

Increasing temperature decreases aluminum concentrations in Central European lakes recovering from acidification

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Abstract

As acidic water percolates through mineral soils, Al is mobilized and transported into streams and lakes. We evaluate the effect of increasing temperature (climate warming) on Al concentrations in four strongly acidified lakes in the Bohemian Forest, southwestern Czech Republic, over a 17-yr period (1984–2001). Pronounced decreases in atmospheric S and N deposition resulted mainly in a sharp monotonic decrease in lake water concentration of total Al (-0.49 to $-1.38 \mu\text{mol L}^{-1} \text{yr}^{-1}$). Residuals from the linear relationship between total Al concentrations and the sum of strong acid anions were inversely correlated with air temperature and the North Atlantic Oscillation index. An increase in the average annual air temperature of $+1.27 \pm 0.49^\circ\text{C}$ between 1984 and 2001 was correlated with decreases in toxic Al^{3+} and explained, on average, 13% of the total Al decrease in three lakes and 11% of the Al^{3+} decrease in Černé Lake. The inverse relationship between Al solubility and temperature caused lower Al mobilization in soil horizons and/or enhanced precipitation of Al in the lakes at higher temperature but otherwise similar conditions. As a consequence, the recent period of warmer years and mild winters significantly contributed to the trend of decreasing Al in lakes recovering from acidification.

Lake water chemistry in areas without direct human influence is controlled mainly by watershed characteristics and atmospheric deposition. Recent studies have shown several climate-related factors to affect concentrations of water constituents, including dissolved organic carbon (DOC; Schindler 1997), SO_4 (Webster and Brezonik 1995; Dillon et al. 1997), pH (Psenner and Schmidt 1992), NO_3 (Mitchell et al. 1996), and Al (Lydersen 1995). Climate also affects terrestrial processes such as the mineralization of soil organic matter, the input of marine aerosols (Evans et al. 2001b), and soil CO_2 and alkalinity production (Norton et al. 2001).

Many of the climate-driven effects on lakes can be linked to winter climate. In the northwestern part of Europe, winter climate is regulated by large-scale atmospheric circulation linked to the North Atlantic Oscillation (NAO) index (Rodwell et al. 1999). The NAO is the difference in surface pressure between the Azore Islands (or some nearby station) and Iceland (Hurrell 1995; Jones et al. 2001). The positive phase of the NAO corresponds to stronger winter westerlies tra-

versing the Atlantic further north toward Europe, and the negative phase brings more continental weather to Central Europe.

The winter NAO values and temperature are negatively correlated with NO_3 concentrations in upland lakes and streams of the United Kingdom (Monteith et al. 2000). Similarly, fluctuations in Cl in mid-Wales lakes have been linked to the NAO and exhibit a similar, decadal periodicity (Evans et al. 2001b). The NAO effects on lake water temperature and ecology are more pronounced in a deep dimictic lake with stable summer stratification than in a circulating polymictic lake because of a longer persistence of the winter signal in the hypolimnion until the following fall turnover (Gerten and Adrian 2001).

Lydersen et al. (1990) suggested a possible negative correlation between water temperature and Al concentrations in Norwegian lakes. Such a link is of great interest in areas recovering from atmospheric acidification because of the toxicity of Al to freshwater biota (e.g., special issue of *Environmental Pollution* 71 (2–4) 1991) and its ability to immobilize P and thereby influence trophic status (Kopáček et al. 2000a).

Total (acid-soluble) Al in water (Al_T) includes (1) dissolved inorganic Al (Al_i), (2) particulate Al ($>0.45 \mu\text{m}$) present as suspended matter (minor or short-lived in acidic waters and lakes), and (3) organically bound Al (Al_{org}). Concentrations of Al_T increase with increasing F^- and organoligands (DOC) and with decreasing pH. The mechanisms that regulate the release of Al are still unclear (Berg-

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gren and Mulder 1995; LaZerte and Findeis 1995). There is no precise relationship between pH and the terrestrial transport of Al to freshwaters (Cronan and Schofield 1990; Norton et al. 1990). Aluminum concentrations in the Central European lakes that are recovering from acidification decreased synchronously with concentrations of strong acid anions (SAAs) rather than with pH (Kopáček et al. 1998; Veselý et al. 1998a,b).

Here we evaluate Al concentrations in four Bohemian Forest lakes (southwestern Czech Republic) during 1984–2001 with respect to regional climatic change (Weber et al. 1997) and the decreasing trend in S and N emissions in this region. We evaluate whether and why the recent increase in temperature contributed to the decreasing Al_T and Al^{3+} concentrations.

Methods and site description

The Bohemian Forest lakes (Černé, Čertovo, Prášílské, and Plešné) are situated between $13^{\circ}11'$ and $13^{\circ}52'E$ and $48^{\circ}47'$ and $49^{\circ}11'N$ at elevations of 1,008–1,090 m a.s.l. in the southwestern Czech Republic. These small lakes (4.2–18.4 ha) have retention times of 0.5–2 yr. Acidification peaked during the mid-1980s and gradually decreased until 2001, mainly because of reductions in the atmospheric deposition of S and N (Veselý et al. 1998b; Kopáček et al. 2001).

Water samples were collected from the epilimnion of each lake at the end of July and in the second half of October, ~100 and ~190 d after the spring thaw during the 1984–2001 period. Four to six water samples were collected during each visit. Numerical average concentrations from each visit were used for the present article. This substantially enhanced the accuracy of data and permits the study of secondary effects on Al_T variability in lake water. During stratification in July, samples were also collected from 4–6 depths between the surface and bottom at the deepest points in the lakes, and volume-weighted mean concentrations were obtained by linking the concentrations to the volumes of individual water layers. Sulfate, Cl^- , and NO_3^- were analyzed by ion chromatography, Al by inductively coupled plasma-atomic emission spectrometry, base cations (BCs , Ca^{2+} , Mg^{2+} , Na^+ , and K^+) by flame atomic-absorption spectrometry, and F^- and pH by ion-selective electrodes. The pH was measured on the day of sample collection at sample temperatures between $15^{\circ}C$ and $20^{\circ}C$. Analytical methods were identical through the study. The details of analytical methods and site description are in Veselý et al. (1998b). Concentrations of DOC were determined by high-temperature catalytic combustion with infrared detection of CO_2 . Positively charged Al (Al_i) and uncharged or negatively charged Al (Al_o) were separated using a cation exchange resin (Driscoll 1984). We analyzed Al_o and Al_T and calculated Al_i by difference. DOC, Al_i , and Al_o were not determined regularly. Inorganic species of Al were calculated in Černé Lake on the basis of Al_i by MINEQL+ (Schecher and McAvoy 1998).

Data on average monthly temperature and precipitation at Churáňov station (at 1,118 m a.s.l. midway in the Bohemian Forest) are from the Czech Hydrometeorological Institute.

The NAO index was obtained from Climate Research Unit (www.cru.uea.ac.uk/cru/data/nao.htm). Ambient concentrations of SO_2 are from the Brotjackriegel station (Umweltbundesamt, Berlin), which is upwind of the Bohemian Forest lakes at a similar elevation.

Concentrations of Al_T , BCs , and H^+ were linearly regressed against the sum of SAAs, where $SAA = (SO_4^{2-} + NO_3^- + Cl^-)$. Residuals of Al_T (ΔAl_T) (and ΔAl^{3+} for Černé Lake), H^+ (ΔH^+), and BCs (ΔBCs) were calculated as the difference between the measured values and the regression line of each parameter against SAAs. Then, linear regression analysis was used for the investigation of relationships between the residuals (ΔX values) and air temperature. Two types of temperature values were used in the analysis: (1) average air temperature for the 12-month period prior to sampling (T_{12}) and (2) average air temperature of the previous winter (December, January, and February; T_{DJF}).

Using factor analysis with software S-plus (MathSoft 1997), we searched for critical factors controlling variability in the residuals (ΔX values) of the linear decrease of Al_T , Al^{3+} , H^+ , and BCs with SAAs. By analyzing the matrix of correlation coefficients for all pairs of variables, a matrix of factor loading was obtained. The total variance is expressed by some low number of hypothetical variables (factors) that may be more suitable for interpretation than the original data. A factor can represent an unmeasured variable or a process that is difficult to identify or characterize.

Finally, we investigated the statistical significance of insertion of climatic variables (T_{12} or T_{DJF} , NAO_{DJF} , and precipitation amount for 12 months before sampling, or P_{12}) into linear regressions with SAA by stepwise linear multiple regression (S-plus 4; MathSoft 1997). The model assessed the response of the variable Al_T against a combination of SAAs, T_{12} (or T_{DJF}), NAO_{DJF} , and P_{12} . Any forcing variable was automatically deleted if the model was not improved by its inclusion in the multiple regression.

Results

Climate variability—The average air temperature (T_{12}) in the year preceding water sampling ranged 3.28 – $5.72^{\circ}C$ in the Churáňov site from 1984 to 2001 (Fig. 1) and increased irregularly according to the equation

$$T_{12}(^{\circ}C) = -144.0(\pm 59.6) + 0.075(\pm 0.029)(\text{year AD})$$

$$(P = 0.024) \quad (1)$$

Equation 1 yields an increase of $1.27 \pm 0.49^{\circ}C$ during the 17 yr of observation. The years 1990, 1998, and 2001 were warmer; 1984–1987 and 1996 were cooler. The warming trend for the station is consistent with that in Central Europe over the past 2 decades (Weber et al. 1997).

The values of winter T_{DJF} ranged between -5.93 and $+0.17^{\circ}C$ from 1984 to 2001 (Fig. 2). Four cold winters at the beginning of study were followed by mild winters from 1988 to 1990. High variability in winter temperature makes the temporal increase in T_{DJF} ($\sim 0.11^{\circ}C \text{ yr}^{-1}$) between 1984 and 2001 statistically insignificant ($P = 0.16$). However, T_{DJF} and T_{12} were closely related:

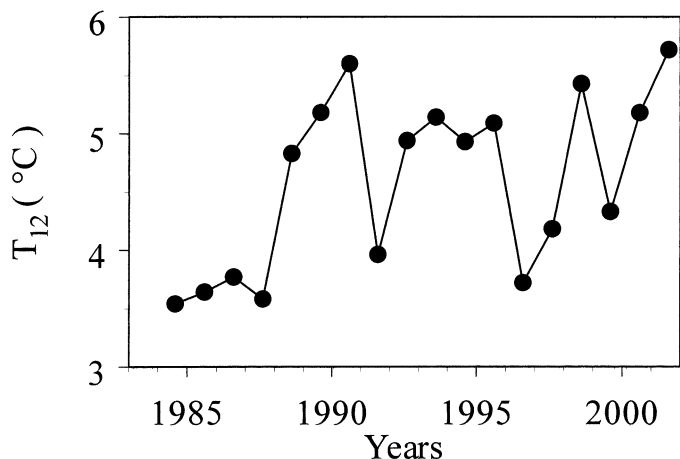


Fig. 1. Mean annual temperature for the 12 months preceding summer sampling (T_{12}) at Churáňov station in the Bohemian Forest for 1984–2001.

$$T_{\text{DJF}}(^{\circ}\text{C}) = -12.4(\pm 1.1) + 2.05(\pm 0.24)T_{12}(^{\circ}\text{C})$$

$(P < 0.001)$ (2)

T_{DJF} and winter NAO (NAO_{DJF}) were also related:

$$T_{\text{DJF}}(^{\circ}\text{C}) = -3.55(\pm 1.45) + 0.68(\pm 0.25)\text{NAO}_{\text{DJF}}$$

$(P = 0.014)$ (3)

and the winter temperature in the region was related to large-scale atmospheric circulation characterized by the NAO (Fig. 3). The 1995/1996 winter was particularly cold ($T_{\text{DJF}} -5.2^{\circ}\text{C}$), with the most negative NAO index value (-2.1) in the past decade. During that winter, the concentration of SO_2 was exceptionally high ($10.4 \mu\text{g m}^{-3}$), whereas it was $<4 \mu\text{g m}^{-3}$ during the four preceding winters (Fig. 2). The SO_2 concentration in winter air not only sharply decreased with time, in parallel to the decline in S emissions, but also was inversely related to T_{DJF} over 1984–2000:

$$\text{SO}_{2(\text{DJF})} = -1.7(\pm 4.7) - 3.17(\pm 0.68)T_{\text{DJF}}(^{\circ}\text{C})$$

$(P < 0.001)$ (4)

Precipitation during the 12-month period preceding the sample (P_{12}) ranged from 84.6 cm (1991/1992) to 135.2 cm (1994/1995), with no significant trend. Winter precipitation correlated positively with the NAO during the past 40 yr ($P = 0.002$), as it has in northwestern Europe (Hurrell 1995; Rodwell et al. 1999). However, the trend during our study was not statistically significant ($P = 0.11$). Continuous snow cover ranged from 95 d during the 1989/1990 winter to 172 d in 1995/1996, with no trend during the study period.

Lake water chemistry—Except for Si and DOC, concentrations of all major components in all four acidified lakes decreased monotonically from 1984 to 2001. The decrease in Al_T concentrations ranged $8.3\text{--}23.4 \mu\text{mol L}^{-1}$ ($224\text{--}632 \mu\text{g L}^{-1}$) over 17 yr (Fig. 4) and was among the largest reported in Europe (Veselý et al. 1998a; Evans et al. 2001a). Because $\text{DOC} \leq 400 \mu\text{mol C L}^{-1}$ for the lakes, Al_i was much higher than Al_o . Concentrations of Al_o were relatively

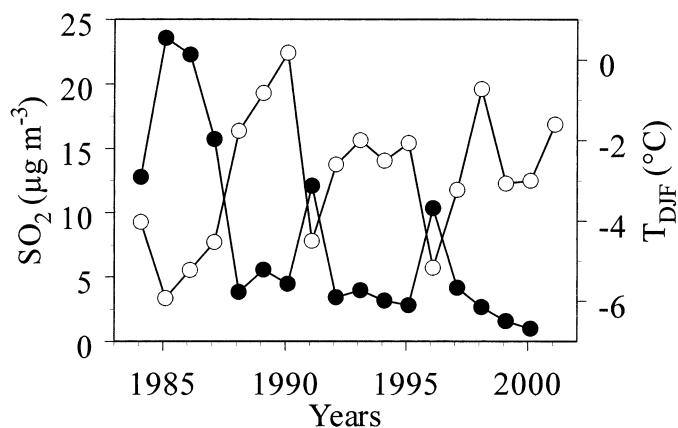


Fig. 2. Relationship between average winter (December–February) concentrations of SO_2 (filled circles) at Brotjackriegel station (data of Umweltbundesamt Berlin) and average winter temperature, T_{DJF} (open circles), in the Bohemian Forest for 1983/1984–2000/2001.

low (on average, 2.0, 2.4, 6.4, and $4.8 \mu\text{mol L}^{-1}$ in Černé, Čertovo, Plešné, and Prášílské lakes, respectively) and were either stable or increased in agreement with the increase of DOC ($\sim 13 \mu\text{mol C yr}^{-1}$ during the 1992–2001 period; authors' unpubl. data).

Residuals of Al_T (ΔAl_T) from the linear regression of Al_T with SAAs (Fig. 5) were related inversely to T_{12} (Fig. 6). The relationships between ΔAl_T and antecedent temperature (-1.1 to $-2.7 \mu\text{mol per }^{\circ}\text{C}$) were similar for the four lakes, highly statistically significant, and substantially greater than the mean standard error of the Al_T determination ($0.41\text{--}0.46 \mu\text{mol}$). ΔAl_T was inversely correlated also with T_{DJF} (Fig. 7). The residuals from the linear regression of H^+ against concentrations of SAAs (ΔH^+) also related to (T_{DJF}) for three lakes (Table 1). However, the effect between T_{12} and ΔH^+ was weaker and was significant only for Černé Lake. There was no temperature effect on residuals from BCs regressed against SAAs.

The temperature effect on Al^{3+} concentrations was eval-

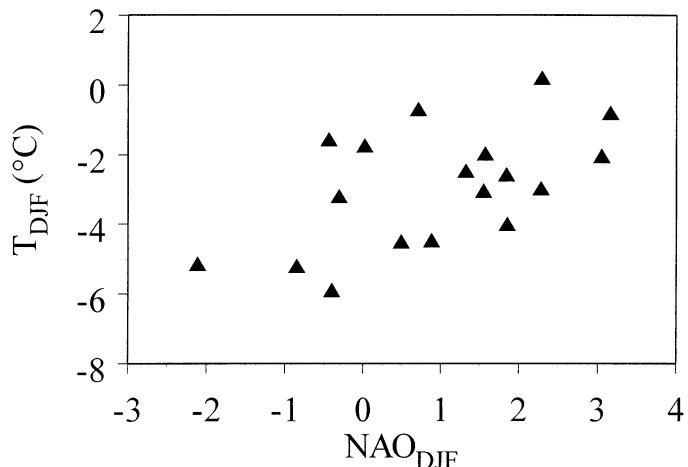


Fig. 3. Relationship between winter air temperature (T_{DJF}) at Churáňov station and winter NAO index for 1983–2001.

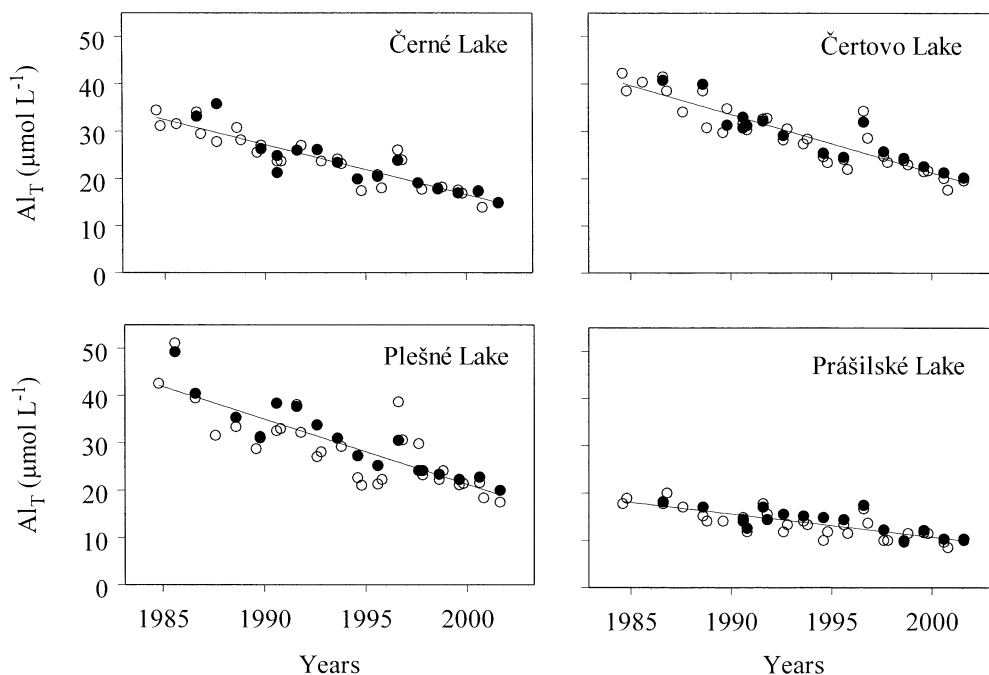


Fig. 4. Changes in Al_T concentrations with time in acidified Bohemian Forest Lakes for 1984–2001. Open circles: epilimnetic concentrations, filled circles: volume-weighted concentrations, and solid lines: linear regression.

uated for Černé Lake, which had the lowest DOC concentrations ($70\text{--}140\ \mu\text{mol C L}^{-1}$) and the lowest Al_o of the four lakes. Under the assumption of $13\ \mu\text{mol}$ of Al_o per mmol of DOC in Černé Lake tributaries (Kopáček et al. 2000b), we calculated concentrations of Al^{3+} using MINEQL+. We used

the relationship $Al_T = Al_i + 1.7 (Al_o)$ (μmol) if DOC had not been determined. The calculated Al^{3+} in Černé Lake decreased by $0.77 (\pm 0.05)\ \mu\text{mol L}^{-1}\ \text{yr}^{-1}$ ($P < 0.001$). If the solubility constant of solid $Al(OH)_3$ in Černé Lake was $\leq 10^{8.50}$ (25°C), concentrations of Al^{3+} were sufficiently high

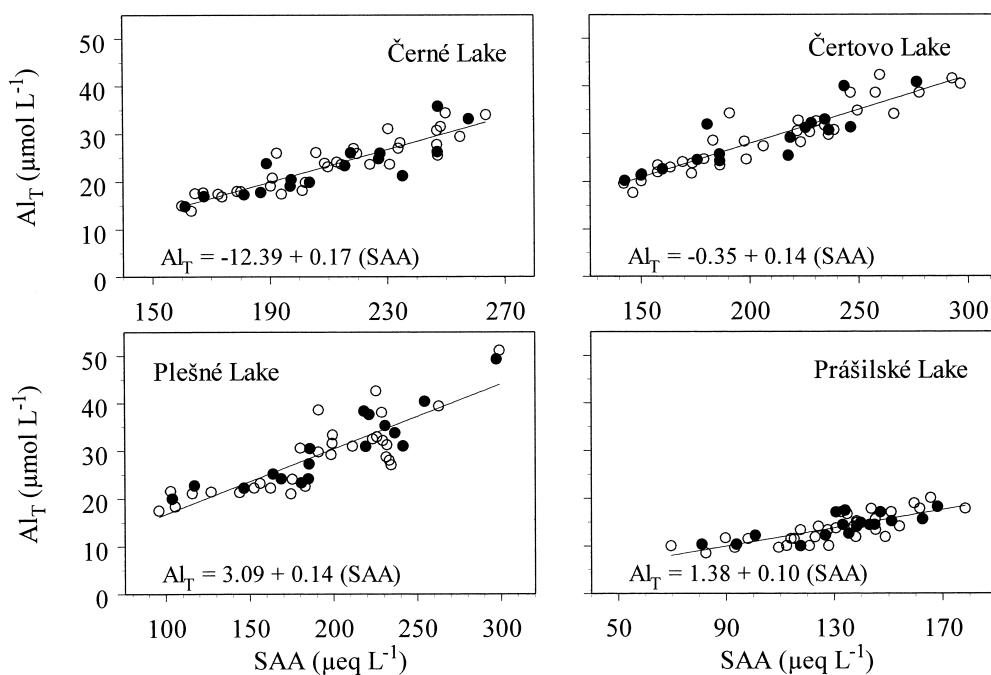


Fig. 5. Changes in Al_T concentrations with SAAs in acidified Bohemian Forest Lakes for 1984–2001. Open circles: epilimnetic concentrations, and filled circles: volume-weighted concentrations. For all regression equations, $P < 0.001$.

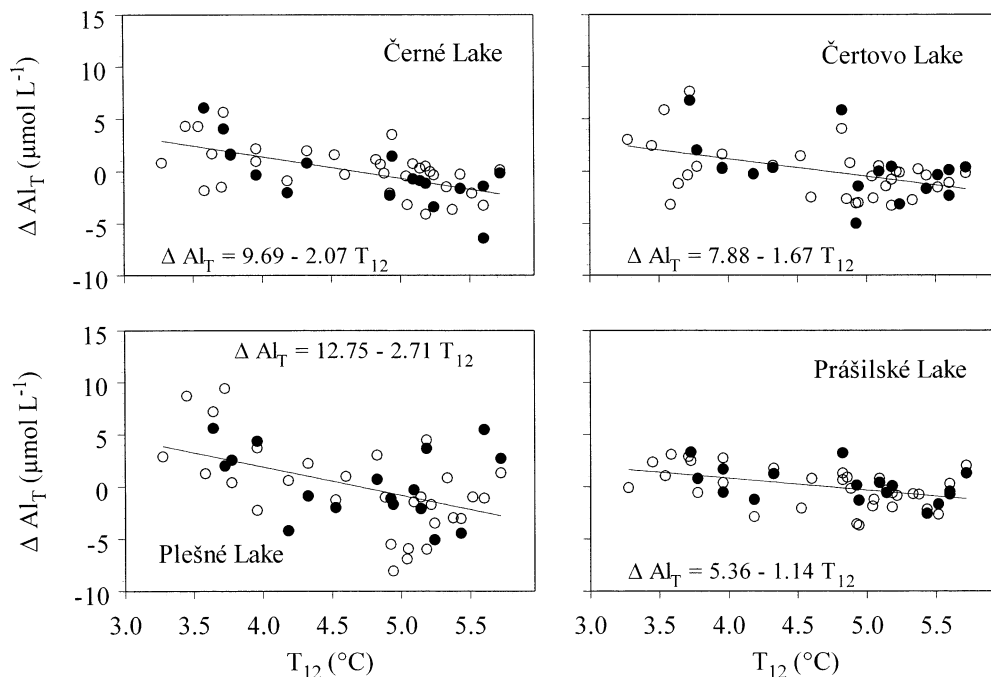


Fig. 6. Relationship between Al residuals (ΔAl_T) and average air temperature during 12 months preceding sampling (T_{12}). Open circles: epilimnetic concentrations, filled circles: volume-weighted mean concentrations, and solid line: linear regression. For all regression equations, $P < 0.01$.

for precipitation to occur in the water column. Residuals of Al^{3+} (ΔAl^{3+}) from linear regression of Al^{3+} against SAAs in Černé Lake also were related to T_{12} and T_{DJF} (Fig. 8):

$$\Delta\text{Al}^{3+} = -6.80(\pm 1.45) - 1.45(\pm 0.31)T_{12} \quad (P < 0.001) \quad (5)$$

$$\Delta\text{Al}^{3+} = 1.78(\pm 0.43) - 0.65(\pm 0.13)T_{\text{DJF}} \quad (P < 0.001) \quad (6)$$

These relationships did not change for the subset of data that excluded years following severe winters with anomalously high SO_2 concentrations (i.e., 1985–1987, 1991, and 1996; Fig. 2). The significance of ΔAl^{3+} versus T_{12} and T_{DJF} remained below $P < 0.001$.

Factor analyses—The variability of ΔAl_T , ΔAl^{3+} in Černé Lake, ΔH^+ , ΔBCs , P_{12} , T_{12} , T_{DJF} , and NAO_{DJF} in the lakes for 1984–2001 were analyzed by factor analyses. We identified four factors that together explain between 60% and 68% of the variance in the data (Table 2).

The variability in ΔAl_T and ΔAl^{3+} is explained by two or three factors. Higher temperatures (both T_{12} and T_{DJF}) and higher winter NAO levels were inversely related to ΔAl^{3+} and ΔAl_T in Černé, Plešné, and Prášílské lakes (factor 1). The NAO effect was more important than temperature for variability of ΔAl_T in Čertovo (factor CT2) and Prášílské (factor PR3) (Table 2). At high winter NAO, Al_T tends to be below the linear regression line between Al_T and SAAs. The factors CN3, CT3, PL2, and PR2 (Table 2) explain the most variability in ΔH^+ and are positively related to precipitation amount for Čertovo (CT3) and Prášílské (PR2) lakes. This factor may include the effect of DOC. Increased pre-

cipitation may increase DOC concentrations, thereby increasing Al_T and H^+ and causing positive residuals for Al_T and H^+ versus SAAs.

Stepwise linear regression—We searched for effects of climate variables (P_{12} , T_{12} , T_{DJF} , and NAO_{DJF}) on Al_T using stepwise linear regression. The multilinear regression

$$\text{Al}_T = a + b(\text{SAA}) + c(T_{12}) + \dots \quad (7)$$

explained more variance than the simple linear regression

$$\text{Al}_T = a + b(\text{SAA}) \quad (8)$$

for all lakes (Table 3). P_{12} was a significant contributor only for Prášílské Lake (with the shortest residence time), and NAO_{DJF} was a significant contributor only for Čertovo Lake, a deep dimictic lake with persistent summer stratification and retention of the winter NAO signal (Gerten and Adrian 2001). The relative decrease in the value of b in the multiple regression (Eq. 7) compared with the simple regression (Eq. 8) can be used to isolate the effect of T_{12} on the Al_T decrease. On the basis of this method, T_{12} is responsible for 12.4% (11.2% for Al^{3+}), 8.5%, 12.4%, and 17.9% of the total Al decrease in Černé, Čertovo, Plešné, and Prášílské lakes, respectively. The substitution of T_{DJF} for T_{12} in the regressions produces smaller decreases in b , which suggests that the whole-year temperature (T_{12}) is more important than the previous winter temperature (T_{DJF}).

Discussion

The chemical recovery of surface water from acidification has been strongly correlated with decreased atmospheric

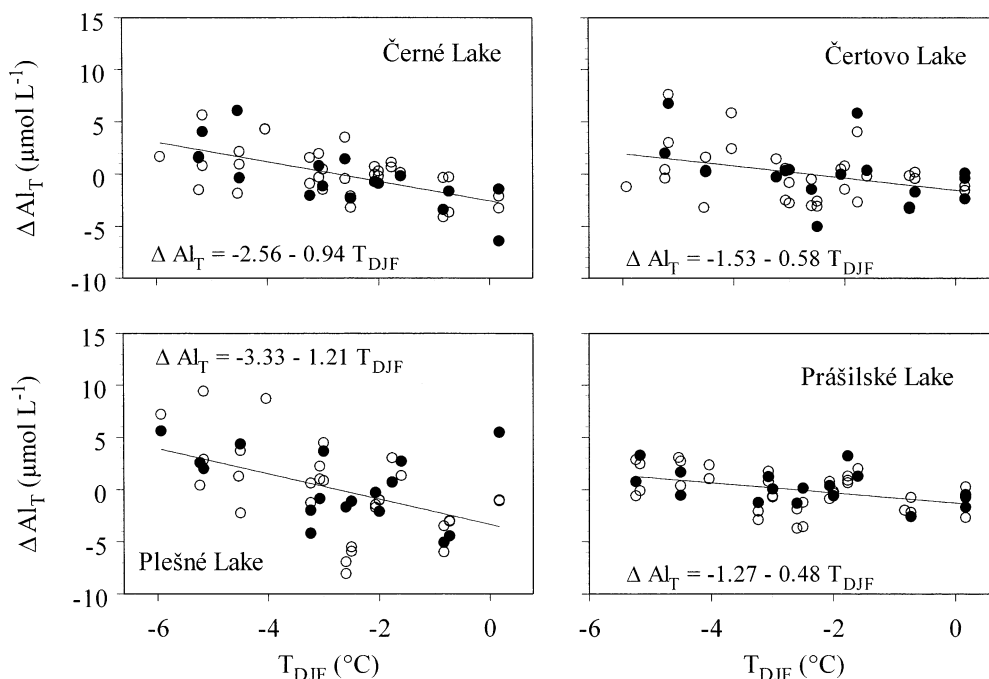


Fig. 7. Relationship between Al residuals (ΔAl_T) and air temperature during the preceding winter (T_{DJF}). Open circles: epilimnetic concentrations, filled circles: volume-weighted mean concentrations, and solid line: linear regression. For all regression equations, $P < 0.01$.

emission of S ($\sim 85\%$) and N (by $\sim 30\%$) in Central Europe (Kopáček et al. 2001; Veselý et al. 2002). The decrease in Al_i concentrations has compensated for $\sim 65\%$ of the decrease in SAAs in the acidified Bohemian Forest lakes over the first 11 years of study (Veselý et al. 1998a). However, climate change has contributed significantly to the variability in Al_T in these lakes. The temperature increase of $1.27 \pm 0.49^\circ\text{C}$ over 17 yr had a statistically significant effect on the decrease in Al_T concentrations, accounting for 9%–18% (11% for Al^{3+} in Černé Lake) of the Al_T decline from 1984 to 2001. The inverse relationship between equilibrium concentrations of Al at saturation and temperature is the likely cause of this trend.

Equilibrium concentrations of Al^{3+} with secondary hydroxylated Al-bearing minerals should decrease with increasing temperature (Lydersen 1990; Tipping et al. 1995). The solubility equilibrium constant (K) is related to temperature by the Van't Hoff relation:

$$d \ln K/dT = \Delta H_r/RT^2 \quad (9)$$

where $R = 8.314 \text{ kJ}^{-1} \text{ mol}^{-1}$, T is the temperature in K, and ΔH_r is the enthalpy change of the reaction. Equilibrium concentrations will decrease with temperature if ΔH_r is negative. The enthalpy change ΔH_r for the dissolution of gibbsite is $-95.5 \text{ kJ mol}^{-1}$ (Robie et al. 1978), -117 kJ mol^{-1} for amorphous $\text{Al}(\text{OH})_3$ (Lydersen 1990), and -148 kJ mol^{-1} for kaolinite (Robie et al. 1978). For a temperature increase from 0 to 10°C , solubility decreases by $\sim 70\%$ (Fig. 9). The average air temperature increase of $1.27 \pm 0.49^\circ\text{C}$ for the lake region from 1984 to 2001 would decrease the Al^{3+} equilibrium concentrations by 17%, 20%, and 26% for kaolinite, gibbsite, and amorphous $\text{Al}(\text{OH})_3$, respectively. The theoretical decrease in solubility of kaolinite was the nearest to the average calculated value from field data (13%). The Al_T includes soluble complexes—for example, $\text{Al}(\text{OH})_2^+$, AlOH^{2+} , AlF^{2+} , $\text{Al}(\text{SO}_4)^+$, organically bound Al_o , and particulate Al—which makes interlake comparison imprecise. The slightly increasing DOC concentrations in the four lakes (Kopáček et al. 2003) likely causes a small increase in concentrations

Table 1. Slope and statistical significance of relationships between residuals of Al_T , H^+ , base cations (BCs), and temperature ($\mu\text{mol L}^{-1}$ per $^\circ\text{C}$).

Lake	Winter air temperature (T_{DJF})				Annual air temperature (T_{12})			
	ΔAl_T	ΔAl^{3+}	ΔH^+	ΔBCs	ΔAl_T	ΔAl^{3+}	ΔH^+	ΔBCs
Černé	-0.94***	-0.65***	-0.67**	—	-2.07***	-1.45***	-1.34**	—
Čertovo	-0.58***	—	-0.88*	—	-1.67***	—	—	—
Plešné	-1.21***	—	-0.65*	—	-2.71***	—	—	—
Prášílské	-0.48**	—	—	—	-1.14**	—	—	—

Residuals were calculated as the deviation from linear decrease of variable with the sum of strong acid anions over the whole study period. * $P < 0.05$; ** $P < 0.01$; *** $P < 0.001$. The range of T_{DJF} is larger, so lower slopes do not mean lower sensitivity.

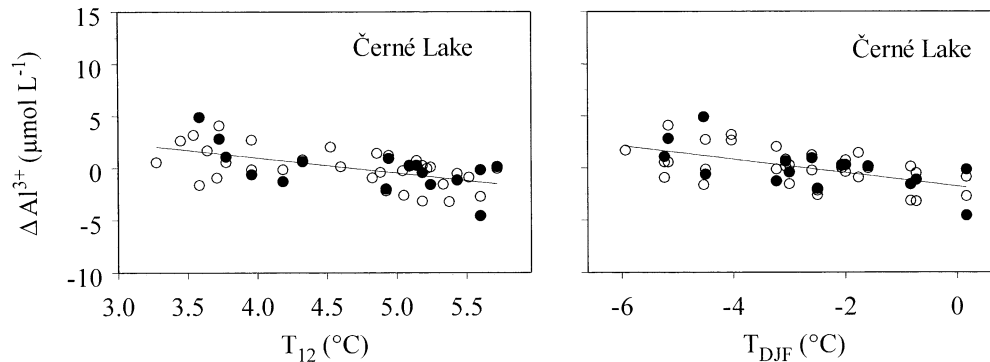


Fig. 8. Relationship between Al^{3+} residuals (ΔAl^{3+}) and average air temperature during 12 months preceding sampling (T_{12}) and preceding winter (T_{DJF}) in Černé Lake. Open circles: epilimnetic concentrations, filled circles: volume-weighted mean concentrations, and solid lines: linear regression.

of Al_o , partly offsetting any decrease in Al_T concentrations. However, the effect of temperature on the Al^{3+} decrease (11%) was also below the theoretical decrease for kaolinite, gibbsite, and amorphous aluminum hydroxide. It is possible that the changes in soil and lake water temperature may be less than and lag air temperature, thus producing a smaller effect on Al solubility. Regardless, temperature effects on Al solubility partly control the variability, trend, and concentrations of Al in these acidic lakes.

The winter climate is controlled by the NAO index (Rodwell et al. 1999). The NAO index was identified as important for Al variability by factor analysis (factor/process 1 and other individual factors in Čertovo and Prášílské lakes) (Table 2). Winter temperature has increased, as has the NAO,

coincident with (and likely related to) the global surface warming (Jones et al. 2001; Zwiers 2002). The winter of 1995/1996 marked a dramatic switch in the NAO index from positive to negative values. Subsequently, Al_T (Fig. 4) concentrations distinctly increased. The relationship between the NAO index and local climate differs between northwestern and southern Europe (Hurrell 1995; Rodwell et al. 1999). Hence, climate effects on freshwater chemistry likely differ across Europe.

Understanding the interactions of climatic variability and human stresses on freshwater chemistry is important for the prediction of future changes in aquatic ecosystems (Norton and Veselý in press). The study period (1984–2001) was a period of very sharp decline in the atmospheric deposition

Table 2. Results of factor analyses (S-plus; MathSoft 1997).

Variable	Černé Lake Factor				Čertovo Lake Factor			
	1	2	3	4	1	2	3	4
P_{12}		-0.67		-0.32		0.41	0.78	-0.64
T_{12}	0.70		-0.26	0.66	0.92	0.27		
T_{DJF}	0.73		-0.33	0.47	0.88	0.26		
NAO_{DJF}	0.80				0.45	0.71		
$\Delta\text{Al}^{3+}; \Delta\text{Al}_T$	-0.38		0.61			-0.67	0.30	0.28
ΔH^+			0.61				0.52	-0.31
ΔBCs		0.73						0.60
V%	0.27	0.15	0.14	0.12	0.27	0.16	0.15	0.09
	Plešné Lake				Prášílské Lake			
	1	2	3	4	1	2	3	4
P_{12}			0.55			0.49	-0.29	
T_{12}	0.91		-0.28		0.91			
T_{DJF}	0.88	-0.28			0.92		0.25	
NAO_{DJF}	0.75				0.49		0.52	
ΔAl_T	-0.39	0.62			-0.29	0.33	-0.53	
ΔH^+		0.75				0.75		
ΔBCs				0.50		-0.77		
V%	0.33	0.16	0.07	0.06	0.29	0.23	0.10	0.01

Only factors loading ≥ 0.25 are given. P_{12} : precipitation during the 12 months preceding sample collection; T_{12} : average air temperature during the 12 months preceding sample collection; T_{DJF} : average temperature during the preceding winter; NAO_{DJF} : winter NAO index (average for December–February); ΔAl_T , ΔH^+ , and ΔBCs : residuals of Al_T , H^+ , and BCs from linear regression with SAAs (ΔAl^{3+} in the case of Černé Lake); V: variance within data, in %. BC: base cation.

Table 3. Comparison of simple and multilinear regression by stepwise linear regression (MathSoft 1997).

Lake	$Al_T = a + b(SAA)$			$Al_T = a + b(SAA) + c(T_{12}) + d(NAO_{DIF}) + e(P_{12})$				
		b	r^2	b	c	d	e	r^2
Černé	Al_T	0.170	0.80	0.149	-2.38	—	—	0.89
Černé	Al^{3+}	0.125	0.80	0.111	-1.67	—	—	0.87
Čertovo	Al_T	0.141	0.84	0.132	-1.22	-0.57	—	0.89
Plešné	Al_T	0.137	0.74	0.120	-3.17	—	—	0.81
Prášílské	Al_T	0.095	0.62	0.082	-1.15	—	0.003	0.73

$n = 49$ for Černé and Prášílské lakes; 50 for Čertovo Lake, and 47 for Plešné Lake. Strong acid anions (SAA) in $\mu\text{eq L}^{-1}$, T_{12} in $^{\circ}\text{C}$, and P_{12} in L m^{-2} .

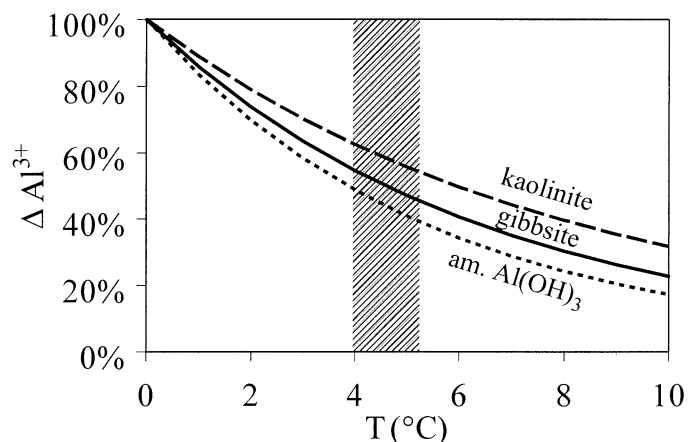


Fig. 9. Calculated percentage changes for equilibrium concentrations of Al^{3+} from 0 to 10°C for amorphous $Al(OH)_3$, gibbsite $Al(OH)_3$, and kaolinite $Al_2Si_2O_5(OH)_4$. Shaded area: the increase of air temperature (T_{12}) in the region during the study.

of sulfur and an increase in temperature. In the four lakes, recovery from acidification, including a decreased Al_i level, was primarily a response to lower concentrations of SAAs. However, residuals from a linear monotonic decrease of Al_T with SAAs in lake waters, in parallel to the decline in acidic deposition, were related to temperature and the NAO index, indicating an effect of climate on lake Al concentration. The relationships observed between temperature (annual and winter) and Al concentrations were probably a result of the inverse relation between solubility of Al-hydroxide phase(s) and temperature. Higher temperatures should reduce the leaching of Al in upper soil horizons and cause lower equilibrium concentrations in slowly moving groundwater. The precipitation of Al in lakes saturated with respect to an Al-phase should increase at higher temperature.

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