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## Special Section: Hydrological Observatories

### Core Ideas

- We present a general overview of research performed at the Strengbach catchment.
- Multidisciplinary studies are performed to understand export of water and associated fluxes.
- Long-term data study disturbances such as acid rain, forest management, and climate change.

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# The Strengbach Catchment: A Multidisciplinary Environmental Sentry for 30 Years

Marie-Claire Pierret,\* Solenn Cotel, Philippe Ackerer, Emilie Beaulieu, Sylvain Benarioumlil, Marie Boucher, René Boutin, François Chabaux, Frederick Delay, Colin Fournier, Pascal Friedmann, Bertrand Fritz, Sophie Gangloff, Jean-François Girard, Anatoly Legtchenko, Daniel Viville, Sylvain Weill, and Anne Probst

Research activity associated with various observations at the Strengbach catchment in the Vosges Massif (880–1150 m) addresses many questions in the domains of hydrology and geochemistry. The catchment is the observation and experimental site of the Observatoire Hydro-Géochimique de l'Environnement appointed by the Centre National de la Recherche Scientifique. It also belongs to the research facilities that form the French Network of Critical Zone Observatories (OZCAR), which supports a network of critical zone observatories. The catchment is small (0.8 km<sup>2</sup>) with steep slopes (20–30%) on granitic bedrock that mainly allow for forestry (spruce and beech stands) as the main land cover. Meteorological, hydrological, and geochemical data have been monitored since 1986. The first studies conducted were dedicated to the elucidation of acid rain effects on forest ecosystems and particularly on forest decline. Multidisciplinary research studies conducted on the Strengbach catchment enable exploration of the following issues: (i) hydrological functioning at the scale of a small catchment and questions regarding the evolution and preservation of the water resources in mountainous environments (stock, recharge, infiltration, and water pathways), (ii) exchange processes observed at the soil–plant–atmosphere continuum and in particular weathering processes and the evolution of soil mineral fertility (Ca, Mg, K, P), (iii) processes responsible for the export of water and for associated fluxes (dissolved chemicals, suspended materials, bed loads) and their dynamic at the outlet, and (iv) responses of the ecosystems to environmental disturbances (acid rain, forest management, and climate change) and their current and future modeling.

Abbreviations: CZ, critical zone; DOC, dissolved organic carbon; MRS, magnetic resonance sounding; OHGE, Observatoire Hydro-Géochimique de l'Environnement; SS, suspended sediment.

Since the beginning of the industrial era, human activity has heavily modified certain environments, resulting in landscape changes, the development of large cities and industrial centers, extensive agricultural management, and the generation of local pollution (e.g., mines, industrial waste, temporary toxicity of drinkable water, incineration residues, and eutrophication of surface water). Global disturbances, such as acid rain, changes in stratospheric ozone and ultraviolet radiation patterns, and climate change, have resulted (e.g., Reis et al., 2012). Understanding how the environment reacts to anthropogenic or natural disturbances at short- and long-term time scales is one of the major future societal and scientific challenges in the field of natural resource management and conservation.

For several decades, sites around the world have been dedicated to environmental observations. For example, several watersheds have been equipped for the monitoring of various parameters (climate, physico-chemistry of waters, hydrology, the biota, etc.) to better understand critical zone structures (in short, the near atmosphere, landscapes, and the near subsurface), processes, and fluxes. The development of such sites has answered demands from the scientific community for long-term time series of variables that characterize the dynamics of the critical

zone, including disturbances on the system (e.g., acidification) and its responses. Such data have also been made available to decision-makers, nongovernmental organizations, and the general public.

The Strengbach catchment, which is the site of the Observatoire Hydro-Géochimique de l'Environnement (OHGE, <http://ohge.unistra.fr>; CNRS/University of Strasbourg), is well suited for addressing such issues owing to (i) a long-term dataset available for the area (from 1985; Probst et al., 1987), (ii) the availability of field equipment and monitoring devices, and (iii) numerous scientific studies that have been performed on the area. Research performed at this catchment, which is located in the Vosges Mountains in northeastern France, has focused on the evaluation of water and soil resources in relation to climatic changes (i.e., rainfall regimes and atmospheric chemistry trends) and anthropogenic patterns (e.g., forest management and acid precipitation) occurring in this moderate-altitude mountainous region.

Usually, in this type of ecosystem, water is scarce and not evenly distributed, and stocks may rapidly vary over time. In addition, although wood production is an economically important activity in these regions, soil degradation (e.g., from acidification and nutrient depletion) modifies soil fertility features and thus affects the development of forests.

The scientific objective of the OHGE is to develop a detailed understanding of water transport processes and of related chemical fluxes (including nutrients and contaminants) through the critical zone extending from the near atmosphere to the near subsurface. The main purposes of the OHGE are (i) to further document the mid-mountain ecosystems; (ii) to model their functioning at the watershed scale (including past records); and (iii) to predict future evolution in response to disturbances resulting from forest management practices, atmospheric pollution, or climate changes. The OHGE is also involved in educational operations and dissemination of public domain data. Therefore, the structure is (i) an open site for multidisciplinary research to improve knowledge of the functioning of the critical zone, eventually in testing and validating in the field new tracers, models, instruments, and methodologies; (ii) a tool for education (primary school to university), for the professional training of academic and nonacademic personnel, and for scientific and nonscientific dissemination and communication; (iii) an observatory and sentry of natural ecosystems that provides public environmental data; and (iv) a tool for the conservation and archiving of natural samples (water, soil, rocks, plant, litter, and drill cores).

## Motivation and Scientific Questions

Because complex physical, chemical, and biological processes in the critical zone (CZ) control the transfer and storage of water and elements, we have developed a methodology based on the tight coupling of several geophysical, hydrological, and bio-geochemical approaches. In addition, understanding the CZ relies on information on spatial and temporal variations of its hydrological, biological, and geophysical properties. Research associated with observations

at OHGE address key questions of the Millennium Ecosystem Assessment (<http://www.unep.org/maweb/en/index.aspx>) concerning (i) the evolution of ecosystems under plausible scenarios and (ii) the knowledge of time scales, inertia, and risks of nonlinear changes occurring in ecosystems. Research conducted on the Strengbach catchment targets two key questions related to major societal challenges: hydrologic patterns and solute transport through the CZ.

## Availability of Water Resources and Impact of Climate Change

In mid-mountain landscapes, long-term water storage cannot occur due to low levels of storage capacity in the subsurface and the fact that slopes rapidly convey surface and subsurface waters out of the system. Because climate change can modify precipitation regimes and lead to a temperature increase (e.g., more frequent and intense droughts and floods and a decline in snow cover), various components of hydrological cycling (e.g., water storage dynamics and capacities and infiltration rates) can be modified (Haddeland et al., 2014; Kundzewicz et al., 2018). The quantification of water dynamics in the studied catchment based on these modifications is of crucial importance for the availability and quality of water in mountainous regions. The scientific strategy in OHGE is to combine field observations with geochemical, geophysical, and modeling approaches.

Long-term time series and studies of extreme events (e.g., storms, flooding, and snowmelt) lead to a better understanding of hydrodynamics at the catchment scale. Watershed boreholes (Fig. 1) allow the exploration of deep zones (deep-water circulation and weathering processes observed within the substratum) by combining the regular monitoring of hydrologic and geochemical parameters (e.g., water level, pH, temperature, and conductivity) with timely field campaigns (e.g., water sampling, geophysical imaging). A major goal is to devise a physically based model for water and solute flux evaluations that would be applicable and adaptable to other climatic, ecological, and geological environments.

## Atmosphere–Water–Plant–Mineral Interactions: The Sustainable Evolution of Soils and Forests

Forests are sometimes viewed as renewable and sustainable energy sources, and wood production and forestry are the main economic activities pursued in medium-altitude mountainous regions (Schulze et al., 2012). However, intensive forest management practices can deplete or limit soil nutrient resources and affect the quality of soil and water. Transfer of elements in the critical zone, including water–rock interactions, atmospheric inputs, and biological exchanges, testifies to the consequences of modern forestry.

Several tree species, such as spruce, which covers 80% of the Strengbach catchment, accelerate the acidification of soils. Plantations also generate organic acid exudation during nutrient root uptake. These two phenomena accelerate the leaching of elements from soils via acidic water and particularly the leaching of essential bionutrients, such as Ca, Mg, or NO<sub>3</sub>. Such processes can

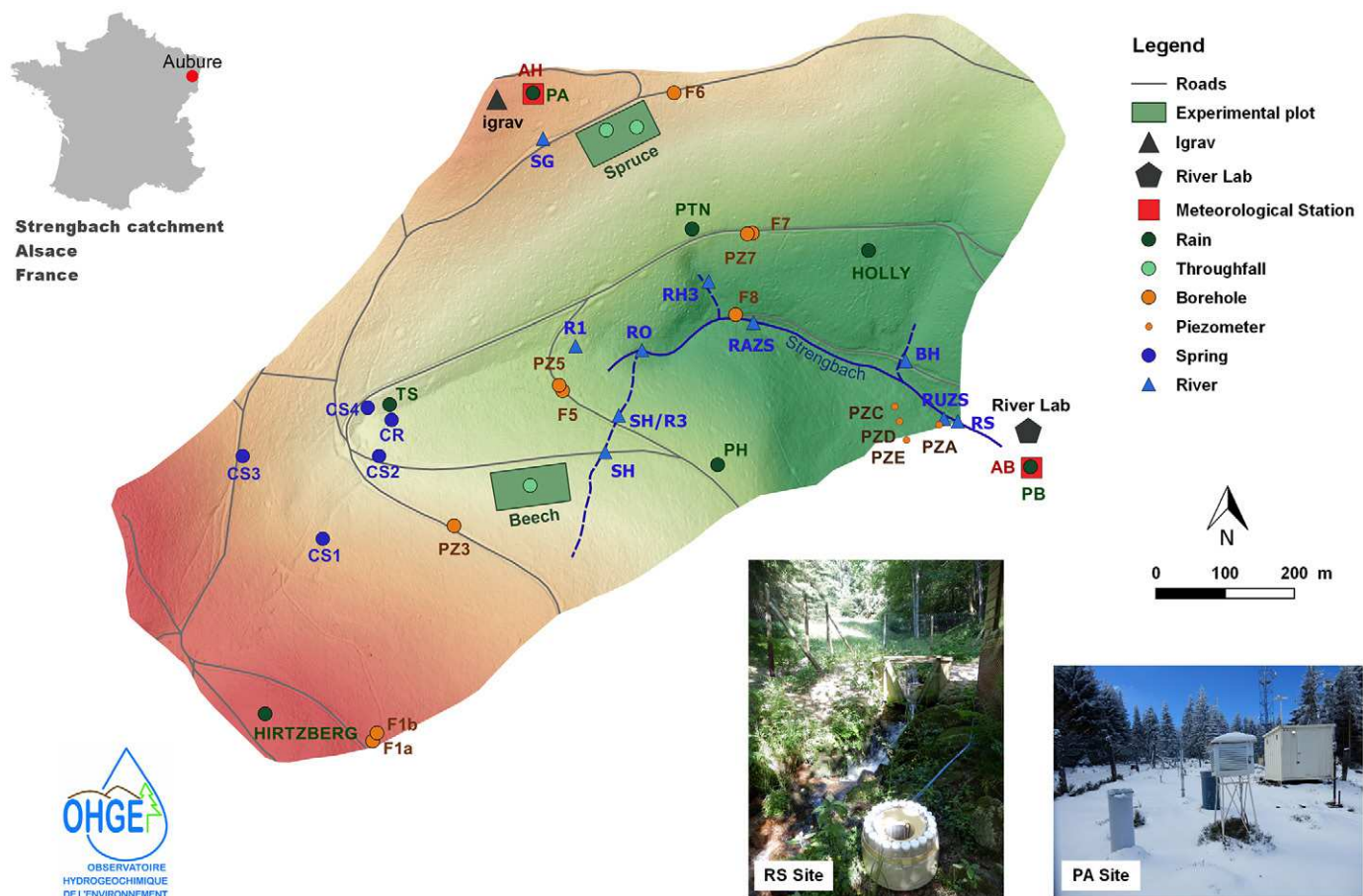


Fig. 1. Map of the Strengbach catchment with equipment used for meteorological, hydrological, and geochemical monitoring. Two experimental tree plots (one located under a spruce plant and another under a beech plant) are used to study throughfall, soil solution, soil, and vegetation levels.

be very critical in acidic bedrock environments, such as granitic environments where reservoirs of Ca and Mg are limited. The consequences of such losses can include the depletion of nutrients in soils, forest decline, the acidification of soil and water, and changes in water–rock interactions (Ackerer et al., 2018; Dambrine et al., 1998a; Fichter et al., 1998a, 1998b; Probst et al., 1990a, 1992a; Prunier et al., 2015; Schmitt et al., 2017).

To summarize, multidisciplinary studies on the Strengbach catchment address the following issues: (i) hydrological functioning at the scale of a small watershed and questions regarding the evolution and preservation of water resources (stock, recharge, infiltration, and water pathways), (ii) exchange processes occurring at the atmosphere–water–plant–soil interface and in particular the evolution of soil mineral fertility (Ca, Mg, K, and P), (iii) weathering process characterization and modeling at the plot and catchment scales and their disturbance by environmental change (acid rain, global climatic changes, forest management, etc.), (iv) processes responsible for export and dynamics observed at stream and subsurface water outlets (e.g., dissolved elements and solid flows); and (v) ecosystem responses to environmental disturbances (acid rain, forest management, and climate change) via present and future modeling.

## Catchment Characteristics

The Strengbach catchment is located in northeastern France on the upper crest of the Vosges Massif, a mountain range oriented north–south along the French and German border 600 km inland from the Atlantic Ocean (Fig. 1) (Probst et al., 1987, 1990a, 1990b). This catchment is a small watershed of 0.8-km<sup>2</sup> surface area with an elevation ranging between 880 and 1150 m asl and with heavily incised side slopes (mean 20–30°) (Fig. 1). It belongs to the municipality of Aubure, a small village with 376 inhabitants in 2011, where economic activity mainly centers on agriculture, forest management, tourism, and a functional re-education and rehabilitation center. Drinking water in the area is fully supplied by mountain water sources. The Strengbach stream is partly fed by several smaller intermittent or permanent streams and springs (Fig. 1). Among them, four permanent springs (called CS1, CS2, CS3, and CS4; Fig. 1) are diverted toward a spring collector. Part of this water is used as drinking water. Since 1986, climatic, hydrological, and geochemical parameters of the Strengbach catchment have been recorded, rendering the watershed one of the longest monitored granitic basements in the world. The first studies were conducted to understand the impacts of acid precipitation on forested ecosystems (Dambrine et al., 1991, 1992; Probst et al., 1987, 1990a, 1992a, 1992b).

## Climate

The local climate is of temperate oceanic mountainous type with a mean annual temperature of 6°C. The coldest month is January, with a mean temperature of -2°C; August is the warmest month, with a mean temperature of 14°C (Probst et al., 1990a; Viville et al., 2012, 2017; OHGE data). Average raw rainfall levels amount to 1380 mm yr<sup>-1</sup> (snowfall occurring 2–4 mo yr<sup>-1</sup>), with interannual variations from 896 to 1713 mm yr<sup>-1</sup> within the period 1986 to 2015 (OHGE data; Fig. 2) (Probst and Viville, 1997).

Mean monthly precipitation for the Strengbach catchment was ~114 mm mo<sup>-1</sup> in the period 1986 to 2015. Less rainfall occurs in April (91.2 mm) and August (110.5 mm), whereas more occurs in October and December (134.8 and 125.7 mm, respectively). Precipitation is characterized by low levels of intra-month variability due to the dominant westerly wind regime that characterizes the area.

Mean annual potential evapotranspiration is estimated at 571 mm yr<sup>-1</sup>, with values ranging between 516 and 729 mm for the period 1986 to 2015.

## Lithology

The bedrock is mainly composed of Hercynian Ca-poor granite (315 ± 7 million yr), which has been subjected to various levels of hydrothermal alterations, with the main hydrothermal alterations occurring 184 million yr ago (El Gh'Mari, 1995). In addition to granite, there exist outcrops of small microgranite and gneiss bodies along the southern and northern slopes, respectively (Fig. 3) (El Gh'Mari, 1995). The gneiss is enriched with Mg, mainly due to the presence of biotite and chlorite (El Gh'Mari, 1995; Fichter et al., 1998a, 1998b). Hydrothermal events have caused the alteration and transformation of albite, K-feldspar, and muscovite into fine-grained illite and quartz; biotite and albite have largely disappeared. The heavily altered granite on the northern slope is characterized by larger amounts of quartz, white mica (muscovite), clay, and Fe oxide; by small amounts of apatite (<1%); and by higher levels of Mg but lower levels of Ca, K, and Na

compared with the less heavily altered granite of the southern slope (El Gh'Mari, 1995; Fichter et al., 1998a, 1998b; Probst et al., 2000).

## Soil and Vegetation Cover

Soils of the watershed are brown acidic to ochreous podzols and are generally roughly 1 m thick. The soils are very coarsely grained, sandy, and rich in gravel (Fichter et al., 1998a). Soil pH levels range from 3.5 at the surface to 4.5 at the 1-m depth. Brown acidic soils are mainly found along the northern slope and are characterized by higher clay, lower K-feldspar, lower albite, higher cation exchange capacity, lower pH, and lower organic matter contents than ochreous podzolic soils, which are mainly found on the southern slope (Dambrine et al., 1998b; Fichter et al., 1998a, 1998b; Probst et al., 1990a). Humus found in the area is moder or mor. The thickness of granitic saprolite varies from 1 to 9 m and is generally thicker on the southern slope (El Gh'Mari, 1995). The two hillsides exhibit distinct climate and topography parameters; the southeast side is mainly north exposed and slopes are smaller, making this part usually colder, more humid, and more rainy than the south-exposed northwest side.

The forest covers 90% of the catchment. The forested area is populated by spruce trees (mainly *Picea abies* L.) (80%) and beech trees (*Fagus sylvatica* L.) (20%). The catchment is used as a commercially managed forest, and plantations range in age from 50 to 145 yr. Lumber density levels range from 430 shafts ha<sup>-1</sup> for a beech plant to 2340 shafts ha<sup>-1</sup> for a young spruce plant. The leaf area index ranges from 2.6 to 3.8 at the catchment scale (Le Goaster et al., 1990). Forest decline related to acid precipitation was observed at the Strengbach catchment in the 1980s, as observed in several areas in northern Europe and the northeastern United States (Dambrine et al., 1998a, 1998b, 2000; de Vries et al., 2014; Paces, 1985; Probst et al., 1990a, 1992b; Ulrich, 1984; Watmough and Dillon, 2003). In particular, the spruce stands underwent ~30% needle loss and needle yellowing due to Ca and Mg deficiencies (Landmann et al., 1995; Probst et al., 1990a).

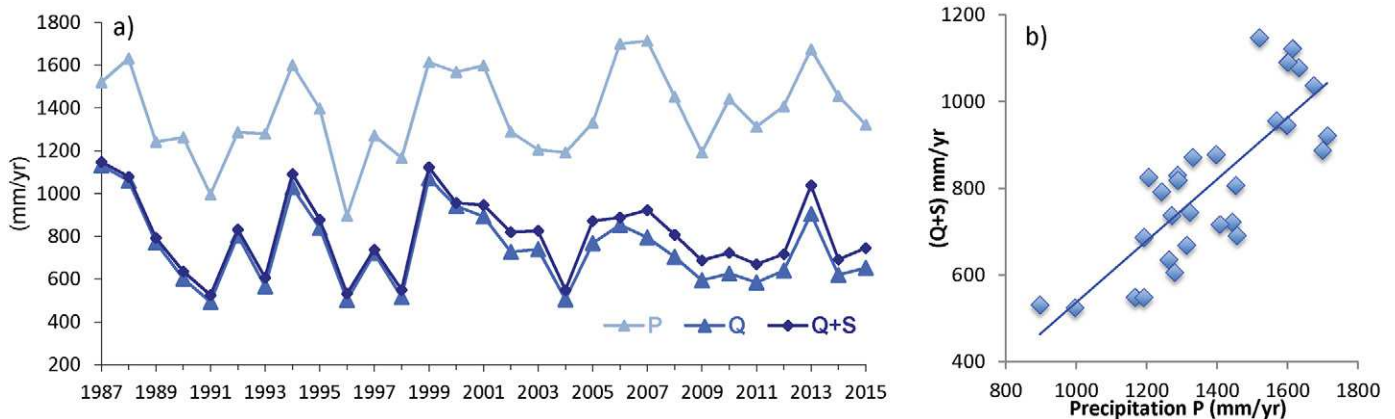


Fig. 2. (a) Variations of annual precipitation ( $P$ ), stream water at the outlet ( $Q$ ), and total flux of water exported at the watershed scale based on discharge levels and drinking water supply ( $Q + S$ ) for 1986 to 2015, and (b) relationship between annual total water output ( $Q + S$ ) and annual precipitation. The Pearson correlation coefficient is 0.81, revealing a significant correlation.

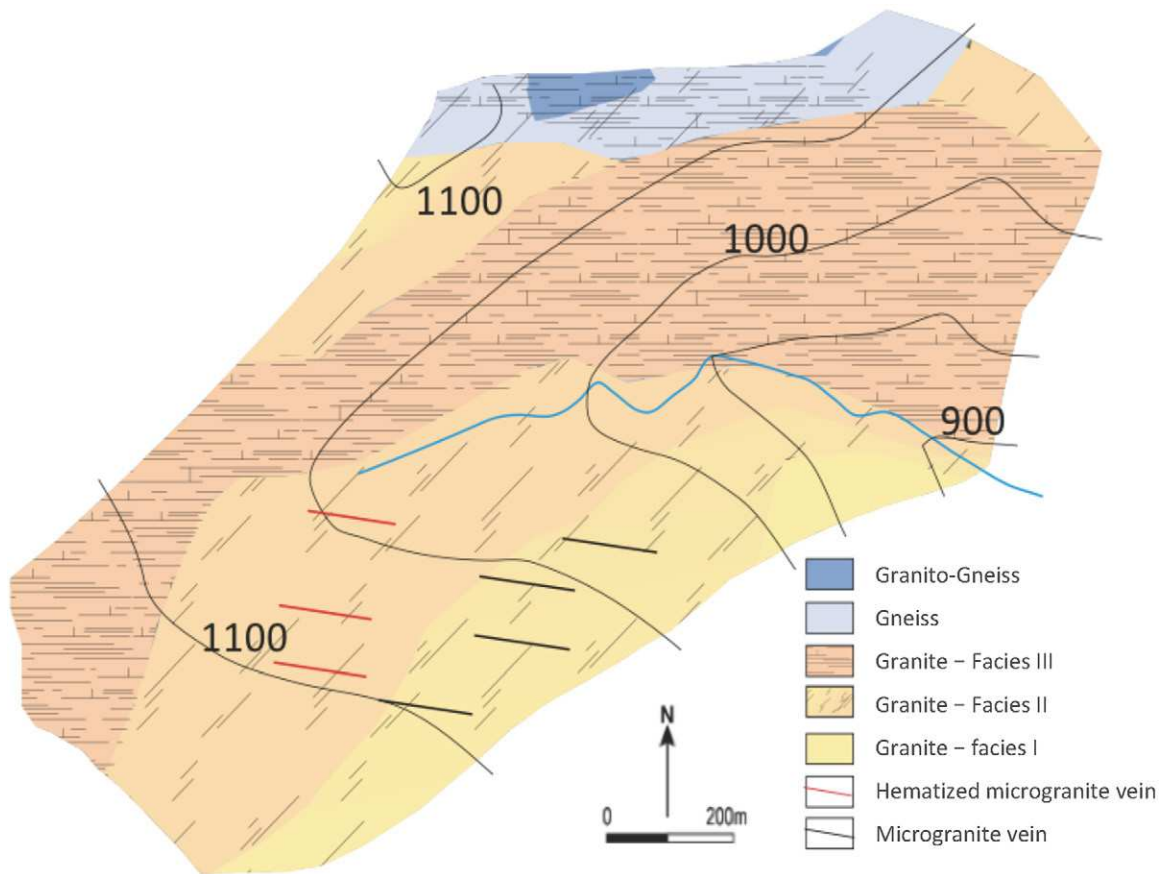


Fig. 3. Geological map of the Strengbach catchment.

## Basic Long-Term Observations

An adapted methodology and sampling locations were carefully set up at the beginning of the catchment survey to perform accurate investigations of the critical zone compartments for hydrological, meteorological, and geochemical data (Dambrine et al., 1998b; Probst et al., 1987, 1990a, 1992a).

## Meteorological and Hydrological Data

Some meteorological parameters (temperature, humidity, wind direction, wind speed, and solar radiation) and rainfall are continuously monitored and sampled every fortnight from a clearing (PA site) (Fig. 1; Table 1).

The minimum rainfall measurement value is 0.1 mm. Snow cover is monitored with a snow depth measurement sensor combined with a direct reading rod. Seven cumulative rain gauges are disseminated on the catchment to assess eventual spatial variability in precipitation (Fig. 1).

The stream flow discharge at the outlet of the catchment has been continuously registered since 1986 (RS site), and the discharges of the four springs (CS1, CS2, CS3, and CS4) located in the spring collector (CR site) are measured every 2 wk (Fig. 1). No other hydrological data were measured on a long-term basis; they were measured only for specific investigations (e.g., Ladouche et al.,

2001). At the outlet, the gauging weir is composed of a calibrated H-flume channel from which the water level is measured with a mechanical water level recorder and an ultrasonic sensor; water level measurements are registered with a precision level of 1 mm. The calibration curve of the relationship between height and discharge has been regularly re-evaluated since 1986.

## Geochemical Data

Stream water at the outlet (RS site), spring water (CR site), and open field precipitation (PA rain) were sampled each week from 1986 to 2005 and have been sampled every 2 wk since (Fig. 1). Stream water was sampled more frequently during flood events using two automatic samplers (one set off at fixed time intervals and a second one set off with stream level variations) to ensure a representative sampling that takes discharge variations into account. From 1986 to 2005, open field precipitation was collected for major-element analysis at four sites positioned in the catchment according to altitude and slope to evaluate chemical variations and the element mass balances (Probst et al., 1990a). All water samples are collected using clean polyethylene bottles (250 mL for major-element analyses) and are filtered through a 0.45- $\mu\text{m}$  pore diameter membrane the same day (Millipore ester cellulose, 142-mm diameter). Values for pH, conductivity, alkalinity, dissolved organic C (DOC),  $\text{NH}_4^+$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,

Table 1. List of data monitored by the Observatoire Hydro-Géochimique de l'Environnement (parameter and sample) with the frequency and the locations.

Parameter	Sample or data	Frequency	Site
Temperature	data	continuously	AH
Humidity	data	continuously	AH
Wind direction	data	continuously	AH
Wind speed	data	continuously	AH
Solar radiation	data	continuously	AH
Rainfall	data	continuously	AH
Rain gauges	data	14 d	PA PH PTN HOLLY TS PB HIRTZBERG
Rain	sample	14 d	AH
Spring	sample	14 d	CR
Stream at outlet	sample	14 d	RS
Stream discharge	data	continuously	RS
Spring discharge	data	14 d	CR

$\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , Si, Al, Mn, and Fe have been measured at the Water Chemistry Laboratory of the Laboratoire d'Hydrologie et de Géochimie de Strasbourg since 1985.

The pH is measured immediately after filtration using a pHM210 MeterLab (Radiometer Analytical) fitted with a Mettler HA405-DXKS8 electrode and calibrated with standard buffer solutions. The precision of the pH measurement is set to  $\pm 0.02$  units. Electrical conductivity and alkalinity levels are determined using a CDM210 MeterLab (analytical radiometer) with a CDC 745-9 electrode (precision of  $0.1 \mu\text{S cm}^{-1}$ ) and with a 716DMS Titrino (Metrohm; precision of  $0.1 \text{ mg L}^{-1}$ , acid–base titration, Gran method), respectively (Gangloff et al., 2014a; Pierret et al., 2014).

Major elements are detected by ionic chromatography, colorimetry, and inductively coupled plasma–atomic emission spectroscopy (ICP-AES). The analytical uncertainty on the concentration of major cations and anions in solution (ionic chromatography; 4000 I, Dionex) is set to  $\pm 2\%$ . The uncertainty of major-element concentrations, such as Fe, Al, Mn, and Si (by ICP-EAS, Jobin Yvon 124) is  $\sim 5\%$ . Since September 1993, DOC has been determined using an organic C analyzer (Shimadzu TOC-5000A) with error levels of 5 to 10% (see further information in Gangloff et al., 2014a; Pierret et al., 2014). The annual average, SD, and minimum and maximum values for major elements are summarized in Table 2.

## Data Management and Policy

The accuracy of chemical analyses is assessed from the ionic balance observed between cations and anions, through the regular analysis of SLRS-4 riverine standards, and through participation in the intercomparison program of the Norwegian Institute for Water Research. Data are stored in the database once a year after their examination and final validation. Available data are

downloadable from <http://ohge.unistra.fr> and <http://bdd-ohge.unstrasbg.fr/index.php/bdd>. Data regarding 22 parameters (e.g., water discharge levels [streams] or volumes [open field precipitation], pH values, conductivity levels, and chemical concentrations) are available. Users can also look at some monitoring devices (as rain PA, spring CR, or stream RS) and associated data for selected periods directly via the web interface (Fig. 4).

## Response To Atmospheric Inputs Case Study of Sulfate and Protons in Open Field Precipitation, Spring, and Stream

Precipitation with pH values  $< 5.6$  is defined as acid rain. Acid rain is mainly associated with industrial releases of  $\text{SO}_2$  in the atmosphere and affects natural and human life in a variety of ways (Charlson and Rodhe, 1982). Acid rain largely appeared in North America and in central and northern Europe in the 1960s (de Vries et al., 2014; Landmann et al., 1995; Norton et al., 2014; Paces, 1985; Ulrich, 1984). The acidification of atmospheric inputs degraded the quality of streams, springs, lakes, and soils. By accelerating the release of base cations and metals (e.g., Al), acid rain led to nutrient imbalances, resulting in forest decline, fish death, eutrophication of surface water, and incidentally to corrosion of monuments and water pipes (Dambrine et al., 1998b; Likens and Bormann, 1974; Likens et al., 1996; Moldan et al., 2004; Probst et al., 1990a, 1990b, 1995c; Watmough and Dillon, 2003; Zhang et al., 2007).

Due to regulation policies (Hettelingh et al., 2008) and economic factors,  $\text{SO}_2$  emissions in the atmosphere have been reduced since the 1980s by 50 to 85% in North America and Europe (Kopáček et al., 2016; Norton et al., 2014) and by 90% in France since the 1990s (CITEPA: <https://www.citepa.org/fr>). However, emissions increased in China until 2007 and continue to increase in India today (Klimont et al., 2013; Smith et al., 2011). That being stated, long-term data obtained from the Strengbach catchment allow the registration of impacts of global and long-range atmospheric pollution to ecosystems located far from direct significant  $\text{SO}_2$  emission regions and sources. Below we focus on pH and sulfate evolution from 1986 as an emblematic case study of acid atmospheric precipitation as a consequence of anthropogenic activities resulting in impacts on stream and spring waters.

The pH and sulfate concentrations in PA, CR, and RS show long-term trends with significant increases and decreases, respectively (Fig. 5a and 5b), as confirmed by *S* score and *P* value data obtained through Mann–Kendal testing (*S* scores of 8.3–15.6 for pH and  $-14.0$  to  $-47.5$  for  $\text{SO}_4^{2-}$ ;  $P < 0.01$  in all cases). Average annual pH levels increased from 4.4 to 5.1, from 6.0 to 6.4, and from 6.1 to 6.5 for PA, CR, and RS, respectively. Average annual sulfate concentrations decreased from 0.028 to 0.007, from 0.110 to 0.044, and from 0.102 to 0.040  $\text{mmol L}^{-1}$  for PA, CR, and RS, respectively.

At the Strengbach catchment scale, annual S deposition decreased from 2 Mg of S in 1986 to 260 kg in 2015 (Pierret et

Table 2. Annual average, standard deviation, and maximum and minimum values of chemical parameters of rain, spring water, and stream water at the outlet of the Strengbach catchment for the period 1986 to 2015 (Observatoire Hydro-Géochimique de l'Environnement data).

	pH	Conductivity	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	H <sub>4</sub> SiO <sub>4</sub>
		μS cm <sup>-1</sup>	mmol L <sup>-1</sup>							
Rain										
Avg.	4.80	15.70	0.012	0.005	0.003	0.008	0.015	0.030	0.017	BDL†
SD	0.68	11.83	0.011	0.007	0.002	0.009	0.012	0.024	0.015	BDL
Max	7.42	86.00	0.078	0.085	0.025	0.065	0.095	0.192	0.121	BDL
Min.	3.78	2.15	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Spring										
Avg.	6.14	34.56	0.088	0.020	0.020	0.077	0.050	0.056	0.076	0.145
SD	0.26	3.38	0.006	0.002	0.002	0.009	0.005	0.017	0.018	0.014
Max.	7.21	46.10	0.124	0.030	0.029	0.113	0.087	0.124	0.110	0.182
Min.	5.40	25.90	0.071	0.012	0.014	0.052	0.034	0.002	0.038	0.017
Stream										
Avg.	6.29	34.04	0.086	0.019	0.025	0.075	0.052	0.036	0.080	0.134
SD	0.21	5.33	0.007	0.006	0.004	0.013	0.011	0.016	0.020	0.012
Max.	7.16	65.00	0.121	0.063	0.039	0.107	0.129	0.093	0.128	0.180
Min.	5.51	21.85	0.061	0.005	0.012	0.036	0.026	0.001	0.032	0.089

† Below detection limit.

al., 2016). However, the mean annual pH of rain for 2015 (5.1) is still lower than the preindustrial pH value estimated at 5.7 to 6 (Kopáček et al., 2016) and shows that steady state and full recovery have not yet been reached. Even though S levels have decreased, N ions have been shown to be the basis of the current levels of precipitation acidity (Pascaud et al., 2016; Pierret et al., 2016).

Stream pH is significantly correlated with precipitation acidity, showing a converging decrease in proton concentrations and thus an increase in pH (Fig. 5a). However, the slope of pH vs. time for open field precipitation ( $8 \times 10^{-5}$ ) is more than twice that for streams ( $3 \times 10^{-5}$ ) (Fig. 5a). Some protons are neutralized while passing through soil, saprolite, and bedrock via exchange and mineral weathering processes (Paces, 1985; Probst et al., 1990a, 1990b). In addition, long-term signal fluctuations for pH in open-field precipitation (SD = 0.61) are significantly dampened during water circulation through the catchment, with a lower SD observed at the outlet (0.17), confirming the neutralization of protons.

Protons calculated from pH (Fig. 5a) and sulfate (Fig. 5b) concentrations are related in precipitation (open field precipitation PA) and in stream water (stream outlet RS) ( $r = 0.88$  and  $r = 0.85$ , respectively). These strong correlations confirm a common origin and behavior. The significant decline of protons and sulfates in atmospheric inputs and in stream water observed at the outlet of the Strengbach catchment can be related to a reduction in anthropogenic SO<sub>2</sub> (proton precursors) emissions from the 1980s in the Northern Hemisphere, as observed from other catchments of northern Europe (Vuorenmaa et al., 2018). These observations reflect the impact of global political decisions on the environment, even in remote regions, because of long-range transport of gaseous pollutants.

## Long-Term Solute Balance

The chemical fluxes of major elements (Na, K, Mg, Ca, Si, Cl, and Seq) at the outlet have been calculated and expressed as is customary as elemental fluxes (Seq for sulfate and Neq for nitrate) (Fig. 6). At the outlet, concentrations of NH<sub>4</sub> and NO<sub>2</sub> fall below detectable limits, and the N flux is controlled by NO<sub>3</sub> concentrations. For the considered period (1986–2015), chemical fluxes can vary by a factor of 2 for Si or Na and by a factor of 4.5 for S (Fig. 6). The relationship between annual chemical fluxes and water fluxes for Na, K, Mg, Ca, Si, Neq, Cl, and Seq during the period 1986 to 2015 at the catchment outlet is significant even for elements undergoing an unstable long-term evolution, such as sulfate. Pearson coefficients ( $r$ ) are 0.99, 0.96, 0.89, 0.88, 1.00, 0.65, 0.95, and 0.80, respectively (Fig. 6). Correlation between chemical and water fluxes does not mean that concentrations at a given time are correlated with the water flux.

These results show that chemical exportation in this type of catchment is partly controlled by water fluxes that influence the time and fluid renewal rate of water–rock interactions. The lower correlation observed for Neq (0.65) with water fluxes could be attributed to additional complex processes implying biological reactions and interactions with vegetation and microorganisms that affect the N cycle. Thus, the effect of hydrological patterns on N export is less significant than it is for other elements. These results also highlight high levels of interannual variability, seasonality, and hydrological and meteorological control over export fluxes. It is therefore of prime importance to monitor streams for several successive hydrological years to make precise and accurate estimations of export fluxes. A short-term analysis could lead to highly uncertain evaluations of weathering fluxes (of >100% in some cases).



RS : Ruisseau à l'exutoire (collecteur global)

date	neRS	débit	NH4	Na	K	Mg	Ca	Al	Mn	Fe	H	Scations	Cl	NO2	NO3	SO4	PO4	alca	alca_H	Sanions	pHlabo	conduc	H4SiO4	NPOC
	m <sup>3</sup> /s	mmol/l	mmol/l	mmol/l	mmol/l	mmol/l	mmol/l	mmol/l	mmol/l	mmol/l	mmol/l	mmol/l	mmol/l	mmol/l	mmol/l	mmol/l	mmol/l	mmol/l	mmol/l	mmol/l	µS/cm	mmol/l	ppm	
2005-01-03 12:45:00	5036	30.92	0.000099	0.086	0.021	0.025	0.073	0	0	0	0.001	0.304	0.06	0	0.069	0.066	0.000099	0.033	0.033	0.328	6.33	30.4	0.134	1.41
2005-01-10 13:50:00	5040	42.6	0.000	0.084	0.021	0.024	0.072	0.000	0.000	0.000	0.001	0.298	0.056	0.000	0.068	0.065	0.000	0.035	0.036	0.326	6.29	31.3	0.134	1.33
2005-01-17 14:05:00	5057	22.68	0.000	0.084	0.020	0.023	0.071	0.000	0.000	0.000	0.001	0.293	0.058	0.000	0.062	0.066	0.000	0.031	0.031	0.315	6.36	33.0	0.136	1.11
2005-01-24 18:20:00	5059	43	0.000	0.083	0.021	0.025	0.073	0.000	0.000	0.000	0.001	0.301	0.057	0.000	0.068	0.067	0.000	0.033	0.033	0.326	6.38	33.1	0.133	1.30
2005-01-31 13:35:00	5086	19.6	0.000	0.084	0.020	0.024	0.072	0.000	0.000	0.000	0.001	0.297	0.057	0.000	0.061	0.068	0.000	0.032	0.032	0.319	6.36	32.9	0.136	1.12

Les données ayant une valeur "null", signifient qu'aucune valeur n'a été renseignée.  
Les données ayant une valeur de 0.00009 signifient que la valeur est en dessous de la limite de détection (LOD).

En cas d'utilisation et/ou d'exploitation de ces données géochimiques, veuillez contacter le responsable de l'OHGE et citer l'observatoire OHGE comme source de données lors de publications scientifiques.

Fig. 4. Example of data obtained directly via web downloading from the Observatoire Hydro-Géochimique de l'Environnement database. This example corresponds to stream water discharge and chemistry for the period from 1 Jan. 2005 to 1 Feb. 2005. The text is in French, but an English version is underway.

## Project-Specific Studies Atmosphere–Water–Plant–Soil Interactions

### Documenting Elementary Mechanisms

Two experimental plots representing specific environmental characteristics of the catchment are equipped for studying atmosphere–soil–plant interactions: one is located under a spruce stand on a south-facing slope and another positioned under a beech stand on a north-facing slope (Fig. 1). In each plot, soil, soil solution, litter, plant, and throughfall have been investigated during the past three decades. Throughfall has been sampled every 2 wk from two gutters (2.0 by 0.2 m) since October 1986 and from five bucket collectors since 1990 for spruce and beech stands, respectively (Fig. 1). The continuous records of open field precipitation and throughfall chemical compositions from 1986 to the present allow the quantification of interactions occurring between atmospheric deposition and vegetation and for inference of long-term evolution patterns subsequent to pollutant emission trends.

The long-term monitoring records reveal some significant temporal trends (pH, conductivity,  $\text{SO}_4^{2-}$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{K}^+$ ). Significant decreases in concentrations and fluxes of several ions have been observed ( $\text{H}^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{Cl}^-$ , and  $\text{Ca}^{2+}$ ) in open field precipitation and throughfall. A regular and strong decline in protons and sulfate has followed a decrease in anthropogenic  $\text{SO}_2$  and  $\text{NO}_x$  (proton precursors) since the 1980s (Probst et al., 1995a). The two tree plots exhibit contrasting effects regarding physicochemical parameters of incoming precipitation with higher levels of water interception and higher chemical concentration observed under spruce trees than under beech trees, highlighting the role of tree species in the transformation of atmospheric inputs reaching soils.

Soil solutions have been sampled with zero-tension lysimeter plates at 5, 10, 30, and 60 cm and at 10- and 70-cm depths for spruce and beech stands, respectively. Analyses are not continuous over time since 1986 because they were performed in association with opportunities brought by different short-term research projects. Data show significant changes in the soil solution chemistry for both tree plots with Ca and Mg depletion. This may be due to a decrease in exchangeable Ca and Mg (Dambrine et al., 1995, 1998b) or to recent modifications of the type of water–rock interactions (Prunier et al., 2015; Schmitt et al., 2017).

The mineral–plant–water interface has been studied at the plot scale using tools such as (i) stable isotopes (Ca, Li, and B) (Cenki-Tok et al., 2009; Cividini et al., 2010; Lemarchand et al., 2010; Lemarchand et al., 2012; Schmitt et al., 2003, 2017), (ii) radiogenic isotopes (Sr, Nd, and Pb; Ackerer et al., 2016; Gangloff et al., 2014b; Poszwa et al., 2003; Stille et al., 2012) and radionuclides (U, Th, and Ra) (Prunier et al., 2015; Rihs et al., 2011), (iii) Mo isotopes (Nägler et al., 2016), and (iv) rare earth elements (REE) (Aubert et al., 2002b, 2004; Brioschi et al., 2013; Stille et al., 2006, 2009). After interception and evapotranspiration processes within the canopy (Viville et al., 1993), the water circulates from the upper soil layer to the deeper zone of weathered cover

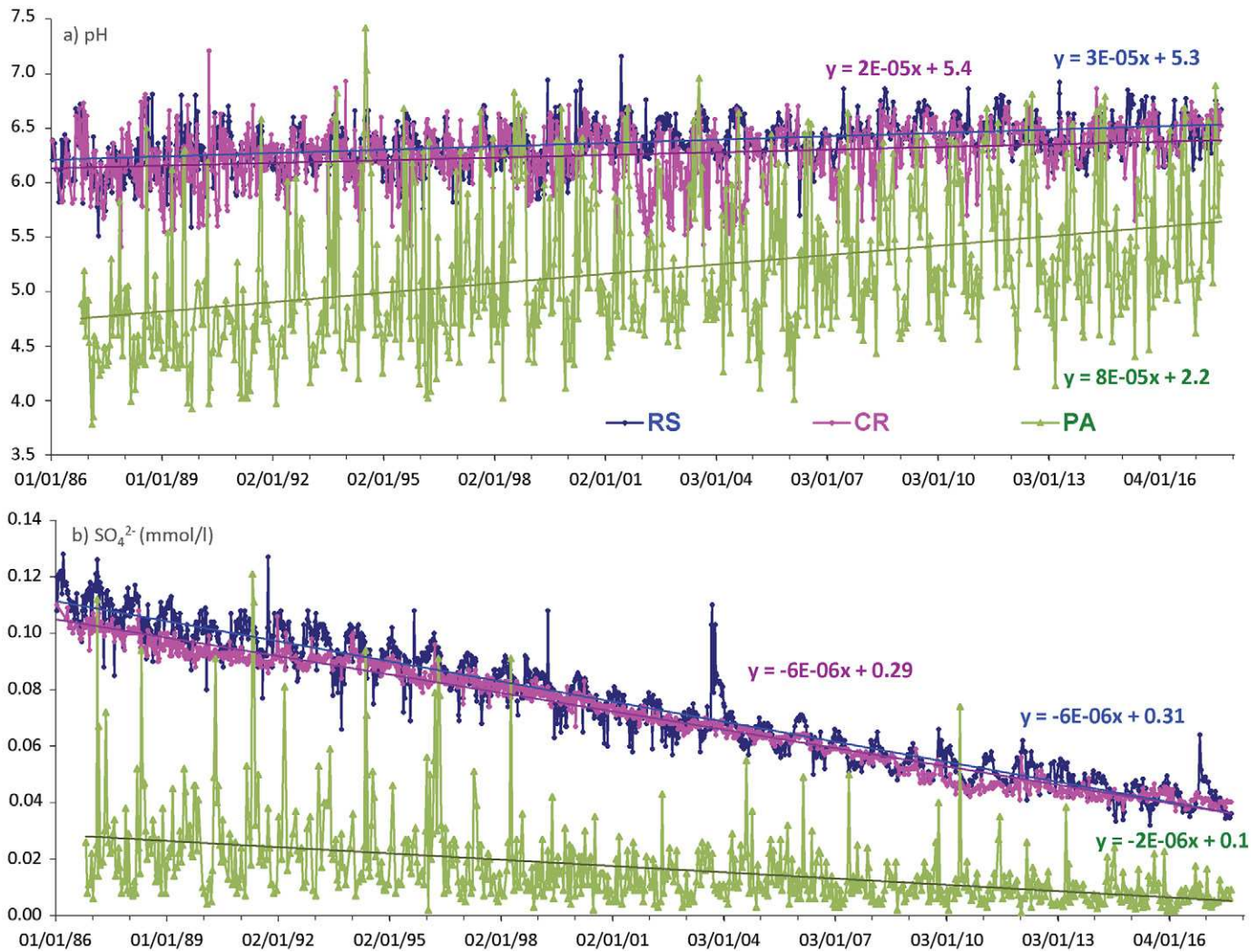


Fig. 5. Evolution of (a) pH and (b) sulfate concentrations for open field precipitation (PA), springs (CR), and stream outlets (RS) for 1986 to 2017 (Observatoire Hydro-Géochimique de l'Environnement, <http://ohge.unistra.fr>).

and the fractured bedrock. Temperature, moisture, and water pressure in the vadose zone have been monitored since 2010 under a representative spruce experimental stand located in the northern upper part of the catchment (Fig. 1) and across depths between 0 and 1.30 m in the subsurface. Five Watermark sensors, including a Campbell 257 for matrix potential and four Campbell 107 probes for temperature measurement, were installed with a sampling time step of 10 min. The collected data serve as conditioning information for modeling water motion through soils (Belfort et al., 2018; Biron, 1994).

### Impacts of Climate Change on Local Vegetation

Climate change impacts on water fluxes and the evolution of soil cover have been analyzed and predicted by Beaulieu et al. (2016), who used a dynamic vegetation model (LPJ) coupled with a three-dimensional hydrogeological model (MODFLOW). The LPJ model and MODFLOW were first calibrated from the observed vegetal cover distribution and from the water cycle during

the period 1987 to 2009 with a 1-mo time step. After calibration, the coupled models were used for long-term simulations (up to the year 2100) using climatic outputs from the Meteo-France climate model ARPEGE/Climate (Scenario A1B; IPCC, 2007). The climate model predicts an increase in temperature of 2.6°C, a rise in atmospheric CO<sub>2</sub> of 80%, and a mean decline in annual precipitation of 4.5% for the region where the Strengbach catchment is located. Subsequent effects on water fluxes remain limited (with values on the order of model uncertainties). In opposition, a significant change in vegetation distributions appears around the year 2085.

### Water Dynamics at the Catchment Scale

#### Water Budget

The mean annual discharge at the stream outlet was about 750 mm (24.5 L s<sup>-1</sup> km<sup>-2</sup>) during the period 1986 to 2015, with variations ranging from 494 to 1132 mm yr<sup>-1</sup> (Fig. 2a). The instantaneous outlet discharge varied between 1 and 400 L s<sup>-1</sup>

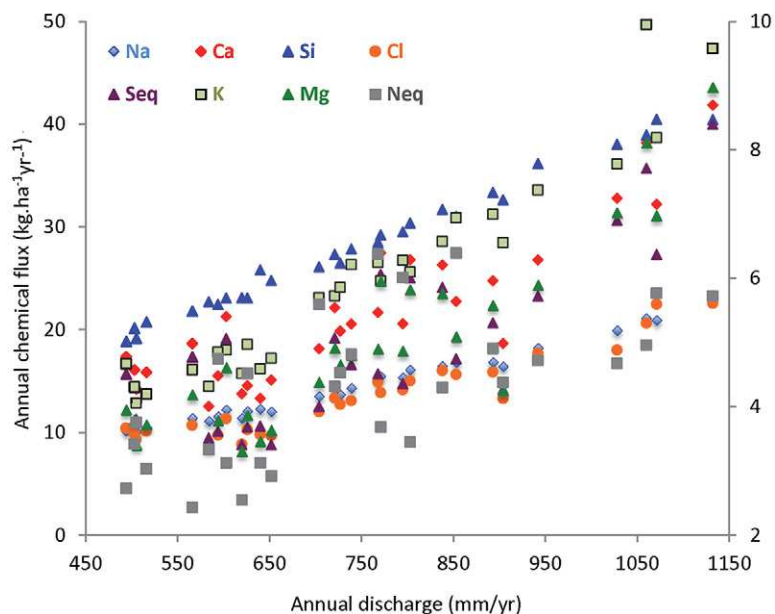


Fig. 6. Relationship between annual chemical flux at the outlet and annual discharge at the outlet. The scale on the right corresponds to K, Neq, and Mg values. Pearson coefficients are valued at 0.99, 0.96, 0.89, 0.88, 1.00, 0.65, 0.95, and 0.80 for Na, K, Mg, Ca, Si, Neq, Cl, and Seq, respectively.

during short periods, mainly associated with snow melting (Idir, 1998; Idir et al., 1999; OHGE data). The amount of water drawn from the spring collector has increased since 1986 (called  $S$ ) from roughly 15 to 20 mm yr<sup>-1</sup> from 1986 to 1989 to 100 mm yr<sup>-1</sup> from 2006 to 2015 (Fig. 2a) due to increasing demand. Total water export levels ( $Q + S$ ) ranged from 525 to 1148 mm yr<sup>-1</sup> from 1986 to 2015 (Fig. 2a).

These data reveal significant variations in cumulated annual discharge and rainfall with strong heterogeneities, illustrated by a factor of 2 between low and high values (Fig. 2a). The variation in the annual water flux exported from the catchment (outflow + drinking water) vs. annual rainfall shows a positive correlation ( $R^2 = 0.81$ ) (Fig. 2b), illustrating that at an interannual time scale, discharge is mainly controlled by rainfall levels. No significant long-term trend has been observed in terms of water input or output, but a periodicity of  $\sim 7$  yr has been recorded (Fig. 2a).

Solar radiation cycles may influence precipitation and hydrological seasonality, with periods between 6 to 7 and 11 yr depending on locations and the processes involved (Mauas et al., 2016; Prokoph et al., 2012).

From the analysis of 29 yr of records, the difference between annual inflows (precipitation) and water outflows traveling through the spring and stream has increased (Fig. 7). Several hypotheses regarding this trend may be considered. Assuming that the characteristic travel time of water in the catchment is roughly a few months (as estimated by the hydrological model), the increasing difference observed can be attributed to abstraction by evapotranspiration processes that have increased based on climate data but also on soil cover and forest management patterns (cutting or tree planting). In addition, the modifications of the rainfall regime and wind mass origins were suggested to explain the evolution of the Na/Cl ratio in atmospheric deposits (open field precipitation and throughfall) of the Strengbach catchment.

### Superficial and Deep Water

Stream and spring water from the catchment have been regularly but not continuously collected during various hydrological periods at high and low water levels in the stream to obtain a precise chemical and isotopic signature ( $\delta^{18}\text{O}$ ,  $\delta\text{D}$ , Sr, U, Li, B, Ca, dissolved inorganic C) (Aubert et al., 2002a, 2002b; Cenki-Tok et al., 2009; Cividini et al., 2010; Ladouche et al., 2001; Lemarchand et al., 2010; Pierret et al., 2014; Probst et al., 2000; Riotte and Chabaux, 1999; Schmitt et al., 2003). Springs denoted SG, ARG, RH, BH, CS<sub>3</sub>, and CS<sub>4</sub> are located on the northern slope, and springs CS<sub>1</sub>, CS<sub>2</sub>, SH, and RUZS emerge along the southern slope (Fig. 1). Spring RUZS exits in the wetland area covered with dense grass vegetation close to the outlet of the catchment (Fig. 1) (Idir et al., 1999; Ladouche et al., 2001; Probst et al., 1990a). This wet zone is equipped with four piezometers for recording groundwater levels and for water sampling (Fig. 1).

The Strengbach catchment drains different sources and streamlets with very different isotopic and geochemical signatures. Various parameters control the diversity of source characteristics. Of importance is the hydrothermal overprint of the granitic bedrock, which is stronger for granite of the south-facing slope than that of the north-facing slope, explaining systematic differences found in Sr isotopic and chemical ratios of water that drains these different slopes (Pierret et al., 2014). Water mean travel time is another important parameter to consider when explaining the chemical composition of springs emerging from a slope of the catchment, as clearly evidenced by reactive transport modeling exercises simulating spatial and temporal variations of Strengbach spring hydrochemistry (Ackerer et al., 2018). In addition, these studies show that the chemical composition of spring water at the Strengbach cannot be explained by a simple scenario of primary bedrock mineral weathering and secondary bedrock material precipitation because chemical signatures must also account for dissolution and precipitation

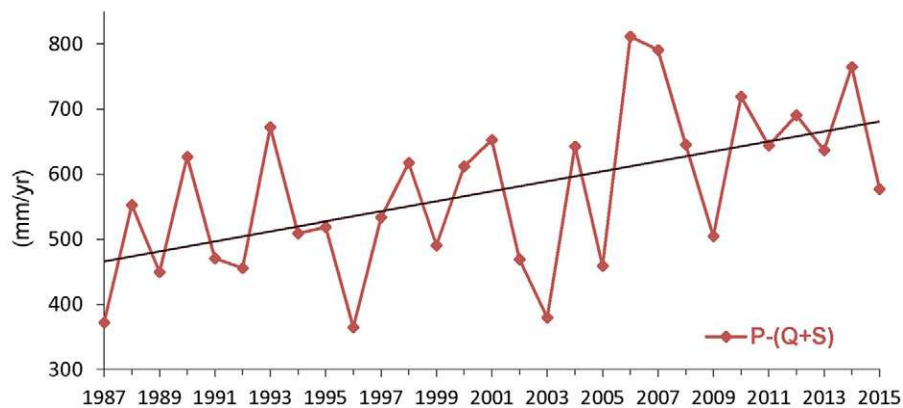


Fig. 7. Variation in the difference between annual water inflows (precipitation  $P$ ) and annual water outflows (discharge at the outlet  $Q$  + drinking water supply  $S$ ).

processes of secondary mineral phases, such as clay minerals (Godd ris et al., 2006). In summary, these results on the chemical signature of spring waters have helped to better constrain the water pathways, the chemical origin, the water–rock interaction processes, and the temporal geochemical variations occurring at the outlet (Aubert et al., 2002a; Cenko-Tok et al., 2009; Cividini et al., 2010; Idir et al., 1999; Ladouche et al., 2001; Lemarchand et al., 2010; Probst et al., 1992a, 1995b, 1995c, 2000; Riotte and Chabaux, 1999; Pierret et al., 2014; Viville et al., 2010).

Seven boreholes (Fig. 1), with two reaching 120 m in depth, have recently been drilled on either side of the watershed. Their drilling has allowed the exploration of the deep critical zone (deep flow and solute transport processes occurring over long travel times). Preliminary results (Chabaux et al., 2017) point out a clear geochemical typology of water based on water pathways (deep vs. hypodermic) in the substratum, with very different mean residence times evidenced for surface and deep-water circulation patterns.

### Solid Export at the Outlet

Effects of punctual storm and flood events have been investigated via the monitoring of the dissolved phase, suspended material, and bedload export occurring at the outlet and using chemicals (DOC, Si, and trace and major elements) and isotopic tracers ( $\delta^{13}\text{C}$ ,  $\delta^{18}\text{O}$ ,  $\delta^2\text{H}$ ;  $^{87}\text{Sr}/^{86}\text{Sr}$ ) (Amiotte-Suchet et al., 1999; Aubert et al., 2002a; Ladouche et al., 2001; Idir et al., 1999; Viville et al., 2012). A first evaluation of solid fluxes exported from the Strengbach catchment was performed via bimonthly sampling and measurement (Viville et al., 2012).

In December 2012, two automatic water samplers were set up at the outlet of the basin to evaluate the eventual bias in calculated solid fluxes generated from the bimonthly sampling. The idea was also to improve measures of suspended sediment (SS) fluxes and to account for the effects of very rapid flooding events and abrupt increases of flow rates in the stream. These two samplers enabled regular sampling over a 16-h time step and high-flow event sampling. The bedload flux was estimated bimonthly by measuring the volume of sediments accumulated in the outlet flume (Cotel et al., 2016). Characteristics of the Strengbach catchment (small surface area, low water levels, low SS concentrations, and mountainous winter climate conditions) required the adaptation of conventional

systems. All of these developments have allowed a better determination of SS fluxes exported from the catchment and setting up of optimal monitoring strategies, resulting in accurate estimations of suspended loads carried by the Strengbach stream at its outlet (Cotel et al., 2016).

During the 4 yr of the study, the annual bedload flux varied between 1.8 and 6.8  $\text{Mg km}^{-2}$ , indicating that solid export is dominated by SS transport in this catchment. Between 2004 and 2010, the mean annual weathering net flux (exports at the outlet corrected for atmospheric inputs) was measured at 2.0  $\text{Mg km}^{-2}$  for basic cations and at 2.9  $\text{Mg km}^{-2}$  for silica (Viville et al., 2012). Thus, in the Strengbach catchment, SS and bedload exports represent a significant proportion of global element exports. In such catchments, these solid fluxes cannot be neglected.

### Hydrological Modeling

The Strengbach catchment is currently targeted as a field system for assessing the capabilities of an integrated hydrological model (Jeannot et al., 2018; Maquin et al., 2017; Pan et al., 2015; Viville et al., 2006; Weill et al., 2017) to cope with water surface and subsurface flow processes occurring in mountainous systems with steep topography. The integrated model is grounded in an innovative low-dimensional subsurface representation that models both vadose and saturated zones using a single two-dimensional vertically integrated flow equation. This approach avoids resorting to the use of the three-dimensional Richards equation, which is computationally demanding and not easy to solve over a distorted computation mesh such as those used to describe complex geometries (including watersheds with steep slopes).

Regarding the Strengbach catchment, the structure of the subsurface compartment was framed on information available on the thickness of weathered material overlying the bedrock. Porous material is only between 1 and 8 m thick across the watershed (El Gh’Mari, 1995), producing a very shallow unconfined aquifer with little storage capacity (Fig. 8). It is assumed that this weathered cover is the main active component of the subsurface compartment, with more deeply connected fracture networks in the bedrock being discarded from any contributions to flows joining the surface river network. From this assumption, various flow simulations show that the annual water budget, as the effective

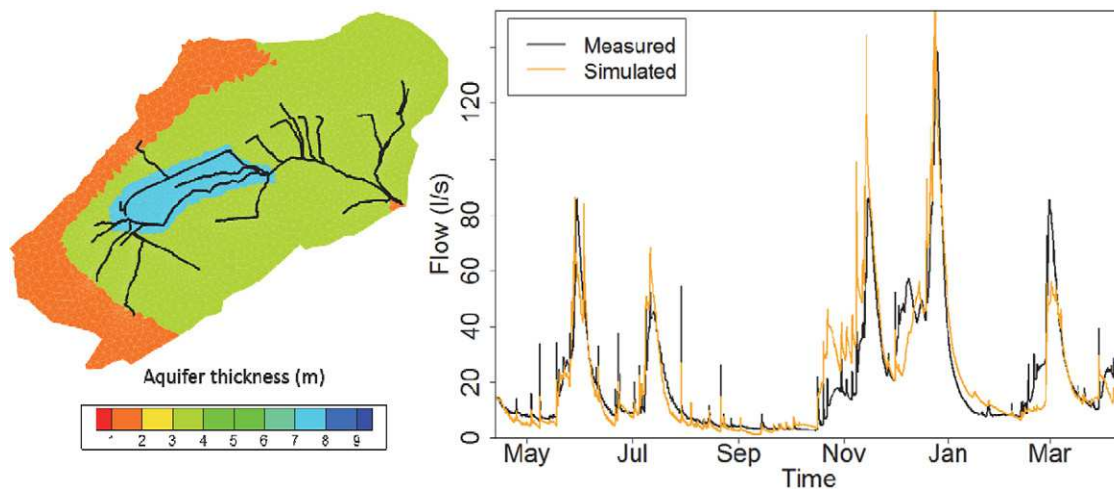


Fig. 8. Thicknesses of weathered materials constituting the shallow unconfined aquifer at the Strengbach catchment (left), and fitting observed flow rates (April 1996–March 1997) at the outlet of the catchment with simulations of flow within the watershed (right). The weak storage capacity of the system and the steep slopes result in very rapid and sharp responses of the stream to hydro-meteorological forcing. For November to March, the model accounts for the eventual melting of intermittent snow cover.

rainfall (including snowmelt) minus the flow rate at the outlet of the catchment, is very close to zero. There is no need for the use of heterogeneous hydrodynamic parameters (porosity = 0.08 and hydraulic conductivity =  $8 \times 10^{-5} \text{ m s}^{-1}$ ) to model the response at the outlet of the watershed (Fig. 8) in a system where infiltration rapidly reaches the river network and mainly via subsurface flow controlled by steep slopes. The watershed model is largely sensitive to the geometry of the unconfined shallow aquifer, for which we simulated various spatial distributions of thickness with good results in terms of fitting the outlet flow rate of the system. These valuable spatial distributions are conditioned by a maximum water storage of  $\sim 2.5 \times 10^5 \text{ m}^3$  (i.e., a subsurface compartment of 4 m, mean thickness over  $8 \times 10^5 \text{ m}^2$ , and a porosity of 0.08).

Water travel times have also been calculated for various periods by backtracking particles initially spread along the river network (Fig. 9). As particles are released from flowing (as opposed to dry) portions of the river network at a given time, the density of streamlines delineated from backtracked particles allows appraisal of areas contributing to the instantaneous flow rate of the river network. These areas are highly variable in time and follow the proportion of flowing vs. dry segments of the river network. Characteristic travel times range from 100 to 200 d, and in general the whole watershed contributes to runoff generation. In certain cases, typically in the summer months, water arriving in the river network at a given time derives from a fraction of the watershed but without important variations in characteristic travel times.

Given the lack of reliable information available on hydraulic head distributions for the subsurface that could have helped us retrieve transmissivity fields via inversion exercises, we turn toward hydro-geophysical measurements to improve our hydraulic understanding of the watershed. Further investigations should look at information provided by electrical resistivity profiles, proton magnetic resonance results, and high-resolution gravimetric anomalies.

## Structure and Characterization of the Surface and Subsurface

### General Approach

Due to questions raised by the characterization of subsurface heterogeneity and related flows, the Strengbach catchment has recently been investigated via various geophysical methods (especially using gravity, electricity [DC], and seismic and magnetic resonance sounding [MRS] methods), offering new insight into weathering structures at depths of 5 to 20 m and especially into differences between the northern and southern areas of the catchment. As an example, gravity measurements offer information on water budgets (Masson et al., 2012).

Several boreholes were drilled in 2012 and 2013, presenting new opportunities in borehole geophysics not only in terms of well logging but also regarding subsurface imaging between two boreholes or between a borehole and the surface. The use of ground-penetrating radar and seismic methods is limited to the ground surface due to the high density of tree roots and the complex topography, but these methods are essential in studying boreholes for characterizing fracture zones with depth as pathways of preferential flow. Borehole data may also serve as local conditioning of the spatial distributions of various parameters, such as the thickness of the weathered cover of the catchment, water storage capacity, transmissivity, and porosity of the saturated and vadose zones.

### Mapping of Water in Soil and Bedrock by Magnetic Resonance Sounding

Magnetic resonance sounding is distinguished from other geophysical tools used for groundwater investigations because it measures a signal directly generated from subsurface water molecules (e.g., Legchenko et al., 2004). An alternating current pulse is injected and cut in a wire loop on the ground surface, and an MRS

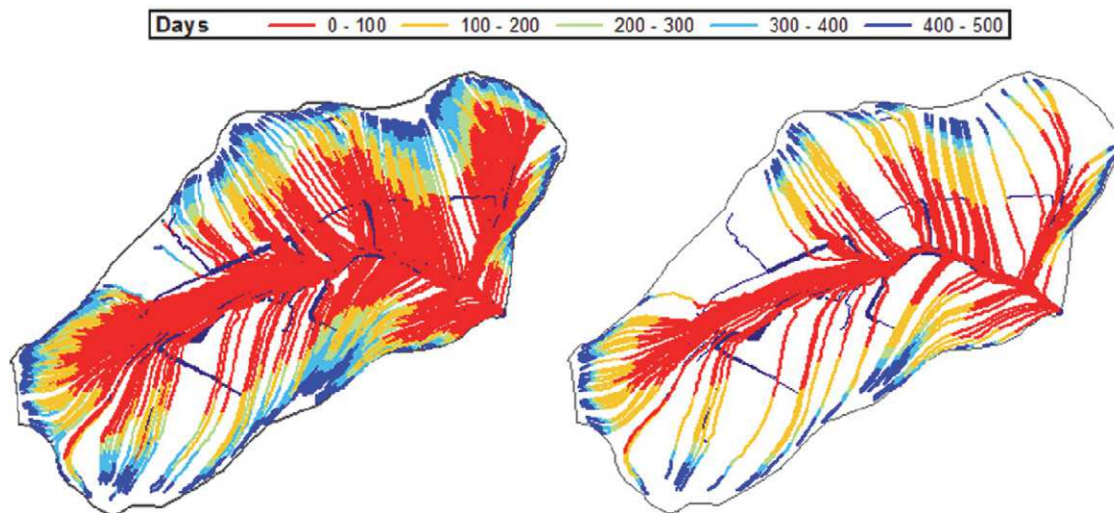


Fig. 9. Streamlines of the subsurface water contributing to the surface drainage network on 1 Mar. 2010 (left) and 1 July 2010 (right). The color scale denotes that a water particle reaching the river on a given date (e.g., 1 March) started its path along the streamline or passed at a given location along the streamline  $x$  days prior. The density of streamlines is associated with the flowing vs. dry fraction of the river network for a prescribed date.

signal is generated. Subsurface water is denoted by the amplitude of the signal: the more water molecules are energized below the loop, the higher the amplitude becomes. For instrumental reasons, MRS is only sensitive to free water and does not detect bound water, typically in clayey material. A magnetic resonance signal lasts longer when pore sizes or fissures are larger, thereby offering insight into permeability properties (Vouillamoz et al., 2014). Measurements made based on varied pulse magnitudes reveal the depth and thickness of water-saturated layers. In our case, MRS enabled us to estimate saprolite thickness, quantity of water in this horizon, and relative transmissivity or permeability from MRS signal decay.

Previous works on basement aquifers (e.g., Vouillamoz et al., 2014) have shown that MRS is poorly sensitive to water in fissured zones and that the MRS signal primarily derives from the weathered zone. Consequently, the bottom of the layer from which MRS water content was detected is interpreted as the transition between weathered and fissured zones. This depth was evaluated for the whole study area. Because MRS is an integrative method that is primarily sensitive to the number of water molecules present, there is a bias between water content and the thickness of saturated layers. To avoid uncertainties related to thickness due to this equivalence issue, it is more robust to consider the volume of water (i.e., the product of water content and thickness) (Legchenko et al., 2004). For the geological context of the Strengbach catchment, this robust indicator is particularly effective when used to map variations of the water-saturated weathered layer.

From 2011 to the present, four major MRS campaigns have been performed at the Strengbach catchment. The first determined feasibility, the next two involved extensive mapping (Boucher et al., 2015), and a last campaign in 2014 collected accurate decay measurements. A total of 56 soundings were performed on 23 locations.

Results allow mapping the volume of water in the weathered zone (Fig. 10). This map (Fig. 10) correlates with the major

geological units. On the northern crest characterized by gneiss (Fig. 3), the weathered zone does not exceed 12 m according to Borehole F6 (Fig. 1). It contains clayey material with mainly bound water, consistent with the lack of detected MRS signals. The northern slope includes fissured granite with low thickness of the weathered zone (<10 m) due to the steepness of the slope. The estimated MRS water volume is intermediate ( $0.20\text{--}0.35\text{ m}^3\text{ m}^{-2}$ ) and may support a steady flow over time. In addition, these values are similar to the free water column estimated from hydrological numerical models (8% of porosity and 4 m of thickness correspond to  $0.32\text{ m}^3\text{ m}^{-2}$ ).

In the central colluvium zone, which is a mostly flat area, the estimated depth of the weathered zone reaches 20 m. The MRS water volume (up to  $0.75\text{ m}^3\text{ m}^{-2}$ ) is high and the MRS decay time is long, suggesting a higher permeability. Across the southern slope, low groundwater volumes are mapped ( $<0.25\text{ m}^3\text{ m}^{-2}$  according to MRS results), which is logical given that this area is less altered and fissured than the northern slope. From decay time and estimations of the weathered zone thicknesses, preliminary MRS results for this watershed reveal relative transmissivity or permeability values between  $5 \times 10^{-6}$  and  $8 \times 10^{-5}\text{ m}^2\text{ s}^{-1}$ .

## Conclusions and Perspectives

The OHGE site is used for multidisciplinary research and has been subjected to a diversity of scientific approaches. Three main future scientific directions are outlined below. Very recent equipment, still in its “breaking-in” period, should also provide new high-frequency data on the geochemistry of stream water and on the structure of subsurface water bodies.

### Deep Critical Zone Exploration

Recent studies have highlighted the major role played by deep horizons in the signature of water. This critical challenge must be

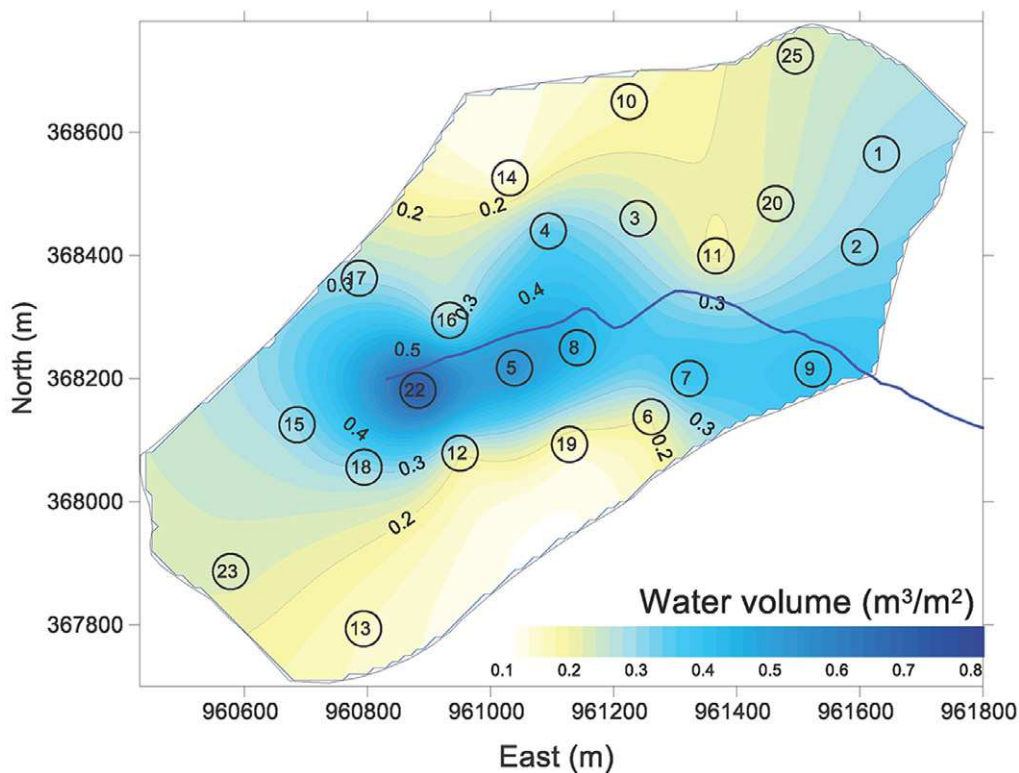


Fig. 10. Map of the magnetic resonance sounding water column (water volume per surface unit), which is an indicator of saturated alterite thickness, interpolated across the whole catchment.

addressed by examining recently drilled boreholes and drill cores for the exploration of deep areas of the critical zone in the catchment. Structural, hydrological, or mineralogical knowledge of the deep zone is generally explored through indirect geophysical studies. In addition, borehole (Fig. 1) and drill core studies provide local but direct in situ measurements, which are a perfect way to calibrate geophysical inversion and data as well as numerical models.

### Role of Vegetation in Water and Solute Transfer

Plants directly influence the water balance, atmospheric deposition of elements, turnover of soil organic matter, weathering rates, and the chemistry of solutions and soil exchange capacity through the uptake and release of nutrients. The impact of vegetation on the chemical balance must be characterized by ecological studies as (i) spatial litterfall evaluations, (ii) stand inventories, (iii) destructive samplings of distributed trees, (iv) fittings of total above-ground mineral mass allometric equations, and (v) quantifications of stand biomass and nutrient content per hectare, whereby net uptake is computed from the difference in immobilized nutrients observed between two consecutive years.

### Laboratory Experimentation to Better Understand Water and Mineral Processes

Primary mineral weathering was not able to fully explain the chemical signature of river waters where the role of secondary phases and especially the dissolution/precipitation of clay minerals have been explored and proposed (Ackerer et al., 2018;

El Gh'Mari, 1995; Lemarchand et al., 2010, 2012; Pierret et al., 2014). Resorting to reactive transport models for simulating the evolution of the fluid chemistry could be useful provided that thermodynamic data and kinetics laws compatible with processes occurring in natural systems are available (Goddéris et al., 2006). Thus, specific laboratory experiments based on the methodological development of isotopic tracer data via reactive transport modeling should outline important new constraints on actual chemical laws to use in codes. In the same vein, weathering experiments based on representative and well-described facies could be used to better measure the solubility products and accurate kinetics laws for primary and secondary minerals by testing values for reproducing the chemical composition of fluids resulting from interactions with materials, which cannot be achieved at the field scale.

### Recent Equipment

In the last 2 yr, new equipment has been installed across the Strengbach catchment. These tools have not provided exploitable data because they are still in their calibration, “breaking-in” period. The observatory benefited from the National CRITEX action targeting innovative equipment and measurement techniques for watersheds to deploy a so-called RiverLab (CRITEX equipment; Fig. 1). It allows the high-frequency analysis (pH, conductivity, alkalinity, silica, DOC, Na, K, Mg, Ca, NO<sub>3</sub>, SO<sub>4</sub>, and Cl) of water at the outlet of the catchment (RS site). When readily available, measurements are expected to provide insights on day–night cycles and on seasonal changes and to capture the dynamics of very transient

events such as those associated with flooding and snowmelt periods (Floury et al., 2017).

CRITEX also funded a new gravimeter Igrav, which has been installed at the top of the catchment (Fig. 1) to monitor the temporal variations in the Earth's gravity field and its fluctuation due to variations in the structure of subsurface water bodies. These data, combined with microgravity measurements over time and space at the catchment scale, should image the water recharge dynamics in the subsurface.

These ongoing research activities and new data should foster the emergence of new knowledge about the structure of the catchment and the water and solute dynamics. The ideas are (i) to improve on and develop tools of subsurface geophysics to manage complex topographies and vegetation, the objective being to create a three-dimensional image of the subsurface that includes different layers of geometry and associated parameters (porosity, permeability, and water content); (ii) to improve the understanding of water-rock processes and of the temporal evolution of the ionic composition of the superficial water through better characterization of reactive phases, including deep zone and fractures; (iii) to further develop the original model for saturated-unsaturated flows with surface water exchange by modeling the impacts of the vegetation and water-rock interactions; and (iv) to calibrate the bio-geohydrological model with long-term data series and to predict the future evolution of the ecosystem.

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