Beryllium Geochemistry in the Lesni Potok Catchment (Czech Republic), 7 Years of Systematic Study

T. NAVRÁTIL[1,2], P. SKŘIVAN[1], L. MINAŘÍK[1] and A. ŽIGOVÁ[1]
[1]*Geological Institute, Academy of Sciences, Rozvojová 135, Praha 6 - Suchdol, Czech Republic;*
[2]*Faculty of Science, Charles University, Albertov 6, Praha 2, Czech Republic*

Abstract. The biogeochemical study was carried out at the Lesni potok (LP) catchment, Central Bohemia, Czech Republic. The ecosystem was impacted by heavy acid deposition during the industrial development in 1980–1990. The catchment is forested mostly by two tree species, Norway spruce and European beech. The Be concentration in the granite bedrock is 12.6 mg kg^{-1}. The plagioclase contains the highest Be concentrations out of the rock-forming minerals. Elevated concentration of Be (5.4 μg L^{-1}) in surface waters is a result of its mobilization from the soils (3.9 mg kg^{-1}) and weathered rock by acid precipitation. As the pH of the precipitation and consecutively pH of the surface waters is increasing in the Czech Republic, the Be concentrations in the surface waters gradually decrease. Groundwater with high pH values contains lower concentrations of Be (0.17 μg L^{-1}) than surface waters. The soils at prone area of the catchment are depleted in Be compared to the soils in the riparian zones. The vegetation located on the prone area contains lower concentrations of the Be than vegetation at riparian zones. The monitoring of Be in the environment is important with respect to its possible harmful effects on aquatic biota and root systems of the plants.

Key words: beryllium, geochemistry, catchment, monitoring, soils, granite, surface water, groundwater, speciation

1. Introduction

Beryllium, metal with atomic number (4) and one stable isotope ^{9}Be, is the first element in the second column of the Periodic Table. It behaves, owing to its atomic parameters and properties, more like a transition metal than might be expected from the position of Be in the Periodic Table.

The average upper continental crust abundance of Be was determined by Wedepohl (1969) to 3.1 mg kg^{-1}. Higher concentrations of Be are associated before all with granitic or pegmatitic rocks. The beryllium concentration in Czech coal reached values up to 85 mg kg^{-1} (Bouška et al., 1997). Deposition of Be originating in coal burning emissions, could cause increased concentrations of Be in surface waters.

Views concerning the toxicity of Be are ambiguous. Higher concentrations of dissolved Be compounds in food and water could induce chronic health effects in

humans. Contamination of drinking water with Be is essentially unknown, although the Czech Republic standard of Be in drinking water is very low (0.2 μg Be L^{-1}). Inhalation of solid aerosol with high concentration of beryllium leads to irritation of the respiratory tract and eventually may cause lung cancer (Bencko et al., 1984; Kuschner, 1981). Higher concentrations of the soluble forms of Be in soil are phytotoxic (Kaplan et al., 1990; Sajwan et al., 1996).

Total annual atmospheric emissions of beryllium (mostly of anthropogenic origin) in Europe are estimated at 50 metric tons. They are much lower than those of most other toxic trace elements (Moore, 1991). The evaluation of the residence time of Be in the atmosphere is based on that of the ^{7}Be radioisotope, which originates in the stratosphere and upper atmosphere. Estimated values for ^{7}Be are 10.3 to 48 days (Baeza et al., 1996; Koch et al., 1996). The value for the stable lithospheric isotope ^{9}Be should be shorter, because it is introduced into the troposphere (mostly with the coal fly ash and terrigenous dust) on coarser particles. The most probable value for residence time of ^{9}Be is about nine days, the same as for atmospheric water. In soils and sediments, beryllium becomes highly mobile under acidic conditions. This behavior of Be determines its sensitivity towards the acidic precipitation. Adsorption-desorption processes play a major role in soil-water transfer affecting the Be mobility in soil and leaching to surface- or groundwater. Increasing soil pH leads to a rapid increase in the net negative surface charge of soil particles (Naidu et al., 1998) causing the enhanced affinity for metal ions and increase in cation exchange capacity. Beryllium is strongly bound to solids under neutral conditions in natural systems. In waters of pH $\approx$ 7.5, practically all Be occurs in/on suspended matter. The partitioning of Be between solid and water phases, characterized by the partition (distribution) coefficient K_D, is a function of charge density of the solid surface, equilibration time, concentration of solid in the mixture, and pH, composition and ionic strength of the solution. Log K_D of Be derived for rivers ranges from 4.20 to 5.30 (Olsen et al., 1986). Lakes are relatively high, 5.30 to 5.95 (Vogler et al., 1996). Besides the adsorption/desorption processes, beryllium may also be liberated from soils and weathering products of the parent rocks during the dissolution of Al or Fe hydroxides (Veselý et al., 1989; Skřivan, 1994).

The objective of this study is to describe and discuss the origin, concentration, speciation and fluxes of Be in precipitation, throughfall and surface/subsurface waters of the Lesni potok catchment. The second objective is to describe and explain the differences in Be geochemistry at two differing Czech sites, the Lesni potok and Lysina catchments (Krám et al., 1998), when possible.

2. Materials and Methods

The Lesni potok catchment (further in text as LP) (0.765 km^2) stretches 30 km southeast of Prague in central Czech Republic. The geographical coordinates of the catchment are 49°58′35″ lat. N, 14°46′40″ long. E (Figure 1). A brief description
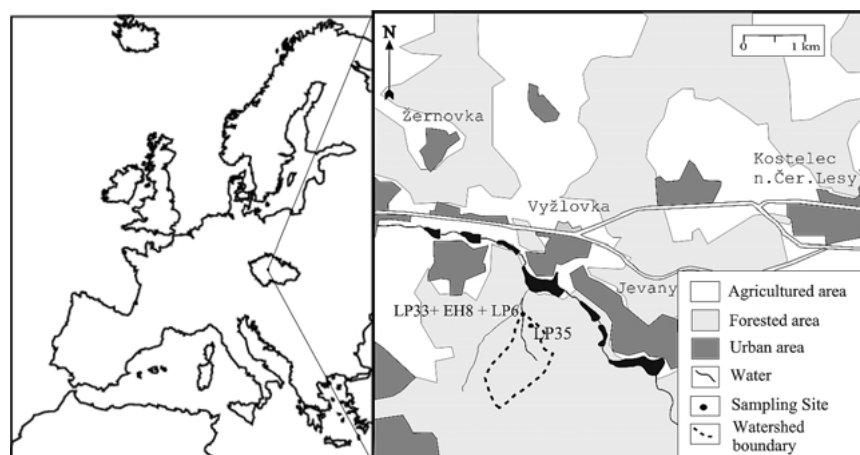
Figure 1. Map of the catchment vicinity.

of the area was presented in (Minařík et al., 1998). The bedrock of the catchment is composed of the Říčany pluton granite. The Thomson weir for sampling of the surface water is at 406 m a.s.l. (site LP6 at Figure 1), adjacent to the weir is situated the EH-8 well (4 m deep, 0.5 m screen at the bottom) for sampling of the shallow groundwater. The highest point of the catchment is at 500 m a.s.l. The mean annual precipitation height is 635 mm and the mean annual temperature is 7.3 °C. The catchment is almost completely forested with deciduous (53%) and coniferous (45%) trees with prevailing European beech (*Fagus sylvatica*) and Norway spruce (*Picea abies*). Surface water, bulk precipitation and beech throughfall have been sampled monthly since 1994. Spruce throughfall has been sampled monthly since 1997 and the subsurface water was sampled for 22 months in 1999 and 2000. The sampling of bulk precipitation and throughfall was conducted in collectors consisting of a 1 L polyethylene (PE) bottle equipped with glass funnel 12 cm in diameter inserted into the cap of the bottle. The funnel was protected from birds by a PE casing with walls cut in a saw-tooth pattern. The mouth of the glass funnel was protected from falling organic debris and insects by a small glass bubble. The PE bottles were exchanged in one month periods. They were sealed in PE bags to prevent their contamination during the transport into the laboratory. All pieces of the equipment were carefully cleaned by diluted nitric acid, hot distilled- and bidistilled water prior to their installation at the locality. The absence of dissolved contaminating ions was controlled by the conductivity measurements of the cleaning liquids. Absence of contamination during the transport of samples between the laboratory and sampling locality, as well as during the sample elaboration was verified by blank procedures with the bidistilled water. More detailed description of sampling and sample elaboration is given in Skřivan et al. (2000). All the liquid samples were filtered (pore size 0.45 μm) in the field or in a laboratory. The filtrates were stabilized with nitric acid, 1 ml of diluted (1:3) HNO_3 per 100 ml of sample.

The soil samples were dried and sieved through a nylon sieve. Fraction of $<$ 1 mm was used for the total Be concentration analyses and also for the leachable Be fractions. The total Be was determined in solution after total decomposition of the sample in mixture of $HF + HNO_3$ and HCl acids. The leachable Be in soil samples was determined after leaching with 0.1 M HNO_3 and 0.5 M NH_4NO_3 ($V/m = 100$, 24 h, fraction $<$ 1 mm, room temperature).

The Be and Al analyses were performed with the graphite furnace AA Spectrometry (GFAAS) at laboratories of the Geological Institute, Academy of Sciences, Prague. Analyses of other cations and anions were performed at laboratories of the Czech Geological Survey, Prague. Concentrations of Be in rock- forming minerals were measured using laser ablation in connection with a mass spectrometer LA-ICP-MS (VG PlasmaQuad 3) at Faculty of Science, Charles University, Prague. The LA-ICP-MS measurement was standardized with NIST-612 standard (Navrátil, 2000). The Be concentration used for calibration with NIST-612 (39 mg kg$^{-1} \pm 5.6$) was published in Jackson et al. (1992).

3. Results and Discussion

3.1. BE IN ROCKS

The parent granite is enriched in Be compared to typical granite. Distribution of Be in the rock is not well known. The data from Hörman in Wedepohl (1969) suggest that Be tends to favor plagioclase over coexisting K-feldspar and biotite.

Results obtained on the basis of mineral separation and its total dissolution are listed in Table I (column GTA-AAS, data from Skřivan, 2000b) together with new set of data acquired by the combination of an electron microprobe and LA-ICP-MS (columns LA-ICP-MS). Results of both methods show that plagioclase is clearly the most important carrier of Be due to its highest Be concentration and considerable mass proportion in the granite. Higher concentrations of the Be in K-feldspar (when determined by mechanical separation and total dissolution) were caused by impurities inside the grains. This fact is supported by the structure of K-feldspar (Figure 2), where the dark areas inside large crystals of K- feldspars are formed by the plagioclase, high in Be. Biotite with high Be concentrations is less important owing to its low abundance in the bedrock granite. Both methods evaluated quartz as a phase with the lowest Be concentration.

3.2. BE IN SOILS

The distribution of Be in the vertical soil profiles was studied in a 1 m deep pits near the weir of the catchment (loc. LP33) and in a prone terrain typical for the major part of the catchment (site LP35, Figure 1). The soil at LP33 was classified as a Gleyic Cambisol and the soil LP35 was classified as Dystric Cambisol (Deckers et al., 1998). The mineralogy of distinguished horizons is nearly identical:

Table I. Be concentration in the bedrock (comparison of the methods).

Method	GTA-AAS		LA-ICP-MS data				
Solid phase	Vol. %	Be (mg kg^{-1})	Be (mg kg^{-1})	(*n*)	StD	MAX	MIN
Plagioclase	27.5	14.8	18.1	37	8.7	40.2	1.4
Orthoclase	32.7	11.9	0.8	37	1.4	6.4	0.2
Quartz	24.8	<0.8	0.3	7	0.6	0.6	0.1
Biotite	8.4	8.3	7.1	8	10.2	34.7	1.4
Bulk rock	100	12.6					
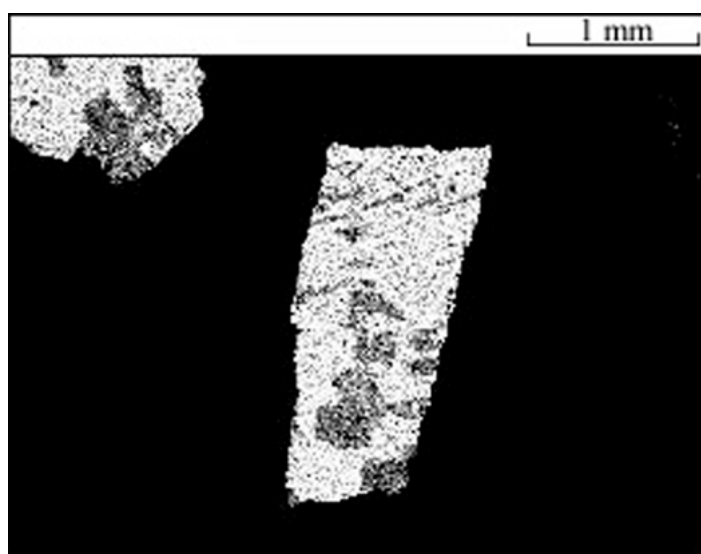
Figure 2. Scattered electron image of the K-feldspar crystal (white) from the Řičany granite, obtained by the scanning electron microscope. Grey areas inside the orthoclase phenocryst are plagioclase inclusions rich in Be.

quartz, kaolinite, illite, orthoclase, plagioclase, and a small amount of smectite. The variations of the total Be concentration with depth are presented in the Figure 3.

Total concentration of Be increases with depth downwards (see Figure 3) and indicates the source of Be – the weathered bedrock. Concentration of leachable Be (portion of the element soluble in 0.1 M HNO_3 and 0.5 M NH_4NO_3) increases also from the A horizon towards the bottom of the profile. Application of more aggressive extraction agent (0.1 M HNO_3) mobilized higher Be concentrations than application of 0.5 M NH_4NO_3. Variations of the Be concentrations were in these cases greater and more dependent on the amount of clayey fraction in the individual soil horizons. The top horizons are leached by acid precipitation and the Be is mobilized from the soil and moved in soil water towards the stream water
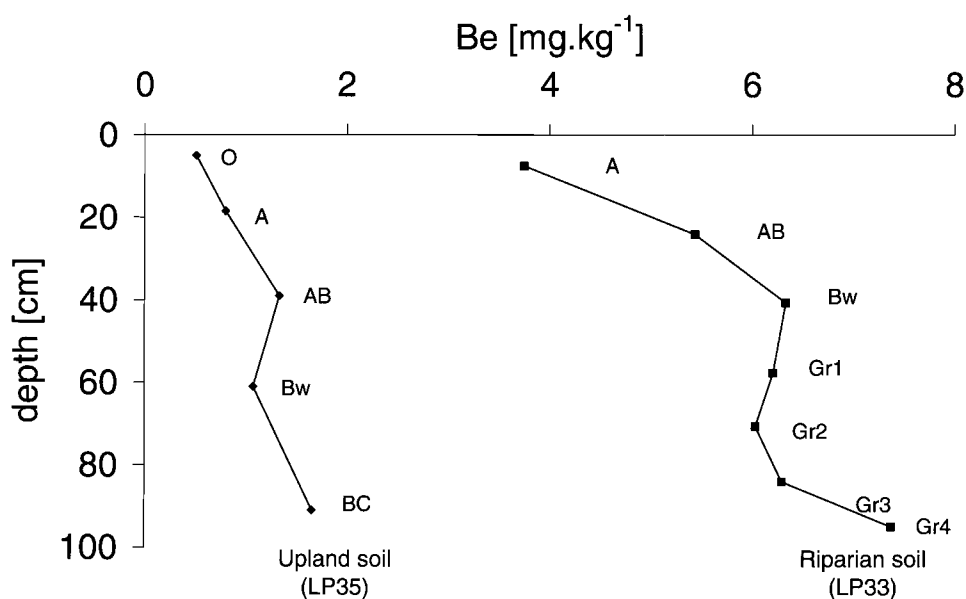
Figure 3. The total Be concentrations in the soil profiles LP33 and LP35.

runoff. Strong acids of anthropogenic origin, which lower the pH of precipitation accelerate this process, as the mobility of Be in soil rapidly increases under induced acidic conditions.

The LP33 profile contains generally higher concentrations of Be than LP35. The profile LP35 is located on the prone terrain with better drainage (soil saturation by water is low), while the LP33 profile is located in the riparian zone of the stream (soil saturation by water is high). Kaste (1999) described the same trend, at the Bear Brook Watershed Maine, USA in more detailed study. He observed the decrease of labile concentrations Be and Al with the distance from the stream in the first 40 cm of soil profile. Concentrations of Be in the riparian zone soils are higher. Changes in bulk chemistry of the ascending soil water caused by degassing (loss of the dissolved CO_2 and associated pH increase, amorphous Al precipitation etc. – Norton, 1983) result in the precipitation of voluminous oxyhydroxides of Al (and Fe, Mn). These phases create, together with the abundant organic matter, favorable conditions for the entrapment of numerous dissolved ions, including those of Be. This fact is documented by a larger share of the labile Be forms at the riparian locality LP33 compared to the prone locality LP35.

3.3. ATMOSPHERIC INPUTS OF BE

Annual volume weighted bulk precipitation flux of Be into the catchment in 2000 was 4.1 μg m^{-2} y^{-1} only, compared to 33.4 μg m^{-2} y^{-1} in 1994. The beech and spruce throughfall fluxes were moderately higher, in 2000 they amounted for

6.6 μg m^{-2} y^{-1} and 8.3 μg m^{-2} y^{-1}, respectively. Higher values of throughfall fluxes should be a result of the stronger entrapment of the near-surface solid atmospheric aerosol or of the leaching of Be incorporated in the assimilatory organs of trees by rainwater. In comparison, the bulk precipitation- and spruce throughfall fluxes at Lysina in 1992 were 45 μg m^{-2} y^{-1} and 42 μg m^{-2} y^{-1}, respectively (Krám et al., 1998). The time-series of Be concentrations in precipitation and throughfall show gradual decrease throughout the years 1994–1999 (Skřivan and Navrátil, 2000). Data of Beneš et al. (1994), where the mean Czech deposition flux of Be in 1990–1992 was 90 μg m^{-2} y^{-1} and those of Berg et al. (1994), 57 μg m^{-1} y^{-1} for rural areas of Norway in 1990, show that the decrease of Be deposition continues for a longer time period. Their deposition fluxes strongly correlate with those of other trace elements As, Cd, Cu, Pb and Zn which are typically present in the fly ash of the Czech low quality lignite coal. These results suggest that significant share of deposited beryllium originates from the coal burning emissions (Navrátil et al. in press). It is evident that the atmospheric inputs of Be in both studied Czech regions follow similar trends and that they are negligible in comparison with its surface stream output, which was at LP 50 times higher than the input (year 2000). The increasing trend of precipitation pH values and consequently of the surface water confirm the declining emissions of anthropogenic acid precursors, as well as the solid aerosol enriched in Be.

3.4. OUTPUTS OF BE THROUGH THE SURFACE- AND GROUNDWATER

Chemical composition of the Lesni potok stream water including Be was already studied by Skřivan et al. (1993, 1994, 1996, 2000) and Navrátil (2000). The average discharge weighted annual concentration of Be in surface water of the Lesni potok catchment changed gradually from 10.5 μg L^{-1} in year 1994 to 4.7 μg L^{-1} in year 2000 (Figure 4). The mean concentration 7.11 μg L^{-1} is high compared to the regional surveys of the Czech freshwaters (Veselý et al., 1998). High concentrations of Be in the surface water of Lesni potok catchment are a result of its abundance in the catchment bedrock and in the weathering products, as well as of high F$^-$, SO$_4^{2-}$ concentrations and occasional low pH values. The observed relationship of pH and Be (Figure 5) concentration is in compliance with previous results (Measures et al., 1983; Veselý et al., 1989; Kaste, 1999; Rissberger, 1993 and Norton et al., 2000). Krám et al. (1998) however, reported no relationship of Be and pH in surface waters of the Lysina catchment. At Lesni potok surface water, pH ranges from 4.7 to 6.8 according to the water discharge. In comparison, pH changes described (Krám et al., 1998) at Lysina catchment are only in the range 3.8 to 4.4. The pH range at Lysina catchment is not wide enough to exhibit the pH dependency of the Be concentrations. Besides this, Krám et al. (1998) attribute this fact to the chemical character of Be (different from the compared Al), which is not a strongly hydrolysing metal.
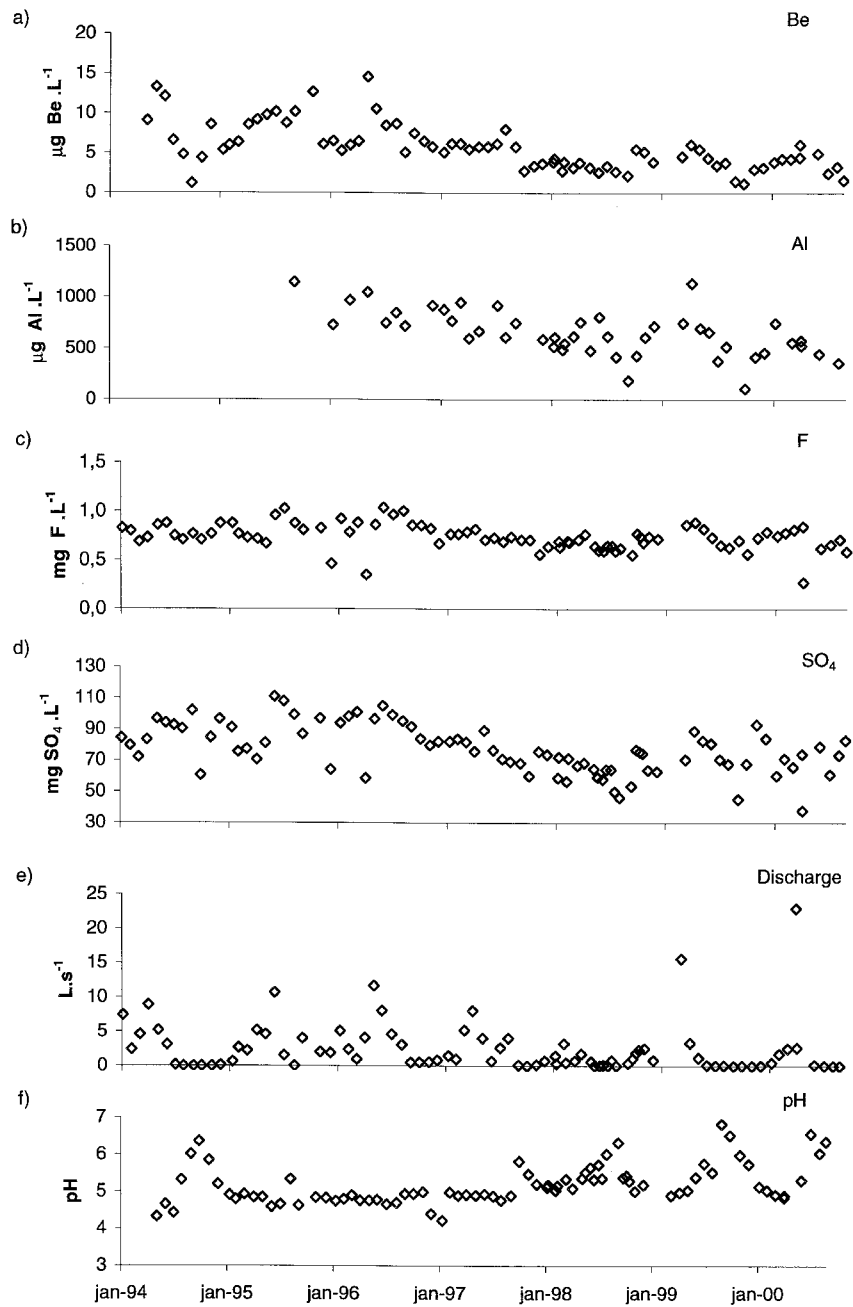
Figure 4. Changes of selected characteristics of surface water from the Lesni potok catchment, years 1994–2000. (a) Concentration of Be, μg L^{-1}, (b) concentration of Al, μg L^{-1}, (c) concentration of F, mg l^1, (d) concentration of SO$_4^{2-}$, mg L^{-1}, (e) discharge, L s^{-1}, (f) pH.
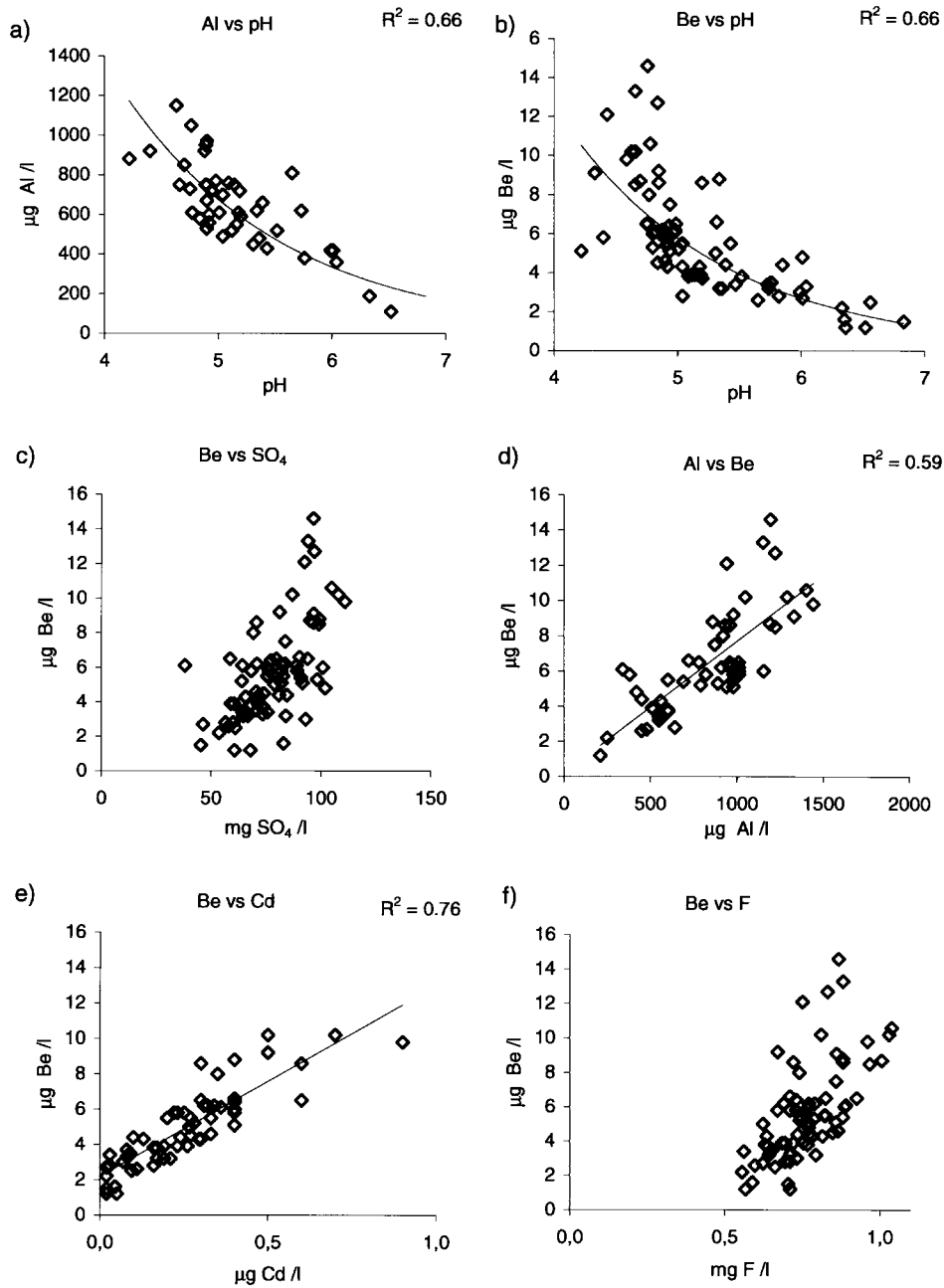
Figure 5. Relationships of the concentrations of selected elements and pH in the surface water: (a) Al vs. pH, (b) Be vs. pH, (c) Be vs. SO_4^{2-}, (d) Al vs. Be, (e) Be vs. Cd, (f) Be vs. F^-. R^2 – squared values of the correlation coefficients.

The major difference between the surface water chemistry is caused by the presence of high DOC concentration (average 16.7 mg L^{-1}) at Lysina (compared with 7 mg L^{-1} at Lesni Potok), which forms complexes with Al, but not with Be. On the other hand, free F^- ions form strong complexes with both Al and Be, depending on the pH value of water (Norton et al., 2000). Competition of Al and Be for fluoride anions is affected by free F^- abundance. Average annual total F concentration in 1999 at Lysina was 0.2 mg L^{-1} in comparison with 0.7 mg L^{-1} at Lesni potok (Navrátil, 2000). Lower fluoride concentrations at Lysina catchment cause full consumption of F^- by Al complexes, so that no free fluoride is available for the complexation of Be. This was confirmed by the dialysis experiments performed by Navrátil (2000).

Concentration of the sum of dissolved forms of Be in surface water is dependent on the bulk chemical composition of the corresponding water (Veselý et al., 1989). On the other hand, resulting concentration of dissolved Be is also controlled by the rate of its dissolution/desorption from the soil profile, stream bed and riparian – or aquatic vegetation.

Significant relationships of the elements concentrations in surface water were found between Be^{2+} and Mg^{2+} ($R = 0.56$, [$n = 55$, $p < 0.001$ for all]), Ca^{2+} ($R = 0.54$), Sr^{2+} ($R = 0.77$), F^- ($R = 0.64$), SO_4^{2-} ($R = 0.64$), Al^{3+} ($R = 0.77$) and Cd^{2+} ($R = 0.75$). Correlation of Be^{2+}, Ca^{2+}, Mg^{2+} and Sr^{2+} originates in similar chemical behavior of the II.A group of elements. Relationship of the Be^{2+} concentration with the F^-, SO_4^{2-} anions (also found at Lysina catchment by Krám et al. 1998) reflects the pH-dependence of the dissolved Be species. However during the low pH values (higher Al and Be concentrations) the Be concentrations may increase faster than those of Al (Figure 5d). This can especially occur during high discharge (episodic events), when other solutes such as SO_4^{2-} and F^- are diluted (Figure 5c and 5f). There is not enough F^- to enable the increased complexation of Al but the pH value enables desorption of Be from the particulate material carried in the stream water. The correlation of Be with Al and Cd actually reflects the well known pH dependence of dissolved Al and Cd ions.

The correlation of Al and Be total concentration in surface waters of the Lesni potok catchment is clear from the Figure 5. The correlation of various Be and Al forms at the Lesni potok surface water was described in (Navrátil, 2000). Krám et al. (1998), however, found no relationship between Al and Be, that was caused by the above mentioned differences in the water chemistries of both catchments (DOC, free F^-, pH range etc.).

The speciation of Be in the surface water of the both catchments was already described in (Navrátil, 2000) using the column exchange techniques and equilibrium environmental software MINEQL+ version 3.01b (Schecher et al., 1992). Main form of Be in the LP surface water is the BeF^+ complex, which always formed >50%. Under low pH conditions portion of free Be^{2+} ion becomes important. But under a low discharge and high pH values $BeOH^+$ and BeF_2 are dominating
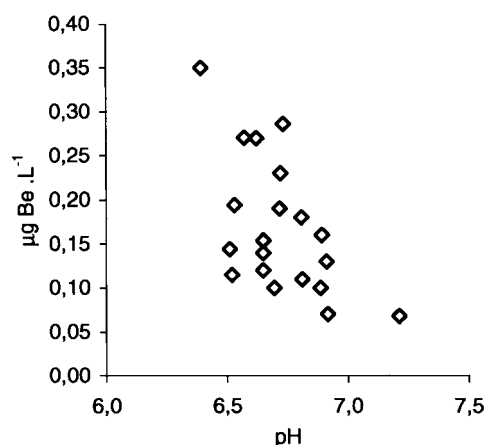
Figure 6. Relationship of the concentration of Be and pH in the groundwater.

over Be^{2+}. The most important factors determining the Be speciation are Al^{3+}, F^-, Be^{2+}, concentrations and pH.

The groundwater sampled from a shallow well (site EH8 at Figure 1) near the weir exhibits high pH values. According to these high pH values (6.4–7.2) the concentrations of Be are very low (0.05–0.35 $\mu g\,L^{-1}$). The average concentrations of the Al^{3+}, SO_4^{2+} and F^- are also lower than in the surface water (10 $\mu g\,L^{-1}$, 0.3 mg L^{-1} and 40 mg L^{-1}). Although the relationship between pH and Be was found (Figure 6) in the groundwater, too. The Be concentrations in the groundwater did not correlate strongly with Al concentrations.

3.5. INTERNAL CYCLING OF BE THROUGH THE FOREST VEGETATION

Atmospheric Be aerosol is scavenged by the forest vegetation, preferably by spruce trees (see the Chapter Atmospheric inputs of Be). On the other hand, part of the lithogenic Be is recycled through the metabolic uptake by rooted vegetation, together with the essential elements such as Ca, Cu, Fe, K, Mg, Mn, Ni, Zn and their homologues, such as Sr, Ba, and Rb (Skřivan et al., 2002). Metabolic root uptake of Be results in its accumulation in various kinds of the tree tissues. Typical concentrations of Be in beech and alder leaves, spruce needles, bark and stem wood of trees growing at various sites in the Lesní potok catchment are presented in Table II. The beech leaves at the LP6 (riparian zone) have higher Be concentrations than those at the LP35 (prone and dry site). These results indicate that the root uptake of Be is governed mainly by the water table level and by the amount of labile Be in the soil profile containing the root system. The relevant Be concentration in the beech tree tissues shows close relation with the Be concentrations in the soil profile (see Figure 3). The highest concentrations were found in the leaves of alder (*Alnus glutinosa*) which is the typical tree in sites with high water saturation of soils. The enhanced uptake of Be at Lesni potok catchment, reflected in its higher

Table II. Be concentrations in the tree tissues.

	Site	Be (mg kg^{-1} d m)
Spruce needles	LP35	<0.1
Beech leaves	LP6	0.024–0.219
Beech leaves	LP35	0.016–0.078
Alder	LP6	0.144–0.418

concentrations in the assimilatory organs, may be caused by the increased acidity associated with the root system of spruce trees.

4. Conclusions

The most important part of the Be in the bedrock is in plagioclases. It contains the highest Be concentrations and its volumetric content in the granite is considerable. Concentrations of total and labile Be in soil profiles increase with depth and explain the source of Be to the watershed as the bedrock and its weathering products. Be concentrations in soils are associated with the amount of clayey fraction and water table level. The Be concentration in the precipitation is decreasing as a result of stricter controls on Czech atmospheric emissions. The main factors affecting the Be concentration and speciation in the surface water are before all Al^{3+}, F^-, DOC concentrations and pH. The Be^{2+} concentrations at Lesni potok surface water have significant relationship with the concentrations of Ca^{2+}, Mg^{2+}, Al^{3+}, Sr^{2+}, F^-, and Cd^{2+}. The Be concentration in the Lesni potok surface water is decreasing according to the changes of the Czech emission budgets and consequently of lower acidic inputs. The Be root uptake is limited mainly by the water table level and by the labile Be abundance in the corresponding soil profile.

Acknowledgments

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