Geochemistry and geophysics of active volcanic lakes: an introduction

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Volcanoes sometimes host a lake at the Earth's surface. These lakes are the surface expressions of a reservoir, often called a hydrothermal system, in highly fractured, permeable and porous media where fluids circulate (Fig. 1). The existence of a volcanic lake depends on a balance between: (1) a seal at the bottom of the lake to prevent water seepage; (2) abundant meteorological precipitation; (3) a sustained input of volcanic fluids; and (4) a limited heat input to avoid drying out of the lake by evaporation (Pasternack & Varekamp 1997; Rouwet & Tassi 2011).

When these conditions are met, volcanic lakes become excellent monitoring targets. First, they integrate the heat flux discharged by an underlying magma body. An increase in magmatic activity can therefore be detected remotely or directly by probing the lake temperature. Second, they condense some volcanic gases, leading to a mixture of dissolved chemical compounds. These solutes are preserved in solution for a long time compared with the gases emitted into the atmosphere through fumaroles. Because they trap volcanic heat and gases, they are excellent tools that can provide additional information about the status of a volcano and the hazards related to a volcanic lake (Rouwet & Tassi 2011; Manville 2015; Rouwet & Morrissey 2015).

Depending on their volume, volcanic lakes can, however, filter and delay the surface expressions of volcanic unrest. Despite only 8% of reported eruptions worldwide having occurred in a subaqueous setting, they have caused 20% of fatalities (Mastin & Witter 2000). One of the most dramatic hazards is a phreatic eruption, which arises from the sudden input of fluids and energy from a magmatic source into a more superficial aquifer (Rouwet & Morrissey 2015). Hydrothermal explosions involve water close to its boiling temperature and are also generated at shallow depths, but are not triggered by an input of mass or energy derived directly from the magma (Montanaro *et al.* 2016). These eruptions, sometimes termed steam-driven eruptions (Montanaro *et al.* 2016), are often more violent than magmatic eruptions and can have ejection velocities $>130\ m\ s^{-1}$ (Rouwet & Morrissey 2015); they may culminate as phreato-magmatic eruptions. Both types of eruption can be accompanied by base surges after column collapse, tsunamis or seiches (Mastin & Witter 2000). They can also generate destructive lahars, which can travel up to tens of kilometres down the slope of the volcano (Manville 2015). Another spectacular and well-studied hazard is limnic gas bursts releasing large amounts of CO_2 (Kusakabe 2015). An indirect impact is the prolonged release of acidic gas species (HC1, SO_2 and HF). Water contamination and wall rock failure after seepage also occur in the direct vicinity of volcanic lake environments (van Hinsberg *et al.* 2010; Delmelle *et al.* 2015).

This volume brings together scientific papers on volcanic lakes, including studies of their structure, hydrogeological modelling, long-term multidisciplinary monitoring and a number of innovative methods of sampling, data acquisition and *in situ* or laboratory-based experiments. Several papers challenge long-established paradigms and introduce new concepts and terminologies. This collection of papers is a useful reference for researchers dealing with volcanic lakes and more generally with hydrothermal systems, phreatic/hydrothermal eruptions and wet volcanoes.

History and state of the art

In the history of volcanology, knowledge about a specific type of phenomena has often undergone an abrupt acceleration only after a catastrophic event (D. Rouwet, https://iavcei-cvl.org/). Investigations on volcanic lakes made dramatic progress

From: Ohba, T., Capaccioni, B. & Caudron, C. (eds) 2017. *Geochemistry and Geophysics of Active Volcanic Lakes*. Geological Society, London, Special Publications, **437**, 1–8.
First published online April 20, 2017, https://doi.org/10.1144/SP437.18

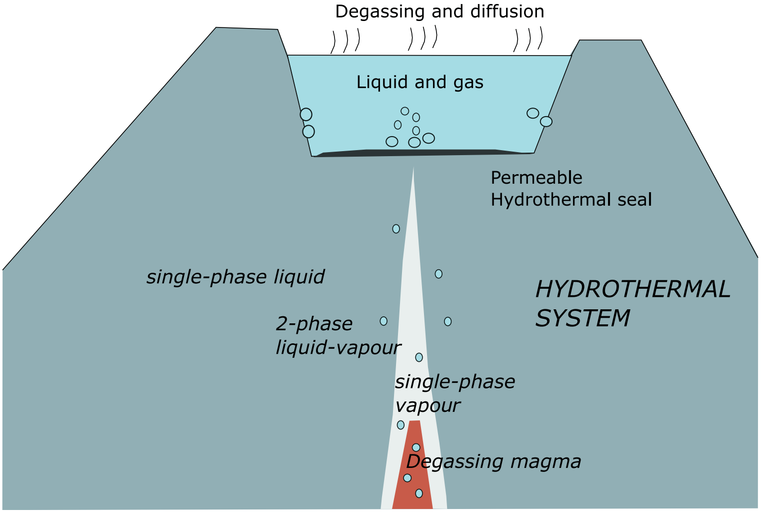

Fig. 1. Schematic cross-section of a typical high-activity volcanic lake and its main features.

after the 21 August 1986 degassing event at Lake Nyos in Cameroon, which killed more than 1800 people. At that time, it became clear that volcanic lakes may constitute a danger in themselves, based on their limnological characteristics and type of interactions with the volcanic system. This has resulted in new investigations and monitoring actions to mitigate the risk inherent in the existence of a crater lake.

In the 1990s, the volcanology community realized that monitoring volcanic lakes could provide additional information about changes in the activity of degassing volcanoes and hydrothermal systems. Research started and improved the forecasting of volcanic eruptions. By the end of the century, different disciplines – including volcanology, geochemistry, hydrology, limnology, microbiology and economic geology – were being used to study intertwined aspects of volcanic lakes.

Since the 2000s, the community studying these ‘blue windows’ (Christenson *et al.* 2015) became broader, with further geophysical and modelling efforts. Volcanic lakes are now not only investigated using state of the art thermal and chemical approaches, but also through geophysical surveys, biological analyses, numerical modelling and multidisciplinary efforts. These 30-year long efforts have led to a better understanding of these environments and a more comprehensive classification.

There are currently 474 lakes listed in the new database of volcanic lakes, VOLADA (Rouwet *et al.* 2014), grouped into six classes (Fig. 2) based on their volcanic activity (following Pasternack & Varekamp 1997): (1) erupting; (2) peak activity; (3) high activity; (4) medium activity; (5) low activity; and (6) no activity. The essential subdivision between the two first classes and the other four relies on the balance between input and output fluxes. The high-activity acidic lakes are further sub-divided depending on their total dissolved solutes (TDS) content and temperature: class 3a, hot, TDS = 150–250 g l^{-1}, $T = 35–45°C$; class 3b, cool, TDS = 40–150 g l^{-1}, $T = 20–35°C$. The medium activity lakes are less acidic and often oxidized (class 4a). Some reduced acid–saline lakes exist (class 4b). Low activity lakes basically consist of steam-heated pools (class 5a) or CO_2-dominated lakes (class 5b). Dissolved CO_2 can reach the surface in shallow lakes (class 5bl) or remain trapped in the hypolimnion forming a stratified lake, also called a Nyos-type lake (class 5b2). Varekamp *et al.* (2000) introduced two physical–chemical classifications based on the sulphate + chloride concentrations and pH values and the percentage residual acidity.

Another physical classification is based on the outlet dynamics: closed lakes, without surface outlets with a variable volume; and open lakes with an outlet and an overall constant volume (Varekamp 2015).

One crucial aspect concerns the residence time (RT), which is defined by the lake volume and the input (or output) fluxes ($RT = V/Q_{input}$); the larger the lake volume and the lower the input fluxes, the longer the RT will be (months to years). Smaller lakes affected by a high fluid input will result in a short RT (weeks to months (Varekamp 2003; Taran & Rouwet 2008; Rouwet *et al.* 2014). The RT therefore determines the lake’s sensitivity to potential changes caused by external processes and

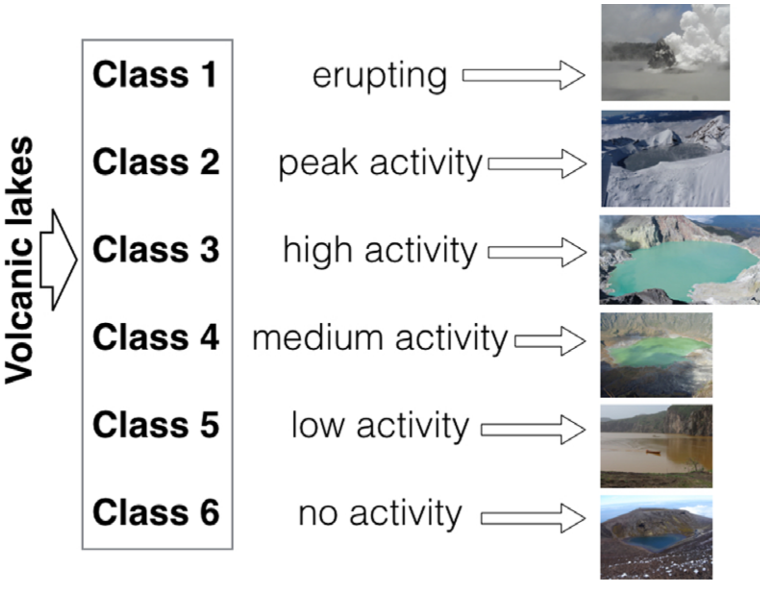

Fig. 2. Classification of volcanic lakes. The different classes of volcanic lakes, simplified from Pasternack & Varekamp (1997) and modified from Rouwet *et al.* (2014), are illustrated with pictures of typical lakes. From top to bottom, these are: Voui, Vanuatu by K. Németh; Ruapehu, New Zealand by V. Chiarini; Kawah Ijen, Indonesia by D. Rouwet; El Chichón, Mexico by D. Rouwet; Nyos, Cameroon by D. Rouwet; and Tristan da Cunha by A. Hicks.

the frequency of monitoring required (Rouwet *et al.* 2014). Recent efforts are further challenging and investigating this crucial aspect, which should drive monitoring strategies (Shinohara *et al.* 2015; Tamburello *et al.* 2015; De Moor *et al.* 2016; Caudron *et al*. 2017). Other studies are presented in this volume and are now briefly introduced.

This volume

Acidic lakes

Several papers concern the Kawah Ijen volcanic lake (East Java, Indonesia). This lake is emblematic and is the largest hyper-acidic volcanic lake on Earth. The volcano-hydrothermal system was studied from a geochemical perspective in the 1990s and the early 2000s (Delmelle & Bernard 1994; Delmelle *et al.* 2000; Takano *et al.* 2004). Caudron *et al.* (2015*a*, *b*, 2017) provide further insights into its volcanic activity using geophysical instruments combined with volcanic lake parameters.

Van Hinsberg *et al.* (2015) constrain the flux emitted by the volcano to the environment for most elements of the periodic table. The compositional signature of emissions is similar to the major volcanic emitters, but the element fluxes are smaller. Importantly, the aqueous flux (i.e. the seepage of volcano-hydrothermal fluids and volcano-influenced groundwater) is at least as important as the gaseous flux. It is therefore of paramount importance to consider the aqueous flux to estimate the impact of volcanoes on their environment and the contribution of volcano-hydrothermal systems to global cycling.

Gunawan *et al.* (2016) report the results from the highly focused international wet volcano workshop that took place at Kawah Ijen in September 2014. Wet volcanoes are volcanoes hosting crater lakes and/or extensive hydrothermal systems. The study provides a detailed description of the different technologies deployed during fieldwork. Measurements include seismic and acoustic recordings, *in situ* and remote thermal measurements of both high-temperature fumaroles and lake water and also multi-gas analyser, differential optical absorption spectroscopy and laser spectroscopy measurements from instruments deployed around and within the crater. All the data and results are combined to build a conceptual model of the Kawah Ijen volcano. This study highlights the similarity of the Kawah Ijen volcano and Ruapehu volcanoes (New Zealand) and improves our understanding of wet volcanoes.

Further insights into the Kawah Ijen volcano are provided by **Caudron *et al.* (2016)**, who present the first comprehensive geophysical investigation. The hydrogeological system surrounding the Kawah Ijen crater lake consists of three different and independent aquifers. The volcanic lake is surprisingly heterogeneous, as illustrated by spectacular subaqueous degassing. These results provide new fundamental knowledge on the hydrothermal system of Kawah Ijen volcano, which may be useful in volcanic surveillance programmes.

Poás volcano (Costa Rica) hosts another hyperacidic crater lake of great importance and is one of the most dynamic wet volcanoes. After 12 years of quiescence, phreatic activities resumed in March 2006 and continue until the day of writing. **Rouwet *et al.* (2016)** describe and analyse the geochemical fluctuations during the period 2006–10. No systematic relationship between geochemical concentrations and phreatic activity exists and a dynamic fluid recycling between the lake and the underlying magmatic–hydrothermal system is suggested. The authors also report phenomena such as the outgassing of chlorine from the lake surface, describe the Laguna Caliente as an 'open fumarole' and provide a stimulating new paradigm: the water chemistry reflects a transient and fast process of gas flushing through the lake, rather than slow fluid inputs/outputs. It therefore discredits water chemistry as an efficient monitoring tool within the classic monitoring frequency.

Capaccioni *et al.* (2016) challenge the assumption that HCl behaves conservatively when dissolved in extremely acidic water. Their laboratory experiments highlight HCl degassing from extremely acidic waters. Two main outcomes arise from this study: (1) Cl^- does not behave conservatively once dissolved in extremely acidic systems; and (2) the presence of very acidic gas species in fumaroles can be associated with a feeding system dominated by liquid water.

Twenty years of geochemical and temperature data provide insights into the volcano-hydrothermal system of Copahue volcano (Argentina) and reveal fundamental changes in the system preceding or accompanying magmatic and phreatic eruptions (**Agusto *et al.* 2016**). The integration of geophysical data allows the development of a conceptual model of this volcanic system and the identification of precursory signals for eruptive activity.

By using an interesting combination of water chemistry and echo-sounding techniques, **Hernández *et al.* (2017)** investigated the less acidic (*c.* pH 2–3), but dangerous, Taal volcanic lake (Philippines). They report changes in the chemical composition and increases in CO_2 emissions associated with volcano-seismic unrest in April 2010–June 2011, attributed to deep fluid injections. They interpret the inflationary periods as due to the faulting of an impermeable cap rock sealing the deeper hydrothermal reservoir in response to degassing and convective movements in the underlying Taal magma chamber.

Rouwet & Iorio (2016) describe two seemingly unrelated phenomena: clays floating on the surface of the El Chichón crater lake resembling Suminagashi patterns, the traditional paper marbling technique. Clays floating on the lake are subject to Marangoni flow when the lake becomes very shallow (a few centimetres) near the lake shore. The flat lake bottom and the dynamic alternation between retreat and incurrence of the lake shore result in precipitation of the clay patterns, preserving the Suminagashi-like designs in the lake sediments. The recognition of similar patterns in a sedimentary record could indicate the presence of a shallow sedimentary environment in the past, possibly pointing to a heated crater lake.

Neutral lakes

Six papers deal with lakes Nyos and Monoun (Cameroon), the iconic volcanic lakes where limnic eruptions, driven by the outgassing of CO_2, occurred in 1986 and 1984, respectively.

Kozono *et al.* (2016) developed a numerical model of CO_2 degassing from the lake waters dedicated to the problem of CO_2 saturation at mid-depths that could lead to a limnic eruption. They modelled the effects of changes in the CO_2 profiles due to the entrance of CO_2-unsaturated fluids from the lake bottom. They emphasize the future possibility of a limnic eruption caused by violent degassing at mid-depth. Their model allows an estimate of CO_2 concentrations at the lake bottom from the heights of fountains observed at the lake surface.

Ohba *et al.* (2015) studied the annual variability of the total amount of CO_2 stored within Lake Nyos since 2011. They point out the effects of the installation of the degassing pipes that led to a rapid decrease in the amount of CO_2 stored within Lake Nyos.

Saiki *et al.* (2016) analyse the vertical sound speed profiles (see **Sanemasa *et al.* 2015** for methodology) measured at lakes Nyos and Monoun in 2012 and 2014. They found a significant linear correlation between the total CO_2 concentrations and the vertical sound speed in the lake water. By performing multi-point measurements, they observed horizontal variations at both lakes. Their results show a stable vertical stratification at Lake Nyos with a decrease in the total CO_2 from 2012 to 2014 and distinct CO_2 concentrations at Lake Monoun due to the presence of three different basins. An increase in CO_2 was further detected in the main basin from 2012 to 2014.

Sanemasa *et al.* (2015) present a new method of measuring dissolved CO_2 concentrations using sound speed and electrical conductivity. It has been tested at lakes Nyos and Monoun and may improve the forecasting of future limnic eruptions as a result of the great spatial and temporal resolutions compared with traditional techniques.

Yoshida *et al.* (2015) describe the results of the installation of a new deep water removal system, operated by a pumping system powered by solar energy, following the loss of capability of the previous systems due to reduced CO_2 partial pressures.

Deep anoxic conditions at Lake Nyos cause the dissolution of iron as Fe^{2+} and the precipitation of siderite. The water transported upwards by degassing pipes undergoes a rapid increase in redox potential, causing the precipitation of iron as $Fe(OH)_3$ and leading to the surface water becoming reddish. **Ozawa *et al.* (2016)** investigate iron dissolution/precipitation using *in situ* experiments at Lake Nyos and laboratory analyses. They describe the dissolution process for siderite at depths <50 m and precipitation at greater depths. By applying interferometric techniques, they highlight the change in the rate of siderite precipitation along the vertical profile, revealing a high increase close to the lake bottom.

Zimmer *et al.* (2015) present another innovative analytical method based on a gas membrane sensor to quantify the concentrations of CO_2 and CH_4 gases in water columns from volcanic lakes. They tested this new set-up at the Monticchio Grande and Piccolo (Mt Vulture, Italy) and Pavin (Massif Central, France) lakes and successfully compare the results with the more conventional single-hose method.

Other neutral lakes were investigated through two studies. **Melián *et al.* (2016)** report the first detailed study of volcanic lakes of São Miguel island, Sete Cidades, Fogo and Furnas, located in the Azores archipelago (Portugal). By combining a floating accumulation chamber and echo-sounding measurements, they detected low surface CO_2 degassing, but dense and moderate subaqueous emissions. Strong dissolution occurs in the water column due to the high pH.

Lefkowitz *et al.* (2016) present the general geochemical features of the unique twin crater lakes on Newberry Volcano Oregon, USA. The lakes are both carbonate-rich, but are different in chemical composition, morphology and sediment composition, although separated by only a narrow volcanic ridge. East Lake shows the features of a terminal lake with a gaseous geothermal input, whereas Paulina Lake has subaqueous hot springs and an outlet. Sudden CO_2 degassing presents a volcanic hazard to be considered in East Lake, as well as the presence of highly toxic components.

Future perspectives

This Special Publication comes at a challenging and exciting time for the volcanic lake community. Multi-disciplinary efforts are becoming increasingly popular, as shown in this volume. They are essential in characterizing a volcano-hydrothermal system. The recent progress made in instrument and storage capabilities has allowed sampling at a much better time resolution. For example, multi-gas analysis or thermal imagery can provide time series with a sampling frequency of a few seconds (e.g. **Gunawan *et al.* 2016**), approaching the sampling rate of most common monitoring instruments, such as seismometers or the global positioning system. This allows the sensing of new processes by integrating a wide range of multi-disciplinary data. It will therefore improve our understanding and forecasting of volcano-hydrothermal systems. However, major questions need to be answered before declaring the usefulness of these techniques in volcano monitoring. Probabilistic methods are also becoming increasingly popular in the field of volcanic lake studies and could facilitate unrest and eruption forecasting (e.g. Tonini *et al.* 2016).

Recent studies have also questioned the traditional monitoring strategies at active crater lakes. The concept of residence time is the key to elaborating an efficient volcano monitoring scheme. The chemical and thermal homogeneity of a lake over time also need to be assessed to obtain representative measurements and valuable monitoring data, even at the most dynamic and active systems (**Rouwet *et al.* 2016**; Caudron *et al.* 2017). This is also crucial in the correct interpretation of remote temperature data from forward-looking infrared or satellite imagery.

A burst of new studies and projects are dealing with active crater lakes following the 2014 Ontake phreatic eruption, which caused the deaths of more than 50 climbers. The filtering and delaying capacity of volcanoes hosting well-developed hydrothermal systems make them very hard to monitor, but the number of variables that can be monitored make them attractive to better understand the generation of these eruptions. Active crater lakes also condense fluids rising from below. They are therefore very sensitive to sudden changes in pressure (fluid injections), which increase the likelihood of a steam-driven eruption (Manville 2015). Sealing has been proposed as the driving force of some phreatic eruptions that have occurred at several different volcanic lakes (Christenson *et al.* 2010; **Agusto *et al.* 2016**; **Caudron *et al.* 2016**; **Rouwet *et al.* 2016**). Several experiments are characterizing the influence of the type and degree of alteration or the host rock lithology on the generation of phreatic eruptions (e.g. Mayer *et al.* 2015) and the partitioning of energy into thermal, mechanical or seismic energy, the fragmentation energy associated with these volcanic events (e.g. Montanaro *et al.* 2016).

This volume shows that the risk at lakes Nyos and Monoun has been reduced by eliminating CO_2 gas dissolved in deep water by 'self-gas lift' as a symptomatic treatment. However, the removal of gas by this method is not perfect, because self-gas lift does not operate when the dissolved gas concentration falls below a certain threshold, preventing the complete artificial degassing of gas-loaded lakes. Such a situation is occurring in Lake Monoun. On Lake Nyos, self-gas lift continues in 2016, but will stop within 10 years. Thereafter, the dissolved gas concentration in the deep water may rise again and observations of the deep waters of the lake will be needed. Local research institutions are responsible for the monitoring of this situation, co-operating with researchers from other countries.

We thank Dmitri Rouwet for reviewing this paper, sharing his thoughts and providing the classification figure. We also thank P.T. Leat for his sound and positive review. C. Caudron is funded through a Chargé de Recherches FNRS postdoctoral grant.

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