Reconstruction of acidic deposition in the catchments of Plešné and Čertovo lakes (the Bohemian Forest)

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Abstract

We revised ten-year old reconstructions of acidic deposition in the Bohemian Forest using (i) newly available measurements of the atmospheric deposition of SO_4^{2-}, NO_3^-, and NH_4^+ in the catchments of Plešné (PL) and Čertovo (CT) lakes, and (ii) new estimates of the historical (1850–2000) and future (2010, 2020) emissions of SO_2, NO_x, and NH_3 in central Europe. The results show that: (1) Acidic deposition has been lower in the PL than CT catchment, probably due to its geographical position further downwind of the major central European emission sources of S and N compounds. (2) SO_4^{2-} deposition was 12–15, 54–66, and 111–135 mmol.m^{-2}.yr^{-1} in the 1850s, 1950s, and 1980s, respectively, and should decline to 13–16 mmol.m^{-2}.yr^{-1} by 2020 (with lower values for PL and higher values for CT). (3) N deposition was dominated by NO_3^- in the 1960–2000 period, while NH_4^+ was the dominant N form from 1850 trough 1960 and will be again after 2010. (4) NO_3^- deposition was only 1–2% higher in the CT than PL catchment, reaching 4, 44, and 99 mmol.m^{-2}.yr^{-1} in the 1850s, 1950s, and 1980s, respectively, and should decline to 35 mmol.m^{-2}.yr^{-1} by 2020. (5) The revised deposition of NH_4^+ was on average 7% higher in the CT than PL catchment, with a Bohemian Forest average of 37, 56, and 85 mmol.m^{-2}.yr^{-1} in the 1850s, 1950s, and 1980s, respectively, and should decline to 61 mmol.m^{-2}.yr^{-1} by 2020. (6) SO_4^{2-} and NO_3^- depositions in the PL catchment were lower than their presupposed values in 2008–2009, due to lower throughfall S and N fluxes beneath canopies damaged by a bark beetle outbreak. (7) The forest decline will probably cause deviations from the recovery of soils and water in the PL compared to the CT catchment due to lower acidic deposition in the early 21st century, but then more rapid soil depletion of base cations and consequent soil acidification will occur due to nutrient uptake by the new forest in the middle of the 21st century.

Key words: Throughfall, precipitation, SO_2, NO_x, NH_3

INTRODUCTION

Atmospheric pollution from anthropogenic sources has become, along with climate changes, a dominant factor affecting the biogeochemistry of aquatic and terrestrial ecosystems in remote areas (PSENNER & CATALAN 1994; NORTON & VESELÝ 2004). Knowledge of the history and possible future development of emission and deposition trends for the major acidifying pollutants, sulphur (S) and nitrogen (N) compounds, is a key factor for understanding changes in these ecosystems and predicting their future development using dynamic modelling (e.g., COSBY et al. 1985). The chemistry of atmospheric depositions has been monitored for a maximum of about 3–4 decades (e.g., BRIMBLECOMBE & STEDMAN 1982), but previous trends in acidic deposition can be estimated from S and N emission trends due to their generally--close relationships (e.g., KOPÁČEK et al. 2001; SCHÖPP et al. 2003; KOPÁČEK & VESELÝ 2005). For example, atmospheric deposition of sulphate, nitrate and ammonium (SO_4^{2-}, NO_3^- and

NH_4^+) in central Europe originates predominantly (65–85%) from the emissions of S and N oxides (SO_2 and NO_x) and ammonia (NH_3) from the same region (BERGE 1997). Historical S and N emissions into the atmosphere can be successfully reconstructed many decades back on the basis of statistical data on the responsible industrial and agricultural activities, which are usually available in national statistical yearbooks (ASMAN et al. 1988; MYLONA 1993; BOUWMAN et al. 1997; OLIVIER et al. 1998).

The changes in central European emission rates of S and N have corresponded closely to parallel changes in their atmospheric deposition (KOPÁČEK & VESELÝ 2005) and the chemistry of central European mountain lakes (KOPÁČEK et al. 1998), which have exhibited a more significant acidification and recovery trends than freshwaters of other European lake districts (EVANS et al. 2001). Attempts to reconstruct historical trends and predict the future development of soil and water chemistry in the Bohemian Forest (BF) (the most atmospherically acidified European lake district) led to the reconstruction of long-term deposition trends of S and N compounds in this area (KOPÁČEK et al. 2001; MAJER et al. 2003). This reconstruction was based on estimated trends in the Czech and central European emissions of S and N compounds to the atmosphere from 1850–2000 (KOPÁČEK & VESELÝ 2005), their probable development presupposed by the Gothenburg Protocol for 2010 (BULL et al. 2001), and measured atmospheric deposition at Čertovo Lake from 1991–2000 (HLAVATÝ 1992; VESELÝ & MAJER 1992; HRUŠKA et al. 2000). In their predictions of future acidic deposition in the BF after 2000, MAJER et al. (2003) assumed 80% and 50% reductions in S and N emissions, respectively, as presupposed by the Gothenburg Protocol in Europe by the year 2010 relative to levels in 1980 (BULL et al. 2001). Moreover, MAJER et al. (2003) used only one deposition trend for the whole BF region. A more detailed evaluation on atmospheric deposition in the catchments of Čertovo (CT) and Plešné (PL) lakes, however, showed that both precipitation and throughfall were less acidic (with significantly higher pH and lower SO_4^{2-}, NO_3^- and NH_4^+ concentrations) in the PL catchment (KOPÁČEK, unpublished data). Moreover, the current models of the 'Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe' (EMEP 2009) have shown that we can expect lower emissions of S and N compounds in central Europe in 2010 and 2020 than the values originally based on the Gothenburg Protocol. The aim of this study is to (1) revise the original reconstructions of S and N deposition trends in the BF (KOPÁČEK et al. 2001; MAJER et al. 2003) for the PL and CT catchments individually, using newly available data on their current and presupposed future emission rates (EMEP 2009), and available trends in their deposition measured directly in both catchments (HLAVATÝ 1992; HRUŠKA et al. 2000; KOPÁČEK et al. 2009), and (2) use the new data to estimate future trends in soil and water recovery from acidification and their possible deviations from the original predictions by MAJER et al. (2003).

MATERIAL AND METHODS

Site description, sampling and analyses

The PL and CT catchments are situated at the north-eastern slope of the BF mountain range at similar elevations of 1090–1378 and 1030–1343 m, respectively. Forest covers ~90% of the PL catchment, is on average 150 years old (in some parts up to 300 years) and is dominated (97%) by Norway spruce. Forest nearly completely covers the entire CT catchment and is dominated by 140-year-old on average Norway spruce (with some trees up to 250 years), with a minor contribution of beech and fir.

Atmospheric deposition is sampled at two throughfall plots and one open area (OA) plot

in each catchment. Throughfall plots are situated at low (PL-L, 1122 m; CT-L, 1057 m) and high (PL-H and CT-H, ~1330 m) elevations. Each of the throughfall plots is equipped with nine bulk collectors. Precipitation (bulk deposition) is sampled in open areas without trees (2 collectors at both the PL-OA and CT-OA plots) at elevations of 1087 and 1180 m, respectively. At each plot, precipitation amounts collected by individual samplers are measured individually, and then combined into one integrated sample for analyses. Rain is sampled in two-week intervals (May to October) and snow in two to four-week intervals (November to April). Sampling has been ongoing at CT-L since 1990 (HLAVATÝ 1992; VESELÝ & MAJER 1992) and at all CT plots since November 1992 (HRUŠKA et al. 2000), and in the PL catchment since November 1997, with the exception of PL-H (since November 2000) (KOPÁČEK et al. 2009).

The plots PL-H and PL-L have been affected by a bark beetle (*Ips typographus*) outbreak since summer 2004 and 2006, respectively, and all trees above collectors died within two years of infestation. Almost all dead trees were still standing in November 2009, but they had lost all their needles, many twigs and some branches. Their specific surface area has thus decreased in the last two years compared to vigorous trees in the CT catchment.

In 1998–2009, concentrations of SO_4^{2-}, NH_4^+, NO_3^-, and other ions (Cl^-, F^-, Ca^{2+}, Mg^{2+}, Na^+, K^+) were determined by ion chromatography (Thermo Separation Products and Dionex IC25, USA), and concentrations of total organic N (TON) and total N (TN) were determined by Kjeldahl digestion according to PROCHÁZKOVÁ (1960) and with a TOC/TN analyzer (Formacs), respectively. The reliability of the analytical results was controlled by means of control samples, an ionic balance approach, and a comparison between measured and calculated conductivities. The differences between the sum of cations and the sum of all anions (including organic acid anions) was <±4% of the total ionic content in the annual volume weighted mean composition of precipitation and throughfall. For details on the sites, sampling, chemical analyses, and quality control of the analytical results see KOPÁČEK et al. (2009). Data on atmospheric deposition in the CT catchment prior to 1998 are from (VESELÝ & MAJER 1992; HRUŠKA et al. 2000; J. VESELÝ – personal communication). The same method (ion chromatography) was used for determination of SO_4^{2-} and NO_3^- concentrations. In contrast, NH_4^+ concentrations were determined by indophenol blue colorimetry prior to 1998. The colorimetric and chromatographic determination of NH_4^+ concentrations provided similar results in a control test in 1998, with the respective annual volume weighted means of 35.5 and 35.4 μmol.l^{-1} at the PL-OA plot and 30.1 vs. 29.0 at the CT-OA plot (KOPÁČEK, unpubl. data).

Total areal atmospheric depositions (*TAAD*, mmol.m^{-2}.yr^{-1}) of SO_4^{2-}, NO_3^-, and NH_4^+ into the PL and CT catchments were calculated from equation (1).

$$TAAD = PF \times X + (TF\text{-}L \times Y + TF\text{-}H \times (1 - Y)) \times (1 - X) / Z \qquad (1)$$

where *PF*, *TF-L*, and *TF-H* are the element fluxes (mmol.m^{-2}.yr^{-1}) deposited to catchments by precipitation and throughfall at low and high elevation plots, respectively. Coefficient *X* is the relative proportion of open area in the catchments and coefficient *Y* is the relative proportion of the forested area in the catchment represented by the low elevation throughfall plot. For a lack of more detailed data, we assumed here that each of the low and high elevation throughfall plots were representative for ~50% of the forested catchment area, and set the *Y* coefficient equal to 0.5. The *X* coefficients were set throughout the analysis at 0.15 and 0.05 for the PL and CT catchments, respectively, as a correction for some parts of the catchments with tree-less and less-dense forest areas compared to the study plots. This calculation probably partly overestimates the actual *TAAD* in the PL catchments in the last two

years, because some dead trees fell down or were cut in 2008–2009. Coefficient Z was used to correct the throughfall data for the effects of canopy processes on the fluxes of individual constituents. The Z coefficient was set at 1.0 for SO_4^{2-} and NO_3^-, with their throughfall deposition roughly equal to the total (wet, dry and horizontal) deposition in the BF, and 0.78 for NH_4^+, because the throughfall flux of ammonium was on average 22% lower than its total flux due to microbial transformation of NH_4^+ into TON in canopies (see later).

Reconstruction of emission trends in sulphur and nitrogen compounds

The historical Czech and Slovak (CS) emission trends for SO_2 and NO_x were reconstructed using the official database of EMEP (2009) for the 1980–2007 period and calculated data for the 1850–1990 period. The 1980–1990 period, with both calculated and official (EMEP 2009) data, was used to compare the two datasets. The emissions reconstruction used in this study differed in several details from the original approach used by KOPÁČEK et al. (2001) and (KOPÁČEK & VESELÝ 2005), which resulted in the following deviations from the original trends:

Anthropogenic emissions of SO_2 were calculated according to VÁRHELYI (1985) on the basis of fuel consumption (lignite, hard coal, oil, and wood), which came from Yearbooks of the CS Statistical Offices for the 1850–1990 period. Historical trends in the average S content of coal and S retention in the ash were obtained from MYLONA (1993). Data on SO_2 emissions from the burning of natural gas were neglected because its contribution to the total CS emissions of SO_2 was low prior to the 1990s. Production of SO_2 from ore smelting before 1900 was estimated from historical records on metal production (Ag, Cu, Pb, and Sn) in the Czech Republic. This estimation was based on metal production, metal and S content of the ore, and S losses during older processing methods (M. HOLUB, personal communication). Emissions of SO_2 from natural sources were estimated in the original study (KOPÁČEK & VESELÝ 2005) as non-sea salt S deposition in remote continental areas (~9 mmol.m^{-2}.yr^{-1}; GALLOWAY et al. 1985), but were neglected as uncertain in this study. When compared to the statistical yearbook data, the calculated CS data on anthropogenic SO_2 emissions were 0 to 14% (on average 7%) higher than the official data in the 1980–1990 period.

Anthropogenic emissions of NO_x from fossil fuel combustion were calculated on the basis of fuel consumption (lignite, hard coal, oil, gas, and wood) in stable and mobile sources (PACYNA et al. 1991) and biomass burning and deforestation (KOPÁČEK & POSCH, in press). The fuel consumption was recalculated to energy production by multiplying with average heating values (Yearbooks of the CS Statistical Offices). NO_x emission factors for major energy sources (electric power plants and heat production) were averages for European data given by PACYNA et al. (1991). The total anthropogenic NO_x emission (the sum of stationary and mobile sources) was estimated from the NO_x emission from stationary sources, which represented 65% of the total anthropogenic CS emission of NO_x in the 1980–1994 period (Yearbooks of the CS Statistical Offices). The contribution of mobile sources to the total anthropogenic emission was considered to be negligible before 1920, 10% between 1920 and World War II, and then to have increased steadily from 10% to 35% in the 1939–1980 period (KOPÁČEK & VESELÝ 2005). NO_x emissions from burning of agricultural waste, biofuel, and deforestation were estimated using a per-capita emission model (KOPÁČEK & POSCH, in press), based on biofuel use, deforestation, and the respective NO_x emission coefficients (OLIVIER et al. 1998; ANDREAE & MERLET 2001). Applying this approach, the NO_x production from biomass burning varied between 1.5–4 mmol.m^{-2}.yr^{-1} in the CS region in 1850–2000, and was lower than the previous value of ~5 mmol.m^{-2}.yr^{-1} (KOPÁČEK & VESELÝ 2005), estimated according to PACYNA et al. (1991) as 3% of anthropogenic NO_x emissions in the 1980s. The total NO_x emission was the sum of its fluxes from both fossil fuel and biofuel burning.

The difference between the calculated anthropogenic CS emission rate of NO_x and the official data varied from –17% to 7%, with an average of –2% in the 1980–1990 period.

The historical trends in NH_3 emission in CS from agricultural sources were calculated for the whole 1850–2007 period from data on livestock production (cattle, pigs, sheep, goats, horses, and poultry; Buijsman et al. 1987), and N-fertilizer consumption and production (Bouwman et al. 2002). In contrast to the original reconstruction, we did not use a constant emission coefficient for cattle throughout the whole period, but rather decreasing values (0.81–1.07 kmol.cow^{-1}.yr^{-1}) back in time as a correction for the lower proportion of dairy cows in total livestock numbers and the N-poorer diet (Kopáček & Posch, in press). Emissions of NH_3 from other (non-agricultural) sources (e.g., humans and pets, sewage sludge, biomass burning and deforestation) were estimated using the per-capita emission model (Kopáček & Posch, in press) on the basis of population, biofuel use, deforestation, and the respective NH_3 emission coefficients (Bouwman et al. 1997; Andreae & Merlet 2001). The NH_3 emissions from non-agricultural sources were 3–8 mmol.m^{-2}.yr^{-1} from 1850–2000, and were lower than the estimate by Buijsman et al. (1987) of ~9 mmol.m^{-2}.yr^{-1} used in previous studies (Kopáček & Veselý 2005). The total NH_3 emission was the sum of agricultural and other sources. The difference between the calculated and statistical yearbook data on the CS emissions of NH_3 varied from –7% to 20%, with an average of 8% in the 1985–2007 period.

Data on central European (Austria, the Czech Republic, Germany, Poland and Slovakia) SO_2 and NO_x emissions and NH_3 emissions in Austria, Germany and Poland after 1980 are from EMEP (2009). Data on central European SO_2 emissions before 1980 are from Mylona (1993). Data on central European NH_3 emissions prior to 1980 were calculated for individual countries using an inventory of livestock production (Asman et al. 1988), the use of synthetic N-fertilizers (FAO 2008), and population (McEvedy & Jones 1978), using the per-capita emission model (Kopáček & Posch, in press). Prognoses of future S and N emissions in Europe in 2010 and 2020 are based on the current models (EMEP 2009).

Revision of trends in sulphur and nitrogen deposition in the Bohemian Forest

Our revision of the original S and N deposition trends in the BF (Kopáček et al. 2001; Majer et al. 2003) was based on the revised historical and presupposed future emissions of S and N compounds in central Europe, and new measurements of their bulk and throughfall deposition in the PL and CT catchments.

The original historical trends in S and N depositions in the BF (Kopáček et al. 2001) were reconstructed, using the original S and N emission trends in CS and central Europe (Kopáček & Veselý 2005), measured average Czech wet-only atmospheric deposition of S and N from 1978–2000 (an average for Svratouch, Košetice, and Hradec Králové; data of the Czech Hydrometeorological Institute), and measured throughfall and bulk deposition of S and N in the CT catchment from 1991–2000 (Hlavatý 1992; Hruška et al. 2000). The reconstruction was done in two steps: (1) Long-term relationships between the respective CS emissions of SO_2, NO_x and NH_3, and the average measured wet-only atmospheric depositions of SO_4^{2-}, NO_3^- and NH_4^+ in the Czech Republic were used to reconstruct historical Czech wet-only depositions of S and N in the 1850–2000 period. The measured Czech wet-only deposition rates of S and N compounds correlated tightly ($P<0.001$) with their emissions in the CS region (Kopáček & Veselý 2005). (2) Relationships between the measured Czech average wet-only deposition and the measured throughfall and bulk depositions of S and N compounds in the CT catchment (1991–2000) were used to recalculate the historical Czech wet-only deposition trends to historical (1850–2000) trends in throughfall and bulk depositions of S and N in the BF. Total atmospheric input of S and N compounds to the BF

catchments was then calculated from the reconstructed throughfall and bulk depositions using equation (1), with the exception of NH_4^+, because the same coefficient $Z = 1.0$ was used for all pollutants. Uncertainty associated with the reconstructed depositions was estimated to be $<\pm30\%$ (for more details and individual relationships see KOPÁČEK et al. 2001).

A similar procedure was used to predict future trends in S and N depositions in the BF from 2000–2010 (MAJER et al. 2003). The expected S and N depositions in 2010 were predicted from their actual trends in the 1990s relatively to the change in S and N emissions presupposed by the Gothenburg Protocol by 2010 (BULL et al. 2001) for the region of the Czech Republic, Germany, Poland, and Slovakia. Deposition data for the 2001–2009 period were then obtained by linear interpolation between the measured 2000 values and the predicted 2010 data.

The original trends in total atmospheric inputs of S and N compounds to the BF catchments were revised in this study, using new measurements of SO_4^{2-}, NO_3^-, and NH_4^+ depositions in the PL and CT catchments (KOPÁČEK et al. 2009), new estimates of historical CS emission trends (1850–1980; this study), actual data on central European S and N emissions (1980–2007; EMEP 2009), and their presupposed values by the years 2010 and 2020 (EMEP 2009). In the first step, the original scenarios of the total SO_4^{2-}, NO_3^-, and NH_4^+ depositions from 1850–2007 were re-calculated proportionally to the difference between the original (KOPÁČEK et al. 2001; MAJER et al. 2003) and new (this study) CS emission trends of SO_2, NO_x, and NH_3, respectively. The estimates of future atmospheric depositions of S and N in the BF ($TAAD_F$) were calculated from the measured S and N depositions in the CT catchment from 1997–2007 ($TAAD_{97-07}$), actual S and N emission rates in the region of Austria, the Czech Republic, Germany, Poland, and Slovakia from 1997–2007 (E_{97-07}; EMEP 2009) and their presupposed future emissions (E_F) in 2010 and 2020 (EMEP 2009), using equation 2:

$$\text{TAAD}_F = (E_F \,/\, E_{97-07}) \times \text{TAAD}_{97-07} \tag{2}$$

Deposition data for 2008–2009 and 2011–2019 were then obtained by linear interpolation between the 2007, 2010, and 2020 data. The CT catchment alone was used in this revision because the original estimate (MAJER et al. 2003) was also based on data from this catchment. In the second step, we calculated the average difference between the revised BF deposition trend and the measured deposition data of each pollutant, individually for both catchments. The total atmospheric deposition of each pollutant was calculated from equation (1), using measured precipitation and throughfall fluxes in the catchments, including the average 22% correction for NH_4^+ transformation to TON in the BF canopies (see later). All available data from the 1992–2009 period (HRUŠKA et al. 2000; KOPÁČEK et al. 2009 and unpublished data) were used for the CT catchment, while only the 1997–2007 period was used for the PL catchment. The PL data for the years 2008 and 2009 were excluded from this calculation, because they were affected by the bark beetle outbreak (see later). The current lower throughfall deposition beneath the damaged canopies compared to that in the unaffected forest would have led to underestimating the historical deposition trends. In the final step, the respective calculated average ratios between the predicted and measured depositions of SO_4^{2-}, NO_3^-, and NH_4^+ (0.92, 0.88, 0.98 in the CT catchment and 1.12, 0.89, and 1.05 in the PL catchment) were used to proportionally recalculate the historical part (1880–2009) and the future part (2010–2020) of the revised BF deposition trends to obtain the "best estimate" S and N deposition trends for the individual catchments.
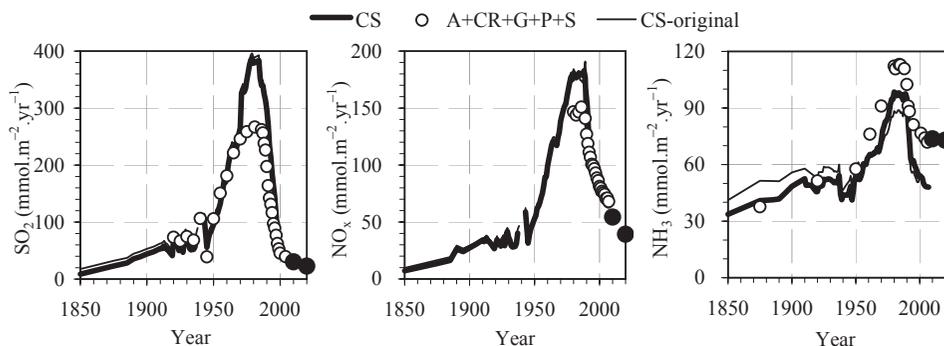
Fig. 1. Trends in areal SO_2, NO_x, and NH_3 emissions in the Czech Republic and Slovakia (CS) and central Europe (Austria, Czech Republic, Germany, Poland, and Slovakia; A+CR+G+P+S). Black points are central European emissions as presupposed by EMEP (2009) models for 2010 and 2020. Original estimates (CS-original) are from Kopáček et al. (2001).

Results and discussion

Central European emissions of SO_2, NO_x, and NH_3

The historical emissions of S and N compounds have exhibited pronounced trends in central Europe over the last two centuries (Fig. 1). The respective rates of total CS emissions of SO_2, NO_x, and NH_3 were <9, <7, and 34 mmol.m^{-2}.yr^{-1} in 1850, and about 23, 14, and 41 mmol.m^{-2}.yr^{-1} in 1880; they then increased slowly until 1950 (96, 53, and 52 mmol.m^{-2}.yr^{-1}) and peaked after a rapid increase in the 1950–1980 period at ~380, ~180 and ~95 mmol.m^{-2}.yr^{-1} in the 1980s. The emissions declined markedly after the political and economical changes in post-communist countries after 1989 (Kopáček & Veselý 2005) to the 2003–2007 averages of 38, 68, and 49 mmol.m^{-2}.yr^{-1}, respectively. The revised trends in SO_2 and NO_x emissions were almost identical to the original trends, with the exception of a smaller contribution of non-industrial sources (Fig. 1). In contrast, the revised emission trend for NH_3 was lower than the original estimates for the 1850–1960s period, and then higher (Fig 1). This difference resulted from different calculations for NH_3 emissions from cattle production and non-agricultural sources than those used in the original estimate (Kopáček & Veselý 2005; Kopáček & Posch, in press).

Comparing emission trends between central Europe as a whole and the CS region, SO_2 declined similarly (86 vs. 90%), while those of NO_x and NH_3 declined less for central Europe (by 51 vs. 64% and 34 vs. 50%, respectively) between the 1981–1985 and 2003–2007 periods (Fig. 1). The large disproportion between declines in the CS and central European emissions of NH_3 was caused by the more stable fluxes in Austria and the former Federal Republic of Germany than in CS, the former German Democratic Republic, and Poland, where larger reductions occurred in cattle production and the application of synthetic N-fertilizers (Kopáček & Veselý 2005; FAO 2008). On a global scale, Europe is the only continent exhibiting a significant decline in NH_3 emissions from agricultural sources within the last two decades (Kopáček & Posch, in press). Most of this change is associated with the post-communist countries (FAO 2008).

Emission controls of S and N oxides started about one decade earlier (since the 1970s) in western versus eastern European countries and were based mostly on reductions in coal use and technical control measures (Schöpp et al. 2003). In eastern European countries, emissions of S and N oxides started declining more abruptly, due primarily to administrative restrictions for the burning of S-rich lignite in the late 1980s and a sharp reduction
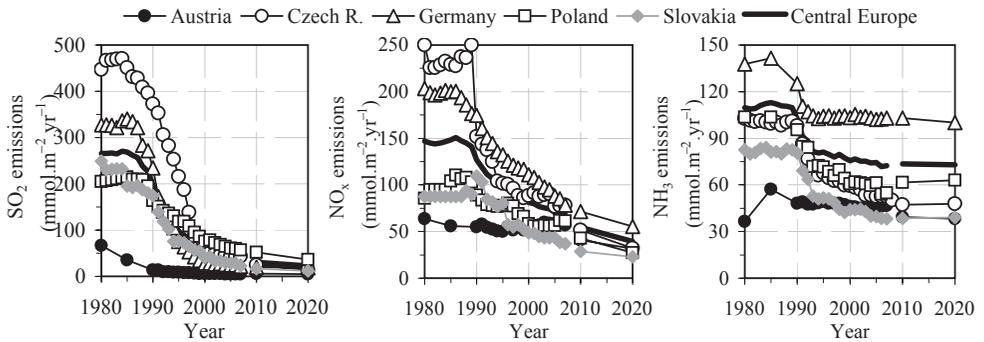
Fig. 2. Trends in areal SO_2, NO_x, and NH_3 emissions in Austria, the Czech Republic, Germany, Poland, Slovakia, and the total area of these countries (Central Europe). Actual values (1980–2007), presupposed values (2010 and 2020). Data are from EMEP (2009), except for NH_3 in the Czech Republic and Slovakia from 1980–2007 (this study).

in energy consumption at the very beginning of the post-communist era (the early 1990s), while technical control measures became more important in the mid 1990s (KOPÁČEK & VESELÝ 2005). In the whole central European region, SO_2 emissions declined steeply and almost linearly between the mid 1980s and 2000, then the decline continued with a slower but steady rate to 34 mmol.m^{-2}.yr^{-1} in 2007. In contrast to S, the NO_x and especially NH_3 emissions declined most rapidly from 1989–1994, and then at slower rates to ~68 and ~72 mmol.m^{-2}.yr^{-1}, respectively, in 2007 (Fig. 2, EMEP 2009). Among central European countries, the highest areal emissions of SO_2 currently occur in the Czech Republic and Poland, those of N (both oxidised and reduced forms) in Germany, while the lowest absolute rates of S and N emissions (as well as their lowest relative changes) have been in Austria (Fig. 2).

The current estimates of possible central European levels of SO_2, NO_x, and NH_3 emissions in 2010 (30, 54, and 74 mmol.m^{-2}.yr^{-1}; EMEP 2009) are 31%, 14%, and 9% lower, respectively, than the values presupposed by MAJER et al. (2003) on the basis of the Gothenburg Protocol (BULL et al. 2001). Further significant declines are expected for SO_2 and NO_x between 2010 and 2020, when their central European emissions should reach 23 and 39 mmol.m^{-2}.yr^{-1}, while a stable emission rate is expected for NH_3 (Fig. 2). The actual trends in SO_2 emissions suggest that the estimate for the year 2010 is realistic on a central European basis (Fig. 1), but probably unrealistically low for the Czech Republic (Fig. 2). Similarly, the presupposed future NO_x emissions are probably too low for the Czech Republic and Poland (Fig. 2), but are only slightly (~10 mmol.m^{-2}.yr^{-1}) below the actual central European trend (Fig. 1). In contrast, the expected central European emissions of NH_3 are ~5 mmol.m^{-2}.yr^{-1} higher than would be expected from the actual trend. Consequently, the future emissions of total inorganic N (= NO_x-N + NH_3-N) could be slightly higher than the values expected for central Europe by EMEP (2009) models. Nevertheless, these new estimates seem to be more realistic than the original estimates (MAJER et al. 2003).

According to the new scenarios (Fig. 1), we can expect that the central European S and N emissions could reach their levels typical for ~1880 (SO_2), the 1950s (NO_x), and the 1960s (NH_3) by the year 2020. In CS and other post-communist countries, NH_3 emissions will probably be even lower than at the beginning of the 20th century, due to continuing reductions in cattle production. For example, the current cattle numbers in the Czech Republic (1.4 million heads) are only 43% of those in 1900, and are even lower than in 1830 (~2 million heads) in the same geographical region (KUSKOVÁ et al. 2008), despite the fact that the human population grew from 6 to 10.4 million between 1830 and 2008.

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Table 1. Average (± standard deviation) atmospheric fluxes of sulphate, nitrate and ammonium to the Čertovo (CT) and Plešné (PL) lake catchments in precipitation (PF), at low and high elevation throughfall plots (TF-L and TF-H, respectively), and in total areal deposition (*TAAD*, calculated from equation 1) from 1998–2009. Units: mmol.m^{-2}.yr^{-1}.

		CT	PL	*P**
SO$_4^{2-}$	PF	17±5	14±3	0.004
	TF-L	22±6	21±7	n.s.
	TF-H	36±9	25±6	0.0007
	TAAD	28±7	23±7	0.0001
NO$_3^-$	PF	39 ±11	34±5	0.03
	TF-L	47±7	56±16	0.04
	TF-H	78±10	65±14	0.004
	TAAD	62±7	57±13	0.04
NH$_4^+$	PF	39 ±10	35±8	n.s.
	TF-L	37±8	37±11	n.s.
	TF-H	65±12	55±13	0.0008
	TAAD	64±10	55±13	0.002

*P-value is the significance of the paired *t*-Test (Microsoft Excel Analysis ToolPack; Paired Two Sample for Means, two-tail) with a significance level of 0.05 to test for differences in element inputs to the catchments (*n* = 12). The null hypothesis was that the sample means were equal; n.s., non-significant.

SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$ deposition in the Bohemian Forest

The average amounts of precipitation (averaged for PL-OA and CT-OA) and throughfall (averaged for PL-L, PL-H, CT-L, and CT-H) were 1416 and 1394 mm in the BF in the 1998–2009 period. The average respective SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$ concentrations were 11, 25, and 26 µmol.l^{-1} in precipitation, and 19, 45, and 35 µmol.l^{-1} in throughfall. The average precipitation fluxes of SO$_4^{2-}$, NO$_3$-N, and NH$_4$-N were 16, 36, and 37 mmol.m^{-2}.yr^{-1} (i.e., 15, 5.1, 5.2 kg.ha^{-1}), respectively, and were lower by factors of 1.3–1.7 than their throughfall fluxes (26, 62, and 49 mmol.m^{-2}.yr^{-1}; 25, 8.6, 6.8 kg.ha^{-1}), respectively. Deposition of TON contributed 25 and 58 mmol.m^{-2}.yr^{-1} (i.e., 3.5 and 8.2 kg.ha^{-1}) to the TN precipitation and throughfall fluxes, respectively. A significant portion of the TON throughfall fluxes (14 mmol.m^{-2}.yr^{-1}) originated from microbial transformation of NH$_4^+$ in canopies (KOPÁČEK et al. 2009). Consequently, the TF:PF ratio was higher for TON (2.3) than for SO$_4^{2-}$ and NO$_3^-$ (1.6 and 1.7, respectively), while that for NH$_4^+$ was lower (1.3). KOPÁČEK et al. (2009) estimated that microbial transformation of NH$_4^+$ reduced its throughfall flux by on average 22% compared to the total atmospheric NH$_4^+$ input into the BF canopies from 1997–2007. The SO$_4^{2-}$ and NO$_3^-$ throughfall fluxes were assumed to be conservative and reasonably represent their total (wet, dry, and horizontal) deposition to the study forest plots, similarly to other European mountain forest areas (e.g., ZIMMERMANN et al. 2006; BALESTRINI et al. 2007). Consequently, we set the *Z* coefficients as 1.0 for SO$_4^{2-}$ and NO$_3^-$ and 0.78 for NH$_4^+$ in equation (1) when calculating their total deposition fluxes to the PL and CT catchments from the respective precipitation and throughfall fluxes.

Atmospheric deposition of acidifying pollutants (SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$) was higher in the CT than PL catchment from 1998–2009 (Table 1). This difference resulted from higher concentrations of S and N compounds in the CT than PL catchment, while precipitation and throughfall amounts were similar in both catchments (KOPÁČEK et al. 2009). The different

Table 2. Comparison of the original (KOPÁČEK et al. 2001; MAJER et al. 2003) and revised cumulative depositions of S and N compounds in the catchments of Čertovo (CT) and Plešné (PL) lakes from 1850–2050. Units mol.m^{-2} per the period shown.

|		1850–1900	1901–1950	1951–2000	2001–2050*
Original	SO$_4^{2-}$	0.9	1.6	4.6	1.2
CT	SO$_4^{2-}$	1.0	1.7	5.0	0.9
PL	SO$_4^{2-}$	0.9	1.4	4.1	0.8
Original	NO$_3^-$	0.5	1.0	3.4	2.3
CT	NO$_3^-$	0.5	1.0	3.7	2.1
PL	NO$_3^-$	0.5	1.0	3.7	2.0
Original	NH$_4^+$	2.6	2.8	3.3	3.0
CT	NH$_4^+$	2.2	2.6	3.6	3.1
PL	NH$_4^+$	2.0	2.4	3.4	2.9

*The same deposition rate as that for 2020 was used for the 2021–2050 period in the revised estimates, while the value for 2010 was used for the 2011–2050 period in the original estimate (MAJER et al. 2003).

levels of areal emissions of S and N compounds in Austria, the Czech Republic, and Germany (Fig. 2) were the probable reasons for this difference. The CT catchment is situated close to the Czech-German border, while the PL catchment at the Czech-Austrian border. The dominant wind directions are south, south-west, and west (59% on an annual basis and 66% in winter), with the median value at 220° in the BF (Churáňov Station, data from the Czech Hydrometeorological Institute; see Appendix 1). The great differences in spatial distribution of pollutant emissions (Fig. 2) suggest that air masses transported from the north (north-west to north-east, 21%) are the most enriched with S (Czech and Polish sources), those from south-west and west (46%) are most enriched with N from German sources, and both these directions bring more S and N pollution than air masses coming from south-east and south (16%) from Austria. The PL catchment's greater distance from Germany and closer vicinity to Austria probably results in lower deposition of S and N pollutants in this catchment compared to the CT catchment.

The revised trends in sulphur and nitrogen deposition in the Bohemian Forest

The revised trends in S and N deposition in the BF (Fig. 3) differed from their original estimates (KOPÁČEK et al. 2001; MAJER et al. 2003) more than the revised CS emissions differed from their original trends (Fig. 1). The reason for this larger difference was the spatial difference in atmospheric fluxes between the CT and PL catchments (Table 1). Consequently, the revised cumulative deposition of SO$_4^{2-}$ was on average 9% higher in the CT catchment and 11% lower in the PL catchment than the original estimate for the 1850–2000 period (Table 2), with the highest average absolute deviations of 11 and –14 mmol.m^{-2}.yr^{-1}, respectively, in the 1980s (Fig. 3A). The revised deposition of SO$_4^{2-}$ was 15 and 12 mmol.m^{-2}.yr^{-1} in the 1850s, 66 and 54 mmol.m^{-2}.yr^{-1} in the 1950s, 135 and 111 mmol.m^{-2}.yr^{-1} in the 1980s, and should decline to 16 and 13 mmol.m^{-2}.yr^{-1} by 2020 in the CT and PL catchment, respectively (Fig. 3A).

Trends in the atmospheric deposition of inorganic N (= NO$_3$-N + NH$_4$-N) differed from S deposition, being markedly higher around 1850 and exhibiting a lower decline from their maxima in the 1980s (Fig. 3B). The revised cumulative NO$_3^-$ deposition was higher than the original trend in both catchments (on average by 7% and 5% in the CT and PL catchments, respectively), with the highest average absolute deviations of ~10 mmol.m^{-2}.yr^{-1} in both
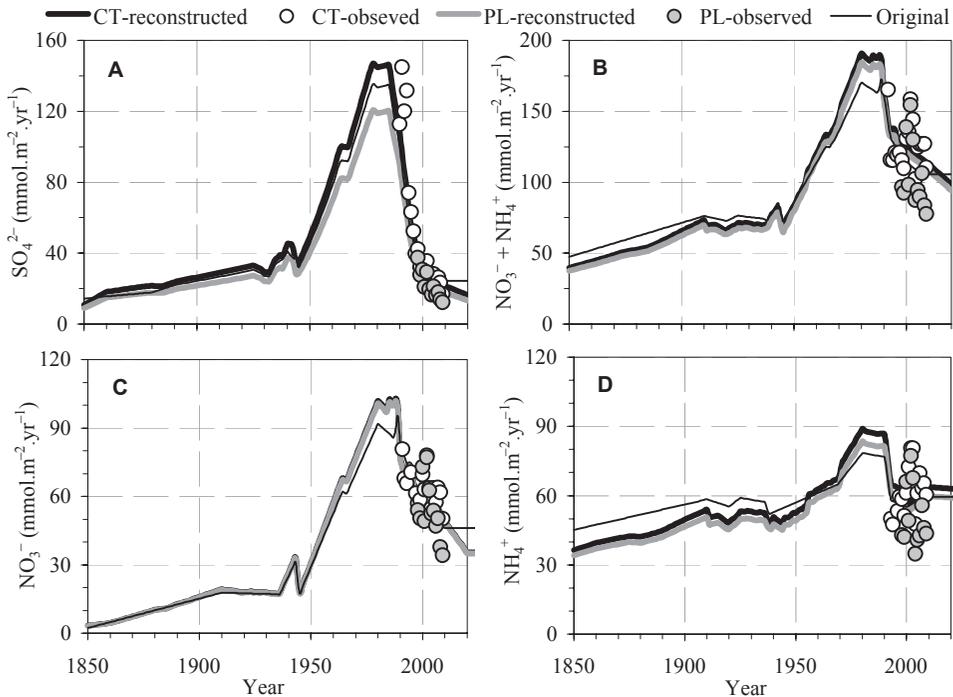
Fig. 3. The revised trends in S and N deposition in the Bohemian Forest and their comparison with measured data and the original trends (Kopáček et al. 2001; Majer et al. 2003).

catchments in the 1980s (Fig. 3C). In contrast to NO_3^-, the revised cumulative NH_4^+ deposition was lower in both catchments than the original estimate (by 5% and 11% in the CT and PL catchments, respectively) from 1850–2000 (Table 2). The revised NH_4^+ deposition was on average ~10 mmol.m^{-2}.yr^{-1} lower than the original estimate in both catchments from 1850–1900, but was 10 and 4 mmol.m^{-2}.yr^{-1} higher in the CT and PL catchments, respectively, in the 1980s (Fig. 3D). Because of the original underestimation of NO_3^- and overestimation of NH_4^+ deposition, the revised cumulative deposition of inorganic N differed only negligibly from the original trends over the 1850–2000 period (Table 2), but its absolute values were on average ~10 mmol.m^{-2}.yr^{-1} lower than the original estimate from 1850–1900, while 21 and 14 mmol.m^{-2}.yr^{-1} higher in the CT and PL catchments, respectively, in the 1980s (Fig. 3B). This suggests that the peak of acidic deposition was probably more severe in the BF than assumed in previous studies (Kopáček et al. 2001, 2002; Majer et al. 2003). The atmospheric deposition of inorganic N was dominated by NO_3^- from 1960–2000, while NH_4^+ was the dominant N form from 1850 to 1960 and will be again after 2010. The revised deposition of NO_3^- was only 1–2% higher in the CT than PL catchment, reaching 4, 44, and 99 mmol.m^{-2}.yr^{-1} in the 1850s, 1950s, and 1980s, respectively, and should decline to 35 mmol.m^{-2}.yr^{-1} by 2020 (Fig. 3C). The revised deposition of NH_4^+ was on average 7% higher in the CT than PL catchment, with the BF average of 37, 56, and 85 mmol.m^{-2}.yr^{-1} in the 1850s, 1950s, and 1980s, respectively, and should decline to 61 mmol.m^{-2}.yr^{-1} by 2020 (Fig. 3D).

In contrast to the underestimation of acidic deposition in the 1980s, the previous study of Majer et al. (2003) overestimated it after 2000, presupposing SO_4^{2-} deposition of 24 mmol. m^{-2}.yr^{-1} from 2010–2050, while the respective current estimates for the CT and PL catch-

Table 3. Cumulative emissions in the region of the current Czech Republic and Slovakia (CS) and cumulative values of total areal atmospheric deposition (calculated from equation 1) in catchments of Čertovo Lake (CT) and Plešné Lake (PL) of S (SO_2, SO_4^{2-}), oxidised N ($N_{ox} = NO_x$, NO_3^-), and reduced N ($N_{red} = NH_3$, NH_4^+) compounds over the 1850–2010 period.

	S	N_{ox}	N_{red}	S	N_{ox}	N_{red}
	mol.m^{-2}	mol.m^{-2}	mol.m^{-2}	t.ha^{-1}	t.ha^{-1}	t.ha^{-1}
Emission in CS	17.0	9.5	8.7	5.5	1.33	1.22
Deposition in CT	7.9	5.7	8.8	2.5	0.80	1.24
Deposition in PL	6.5	5.6	8.3	2.1	0.79	1.16

ments are 22 and 18 mmol.m^{-2}.yr^{-1} in 2010, and 16 and 13 mmol.m^{-2}.yr^{-1} in 2020. Almost similar depositions of NO_3^- and NH_4^+ as those presupposed by MAJER et al. (2003) will probably occur in the CT catchment in 2010 (~49 vs. 46 and 64 vs. 60 mmol.m^{-2}.yr^{-1}, respectively). Deposition of NO_3^- lower than the original estimate can be expected in 2020 (~35 vs. 46 mmol.m^{-2}.yr^{-1}), while NH_4^+ deposition will remain almost stable (Fig. 3). However, the prognosis of future NH_4^+ deposition is based on presupposed NH_3 emissions (EMEP 2009) that assume stable agricultural production. But, in central Europe, the current actual NH_3 emission has already been lower than the values presupposed for 2010 (Fig. 2), and the decline in NH_3 emissions (Fig. 1) may further continue due to the ongoing decrease in European cattle production (FAO 2008).

The respective cumulative fluxes of SO_4^{2-}, NO_3^- and NH_4^+ deposition in the BF represented on average ~42%, 59%, and 98% of the cumulative areal CS emissions of SO_2, NO_x, and NH_3 over the 1850–2010 period (Table 3). The greater similarity between emission and deposition rates of the reduced versus oxidised N forms results from a shorter residence time of NH_3 in the atmosphere compared to N and S oxides. Due to the relatively short residence time in air (hours to days) of NH_3, a large fraction of emitted NH_3 is deposited near the sources, while NO_x (with a longer lifetime in the atmosphere; 1–10 days) can be transported far from its sources (e.g., ASMAN et al. 1998; SEINFELD & PANDIS 1998). The revised estimates suggest that cumulative S deposition should be ~10% lower from 2001–2050 than it was over the 1850–1900 period (Table 2). In contrast, the deposition of both N forms will remain high from 2001–2050, despite their significant reductions compared to the 1951–2000 period, and their cumulative values will be 44% (NH_4^+) and ~350% (NO_3^-) higher than in the 1850–1950 period (Table 2). However, all these future predictions are based on the assumption that central European emissions of SO_2, NO_x, and NH_3 will not change markedly from 2020–2050 and will remain at their 2020 levels.

Implications for the future recovery of Bohemian Forest soils and lakes from acidification

Despite the deviations of the revised trends in acidic deposition from the original trends (Fig. 3), the general conclusion by MAJER et al. (2003) regarding the potential recovery of BF catchment-lake ecosystems from acidification would not significantly change for the CT catchment (or for the neighbouring Černé Lake), because the differences between the revised and original cumulative acidic deposition are small (Table 2). Due to the lower than originally expected acidic deposition after 2010, the recovery of the CT system from acidification may be slightly faster, but acidic deposition will still be too high to prevent the continuing depletion of soil base cation pools for the next several decades.

The future development of soil and water chemistry in the PL catchment, however, may differ from the original prognosis by MAJER et al. (2003) for two reasons. First, according

to the revised trends the cumulative acidic deposition (26 eq.m^{-2} of SO_4^{2-}, NO_3^-, and NH_4^+ in 1850–2000; one eq is one mole of charge) was 8% lower than the originally assumed fluxes (Table 2). The cumulative effect of acidic deposition on the PL system was thus lower, and recovery should be faster than originally expected. Second, the actual atmospheric deposition of S and N compounds in the PL catchment has been even lower than the trends suggested by Fig. 3 due to the forest decline. The continuous loss of needles and twigs has reduced the canopy surface area since 2004 and caused lower horizontal deposition compared to the unaffected forest in the CT catchment. This effect was already evident in 2008–2009, when the observed SO_4^{2-} and NO_3^- depositions were markedly below the PL trend-line (Fig. 3A, C). This lowered acidic deposition may last several decades as observed and modelled elsewhere (Oulehle et al. 2007), depending on the rapidity of reforestation and density of new forest. The forest decline will probably cause the following two deviations from the recovery of soils and water in the PL catchment presupposed by Majer et al. (2003): (1) Acidic deposition will be lower than expected in the early 21st century, but then (2) more rapid soil depletion of base cations and consequent soil acidification will occur in the middle of the 21st century due to nutrient uptake by the new forest. This uptake usually reaches a maximum in 40–50-year-old spruce forests (Hruška & Cienciala 2005). The net effect of the latter process, however, will not be as pronounced as if the decaying forest biomass had been extracted from the catchment, and will depend on the rapidity of nutrient liberation from the dead biomass and the extent of their leaching from soils to surface waters. Both processes, including enhanced S and N deposition onto a newly growing spruce canopy and subsequent uptake of base cations by growing forest will probably result in water acidification in the future (within 30–50 years). Nevertheless, the extent of this likely acidification is difficult to predict.

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Appendix 1. Percent distribution of wind directions at the Churáňov station from 1961 to 2009. Data are based on 3 readings per day (at 7, 14, and 21 hours), provided by the Czech Hydrometeorological Institute. Winter, December-February; spring, March-May; summer, June-August; autumn, September-November; year, January-December.

Direction		Year	Winter	Spring	Summer	Autumn
Calm	(no wind)	3.6	3.3	3.0	4.5	1.8
North	337.5°–22.5°	8.3	6.5	10.3	10.3	5.0
North-east	22.5°–67.5°	7.5	4.6	9.7	9.5	1.1
East	67.5°–112.5°	13.5	11.4	16.1	12.2	9.8
South-east	112.5°–157.5°	3.2	3.5	3.3	2.8	12.4
South	157.5°–202.5°	12.4	14.4	10.8	9.1	9.0
South-west	202.5°–247.5°	17.1	19.3	14.1	15.8	17.8
West	247.5°–292.5°	29.0	32.6	27.0	29.4	22.3
North-west	292.5°–337.5°	5.3	4.5	5.8	6.4	20.8