Water-quality genesis in a mountain catchment affected by acidification and forestry practices

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Abstract: Effects of changes in air pollution and forest cover on the acid atmospheric deposition and runoff were studied in the Jizerka experimental catchment (Czech Republic), a sensitive mountain environment of low buffering capacity. From 1982 to 2015, resident scientists and volunteers measured water quality of precipitation, fog, and stream samples at the watershed level. Archived LANDSAT imagery was used to reconstruct changes in forest composition in the watershed based on a detailed ground inventory done in 2010 to 2012. Spatial interpolation was used to approximate atmospheric deposition of water and SO_4^{2-}, NO_3^-, and NH_4^+ over the watershed area. The open-field load of S peaked in 1987 to 1988 and dropped substantially in the 1990s, but inorganic N did not show a significant trend. The N : S deposition ratio increased from 0.37 to 2.83. Mean annual stream-water pH increased from 4.2 to 5.9, and concentrations of SO_4^{2-} and NO_3^- decreased by 55 and 53%, respectively. Seasonal acidification of stream water was observed during snowmelt (March, April) and episodic summer rainstorms. The relatively rapid response of stream-water quality to reduced deposition corresponded with subsurface runoff generated in a shallow podzolic soil. Relatively high leaching of NO_3^- in the 1980s followed limited N uptake in damaged spruce stands and clear-cut areas. Recovery of stream-water chemistry followed the drop in the acid atmospheric deposition by ~5 y, and stream biota revived after 10 to 15 y. Removal of spruce forest and reduced air pollution caused faster recovery from acidification than expected from pure air-quality improvement. Reduced atmospheric deposition and fog-drip interactions caused by lower canopy area suggest that modified forestry practices can affect deposition rates and stream-water quality. Deciduous or mixed forests could decrease the acidic atmospheric load by reducing leaf area and surface roughness.

Key words: mountain catchment, acid atmospheric deposition, forestry practices, runoff genesis, citizen science

Mountainous parts of many river basins provide 40 to 80% of the water that is available to lowland users (Messerli et al. 2004). The importance of mountain catchments as water resources will increase with population pressure (UNEP 2007, Viviroli et al. 2007) and effects of expected climate change (Christensen 2005). Leopold (2006) emphasized the role of headwater mountain streams in river system development, and Körner and Ohsawa (2005) considered the recharge of water resources as the most important environmental benefit of mountain regions. Mountain watersheds in central Europe are mostly forested, and their sustainable environmental benefits are guaranteed by forestry practices (FAO 2008).

Biswas et al. (2014) suggested that water-quality deterioration at the global scale is attributable mainly to poor management of water resources. The European Commission (2012) recommended application of a multidisciplinary approach to watershed management and revision of stream water-quality regulations. In populated regions, the quality of natural fresh waters is degraded mostly by point-source pollution, whereas distant mountain catchments are particularly affected by large-scale air pollution (emissions of SO_2, NO_x, NH_4^+) and atmospheric acid deposition (Reuss and Johnson 1986, Baldigo and Lawrence 2001, Schöpp et al. 2003, Kopáček et al. 2016).

Anthropogenic emissions of acidic precursors have been increasing since the industrial revolution and peaked in the late 1980s. International cooperation to reduce atmospheric emissions (the 1985 Helsinki Protocol on the Reduction of Sulphur Emissions or their Transboundary Fluxes by ≥30%)

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has led to signs of recovery in acidified European headwater regions (Křeček and Hořická 2001, Holen et al. 2013). Falkenmark and Allard (2015) called for a detailed analysis of natural waters from a dynamic perspective, but too many studies in headwater catchments have investigated only baseflow conditions. Thus, studies are needed of water movement through the surface and subsurface environments combined with chemical reactions taking place along their pathways (Bolstad and Swank 1997, Takagi 2015).

Lumb et al. (2011) reviewed methods of indexing with a numerical value based on physical, chemical, and biological indicators, including especially pH and NO_x loading. For larger-scale investigations, Rapport et al. (1998) emphasized the important role of citizens in monitoring indicators to assess water quality, and the USEPA (1997) developed detailed methods of volunteer water monitoring. Since the 1970s, several nonprofit organizations have been founded to promote participation of lay volunteers in environmentally sound field research (Hand 2010). Irwin (1995) and Silvertown (2009) see involvement of volunteers in collecting and processing the field data as an important part of scientific inquiry and environmental education.

Czech Republic was graded above average relative to 147 countries based on the Water Poverty Index (Lawrence et al. 2002). However, the Czech Republic received lower values for environmental indicators associated with the risk of water pollution. In the headwaters of the Jizera Mountains (northern Bohemia, Czech Republic), water acidification began to be visible in the 1950s and peaked in the mid-1980s (Křeček and Hořická 2006). Its consequences were a large-scale (40–80%) die-back of spruce stands and their subsequent removal particularly in headwater catchments, a decrease in water pH, and degradation of life in streams and water reservoirs (Stuchlik et al. 1997). The number of species in planktonic and benthic communities was significantly reduced, and fish became extinct in the late 1950s.

Our objective was to analyze long-term (1982–2015) changes in water quality in the Jizerka experimental catchment and link these data to changes in atmospheric acid deposition and forest cover. We hypothesized that on a catchment scale, the acid atmospheric load and streamwater quality in mountain regions could be ameliorated by forestry practices.

METHODS

Study site

The study was performed in the upper plain of the Jizera Mountains (Fig. 1). In the 1980s, this area was strongly affected by acidic atmospheric deposition and die-back of spruce plantations (*Picea abies*). After the clear-cut of damaged spruce stands, grass-dominated *Junco effusi–Calamagrostietum villosae* became a new dominant commu-

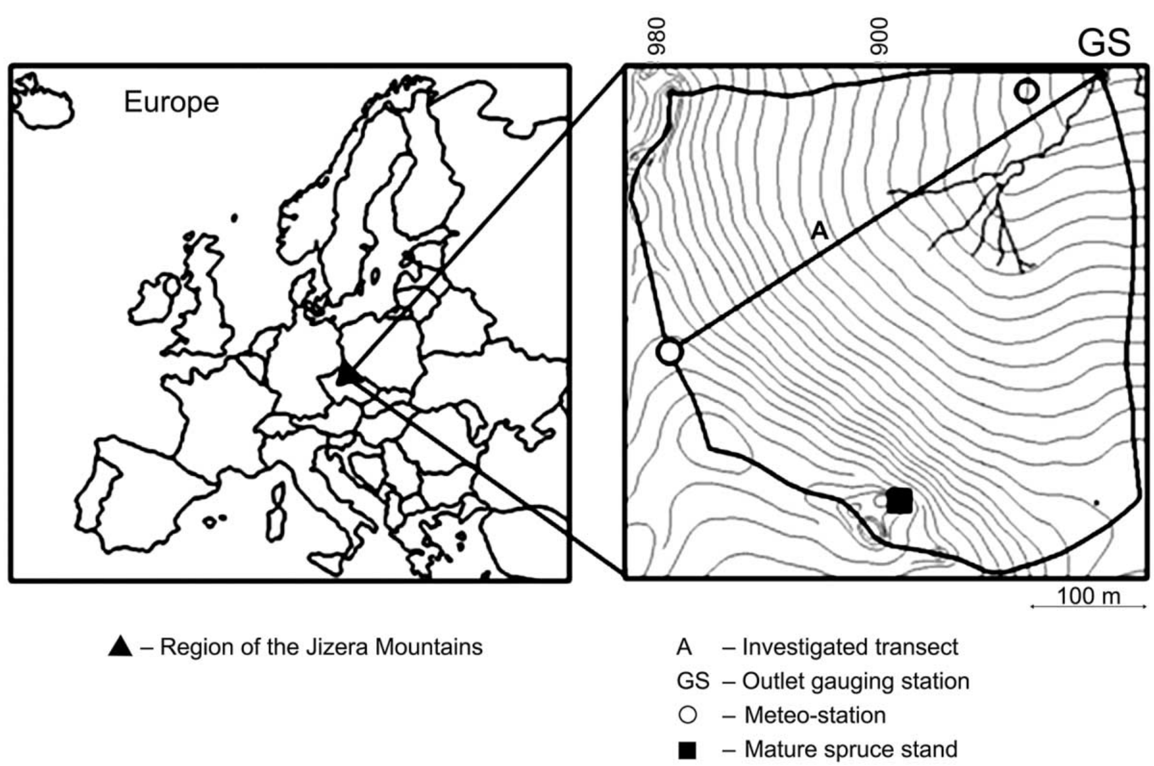

Figure 1. The Jizerka experimental basin in Europe.

nity (Křeček et al. 2010). The Jizerka experimental catchment (lat 50°48′21″–50°48′59″N, long 15°19′34″–15°20′48″E, Elbe river district 1-05-01-004; Table 1) has operated since 1981. Characteristics of the recent climate (1961–1990) are: north temperate zone, Kőppen Dfc subarctic region, mean annual precipitation = 1400 mm, air temperature = 4°C, average maximum snowpack = 120 cm (snow cover usually lasts from the beginning of November to the end of April; Tolasz 2007). Here, low-base-status soils (sandy–loamy podzols) between 0.5- and 1.2-m depth have developed above porphyritic granite bedrock. Topsoils are dominated by grass root systems to depths of 15 cm. The topsoil is composed of litter (O_l, depth = 0–2 cm), humus layer ($O_f + O_h$ = 2–10 cm), and leached horizon (A_h= 10–15 cm). Mor is the most common humus (2–5 cm). The area is characterized by rapid subsurface runoff where the ground is restricted to shallow, weathered rock formations. In this basin, climax forests include Norway spruce (*Picea abies*) and common beech (*Fagus sylvatica*), but, since the end of the 18th century, spruce plantations have dominated the landscape. In 1984–1988, mature spruce stands (showing ~30% defoliation) were harvested by clear-cutting followed by reforestation with coniferous stands.

Instrumented catchment

The experimental basin (Fig. 1) was instrumented in 1982. The outlet is equipped with a sharp-crested v-notch weir with an automatic water pressure and temperature recorder ALA 4020 (ALA, Bučovice, Czech Republic) logging every 10 min. In situ monitoring of stream waters, including temperature, pH, and conductivity, was done with the field multimeter WTW-350i (WTW, Weilheim, Germany). Meteorological observations were made along transect A (established for hypsometric studies) at 875 and 975 m asl. Two Czech Hydrometeorological Institute climate stations (Kořenov-Jizerka and Desná-Souš, elevation = 772 and 850 m asl) are ~2500 and 300 m from the catchment boundary.

Table 1. Geomorphology of the Jizerka catchment.

Variable	Value (range)
Area (km^2)	1.03
Elevation (m)	927 (862–994)
Slope (%)	7.52 (0.02–24.33)
Shape index	0.69
Length of streams (m)	1490
Drainage density (km/km^2)	1.45
Length of the main stream (m)	657
Slope of the main stream (%)	5.98
Strahler stream order	2

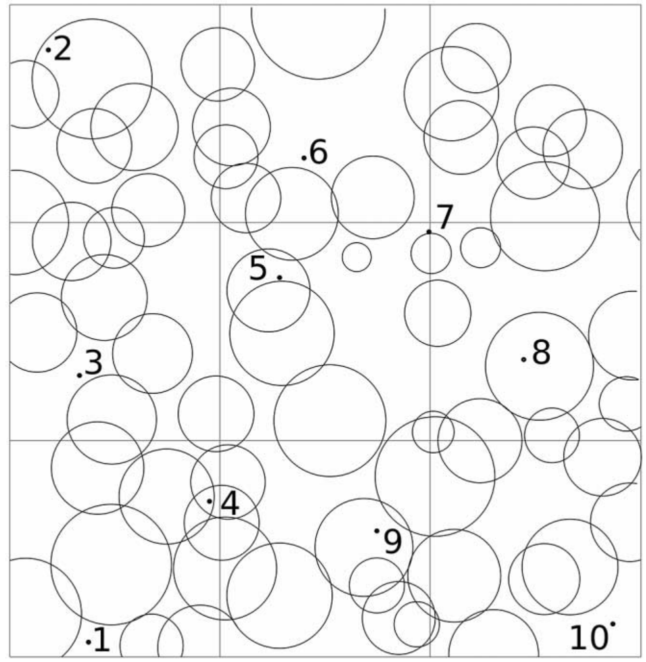

Figure 2. Locations of rainfall collectors (1–10) and the horizontal canopy projection of the mature spruce stand.

Along the vertical transect A (harvested in 1984), an additional 3 modified Hellmann rain gauges (area = 200 cm^2, plastic collectors with a shield against bird contamination, 1-L sampling bottles) were placed at elevations of 862, 899, and 975 m asl in 1991. In the mature spruce stand (plot area = 30 × 30 m, elevation = 975 m), through-fall under the canopy (Fig. 2) was sampled with 10 rainfall storage gauges (200 cm^2), and stemflow was collected at 2 average trees. This method of through-fall observation is recommended as the most appropriate approach to identify fog and cloud water deposition (Lovett 1988). Sets of modified storage gauges (200 cm^2) were installed in the soil to collect through-fall under the ground vegetation in 2 harvested (and reforested) sites at elevations of 862 and 975 m asl. To identify the evidence of fog drip, 12 passive fog collectors were installed along transect A (862–994 m). At each collector, the fog drip was generated by 400 m of Teflon line (diameter = 0.25 mm, surface area index [SAI] = 5) exposed at the height of 1.7 m above the ground. Sample bottles were protected against direct rainfall access by a wide-brimmed cover that overlapped the fog collector at an angle of 34°.

Water and biota sampling, analytical methods

Stream waters at the catchment outlet were sampled weekly with more frequent sampling during flood events, and deposited rain/fog drip was sampled monthly with more frequent sampling (weekly or after individual rain events) during the field expeditions. Samples (rain/fog drip, through-fall, stemflow, stream water) were filtered through

40-μm inert mesh, stored in the refrigerator, and analyzed in the laboratory at the Hydrobiological Station Velký Pálenec (Charles University, Prague). Concentrations of all ions were determined by ion chromatography with conductometric detection; pH was measured with radiometer combination electrodes, and conductivity determined by radiometer conductometric sensor (Stuchlík et al. 2006).

The chemical composition (especially N) of the rain/fog drip samples can be altered by bacterial activity at the time of collection. However, in our study area, bacterial activity is greatly reduced by a combination of the relatively cold mountain climate (subarctic region), relatively high concentrations of N, and low values of pH and dissolved organic C (Cape et al. 2001). The potential growth of algae was reduced by the dark sampling bottles and shelters.

Benthic macroinvertebrates were sampled near the outlet of the Jizerka catchment (a low-gradient stream channel with only sand and gravel substrate types, depth < 0.5 m) in May (after snowmelt), July/August (high summer), and in September/October (a relatively dry period). In 2004–2005, this sampling was done in the framework of a Czech regional campaign (Horecký et al. 2013). A kick-net sampling technique was used (Rosenberg and Resh 1993). The invertebrates were collected with a hand-net (mesh size = 500 μm) then sieved through a 300-μm net and preserved with an 80% ethanol solution. Accurate counts of each taxon were undertaken in the laboratory (by eye and under a binocular microscope at 12–16 × magnification) by trained professional staff.

Catchment inventory

The archive of LANDSAT imagery (NASA 2014) was used to detect development of the vegetative cover (1984, 1992, and 2010). This imagery has a resolution of 30 m. Only data from clear-sky summer seasons (June–August) were used. The normalized difference vegetation index (NDVI) was calculated for the spectral reflectance registered in the visible (red) and near-infrared bands according to Weier and Herring (2000):

$$\mathrm{NDVI} = (NIR - VIS)/(NIR + VIS), \qquad \text{(Eq. 1)}$$

where NIR is near-infrared radiation (0.7–1.1 μm), and VIS is visible radiation (0.4–0.7 μm).

Supervised classifications of multiband raster images (Landsat 4 and 5) were used simultaneously, and images representing distinct sampling areas of the different canopies were classified with the image analyst tool in ArcGIS (version 10.2; Environmental Systems Research Institute, Redlands, California; Nagi 2011).

Since 1991, detailed forest inventories have been conducted during field surveys of 12 (20- × 20-m) fixed quadrats situated at 100-m incremental altitudinal steps along transect A. Basic forestry variables (tree species, age, basal area, tree height, timber volume, and horizontal canopy density) were evaluated by standard techniques (Watts and Tolland 2005). Complementary detailed botanical investigations included phytosociological relevés (4 × 4 m) and seasonal development of herbaceous canopy (height, canopy area). The assimilating area of grass was measured with a portable leaf area meter LI-3100C (LI-COR, Lincoln, Nebraska). In 2012, the leaf area of spruce stands was estimated by direct ground-based measurements (Breda 2003). The definition of leaf area index (LAI) was interpreted as ½(total green leaf/needle area per unit surface area), as recommended by Chen and Black (1992).

Five canopy classes were identified from the multiband raster images in the years 1983, 1985, 1992, 2002, and 2010. These 5 classes were: 1) mature spruce forests, 2) stands with crown closure >0.3, 3) reforested plots with crown closure < 0.3, 4) areas covered by herbaceous communities only, and 5) clear-cut (Křeček and Krčmář 2015). These classes correspond with definitions of forest used by the United Nations Framework Convention on Climate Change (crown closure > 0.3, height >2–5 m at maturity; Sasaki and Putz 2009).

Atmospheric deposition and runoff genesis

According to the findings of Krečmer et al. (1979), Wrzesinsky and Klemm (2000), and Křeček et al. (2017), atmospheric precipitation is affected by both elevation and vegetative canopy. The hypsometric method was used to assess the effect of elevation on precipitation, canopy throughfall, and deposition of SO_4^{2-}, NO_3^-, and NH_4^+ under the canopy (Křeček et al. 2017) using the same 5 canopy classes used by Křeček and Krčmář (2015). Seasonal atmospheric loads were estimated as (Křeček et al. 2017):

$$m = (bE + b_0)F_c/A_{er}, \qquad \text{(Eq. 2)}$$

where m = seasonal load (summer and winter), b and b_0 = empirical hypsometric coefficients, E = elevation, A_{er} = effective receptor area, and F_c = fog-drip coefficient.

Methods of spatial interpolation (ArcGIS) were used to approximate the catchment deposition of water and acidifying substances (SO_4^{2-}, NO_3^-, and NH_4^+), and their runoff was estimated from concentrations and discharge (Q) measured at the catchment outlet. Mean annual concentrations were calculated by weighted averages, and mean pH values were recalculated from converted values of H^+. The method of local minima in the hydrograph separation was applied to detect fast (direct) runoff in the catchment (Sloto and Crouse 1996).

Standard descriptive statistics and 1-way analysis of variance (ANOVA) was applied to analyze the data sets and to identify relationships between the groups of data (Motulsky and Searle 1998). Trends in the time series data (and a change in trends) were detected by the Change and Trend Problem Analysis (CTPA) programme (WMO 2001).

Participation of citizen scientists

From 1991 to 2012, voluntary citizen scientists participated in ground observations. This participation made possible extra and time-sensitive sample collection during critical hydrological events. Each year, 4 to 5 teams each involving 4 to 8 volunteer participants spent 2 wk engaged in supervised field surveys organized by the Earthwatch environmental program (Earthwatch Institute 2012). After the standard preliminary selection done by Earthwatch, volunteers were instructed and trained in the field. The accuracy of their results was assessed daily. This project also was focused on the environmental education of volunteers, so after the 2-wk field activities and thematic discussions, their knowledge and skills were evaluated by specific tests.

According to findings of Robson et al. (1993) or Hodgson and Evans (1997), stream-water observations in upland watersheds require sampling at hourly or shorter intervals to provide good temporal resolution. Therefore, volunteers also gathered more detailed information on stream-water characteristics (temperature, pH, and conductivity) and collected water samples during some snowmelt and rain events. Data measured by volunteers in situ were controlled under laboratory conditions.

RESULTS

Changes of the canopy

The Jizerka catchment was covered by mature spruce stands. In 1984, 62% of the catchment area was harvested by clear-cutting, and ~88% was harvested by the end of the 1980s. Reforestation was complicated by establishment of invasive grass communities (*Calamagrostis* sp., predominantly *Calamagrostis villosae*) that spread over the basin area. Characteristics of the monitored reforested stands along transect A (A1–A12) and the mature stand (Fig. 1)

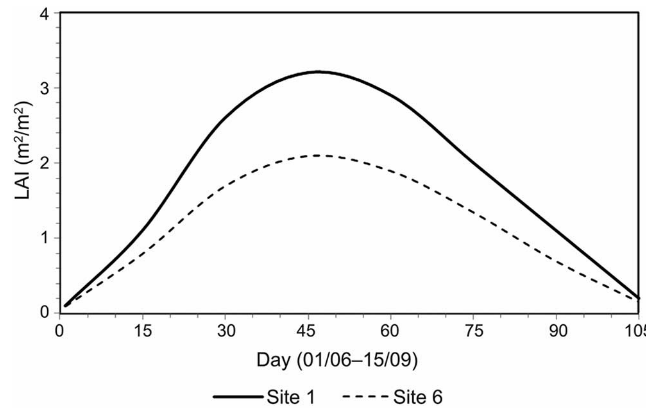

Figure 3. Seasonal changes in the living canopy of grass as leaf area index (LAI) along the A transect in 2012. Days are formatted dd/mm.

are presented in Table 2. In the reforested area (2012 inventory), blue spruce (*Picea pungens*) and Norway spruce (*Picea abies*) predominated with areal coverage of 48 and 32%, whereas the areal percentage of deciduous species, mountain-ash (*Sorbus aucuparia*) and silver birch (*Betula pendula*), was only 10%.

In 2012, 25 y after the clear-cut, LAI of reintroduced trees was 0.11 to 2.76 (mean = 1.31 ± 0.26). In addition, the seasonal assimilating surface of the herbaceous canopy (Fig. 3) reached maximum LAI values between 2.1 and 3.2 in high summer. The corresponding values of NDVI varied from 0.66 to 0.76 (mean of 0.72 ± 0.1) and were relatively insensitive to changes in the canopy when LAI was >2. Thus, the supervised classification of multiband raster images (Landsat 4, 5) was a more efficient indicator of canopy classes according to the crown closure of trees. In 1982 to

Table 2. Characteristics of the monitored stands in 2012. N_t = number of trees, CD = canopy density (crown closure), H = average tree height, LAI = leaf area index of trees, NDVI = normalized difference vegetation index.

Stand	Elevation (m)	N_t	CD (m^2/m^2)	H (m)	LAI (m^2/m^2)	NDVI
A1	862	107	0.38	3.62	1.9	0.68
A2	870	416	0.62	2.33	2.63	0.66
A3	878	317	0.55	2.61	2.07	0.69
A4	885	192	0.28	2.74	1.28	0.68
A5	891	324	0.59	3.96	2.76	0.70
A6	899	99	0.31	5.42	1.56	0.71
A7	907	72	0.06	0.50	0.11	0.75
A8	918	56	0.16	1.48	0.37	0.76
A9	930	102	0.21	2.61	1.06	0.75
A10	942	49	0.20	3.86	1.08	0.75
A11	961	146	0.15	2.39	0.5	0.75
A12	975	90	0.17	2.41	0.42	0.75
Mature stand	975	68	0.78	23.0	6.7	0.90

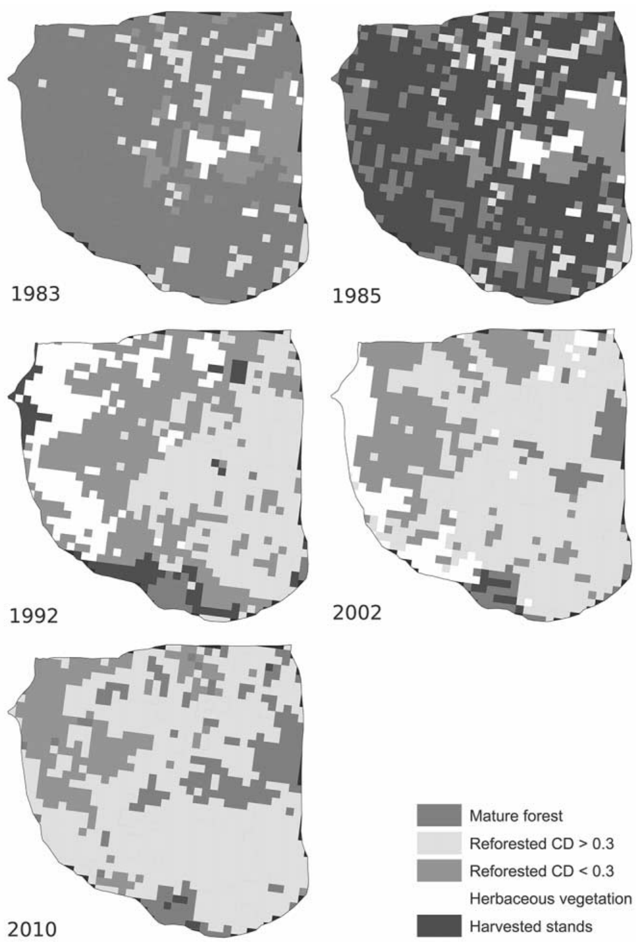

Figure 4. Canopy crown closure of trees (CD) in the Jizerka catchment from 1983 to 2010.

2015, canopy changes were reconstructed from the multiband imagery (Fig. 4). The areal percentage of the 5 canopy classes is presented in Table 3.

Acidic atmospheric deposition

In 1982 to 2015, open-field deposition of SO_4^{2-}-S and N (NO_3^--N and NH_4^+-N) recorded in most of the Jizerka catchment correspond with content of SO_2 and NO_x in the air ($r = 0.87$ and $r = 0.48$, respectively; $r_{crit} = 0.32$, $n = 34$, $p = 0.05$; moving averages of order 3 are presented in Fig. 5). Atmospheric concentrations of SO_2 and NO_x were retrieved from the standard observation network of CHMI (2016): AIM (Ambient Ion Monitor) stations Desná-Souš (LSOU-1022) and Kořenov-Jizerka (LJIZ-1047). SO_2 was measured by UV fluorescence over 10 min, and NO_x was measured by chemiluminescence in hourly intervals.

Table 3. Areal percentage of the canopy classes in the Jizerka basin (1982–2010). CD = canopy density (crown closure).

Stands	1983	1985	1992	2002	2010
Mature forests	71	9	2	7	16
Reforested CD > 0.3	14	14	38	58	65
Reforested CD < 0.3	11	11	38	22	18
Grass	4	4	16	11	0
Clear-cut	0	62	6	2	1

In 1982 to 1992, mean annual concentrations of SO_2 exceeded the threshold for forests (20 µg SO_2/m^3; Posch et al. 2001), whereas concentrations of NO_x were below the critical value (30 µg NO_x/m^3). The deposition of S showed a decreasing trend with gradient of −0.12 (t = 20.3, t_{crit}= 2.1, p = 0.05; WMO 2001). However, the trend in the deposition of N was not quite significant (t = 0.69). In 1982 to 2015, mean annual pH of precipitation increased from 4.2 to 5.3, and a pH-relevant annual open-field flux of H^+ in precipitation decreased from 90 to 5 mg m^{-2} y^{-1}. However, based on the deposition of SO_4^{2-}, NO_3^-, and Cl^- (Yang et al. 2010), the open-field H^+ flux decreased from 265 to 86 mg m^{-2} y^{-1}, and the total H^+ flux in the Jizerka catchment decreased from 325 to 93 mg m^{-2} y^{-1}. The annual open-field flux of buffering basic cations (Ca^{2+}, Mg^{2+}, K^+, and Na^+) fluctuated between 1.67 and 3.35 (mean = 2.86) g m^{-2} y^{-1}, and did not show any significant trend.

Interception loss of spruce stands was modified by the deposition of fog water on the canopy. The observed canopy storage capacity in the 975-m^2 study stand (Figs 1, 2) was 2.3 mm. Given a seasonal rainfall of 683 mm and ~100 rainy days saturating the storage capacity, the total interception loss of the spruce canopy (not affected by fog) was ~230 mm (34% of the gross precipitation). Therefore, the interception loss of 112 mm (16% of gross precipitation) was evidently caused by additional deposition of

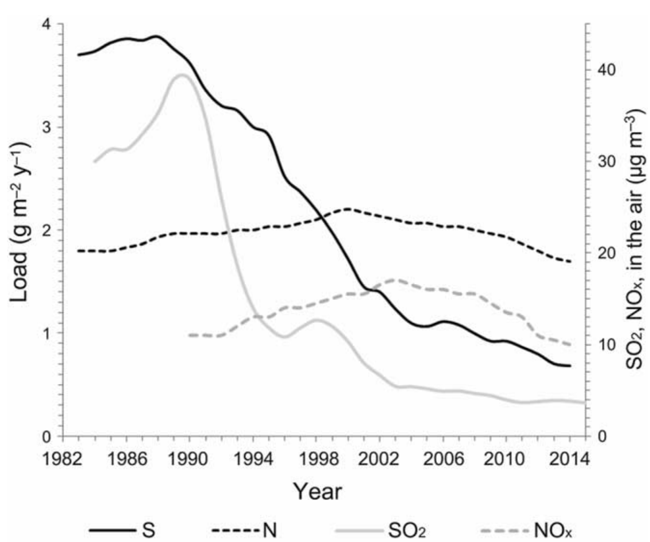

Figure 5. Concentrations of SO_2 and NO_x in the air and the open-field load of S (SO_4^{2-}-S) and inorganic N (NO_3^--N and NH_4^+-N): moving averages of order 3 in the Jizerka catchment from 1982 to 2015.

Table 4. Seasonal coefficients b and b_0 for summer (May–October) and winter (November–April) loads of acidifying substances.

	Summer		Winter	
Chemicals	b	b_0	b	b_0
SO_4^{2-}-S	0.0766	−56.1069	0.1126	−81.7721
NO_3^--N	0.0270	−19.8859	0.0362	−26.2902
NH^{4+}-N	0.0754	−55.5671	0.0940	−68.6638

fog water. The volume of stem-flow was negligible. Along transect A (Fig. 1), measurable volumes of fog water were collected by passive collectors at elevations >900 m (collectors 7–12). Mean monthly fog drip and elevation were significantly correlated in summer (May–October) and winter (November–April) (r = 0.93 and 0.98, respectively; r_{crit} = 0.75, n = 5, p = 0.05). These relationships have slopes significantly different from 0 (p = 0.0082 and 0.0033, respectively) and no significant departure from linearity (F = 39.3 and 73.75, respectively; F_{crit} = 9.78). The load of fog drip was greater in winter than in summer by 23 to 50%.

The empirical coefficients b and b_0 (Eq. 2) based on field observations in 2010 to 2012 are given in Table 4. Fog-drip coefficients F_c were calculated as 1 (dense mature stand), 0.33 (stand of crown closure >0.3), and 0.18 (area overgrown by grass).

The atmospheric load of water and acidifying substances (SO_4^{2-}, NO_3^-, and NH_4^+) in the Jizerka catchment was estimated by spatial interpolation based on the canopy classes and elevation (Eq. 2) in 1982 to 2015. Annual values of fog drip, open-field deposition, and an additional canopy load (total loads are sums of the open-field and the extra canopy values) are shown in Fig. 6A–C. These data reconstruct the atmospheric load at a catchment scale. In 1982 to 2015 (34 y), the mean annual runoff of S (2.96 g m^{-2} y^{-1}) exceeded the open-field deposition (2.14 g m^{-2} y^{-1}) by 38%, but not the total deposition (3.31 g m^{-2} y^{-1}, 89%) (Fig. 6B). The mean annual runoff of N (0.95 g m^{-2} y^{-1}) was less than the total (2.43 g m^{-2} y^{-1}, 39%) and the open-field (1.68 g m^{-2} y^{-1}, 56%) loads (Fig. 6C).

The decreasing trends in the output (runoff) of S and N (slopes = −0.163 and −0.025, respectively; t = 22.49 and 11.2, t_{crit} = 2.1, p = 0.05) exceeded that found in the open-field deposition (−0.12 and 0.002; Fig. 5). Comparing data from 1982 to 1984 (before the forest clear-cut) with 2001–2015 (after the drop in emissions and forest regrowth), mean total annual deposition of S decreased from 8.7 to 1.6 g m^{-2} y^{-1} (the extra loading on the canopy decreased from 60 to 40%, Fig. 6B), whereas N did not change significantly (from 2.62 to 2.74 g m^{-2} y^{-1} with decreasing canopy effect from 45 to 27%; Fig. 6C). Annual fog-drip amounts corresponded to the % forest area covered by stands with crown closure >0.3 (r = 0.79, r_{crit} = 0.32, n = 34, p = 0.05; Fig. 6A). From 1982 to 2015, atmospheric N deposition consisted of 72% NH_4-N and 28% NO_3-N. The ratio between the total N and S loads increased from 0.35 (1982–1988) to 2.0 (2011–2015).

Runoff genesis

Significant correlations were found between mean annual stream-water characteristics (pH, contents of SO_4^{2-}, and NO_3^-) and the air pollution (AP; concentrations of SO_2 and NO_x combined) and mean canopy density (CD) of the Jizerka catchment (Table 5). These characteristics

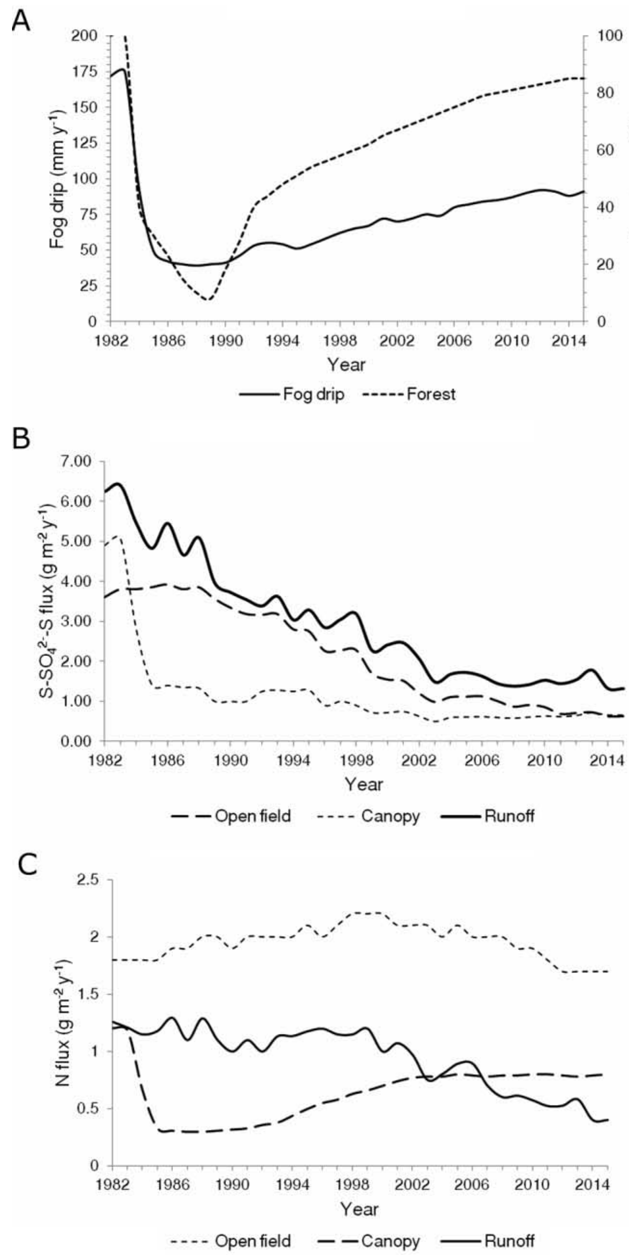

Figure 6. Fog drip (A), the balance of S (B), and the balance of N (C) in the Jizerka catchment (1982–2015).

Table 5. The correlation matrix between mean annual stream-water pH, SO_4^{2-}, and NO_3^-, air pollution (AP; SO_2 and NO_x combined), and canopy density (CD) in 1982–2015 ($r_{crit} = 0.28$, $n = 33$, $p = 0.05$).

Variable	CD	AP	SO_4^{2-}	NO_3^-	pH
CD	1	0.55	0.62	0.38	−0.49
AP	0.55	1	0.84	0.23	−0.61
SO_4^{2-}	0.62	0.84	1	0.53	−0.94
NO_3^-	0.38	0.23	0.53	1	−0.63
pH	−0.49	−0.61	−0.94	−0.63	1

were not well correlated with annual precipitation or runoff ($r = 0.04$–0.06) because of high annual precipitation (mean ± SD, 1398 ± 143 mm). Thus, in the 1990s, the recovery of stream waters from acidification resulted from a synergy of the drop in SO_2 emissions and reduction of the surface area of spruce forests. Later, open-field deposition rates for both elements stabilized, and their atmospheric loads and stream-water chemistry were controlled by regrowth of forest stands (Fig. 7, Table 3).

Fast (direct) flow, estimated from observed hydrographs by local minimum separation (Sloto and Crouse 1996) ranged between 54 and 61% of the annual runoff. Differences in the hydrograph formation are determined by annual snow water volume and frequency of rainstorms and are not affected by changes in forest canopy (Křeček and Hořická 2001). Relationships between stream-water variables (pH, electrical conductivity, concentrations of SO_4^{2-}, NO_3^-) and instantaneous discharge are shown in Table 6. pH was the most effective single variable distinguishing the occurrence of fast direct runoff at the Jizerka catchment outlet.

The recovery of stream-water chemistry (Fig. 8) followed the drop in the atmospheric acid deposition by ~5 y, but a revival of stream biota reflects these changes with a lag period of 10–15 y (Table 7). In 1994, the number of taxa of benthic organisms (36) corresponded to a strongly acidified environment (pH < 4.2; Veselý and Majer 1996), whereas by 2005, the number of taxa had increased to 68, which is more typical of a moderately acidified environment (pH = 5.0–6.3; Horecký et al. 2013). By 2005, several acid-sensitive taxa either had reappeared (Crustacea, Ephemeroptera) or increased significantly in the number of species present (Plecoptera, Trichoptera). The stream investigated at Jizerka was devoid of fish in the 1980s and remained without fish in 1990–2015.

Seasonal and episodic acidification

Annual distributions of mean monthly pH in 1982 (before the forest harvest), 1992, 2002, and 2012 (progressed forest regrowth) are shown in Fig. 8. Monthly pH increased, but seasonal pH minima continued to drop <5.3, which is considered a threshold for the rapid mobilization of toxic Al (Křeček and Hořická 2001). Streamflow Al content decreased from 1 to 2 (1980s) to 0.2 to 0.5 (1990s) and 0.1 to 0.2 mg/L (2010s). Seasonal acidification was driven mainly by direct (fast) runoff from spring snowmelt (Fig. 9A). In summer, stream-water pH decreased during rainstorms (Fig. 9B).

DISCUSSION

In the Jizerka catchment, the open-field deposition of SO_4^{2-}-S peaked in the late 1980s and showed a decreasing trend with the drop in atmospheric emissions of SO_2 during the 1990s (Fig. 5) in response to the 1985 Helsinki Sulphur Protocol (Holen et al. 2013). However, the open-field load of NO_3^--N and NH_4^+-N did not change significantly. Between 1982 and 2015, the N : S deposition ratio increased from 0.37 to 2.83. NH_4^+ and NO_3^- presented 72 and 28% of the long-term load of inorganic N, respectively. Ground observations confirmed linear hypsometric relationships between precipitation, the number of foggy days, and fog drip with atmospheric deposition. Rapidly decreasing trends in catchment runoff of both S and N (Fig. 6B, C) correspond with clear-cutting of spruce stands (1984–1988) and reduction of canopy area. Reforestation (mainly with spruce stands) of the Jizerka basin started only a year after the harvest, but regrowth of forests was relatively slow. Field surveys in 1992, 2002, and 2010 showed that the area dominated by grass (crown closure <0.3) was 62, 37, and 19%, respectively. The invasive grass community (*Calamagrostis* sp.) spread across the catchment with the defoliation of mature spruce stands (Křeček et al. 2010).

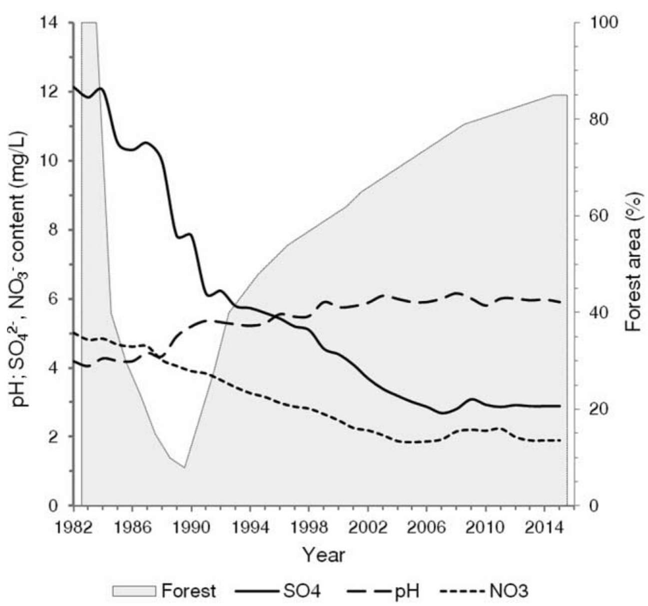

Figure 7. Changes of mean annual pH, SO_4^{2-}, and NO_3^- in stream water as the forest regrew in the Jizerka catchment (1982–2015).

Table 6. The correlation matrix between stream-water discharge (Q), pH, electrical conductivity (EC), and concentrations of SO_4^{2-} and NO_3^- in 2010–2012 ($r_{crit} = 0.20$, $n = 102$, $p = 0.05$).

Variable	Q	pH	EC	SO_4^{2-}	NO_3^-
Q	1	−0.67	−0.46	0.35	−0.29
pH	−0.67	1	0.51	−0.40	−0.33
EC	−0.46	0.51	1	0.31	0.24
SO_4^{2-}	0.35	−0.40	0.31	1	0.30
NO_3^-	−0.29	−0.33	0.24	0.30	1

The Landsat imagery analysis was an effective tool for identifying changes in the canopy and atmospheric depositions in the Jizerka catchment. However, NDVI values were relatively insensitive when canopy LAI was >2, as found previously by Křeček and Krčmář (2015). In contrast, supervised classification of multiband raster images (Landsat 4, 5) was more efficient in detecting differences in crown closure of trees (Fig. 4).

Prošková and Hůnová (2006) regarded an elevation of 800 m as the threshold for significant fog/cloud occurrence in the Czech Republic. In the upper plain of the Jizera Mountains, Křeček et al. (2017) reported significantly modified interception processes by fog deposition at elevations >700 m. In our study stand (elevation = 975 m), interception storage not affected by fog (34% of the gross precipitation) corresponded with the interception loss (30–40%) found in similar spruce forests by Krečmer et al. (1979). The observed interception loss (112 mm; 16% of gross precipitation) was affected by the additional deposition of fog water. The volume of stem-flow was negligible, consistent with findings by Krečmer et al. (1979) and Lovett and Reiners (1986). Compared to an open-field load, the reconstructed estimates of fog drip and additional canopy deposition (Fig. 6A–C) provided more realistic information on the atmospheric load within a mountain catchment and a better view of where this deposition exceeded critical levels of S and inorganic N. In the Jizerka catchment, the critical load of S (75 meq m^{-2} y^{-1}, according to the regional mapping; Schwarz et al. 2009) was exceeded from 1982 (beginning of the study) until 2002 (75–247 meq m^{-2} y^{-1}) in the open-field (herbaceous vegetation), and continues to be exceeded in forest stands (79–553 meq m^{-2} y^{-1}). The critical load of N (55 meq m^{-2} y^{-1}) also continues to be exceeded in both (99–149 meq m^{-2} y^{-1}) and spruce stands (142–206 meq m^{-2} y^{-1}). Bobbink and Roelofs (1995) consider 1 g m^{-2} y^{-1} (71 meq m^{-2} y^{-1}) as an empirical critical deposition of N in forests of central Europe. This threshold has been exceeded by a factor of 2 to 3 throughout the period from 1982 to 2015 in the Jizerka catchment. This greater deposition of nutrient N is particularly important considering the evidence of increased environmental sensitivity and changes in biodiversity (Matzner and Murach 1995).

Prechtel et al. (2001) reported a significant decline of SO_4^{2-} concentrations in European headwater streams in the 1990s (relative to in the 1980s), but less than the decline in input fluxes. The response in runoff increases as soil storage capacity decreases. In the Jizerka catchment, the fast response of SO_4^{2-} runoff relative to the drop in the deposition during the 1990s, reflects the clear-cut of spruce forests (1984–1988) and prevailing fast subsurface runoff generated by relatively shallow podzolic soils of a low SO_4^{2-} storage capacity. The open-field load of N did not change significantly in 1982–2015, but NO_3^- concentrations in stream water decreased by 12% in the 1990s and by 53% after 2010 (relative to in the 1980s). The relatively high leaching of NO_3^- before the forest harvest (1982–1984) corresponds with high atmospheric loads of N and limited N uptake by already damaged spruce stands (defoliation of 30%). Grenon at al. (2004) reported higher NO_3^- leaching from forest floor with forest decline caused by the decreased uptake of N by vegetation and increased microbial release of N. Decreased N uptake by spruce trees contributes to increasing availability of mineral N in the summer, whereas enhanced microbial N release takes place over the whole year. Tahovská et al. (2010) reported increased in situ availability of NO_3^- before the defoliation peaked, and Huber (2005) found a positive correlation between herbaceous ground vegetation and NO_3^- concentration in soil water during the first 2 to 5 y after forest die-back.

Low pH (Figs 7, 8), low hardness (≤10 mg/L Ca^{2+} and Mg^{2+}), and high Al contents (>0.2 mg/L) were observed in surface waters of the Jizera Mountains in the 1980s (Křeček and Hořická 2006). With the recovery of the water environment in the 1990s, pH values increased from 3.3–

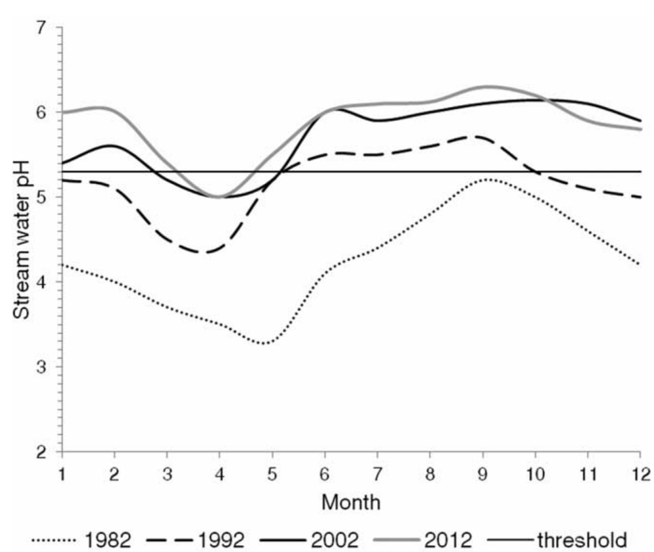

Figure 8. Mean monthly pH of stream water at the outflow of the Jizerka catchment.

Table 7. The number of identified taxa of benthic organisms at the Jizerka outlet.

Taxa	1994	2005
Nematoda	–	1
Oligochaeta	2	4
Hydracarina	–	1
Crustacea	–	1
Ephemeroptera	–	3
Plecoptera	12	20
Megaloptera	1	–
Trichoptera	4	17
Diptera excl. Chironomidae	11	10
Chironomidae	4	5
Coleoptera	2	6
Total	36	68

5.2 to 4.4–5.7, Al content dropped to 0.1–0.2 mg/L and fish (Brook Char and Brown Trout extinct since the 1980s) were reintroduced in selected streams (Křeček and Hořická 2001). No health-based guideline value has been proposed for pH of water, but pH is one of the most important operational water-quality variables (WHO 2004). Without pollution or acidic rain, most lakes and streams would have a pH level near 6.5 (Merilehto et al. 1988). Decreased pH is particularly associated with increased mobility of Al and heavy metals in the podzolic soil layer and has a negative impact on the drinking-water supply and survival of aquatic biota (Křeček and Hořická 2006, Horecký et al. 2013). Mean annual pH at the Jizerka outlet increased from 4.0 (1982–1985) to 5.3 (1990–1994), but repetitive episodic acidification still affects the recovery of the biota, particularly acid-sensitive species. Seasonal pH minima during snowmelt (Fig. 8) are <5.3, a threshold associated with a rapid mobilization of toxic Al (Bache 1985, Veselý and Majer 1996). Water pH seems to be an effective variable of the hydrograph separation (Fig. 9A, B, Table 6) and is more powerful than conductivity recommended by Caissie et al. (1996).

Guerold et al. (2000) considered aquatic invertebrate communities as the best indicator for assessing the negative environmental effect of acidification. Skjelkvåle et al. (2003) found large-scale evidence of chemical recovery from surface-water acidification in Europe, but little evidence of biological recovery. Recovery of stream-water chemistry at the Jizerka outlet (Fig. 7) occurred ~5 y after drop in the acid atmospheric deposition, but recovery of stream biota appeared after a lag of 10–15 y (Table 7).

Acidification of sensitive ecosystems has been a serious environmental problem in Europe in recent decades. Schöpp et al. (2003) estimated the amounts of SO_2, NO_x, and NH_3 emissions in Europe from 1880 to 2030, and Kopáček et al. (2016) modeled the chemistry of precipitation affected by industrial dust in Central Europe since the 1850s. Spruce forests contributed to acidification of headwater catchments in Central Europe by increasing acidic atmospheric deposition. Data from the Jizerka catchment (1982–2015) document that the acidic atmospheric deposition and stream-water quality can be modified at the catchment scale by forestry practices. In comparison with mature spruce stands, the herbaceous canopy that developed on harvested areas decreased the atmospheric acid load by ~40% and mitigated acidification of surface waters. Deciduous or mixed forests could decrease the acidic atmospheric load because of their lower leaf area and surface roughness (Lovett and Reiners 1986), particularly in the dormant season. In the Jizera Mountains, mixed forests of near-native composition can increase the soil buffering capacity (Matzner and Murach 1995) and support ecological stability (Rapport et al. 1998) by a deeper root system

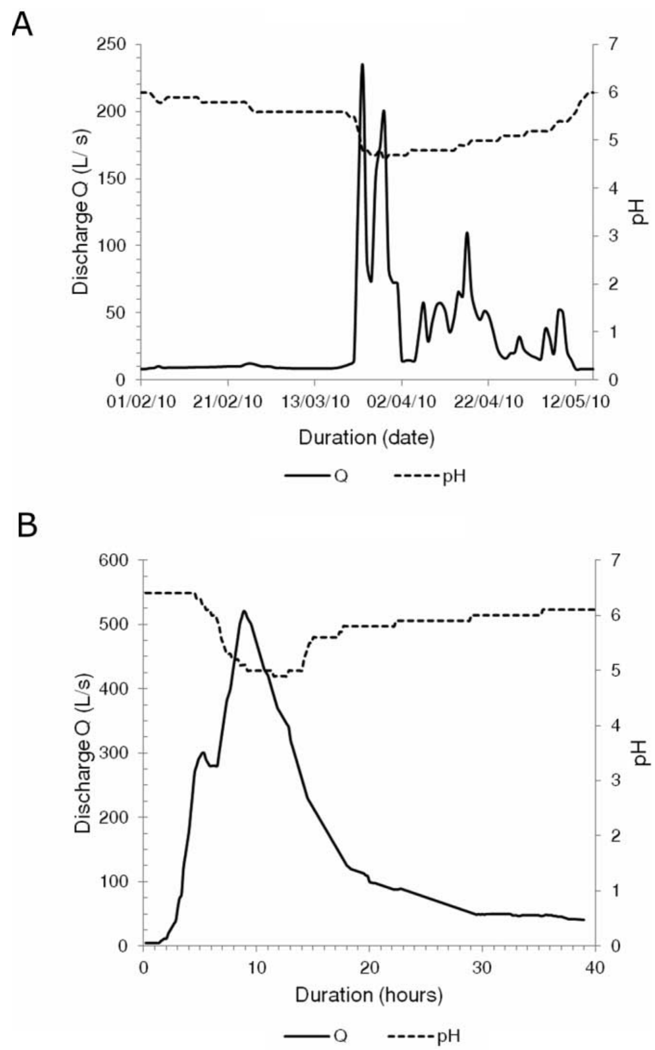

Figure 9. pH and discharge (Q) of snowmelt runoff in the Jizerka basin from March–May 2010 (A) and of summer runoff episode of a frequency > 1 (B) in the Jizerka basin from June 8–9 2010. Dates are formatted dd/mm/yy.

and a higher resistance to air pollution. UNEP (2007) calls for a reduction of the acidic atmospheric deposition in headwater catchments to mitigate the progressive downstream acidification of rivers and the ocean. However, the regrowth of coniferous stands following a reduction in atmospheric emissions could slow the recovery of surface waters (Fig. 7). Uncertainties in predictions of the future of recovery from acidification still depend on rates of production of atmospheric emissions, global climate change, and the long-term behavior of N in forest ecosystems.

Citizen scientists

Citizen scientists of the Earthwatch Institute played an important role in gathering extensive field data. Hodgson and Evans (1997) warned of reduced accuracy and increased response time when measuring pH in waters with naturally low ionic strength. The in situ data (pH, conductivity) collected by volunteers showed a relatively good agreement with values obtained under the laboratory conditions (20 readings tested/expedition, $r = 0.79–0.93$ ($r_{\mathrm{crit}} = 0.16$, $n = 100$, $p = 0.05$) in the years of field expeditions. Rosenberg and Resh (1993) considered the primary role of nonspecialist volunteers to be sampling benthic macroinvertebrates, but their participation in water monitoring and the forest inventory were controlled by the professional project staff in our study. In addition, the volunteers enabled greater temporal resolution in the sampling campaigns and were a source of essential data. The motivation of citizen participants played an important role in their education as evaluated by the Earthwatch Institute (2012).

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Author contributions: JK contributed to the design of this study, analyzed the data, and wrote the initial version of the manuscript, LP and EP contributed to the data processing, and ES to the laboratory analyses and their interpretations.

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