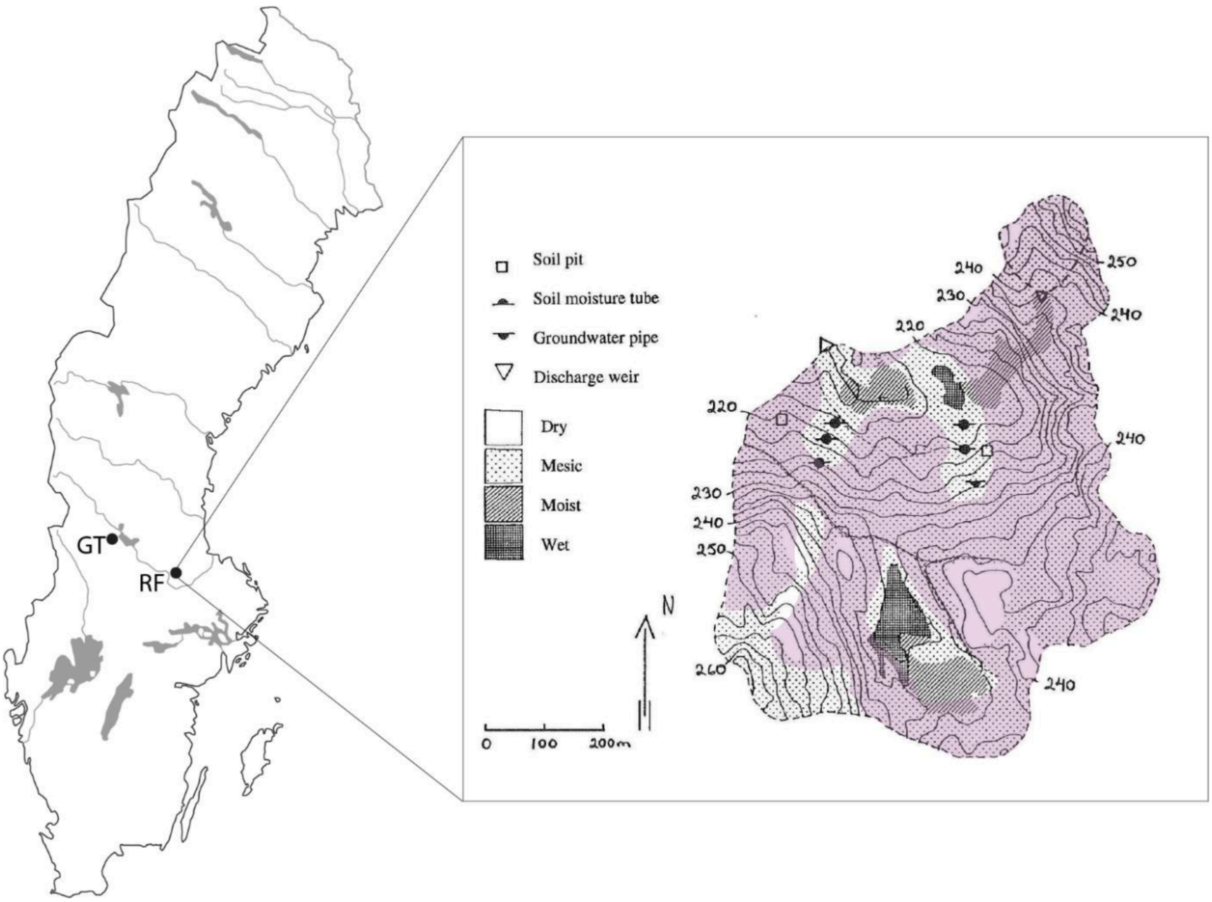

Fig. 1. Left: Map of Sweden showing geographical location of the Gusseltjärn (GT) and Risfallet (RF) catchments. Right: Detailed map of the Risfallet catchment showing hydrological installations, contour lines (m a.m.s.l.) and distribution of moisture classes (Lundin, 1995). Superimposed is shaded violet colour showing the fertilized area. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1
Site characteristics of the Gusseltjärn (GT) and Risfallet (RF) catchments (updated values for 2012 from Maxe ed. 1995). Tree species frequency given as ratio (1/10) of pine, spruce, birch (P/S/B). Climate data (1960–1990) from Raab and Vedin (1995). Table from Lundin and Nilsson (2014).

	Gusseltjärn, GT	Risfallet, RF
Location, Lat./Long.	N 60°35′; E 14°25′	N 60°21′; E 16°14′
Altitude, m a.m.s.l.	265–365	215–270
Area, ha	83	45
Temp. °C	+3.1	+4.0
Temperature sum (>5 °C)	1150	1200
Snow cover duration, days	160	140
Precipitation, mm	660	616
Runoff, mm	380	300
Tree species frequency, P/S/B ratio	9/1/0	8/1/1
Stand age, years	60 (17 ha), >130 (58 ha)	35
Stand stem volume, m^3 ha^{-1}	~200	184
Current stand increment, m^3 ha^{-1} yr^{-1}	4.0	4.6

Discharge from the two catchments was determined by continuous water level recording at V-notch weirs in the outlet, where water was also sampled monthly. Flows of elements were calculated based on daily discharge and interpolated element concentrations in water (Lundin and Nilsson, 2014). The values for the ‘untreated’ RF catchment, i.e., based on the control, were mainly based on RF/GT ratio in the calibration period, when rather similar variation in values occurred. Differences between untreated and treated (measured) RF values were tested using

Table 2
Mean values for stream water pH, colour (mg Pt L^{-1}) and concentrations (mg L^{-1}) in the control Gusseltjärn (GT) and the fertilized catchment Risfallet (RF) in the calibration period (1997–2011), and the first 1.5 years after fertilization (July 2012-December 2013) and 1.5 to 7.5 years after fertilization (January 2014-December 2019). RFcalc – calculated as unfertilized; RFmeas – measured after fertilization. Mean values for an element, within a period, followed by the same letter are not significantly different (T-test, $p < 0.05$). The capital letters defines if there are significant different values for RF-measured between periods (ANOVA-test, $p < 0.05$).

Element	Calibration period 1997–2011 N = 160–180		Periods after fertilization			
			0–1.5 years N = 18–28		1.5–7.5 years N = 63–66	
	GT	RF	RFcalc	RF meas	RFcalc	RFmeas
pH	5.78 a	5.76 a B	5.98 a	5.59 b C	6.01 a	5.90 b A
Colour	38.6 b	52.1 a B	53.3 a	51.7 a B	72.9 b	85.3 a A
DOC	7.86 b	9.72 a B	9.63 a	9.40 a A	9.34 b	10.7 a A
Na	1.53 b	2.22 a B	2.93 a	3.07 a A	2.12 a	2.13 a B
K	0.35 b	0.38 a B	0.47 b	0.61 a A	0.35 a	0.37 a B
Ca	1.59 b	2.98 a B	3.87 a	4.95 a A	2.69 b	2.89 a B
Mg	0.33 b	0.59 a C	0.92 a	1.08 a A	0.65 a	0.64 a B
SO_4	2.36 b	5.91 a A	4.76 a	3.65 b C	4.68 a	4.77 a B
Cl	0.99 b	1.64 a AB	1.98 a	1.53 b B	1.67 a	1.68 a A
NO_3-N	0.036b	0.055 a C	0.090 b	3.66 a A	0.050 b	0.274 a B
NH_4-N	0.011 a	0.014 a C	0.027 a	0.069 a A	0.005 b	0.012 a B
Norg	0.255 a	0.289 a A	0.203 a	0.173 a B	0.193 b	0.268 a A
Ntot	0.303 b	0.368 a C	0.317 b	3.90 a A	0.252 b	0.557 a B
Ptot	0.006 b	0.008 a A	0.006 a	0.007 a A	0.005 b	0.007 a A

students' *t*-test. Error bars (sd) were included in concentration graphs and also in Tables 2 and 3. All statistical analyses were performed in SAS 9.4 (http://www.sas.com). As most of the data for the different elements were not normally distributed, the data were log-transformed before testing the effects. *T*-test, or analysis of variance (ANOVA) were used to examine the effects of fertilization. In all statistical tests the level of significance was set at $p < 0.05$.

2.4. Water sampling and chemical analyses

Water sampling in both RF and GT catchment was carried out monthly except in the first five months after fertilization, when up to four samples per month were taken in RF catchment. Chemical analyses were carried out by laboratories at SLU, Uppsala, using standard techniques according to SWEDAC (Swedish Board for Accreditation and Conformity Assessment). Among the elements assessed in this study, base cations were analysed with optical inductively coupled plasma-optical emission spectrometry (ICP OES), accuracy Ca 0.05 mg L^{-1}, K 0.02 mg L^{-1}, Mg 0.015 mg L^{-1} and Na 0.03 mg L^{-1}. Anions analysed with ion chromatography (Dionex ICS 1100) accuracy SO4 0.29 mg L^{-1} (Metrohm 930), Cl 0.05 mg L^{-1} (Metrohm 881). Total N mainly by persulphate digestion and later by combustion (SS-EN 12260:2004; Shimadzu VCPH; accuracy 15%). Nitrate photometrical, accuracy 0.004 mg L^{-1} and ammonium discrete analysis and photometrical, accuracy 0.004 mg L^{-1}. Boron was analyzed by digestion in 0.5% HNO_3 and measurement by inductively coupled plasma-mass spectrometry (ICP-MS). DOC analyzed by combustion (EN 1484; Shimadzu Instrument; accuracy 10%). pH was measured with combination electrodes, accuracy 0.28 pH units (Metrohm 855; SS-EN ISO 10523:2012). Water colour was measured spectrophotometrically, accuracy 10% (SS-EN ISO 7887:2012). Detection limits summarized; NO_3-N 3 μg L^{-1}, NH_4-N 3 μg L^{-1}, N_{tot} 50 μg L^{-1}, Ca 0.02 mg L^{-1}, Mg 0.01. mg L^{-1}, K 0.04 mg L^{-1}, Na 0.02 mg L^{-1}, SO_4 0.48 mg L^{-1}, Cl 0.25 mg L^{-1}, DOC 0.5 mg L^{-1}, colour 4 mg L^{-1} and pH 3.

3. Results

3.1. Temperature and precipitation

Temperature and precipitation were calculated for two periods based on values from the Swedish Meteorological and Hydrological Institute (SMHI). The first period was before fertilization (1987–2011) and the second period was after fertilization (2012–2019, with the whole of 2012 included). Mean annual temperature in the period before fertilization was 4.9 °C, which was 0.6 °C lower than in the period after fertilization (mean 5.5 °C). The lowest mean annual temperature in the period after fertilization (4.5 °C) was observed in 2012 and the highest in 2014 (6.2 °C).

Mean annual precipitation sum (P) for the period before fertilization (1987–2011) was 696 mm (ranging from 520 mm in 1995 to 879 mm in 2000), while in the period after fertilization it was 694 mm (ranging from 484 mm in 2013 to 942 mm in 2019). P was high (847 mm) in 2012, i.e., the actual year for fertilization, with a very high value (379 mm) in the three-month period following fertilization (June-August)) of 379 mm being 166 mm, 78% higher than the long-term average for these three months on 213 mm.

3.2. Discharge

The average annual discharge pattern for RF catchment mainly reflected the normal pattern for central Sweden, with mean monthly discharge in winter months of around 20 mm, a snowmelt peak in spring and generally low flows in summer (June-September). One shift in pattern seen in recent years is that the spring flood is occurring earlier, with peak flow in March, rather than in April, in the latest 10-year period. The spring peak often reaches 2–3 mm day^{-1} but not uncommonly up to 5–7 mm day^{-1}. High spring values, as seen in 1988, exceed 10 mm day^{-1} and are dependent on the magnitude of snow cover. Summer discharge is generally low, less than 0.1 mm day^{-1}, and in many years there is zero water flow for more than a month in summer. In the years after fertilization of RF catchment, the frequency of zero flow periods was lower than in the years before fertilization, with flow ceasing for consecutive days over one month only in 2013, 2017, and

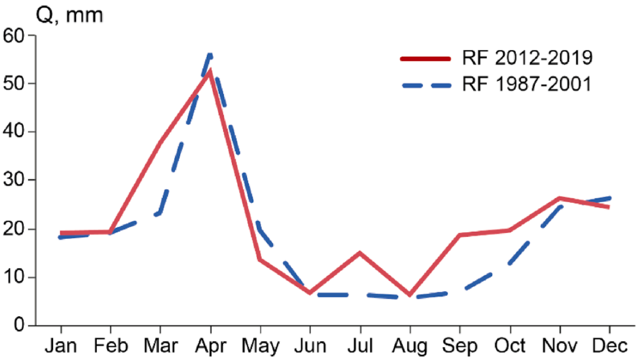

Fig. 2. Monthly discharge (Q, mm) from Risfallet (RF) catchment during the period before fertilization (blue dashed line) and the period after fertilization (red solid line). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 3
Outflow of different elements (for abbreviations, see text) in streamwater from Risfallet (RF) catchment in calibration period, as a mean for 7.5 years, and sum for 1.5 years and 7.5 years. RFcalc – calculated as unfertilized; RFmeas – measured after fertilization. Mean values for an element transport, between periods, followed by the same letter are not significantly different (ANOVA-test, $p < 0.05$).

Element	Calib.period kg/ha/, yr 1997–2011	RF_{calc} mean kg/ha/yr 2012–2019	RF_{meas} mean kg/ha/yr 2012–2019	RF change % 2012–2019	RF_{calc} sum, 1.5 yr kg/ha 2012–2013	RF_{meas} sum, 1.5 yr kg/ha 2012–2013	RF_{calc} sum 7.5 yr kg/ha 2012–2019	RF_{meas} sum, 7.5 yr kg/ha 2012–2019
H	0.0059 a	0.0026 b	0.0069 a	164	0.0037	0.025	0.021	0.055
Na	4.6 a	4.6 a	5.7 a	24	7.3	15.5	36.6	45.5
K	0.8 a	0.80 a	1.0 a	31	1.2	3.3	6.4	8.3
Ca	6.2 a	5.9 a	8.4 a	42	9.8	26.7	47.1	67.1
Mg	1.2 b	1.4 ab	1.8 a	35	2.1	5.8	10.9	14.7
SO4	12.6 a	9.5 b	10.7 ab	12	12.1	15.8	76	86
Cl	3.5 a	3.9 a	4.0 a	3	6	7.2	30.9	31.9
NO_3-N	0.12 b	0.14 ab	3.03 a	2021	0.35	20.5	1.14	24.2
NH_4-N	0.03 a	0.02 a	0.10 a	451	0.07	0.61	0.14	0.76
N_{org}	0.62 a	0.40 a	0.64 a	59	0.53	1.12	3.06	5.15
N_{tot}	0.78 b	0.54 b	3.77 a	594	0.95	22.2	4.34	30.12
DOC	22.1 a	19.1 a	27.1 a	42	28	49.1	152	217
P_{tot}	0.019 a	0.014 a	0.016 a	13	0.024	0.038	0.11	0.12

2018. Summer discharge in the period after fertilization was higher than in the pre-fertilization period (1987–2011) (Fig. 2).

Mean annual discharge from RF catchment in the calibration period (1987–2011) was 80% of discharge from the reference GT catchment used to calculate the flow in RF (RFcalc) for the period after fertilization. The hydrogeology of the two catchments is similar, with fairly coarse glacial till soils, a thick unsaturated soil moisture zone, rather deep groundwater levels (1–2 m) and considerable soil water storage capacity with 100–150 mm in the top one metre. These conditions give fairly stable hydrological conditions. Estimated discharge from GT catchment (i.e., RFcalc) agreed fairly well with the measured values in RF (RFmeas), but the year 2012 deviated, with clearly higher measured values compared with calculated excess 142 mm) (Fig. 3).

Measured annual discharge from RF catchment was on average 219 mm (range 97–330 mm) in the period before fertilization and 259 mm (range 157–457 mm) in the period after fertilization. The RFcalc value for the period was 233 mm, with higher discharge in 2012 and 2019 comprising the main difference, probably due to heavy rain events in summer differing between the sites. Disregarding these two years, the average runoff for both RFmeas and RFcalc was 205 mm. The range of variation was similar for the periods 1987–2011 and 2013–2019 (100–380 mm).

3.3. *Concentrations of chemical elements in surface water runoff*

Initial impacts of fertilization reported for the first year after fertilization (Lundin and Nilsson, 2014) were followed over the next seven years to determine the duration of changes. Effects of fertilization on streamwater chemistry were estimated by comparison against calculated concentrations for untreated conditions (RF_{calc}) obtained using the control catchment (GT) stream values and the relationships in the calibration period between the catchments. Deviations between the values observed after fertilization and the calculated background values based on the control site showed effects of the fertilization (Table 2). There was good agreement between RF_{calc} and RF_{meas} values for the calibration period, as reported previously (Lundin and Nilsson, 2014). With respect to the elements added with fertilizer, the focus was mainly on N, base cations, and pH, with some additional elements, e.g. DOC, also considered (Table 2).

3.4. *Nitrogen*

Nitrogen fertilization was carried out with NH_4-NO_3. In assessing effects on streamwater, particular focus was placed on nitrate (NO_3), the most active ion and also the most easily mobile. In unfertilized conditions, the two inorganic N ions (NO_3 and ammonium (NH_4) made up 15–25% of total nitrogen (N_{tot}) in streamwater, while organic nitrogen (N_{org}) dominated (75–85%). This represents normal conditions for northern boreal landscapes (Högberg et al., 2017). In the period 1997–2011, prior to fertilization, mean NO_3-N concentration in RF was 0.054 mg L^{-1} (GT 0,036 mg L^{-1}) (Table 2) and only occasionally reached values over 0.1 mg L^{-1}. In July 2012, less than one month after fertilization, the concentration peaked at 8.2 mg L^{-1} of NO_3-N (Lundin and Nilsson, 2014) and NO_3-N dominated N_{tot} content after fertilization. In the second half of 2012, the average NO_3-N concentration was 4.5 mg L^{-1}, which was 4.4 mg L^{-1} significantly higher than the background value. In the calendar year 2013, the average NO_3-N concentration was 2.5 mg L^{-1}, which was 2.4 mg L^{-1} higher (significant) than in unfertilized conditions (RF_{calc}). The NO_3-N concentration declined in the following years, to 0.1 mg L^{-1} in 2016, which was still double the background value (significant). The concentration did not return to the unfertilized level until 2018–2019 (Fig. 4).

Prior to fertilization, NO_3-N made up 15% of N_{tot} and NH_4-N less than 4%. After fertilization, NO_3-N made up 94% of N_{tot} in the first half-year after fertilization and 95% in 2013, while NH_4-N comprised 1.5% and 1.4%, respectively. Before fertilization, N_{org} constituted 82% of N_{tot}. In the first half-year after fertilization N_{org} made up less than 5% of N_{tot} and in 2013 only 3%. Three years after fertilization, N_{org} made up 45% of N_{tot} and seven years after fertilization (2019) values were back to the original over 80%. One year after fertilization, the N_{org} concentration was 0.08 mg L^{-1}, which was less than 36% of the calculated unfertilized value (RF_{calc}) of 0.3 mg L^{-1} of N_{org}. Five years after fertilization, the N_{org} concentration was back at the ordinary level, although it turned to slightly higher than RF_{calc} in the last four years (2016–2019) (Fig. 4).

Before fertilization, NO_3-N outflow was 0.15 kg ha^{-1} yr^{-1}, but it reached 3.23 kg ha^{-1} yr^{-1} after fertilization. In the first 1.5 years, the outflow was 20.5 kg ha^{-1} of NO_3-N, compared with the RF_{calc} value (0.35 kg ha^{-1}). Elevated outflow was estimated to continue for 5.5 years and reached 23 kg ha^{-1} in total. This represented 15% of the N amount added by fertilization. In total for the whole 7.5 year period to 2019, 24 kg ha^{-1} of NO_3-N was leached, with 83% of leaching occurring in the first 1.5 years (Table 3).

Outflow of NH_4-N after fertilization was much lower than NO_3-N outflow, with an average measured value in discharge of 0.1 kg ha^{-1} yr^{-1}. However, compared with the background value RF_{calc} on 0.02 kg ha^{-1} yr^{-1}, this was an increase of 400%. Outflow during the first 1.5 years was 0.61 kg ha^{-1} of NH_4-N, 0.54 kg ha^{-1} in excess of the background level. The excess NH_4-N outflow for the whole period was 0.62 kg ha^{-1}, with 87% occurring in the first 1.5 years. Elevated outflow occurred for the whole period and was ~140% higher than background, including in 2018–2019. For NO_3-N, the excess amount in the final two years was only around 10%.

The N_{tot} concentration in discharge was highest in July 2012 (9.5 mg L^{-1}), while the calculated unfertilized mean (RF_{calc}) was 0.5 mg L^{-1}. In 2013, average N_{tot} was 2.64 mg L^{-1}, seven times higher than the RF_{calc} value and N_{tot} concentration remained significantly higher until 2017. The concentration then declined to reach the original value after about five years (Fig. 4).

Measured mean N_{tot} outflow was 4.02 kg ha^{-1} yr^{-1}, 593% higher than the calculated background value of 0.58 kg ha^{-1} yr^{-1}. Loss in outflow for the whole period amounted to 30 kg ha^{-1} of N_{tot}, which was 25.8 kg ha^{-1} higher than the background and 17% of the amount applied with fertilizer. Outflow in the first 1.5 years dominated the whole period. It was 22.2 kg ha^{-1}, 85% of total excess outflow in the whole period (Table 3).

As mentioned, N_{org} dominated N_{tot} (86%) under ordinary conditions, but in the first five years after fertilization N_{org} constituted <70%. The average RF_{calc} outflow of N_{org} in the first 1.5 years was 0.35 kg ha^{-1} $year^{-1}$, while RF_{meas} outflow was 0.75 kg ha^{-1} $year^{-1}$ of N_{org}, a 114% increase. In this first period, outflow reached 1.12 kg N_{org} ha^{-1}, a 111% increase compared with the background of 0.59 kg ha^{-1}. Outflow in the first 1.5-year period made up 22% of total N_{org} outflow in the 7.5-year

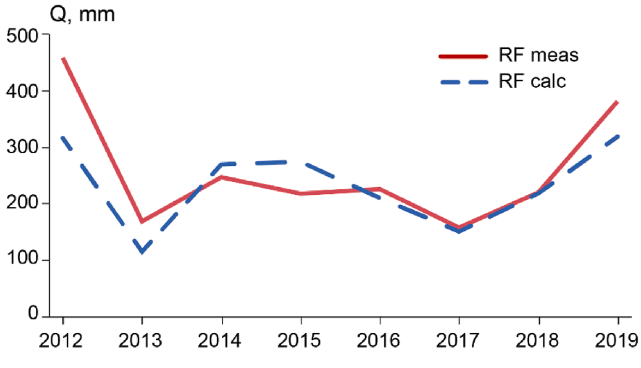

Fig. 3. Measured annual runoff (R, mm) from Risfallet catchment (RFmeas; red solid line) and calculated runoff based on the unfertilized control catchment GT (RFcalc; blue dashed line) in the period after fertilization (2012–2019). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

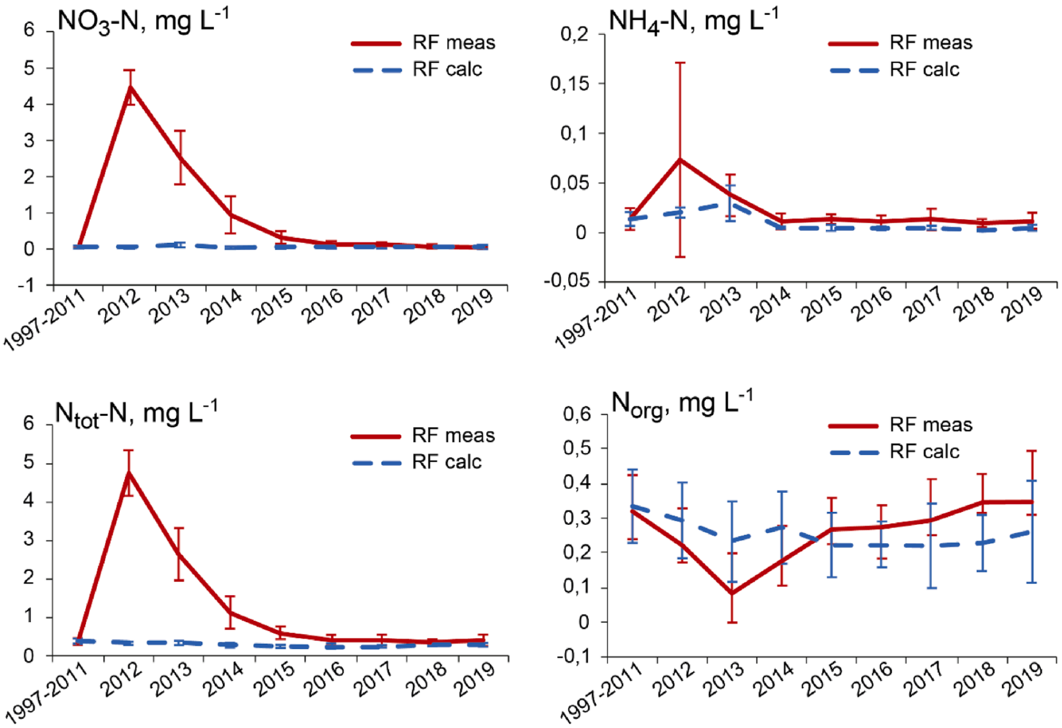

Fig. 4. Concentrations with error bars (mean for 1997–2011 and annual means for 2012–2019) of nitrate-N (NO_3-N), ammonium-N (NH_4-N), organic N (N_{org}) and total N (N_{tot}), in stream water for unfertilized background conditions (RF_{calc}, blue dashed line) and measured value after fertilization (RF_{meas}, red solid line). NB: Different Y-axis scales. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

period 2012–2019. Increasingly higher N_{org} outflow was observed for the last four years of the period, with an increase in 2019 to 0.70 kg ha^{-1}, 108% of the calculated background value (Table 3).

3.5. Dissolved organic carbon (DOC)

The general patterns of N_{org} and DOC were similar, with mainly lower concentrations than background values in the first years after fertilization but in 2015, three years later, with values for RF_{meas} and RF_{calc} being similar. In the following four years (2016–2019), N_{org} and DOC altered the relation between observed and calculated concentrations with higher RF_{meas} than the background values (Figs. 4 and 5). Mean measured DOC concentration in the first three years was 9.3 mg L^{-1}, similar to the calculated background value (9.8 mg L^{-1}). After 4.5 years, measured DOC began to exceed the background value and in 2019 it was 12.8 mg L^{-1}, 3.3 mg L^{-1} higher than the background level (Fig. 5). DOC affects water colour that changed after fertilization. The year of fertilization colour at RF was slightly lower than calculated but already after 2.5 years colour was higher and successively increasing to 73 mg Pt L^{-1} in 2018 and 103 mg Pt L^{-1} in 2019, i.e. 18 mg Pt L^{-1} and 25 mg Pt L^{-1} increase, respectively. Measured DOC outflow from RF catchment for the whole period was 217 kg ha^{-1} year^{-1}, 65 kg ha^{-1} year^{-1} higher than the calculated control value. However, due to high discharge in the initial 1.5 years, the measured amount was 29 kg ha^{-1}, 42% higher than the background value of 20 kg ha^{-1}. Also the final four years contributed 43.5 kg ha^{-1} excess outflow from RF catchment, while years 3–4 had slightly lower outflow (Table 3).

3.6. pH

Annual mean pH in discharge did not vary very much (range 5.5–6.1) (Table 2). However, the lowest pH occurred in 2012, the year of fertilization. In the first month after fertilization (July) pH was 4.8, while mean pH for the second half of 2012 was 5.5, which was significantly lower than the calculated unfertilized value of pH 5.9. In the period 2013–2019, mean annual pH varied between 5.8 and 6.1, with the

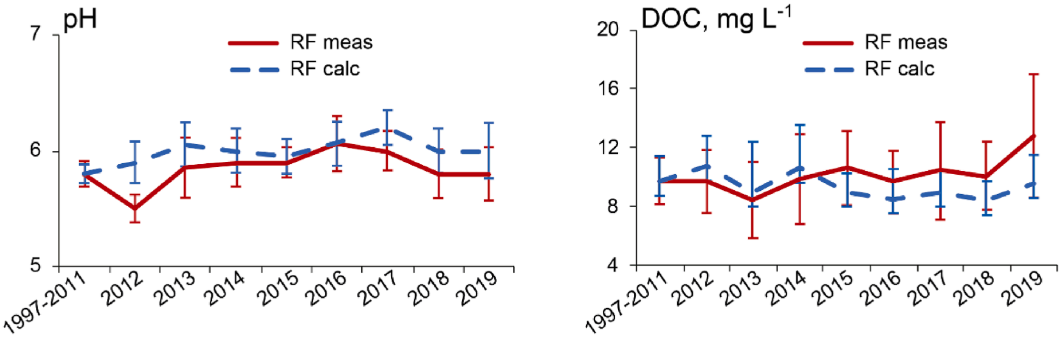

Fig. 5. Dissolved organic carbon (DOC) and pH with error bars in stream water before forest fertilization (1997–2011) and in the seven years following fertilization (June 2012) in unfertilized background conditions (RF_{calc}, blue dashed line) and measured value after fertilization (RF_{meas}, red solid line). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

fertilized value generally being 0.2 pH units lower than the calculated background value (Fig. 5). Calculated unfertilized outflow of hydrogen (H^+) was 0.28 mg m^{-2} year^{-1} on average for the 7.5 years after fertilization and the measured amount was 0.73 mg m^{-2} year $^{-1}$ of H^+, an increase of 161%. The first 1.5 years contributed most to excess outflow (2.1 mg m^{-2} of H^+, 5.8-times higher than unfertilized value) (Table 3). High discharge in 2012 contributed to this difference.

3.7. Base cations, (BC)

Addition of Ca and Mg to N fertilizer to mitigate acidification affects soil base cation (BC) exchange pool and leaching. In streamwater from RF catchment, concentrations were mainly only elevated in the first year after fertilization (Table 2 and Fig. 6). In January through July 2013, the concentrations in RF streamwater were significantly higher than calculated unfertilized values, but for the whole of 2013 RF_{calc} and RF_{meas} values were similar, possibly with the exception of potassium (K) showing somewhat higher concentrations in 2013–2016. The high RF_{calc} values for 2013 were due to unusually high concentrations in the control (GT) stream in autumn 2013. Separate calculations for only the first nine months of 2013 gave lower values that were in line with values for the whole period 1997–2019 with autumn 2013 excluded. For Ca, this resulted in a RF_{calc} concentration of 4.4 mg L^{-1} for the last three months of 2013, compared with 1.6 mg L^{-1} as a mean for the first nine months, a similar concentration as in the calibration period 1997–2011. Mean RF_{calc} concentration for the years 2014–2019 was also lower, 1.5 mg L^{-1}. This means that autumn 2013 differed considerably from all other years, resulting in deviating values in 2013. However, from 2014, background and measured values were similar, showing no excess fertilized BC concentrations after 2014 except for Ca. All four base cations showed deviating high RF_{calc} concentrations during autumn 2013. Autumn 2013 omitted, RF_{calc} for the calibration period and the period 2014–2019 was 3 mg L^{-1}, compared with RF_{meas} value of 4.1 mg L^{-1} (1.1 mg L^{-1} higher than background) in 2013.

Similar calculations for the other base cations in 2013 gave Mg concentrations of 0.64 mg L^{-1} and 0.93 mg L^{-1}, K concentrations of 0.35 mg L^{-1} and 0.59 mg L^{-1} and sodium (Na) concentrations of 2.2 mg L^{-1} and 2.9 mg L^{-1} for RF_{calc} and RF_{meas}, respectively. In 2013, the concentration after fertilization was 0.3, 0.24, and 0.7 mg L^{-1} higher for Mg, K, and Na, respectively. Base cation concentration increased after fertilization, significantly (95%) for Ca in 2012–2015, Mg in 2012–2013, K in 2012–2013, and Na 2012 with autumn 2013, control values excluded (Fig. 6).

Calculated Ca outflow from RF catchment was ~6.3 kg ha^{-1} year^{-1} as a mean for the 7.5-year period studied and measured outflow reached 8.9 kg ha^{-1} year^{-1}, an excess of 2.6 kg ha^{-1} year^{-1}, 41% and 91% of Ca added with fertilizer. The first 1.5 years mainly contributed to the higher Ca outflow, with almost 17 kg ha^{-1} out of the total excess outflow for the 7.5 years of 20 kg ha^{-1} (Table 3).

For Mg, the mean background control value for the 7.5-year period was 1.4 kg ha^{-1} year^{-1} and measured mean outflow was 1.8 kg ha^{-1} year^{-1}, a 29% increase. The first 1.5 years contributed 3.7 kg ha^{-1}, a 176% increase, which represented 31% of the applied amount (12 kg ha^{-1} of Mg and 97% of total outflow in the 7.5-year period (Table 3).

Potassium (K) showed background outflow of 0.85 kg ha^{-1} year^{-1}, compared with the measured 1.1 kg ha^{-1} year^{-1} of K from the fertilized RF catchment as a mean for the 7.5 years after fertilization. In total for the period, excess outflow was 2 kg ha^{-1} of K, a 30% increase. The first 1.5 years contributed almost 100% of the excess outflow in the 7.5-year period (Table 3).

Sodium (Na), was also influenced by the fertilizer application and partly released from the soil stores. The Na concentration in streamwater was elevated from 2012 to 2014 and outflow increased. Background mean outflow was 4.9 kg ha^{-1} year^{-1} and measured outflow from RF catchment was 6.1 kg ha^{-1} year^{-1}, an excess outflow of 1.2 kg ha^{-1} year^{-1} (24% increase). In total for the 7.5-year period after fertilization, the excess flow was 8.9 kg ha^{-1} of Na (24% increase). The first 1.5 years contributed most to elevated outflow, 8.2 kg ha^{-1} or 93% of the total for the 7.5-year period (Table 3).

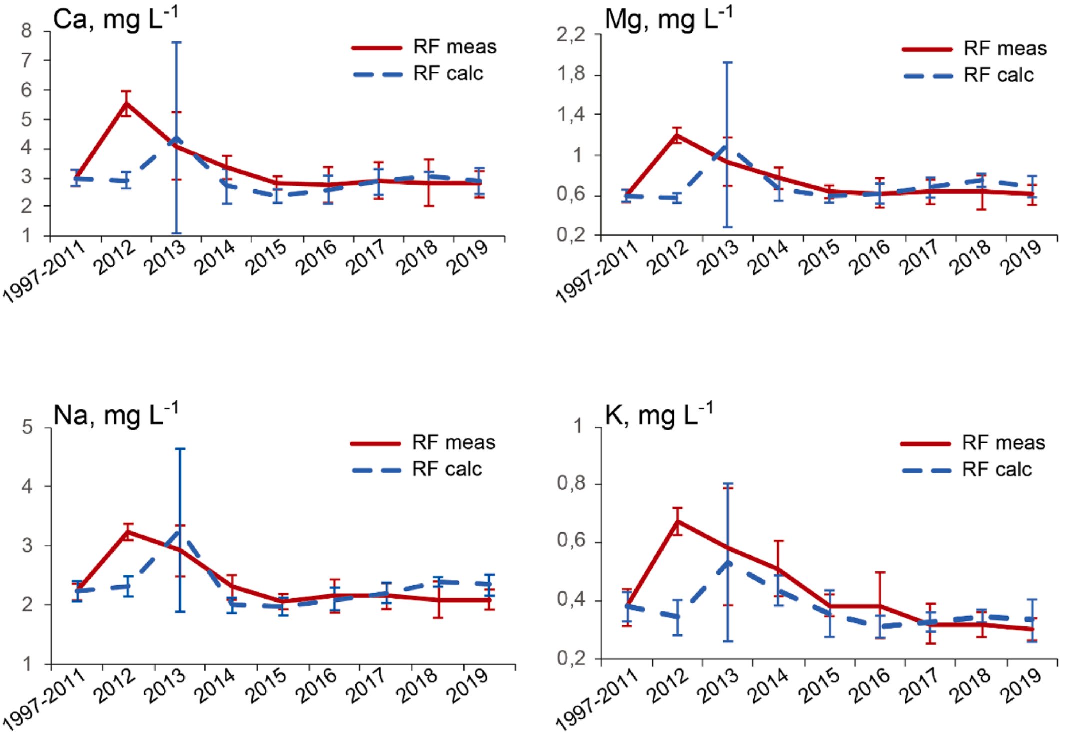

Fig. 6. Concentrations with error bars (mg L^{-1}) of the base cations calcium (Ca), magnesium (Mg), potassium (K), and sodium (Na) in stream water from Risfallet (RF) catchment before forest fertilization (mean 1997–2011) and the background concentration (RF_{calc}, blue dashed line) and measured concentration (RF_{meas}, red solid line) after fertilization (2012–2019). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3.8. Sulphfate, chloride, boron, and phosphorus

Anions such as sulphate (SO_4) and chloride (Cl) are affected by the ion balance change in the soil and soil water system (Table 2). In outflow from RF catchment, SO_4 showed only small changes, with the RF_{meas} concentration of 2.8 mg L^{-1} being 1.0 mg L^{-1} lower than the background in 2012 but later varying between 4 mg L^{-1} and 6 mg L^{-1}, with only 0.6 mg L^{-1} difference, and lower in the last two years (2018–2019). Sulphate outflow as a mean was 11.5 kg ha^{-1}, +14% for the whole period after fertilization with 3.7 kg ha^{-1} (30% excess) compared with calculated amount in the first 1.5 years. This was 4% of the total for the whole 7.5 years. Elevated SO_4 outflow was seen mainly in the first three years.

In the first 1.5 years, Cl concentration also showed only small changes (+0.1–0.5 mg L^{-1}), with measured concentrations mainly being 1–2 mg L^{-1} (Table 2). Mean Cl outflow for the whole period was 4.3 kg ha^{-1} $year^{-1}$, 0.2 kg ha^{-1} $year^{-1}$ higher than the background level (5% increase), and mainly occurred in the first 1.5 years (Table 3).

Boron was measured only in the first year after fertilization. It had background values of 1–2B μg L^{-1} and mean concentration in the first five months after fertilization was 16 μg B L^{-1}. By July 2013 this had levelled out to 5–6 μg L^{-1}, 3–4 μg L^{-1} higher than the background. Outflow of B in the first year after fertilization was 10 times higher than the background outflow (Lundin and Nilsson, 2014).

Phosphorus, not added with fertilizer, was expected to decrease in concentration with increasing forest growth, due to increased nutrient uptake. Total phosphorus (P_{tot}) in RF streamwater showed concentrations of 4–8 μg L^{-1} and the background concentrations and those measured in the first three years after fertilization were similar (Table 2). Later, in line with the trend for DOC, P_{tot} was 2.5 μg L^{-1} higher (by 50%; not significant) in RF until the end of 2019 (Fig. 7). No changes observed for PO_4-P, with measured concentrations that were similar to background concentrations in most years and generally very low (1–5 μg L^{-1}, or even lower).

Mean measured P_{tot} outflow for the whole period after fertilization was 0.016 kg ha^{-1} $year^{-1}$, slightly higher than the background (0.015 kg ha^{-1} $year^{-1}$). For the whole period, outflow reached 0.12 kg ha^{-1} of P_{tot} compared with a background value of 0.11 kg ha^{-1}. The first 1.5 years showed a 0.001 kg ha^{-1} increase, which was 10% of total outflow, reflecting the higher concentrations noted for the final five years (Fig. 7).

4. Discussion

Forest fertilization with N is a long-term measure to enhance biomass production as N is the main limiting nutrient for boreal forest growth (Nohrstedt, 2001). However, anthropogenic atmospheric deposition provided high N input for 20–30 years avoiding the need for fertilization (Vuorenmaa et al., 2009). In recent decades, atmospheric N deposition has declined (Forsius et al., 2020). This has renewed interest in forest fertilization especially as demand for wood products is high with, also the need to turn away from fossil fuels in a climate change perspective.

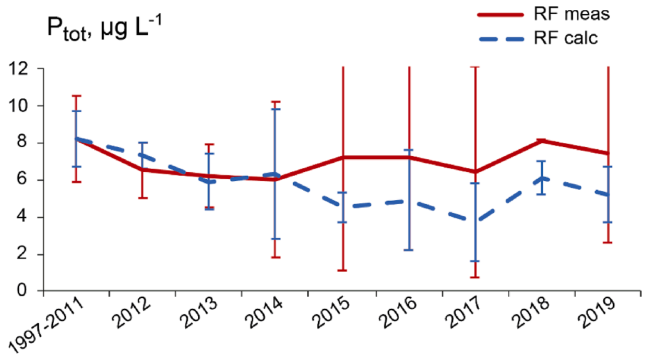

Fig. 7. Total phosphorus (P_{tot}) concentrations with error bars in Risfallet (RF) catchment stream water in 1997–2011 (mean), and the calculated background concentration (RF_{calc}, blue dashed line) and measured concentration (RF_{meas}, red solid line) after fertilization (2012–2019). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Catchment investigations of forest fertilization have been carried out previously but often the treated part of the catchment has been rather small (20–50%) (Grip, 1982) giving an uncertain signal from the treatment. In RF catchment fertilization encompassed almost 80% of the catchment area furnishing a strong signal. Furthermore fertilization today is mainly conducted on well-drained soils i.e. with the groundwater mainly below 1 m depth allowing a considerable part of the fertilizer to be stored in the unsaturated upper soil where it is available for tree roots but not easily transported to surface waters. This gives fairly stable hydrological conditions compared with the more flashy discharge pattern in catchments with dense glacial till with high groundwater levels often close to the surface. To avoid direct input into the streams in the RF catchment a ground tractor was used for spreading contrary to aerial fertilization that often involves direct inputs to streams (Lundin and Bergquist, 1985). However, first-year mean N_{tot} in streamwater from RF catchment reached 3.25 mg L^{-1} compared with a previously reported highest concentration of 4 mg L^{-1} (Binkley et al., 1999). The highest occasional N_{tot} concentration was 9.4 mg L^{-1} compared with a background concentration of 0.4 mg L^{-1}.

A study on the initial effects of forest fertilization in RF catchment showed considerable leaching for several elements in the first year after fertilization (Lundin and Nilsson, 2014). However, measurements of hydrology and streamwater chemical conditions have continued for a longer period now embracing over seven years until the end of 2019. This allowed determination of the duration of leaching after fertilization. Leaching of N in unfertilized boreal forests is usually <2 kg N ha-1 year −1, and N_{org} the main N species (Högberg, 2017).

In RF catchment initial N leaching was 14% of the applied amount in the first year after fertilization and elevated leaching occurred for five years with total excess outflow of 25.8 kg ha^{-1} 17% of the applied amount. The N fertilizer used provided equal amounts of NO_3-N and NH_4-N but the N outflow was dominated by NO_3 as NH_4 was nitrified and only limited excess NH_4-N was seen in outflow. This transformation contributed to other soil chemical processes such as hydrogen increase and base cation exchange.

In earlier studies of forest fertilization it was found a rate of 20%, and aerial application to peatlands resulted in 22% of N being lost in outflow (Grip, 1982; Lundin and Bergquist, 1985). Low leaching losses (0–5% of added N, 1–2 years after fertilization) have been reported by Flint et al. (2008) and Davis et al. (2012). Other studies have reported N leaching values of 7–15 kg ha^{-1}, 5–10% of added N (Melin and Nômmik, 1988; Ring, 2007). A very high N outflow (91 kg ha^{-1}, or 45% of applied N) during the first 2 years after fertilization was reported by Davis et al. (2012) for a young Pinus radiata plantation in New Zealand. There was a coarse coastal sand soil and an average annual precipitation amount of 1016 mm. Coarse soils and high precipitations amounts are of course two important factors for high N leaching after N addition. Forestry operations such as tree harvesting also increase N outflow by 2.7–9.5 kg N ha^{-1} (Wiklander et al., 1990; Lundin, 1999; Löfgren et al., 2009). Other important factors are fertilizer dose and type of fertilizer, time of application, single or repeated N applications, topography and ground vegetation (Saarsalmi and Mälkönen, 2001; Hedwall et al., 2018). One extraordinary condition in the present case was high discharge in the first five months after fertilization. The combined amount was 321 mm compared with normal runoff in those months of 66 mm. This contributed to high N outflow to surface waters. The commonly used 150 kg N ha-1 has been discussed and repeated smaller additions mentioned. However, it has been noticed higher growth effects from one high dose compared to several small additions (From, 2014).

Nitrogen addition to a soil system often lowers pH and contributes to soil exchange of base cations (Perakis et al., 2013). In streamwater from RF catchment the initial effect of forest fertilization was increased acidity. The decrease in pH persisted for the first three years, but pH was lower than the background level even in also the last three years of the study period. Nitrification and soil exchange processes together with elevated groundwater levels contributed to the lower pH.

Since N fertilization increases soil acidity base cations were added with the fertilizer as dolomite ($CaMg(CO_3)_2$) to mitigate acidity. Outflow of Ca and Mg increased as a result and represented substantial losses to surface waters. This partly also influenced release from the soil exchange pool with K and Na showing excess outflow compared with background values. Cations balanced the considerable outflow of NO_3-N partly including cations released from the soil pool. Calculated ion balance showed anions of 2.0 kEq ha^{-1} and base cations of 2.5 kEq ha^{-1} for the first 1.5 years without organic anions included. Including the contribution of organic anions from DOC added 0.5 kEq ha^{-1} bringing anion levels up to cation levels (Lundin and Nilsson, 2014). Similar calculations for the whole 7.5-year period gave 6.6 kEq ha^{-1} and 6.7 kEq ha^{-1} for anions and cations respectively. Altogether ion balances showed reasonable agreement for the values estimated.

Organic matter content in streamwater increased over time with additional DOC and N_{org} especially in the last five years (2015–2019). Also water colour increased contributing to brownification. This was possibly because fertilization increased forest growth providing more litterfall with subsequent decomposition adding organic substances to topsoil layers and increasing leaching to surface waters. There are studies that found increased DOC concentrations (Pregitzer et al., 2004; Fröberg et al., 2013; Shi et al., 2019). However, other field studies did not find effects on DOC from N applications (Magill et al., 2004; McDowell et al., 2004; Rappe-George et al., 2013). In fact the conventional concept for forest fertilization is to repeat application after around seven years which corresponded well to the duration of fertilization effects seen in this study.

5. Conclusions

Forest fertilization encompassing 80% of a Swedish catchment influenced the chemical composition of streamwater particularly in terms of elements added with the fertilizer (N, Ca, Mg and B) but also of other soil chemical compounds (major cations and anions, N_{org} DOC and H). Nitrogen showed the largest changes with 15% of the applied amount of N_{tot} lost in outflow in the first 1.5 years and 20% in the 7.5-year study period. Nitrate dominated N_{tot} outflow making up 80%. Duration elevated N concentrations in outflow persisted for 5.5 years after fertilization. In the first 1.5 years after fertilization hydrology and meteorological conditions most likely had considerable impact on outflow since unusually heavy rainfall resulted in high groundwater levels and high discharge from the study catchment. In the last years of the study period N_{org} dominated N_{tot} as also seen before fertilization and in the control where N_{org} made up 86% of N_{tot}. Increased outflow in the last four years of the period was seen for organic matter (DOC) and thus also for N_{org}. In addition, water colour increased a few years after fertilization. Fertilizer effects on tree growth probably contributed to increased litterfall organic matter storage in the humus layer and subsequent decomposition increasing organic matter leaching.

Nitrogen fertilization can affect soil and surface water acidity so dolomite was added to the fertilizer. This gave a considerable increase in Ca outflow representing 82% of the applied amount. However some leaching was probably related to soil exchange processes. Magnesium outflow was 32% of the applied amount but addition of dolomite mitigated proton release. However pH was on average 5.5 in the first half-year after fertilization 0.4 units lower than in the calculated unfertilized control. Over the whole study period (7.5 years) average pH was 5.9 which was 0.2 units lower than the background, i.e. not a hazardous change.

Ground spreading instead of aerial application was beneficial as it avoided direct inputs of fertilizer to surface water. However weather conditions represented probably the worst case as regards leaching with very heavy rain and high discharge in the year directly after fertilization. The till soil depth stratified hydraulic conductivity with high values in upper soil layers and relatively high groundwater levels caused by high precipitation in the initial period after fertilization provided high groundwater flows. If instead, it would have been drier with deeper groundwater levels, fertilizer would have percolated to deeper soil layers with lower conductivity and more fertilizer stored longer in the soil.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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