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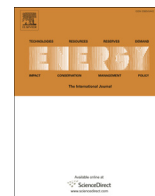
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Energy efficiency and sustainability assessment for methane harvesting from Lake Kivu



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ABSTRACT

Lake Kivu is a great environmental and economic resource in Rwanda. Its deep-water methane reservoir can help the country to narrow its energy supply gap. However, mishandling of the lake could lead to devastating consequences, from potable water contamination to limnic eruption. To evaluate the lake's potential for energy harvesting, we have developed a numerical model and validated it experimentally. Based on this model, we propose an optimal methane harvesting strategy. The harvesting efficiency improvement is from 4 to 6% relative to the alternatives. While seemingly insignificant, a 1% improvement of harvesting efficiency extends the operational time of a gas power plant by ~5%. With these improvements, the lake will sustainably supply 100 MW of electricity for up to 100 years. Potential CO₂ emissions are negligible in comparison with the low-emitting developed countries. We conclude that forestry and agroforestry can mitigate CO₂ emissions and reduce currently widespread deforestation. The degassed water after methane extraction poses another environmental concern. It must be reinjected at the depth of 190–250 m to minimize the environmental impacts on the lake and allow for continuous methane harvesting.

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1. Introduction

Lake Kivu is a large lake in East Africa, located between Rwanda and the Democratic Republic of Congo in an active volcanic region of the East African Rift System [1,2]. A special characteristic of this lake is the existence of a gas reservoir rich in carbon dioxide (CO₂) and methane (CH₄), which are generated by the volcanic activity near the lake and the anaerobic bacterial activity at the bottom of the lake, respectively. The dissolved gas volumes are estimated at 60 billion m³ of CH₄ and 285–300 billion m³ of CO₂ [3–6].

The unique architecture of Lake Kivu has been the focus of several studies. Gas concentration measurements date back to 1937, with several campaigns in the 50s, 70s and 90s [3,7–9]. In 2005, 4 reported that the concentration of CH₄ had increased by 20%, and CO₂ increased by 10% since the 70s. However, the data were still insufficient and tainted with high uncertainty [6]. The most recent measurements in 2018, suggest constant aqueous concentrations of CH₄ and CO₂ [10].

Chemical profiles of Lake Kivu water have been described in

several studies [4,11,12]. The water temperature, concentration gradients, dissolved gases, and lateral inflow result in a stratified structure and formation of chemoclines (*i.e.*, layers with strong salinity gradients). Mixing between the chemocline layers is limited by a double diffusion-convection mechanism [4]. Double-diffusion convection and the lateral inflow have been raising the chemoclines and could induce a limnic eruption [13]. Numerical methods were used to reproduce the double-diffusive interfaces in the lake [14]. Subaquatic inflow was shown to play an important role in the formation of a stratified water structure [15]. The most recent studies aim to explain how the double-diffusion convection affects the heat and mass balances of lake water [16]. Hirslund [17,18] introduced a novel approach to account for heat and mass transport in the lake and explain the formation of the stratified lake structure. In a recent study, Brenbold et al. [10] suggested a low but nonzero probability of limnic eruption due to natural causes.

Rwanda is a sub-Saharan country that is developing at a fast rate. In the last decade, the gross domestic product (GDP) per capita grew by an average of 5% per annum [19]. Electricity generation sector is crucial to the development of any nation. Rwanda has an installed capacity of 218 MW, with the energy mix composed of hydro, thermal (diesel and heavy fuels), methane, peat, and solar

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[20,21]. In 2016, only 51% of households had access to electricity, but the government plans to achieve a 100% access by 2024 [21]. This is an ambitious target for a country where 80% of the population is rural. It requires massive investment in the expansion of the national grid and/or implementation of the off-grid systems. Universal access to electricity is an essential step for the development of Rwanda. The main challenges are to strike a balance between the power supply and demand and provide electricity at an affordable price [20].

Methane dissolved in Lake Kivu can boost power generation in Rwanda. Rwanda's government estimates that the methane reserves are sufficient to generate 280–360 MW for over 50 years [21]. There are already “methane-to-power” projects such as KivuWatt that aims to install a nominal capacity of 100 MW with a current installed capacity of 26.4 MW. Kibuye Power (KP1) with 3.6 MW of capacity was recently acquired by Symbion Power that plans to install additional 50 MW [22].

Methane, the most abundant hydrocarbon gas on the Earth, plays an important role in atmospheric chemistry and climate [23]. However, it also contributes to the greenhouse effect. On the mass basis, CH₄ is about 70 times more harmful to the atmosphere than CO₂ (25 times on the molar basis) [24]; The social costs of CH₄ are 50 times greater than those of CO₂ [25].

Harvesting methane from the lake for electricity generation is a complex engineering and environmental problem that requires consideration of the following points:

- Water solubilities of methane and CO₂ depend differently on pressure. High enrichment of produced methane with CO₂ reduces energy production efficiency, because part of the recovered energy must be spent on gas separation. There are different methods of removing CO₂. Each method has its benefits and drawbacks, but all require energy investment.
- CH₄/CO₂/air gas mixtures can ignite with up to 70% of CO₂ [26]. Depending on the power plant design, requirements for CO₂ content may vary considerably. If a pipeline is to be used, stricter requirements on methane purity must be followed. On the other hand, on-site gas “washing” is less demanding. However, care must be taken to dry (another energy investment) the resulting gas mixture, because water-CO₂ mixtures will likely corrode power plant turbine elements.
- Power plant can be single or dual cycle. While the former is simpler to operate, the latter is more energy efficient. The dual cycle turbines are more expensive and require highly qualified staff to operate.
- Methane accumulation rate in the resource zone is low or zero, which has only been confirmed recently. For a long-term energy forecast (50+ years), it is necessary to know methane accumulation rate, (*i.e.*, net recharge rate) in the lake's resource zone. However, there is no agreement on the magnitude of methane recharge [4,6,10,12].
- Poor management of produced water (*i.e.*, uncontrolled reinjection of degassed water into the lake) may disturb the lake equilibrium and initiate a limnic eruption. Although the likelihood of this eruption is low, the risks are catastrophic. Also, the lake is used as a source of drinking water, and the harvesting activities must not compromise water quality [27].

This paper addresses most of the points above. We start with an experimental validation of our physico-chemical numerical model of the lake, and calculate gas solubilities at different pressures (Section 3.1). Then, by varying the produced gas ratios and energy separation costs, we arrive at the optimal strategies for methane harvesting from different lake depths (Section 3.2). What follows is the global efficiency analysis of a power plant in terms of produced

energy, CO₂ emissions, recirculated water *etc.* (Section 3.3). We then assess environmental impacts of methane harvesting activities, in particular, the accompanying CO₂ releases, produced water reinjection into the lake, and their potential mitigation solutions (Sections 3.4 and 3.5). Finally, we present and discuss a long term lake development strategy that aims at a sustainable methane harvesting (Section 3.6).

2. Methodology

2.1. Optimization of produced power

A numerical model was developed with Phreeqc, and validated experimentally to estimate the gas recovery, see section 2 in Supporting Information. To analyze the water degassing efficiency based on the Phreeqc model, and the lake water composition at various depths, we have developed an in-house algorithm using MATLAB® (R2018a) that calculates the water energy and mass balances.

The studied degassing pressures were: 1) complete degassing to 1 atm of the saturated gas pressure; 2) the pressure needed to achieve 90% methane recovery; and 3) the pressure that maximizes energy output. For the optimization, we considered three different energy costs of CO₂ separation from CH₄, which are 2, 4, and 6 MJ kg⁻¹ of CO₂ separated. These costs are selected based on the average values reported for aqua-ammonium, amine, and zeolite separations, respectively [28–30]. The net produced power is

$$\dot{E}_{net} = \dot{E}_{CH_4,primary} - \dot{E}_{CH_4/CO_2,sep} \quad (1)$$

where $\dot{E}_{CH_4,primary}$ is the primary power contained in CH₄, and $\dot{E}_{CH_4/CO_2,sep}$ is the power spent to remove CO₂ completely from the stream. The lower heating value of 50 MJ kg⁻¹ for CH₄ is used, because some of the separation processes require temperatures above 100 °C (*e.g.*, amine regeneration).

The technological limit of CH₄ extraction is 5 mol m⁻³ of residual CH₄ concentration [31]. Therefore, at the current concentration values, approximately 30% of CH₄ will be left behind. Additionally, we neglect the energy required to pump and reinject water, because the flow is driven mainly by a difference in buoyancy and siphoning effects (see Supplementary Information).

2.2. Environmental impacts: water disposal

2.2.1. Disposal at the surface

Surface disposal assumes that the degassed water will only be mixed with the oxic layer (0–60 m depth). We analyzed the chemical oxygen demand to oxidize the dissolved organic carbon, ammonium, hydrogen sulfide, and methane using the concentration profiles made available by Morana *et al.* [32], Pasche *et al.* [33], Schmid *et al.* [34]. For current phosphate concentrations, we used the data available in Morana *et al.* [35]. The phosphate limit of 30 µg L⁻¹ is based on the criteria available in Yang *et al.* [36], Xiaoying and Shijie [37].

2.2.2. Disposal at depth

The stability of the stratified structure of the lake that prevents the release of CH₄ and CO₂ to the surface can be evaluated with different approaches.

A commonly used method is the dimensionless density ratio (R_ρ) proposed by Turner [38], which calculates explicitly the ratio of stabilizing and destabilizing forces. This method identifies the regime where double diffusion convection occurs. This approach has recently been criticized for its limited capacity to identify

changes that occur in the middle of the chemocline. Hirslund [17] suggests a simplified version of the buoyancy frequency instead. Similarly, a more general approach widely used across fields is the buoyancy frequency (also known as Brunt-Väisälä). It gives information about the water column stability and has been used in previous studies, but it cannot identify the double diffusion convection (DDC) regime [1,31]. A detailed comparison and discussion of these three methods can be found in the Supplementary Information. Below we use the buoyancy frequency approach.

The buoyancy frequency can be calculated using an equation derived by 1

$$N^2 = \frac{g}{\rho} \frac{d\rho}{dz} \left[\text{s}^{-2} \right] \quad (2)$$

where g is gravitational acceleration in m s^{-2} , ρ is the water density in kg m^{-3} , and z is the lake depth in m .

The stability criteria were derived for three conditions. First, when $N^2 > 0$ the system is stable. A neutral equilibrium state occurs for $N^2 = 0$. The system will be unstable when $N^2 < 0$; in this case, the buoyancy force reinforces fluid displacement.

The water density was calculated according to the 1990 International Temperature Scale (ITS-90) [39], and then corrected for salinity, and dissolved CO_2 and CH_4 [1].

$$\rho(T, S, \text{CO}_2, \text{CH}_4) = \rho(T) (1 + \beta_S S + \beta_{\text{CO}_2} C_{\text{CO}_2} + \beta_{\text{CH}_4} C_{\text{CH}_4}) \quad (3)$$

where the contraction coefficients for the dissolved salts, CO_2 and CH_4 are, respectively, $\beta_S = 7.5 \times 10^{-4} \text{ L g}^{-1}$, $\beta_{\text{CO}_2} = 1.25 \times 10^{-2} \text{ mol L}^{-1}$, and $\beta_{\text{CH}_4} = -0.020 \text{ mol L}^{-1}$ [40].

The stability analysis was performed for the degassed water reinjection at 150 and 190 m depth, with the impacted zones at 130–200 m deep and 190–250 m deep, respectively. A third scenario was calculated with the reinjection right below the main chemocline at 270 m depth with the impacted region between 270 and 320 m of depth.

Two conditions were considered: 1) partial replacement (50%) and 2) complete replacement (100%) with degassed water. The degassed water composition was based on the lake water composition at 350 m depth and degassed at 5 atm of head pressure (matching the optimum condition). This composition was used for the disposal at depth and at the surface. It was assumed that the reinjected water and lake water were at the same temperature.

2.2.3. Lake block exploitation

To evaluate duration of the optimal harvesting time at each location, we broke down the lake into blocks with a surface area of 5 by 5 km^2 , and depth from the lake surface to the bottom. For these blocks, we analyzed the time required to reach one of the critical conditions: 1) limiting phosphate concentration or anoxic condition (see section 2.2.1); 2) depletion of the resources ($\sim 70\%$, see section 3.3); and 3) limiting reinjection volume (see section 2.2.2). This analysis accounts for the lake water influx of $7 \text{ m}^3 \text{ s}^{-1}$, to supply a 25 MW of nominal power.

The technological limit of CH_4 extraction is 5 mol m^{-3} of residual concentration [31], which implies that at the current concentration values, approximately 30% of CH_4 will be left behind.

3. Results and discussion

3.1. Quantification of methane release

According to our model, which was verified experimentally, the amount of CO_2 released during a complete degassing process is approximately four times greater than that of methane. This finding

is in agreement with the observed values measured *in-situ* [3,34]. Such degassing ratio is in part caused by intrinsic CO_2 solubility in water as well as its chemical equilibrium with other species. In contrast, methane is a relatively inert gas at the lake conditions. The gas-phase starts evolving at different hydrostatic pressures, depending on where the water was sampled from. For example, the dissolved gases start evolving at 17 atm from the water sampled at 350 m, at 14 atm for water at 300 m, at 11 atm for water at 250 m, and 3.5 atm for 200 m of depth. Thus, the amount and composition of the gas-phase evolved from each depth and pressure is different. At the same head pressure, the total amount of gases is higher in the resource zone, while it decreases in the potential zone, and it is the lowest in the transition zone [33,34]. The recovery of the gases stops at the lake's surface, where the saturated gas phase pressure is 1 atm.

Fig. 1 shows moles of CH_4 and CO_2 evolved from a liter of lake water degassed at a given head pressure. CH_4 has an almost linear degassing concentration dependence on pressure, while CO_2 degasses exponentially. For example, at a head pressure of 5 atm, 22.4% of the dissolved CO_2 is released from the water 350 m deep.

In Fig. 1a, the left y-axis shows as the amount of CO_2 released per liter of water, and the right y-axis gives the mass equivalent of CO_2 in kilograms per cubic meter of water pumped. The degassing profile of CO_2 is steep when the head pressure is below 5 atm. The profile is almost exponential, and the transition zone has a concentration difference of 75% between the upper and lower limit. In addition, the right axis of Fig. 1a allows estimating the environmental impacts in terms of the amount of CO_2 that will be released to the atmosphere after methane extraction.

In Fig. 1b, the recovered methane concentration drops approximately by 15% for every 50 m of lake depth, except for the 200 m, where it decreases by 70% compared with 250 m. The right y-axis in Fig. 1b shows the value of CH_4 as primary energy gained for each m^3 of water degassed. This gain follows a linear trend. The maximum energy is extracted at the 350 m depth. In comparison with the 350 m depth, 87.5% and 75% of the energy is recovered from the 300 m and 250 m, respectively. At 200 m, the same amount of water provides only 20% of the energy in comparison with the 350 m. The more water is extracted, the larger the perturbation to the lake and the higher the cost.

The least desired component in the produced gas phase is hydrogen sulfide, H_2S , which has a degassing profile similar to CO_2 , but its concentration is much lower. Even at complete degassing, H_2S content is within the pipeline requirement margins [41]. The analysis is presented in Supporting Information (Fig. 1S shows H_2S evolution profile in ppm for direct comparison with the threshold values. The red dashed line designates the threshold concentration at which H_2S is harmful to humans [42]).

We conclude that the differences in physical and chemical properties of CO_2 and CH_4 result in the drastically different evolution profiles for these two gases upon lowering the head pressure at the surface. The depth of extraction affects when the gas phase evolution commences. Therefore, for subsequent energy production it is important to find the head pressure that yields the optimal energy output.

3.2. Optimization of methane harvesting

Different steps along the extraction and production pathways consume energy, and thus reduce the net produced energy. We estimate that the largest losses are caused by the separation of CO_2 and CH_4 from the gas mixtures. Removal of CO_2 can be achieved using several technologies, such as amine scrubbing or adsorption on zeolites [43]. This is an energy penalty on the energy production processes. The above-mentioned technologies have energy

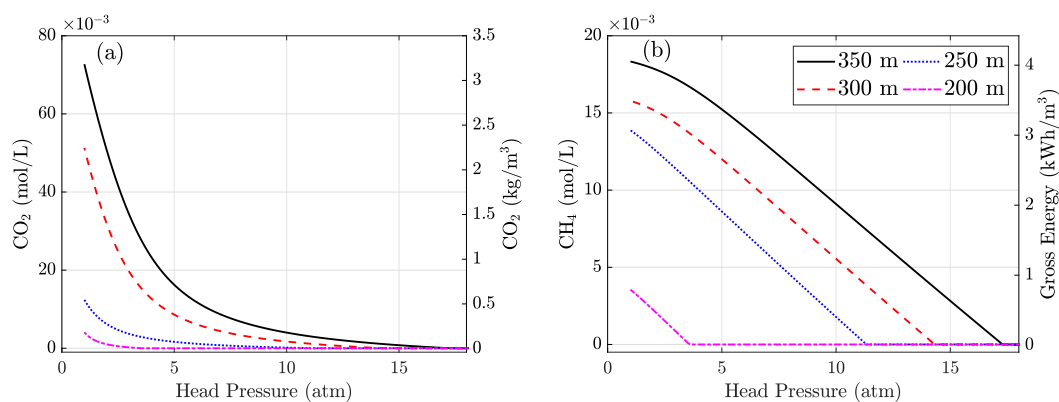


Fig. 1. Quantification of CO₂ (a) and CH₄ (b) release at different head pressures and from different Lake Kivu zones. The left y-axis gives the moles of gases recovered from a liter of water as a function of head pressure. For a), the right y-axis quantifies the amount of CO₂ in a kilogram of cubic meter of water pumped and for b) the energy recovered, which is the total energy available as fuel. The legend shows the reference depths at which the water was collected.

requirements in the range of 2–6 MJ kg⁻¹ of CO₂ removed [28–30]. We neglect the energy needed to pump and reinject water, assuming that it is driven mainly by a difference in buoyancy and siphoning effects. The detailed analysis of the siphoning effect and gravity reinjection is presented in Supporting Information. Following Zhang [44], we estimate the pipe cross-sectional area to be 1.3 m² for a volumetric flow rate of 7 m³ s⁻¹, required to generate 25 MW_e, which is technologically feasible. Using our numerical model and the above results, we find the most energy-efficient extraction strategy.

Fig. 2 graphs the net energy produced (Equation (1)) from four different waters sampled at 200 m (a), 250 m (b), 300 m (c), and 350 m (d) depths. We study the net energy dependence on the head

pressure and separation technology. The green line points to the optimum condition, which depends on the separation technology used. The optimum recovery of methane fluctuates around 90±7.5% for the different depths. Given the uptake at 200 m, a complete degassing is the best option. For the gas phase harvested at 250 m depth, the optimum condition is degassing up to 2 atm of head pressure. At 300 m depth, the optimum is between 3 and 4.6 atm. For the uptake at 350 m of depth, the optimum conditions are at 3.9, 5.1, and 6 atm for each separation technology, respectively. The trend common to all water compositions is that as the energy cost increases, the optimum head pressure for degassing also increases, which lowers recovery of the two gases. This lowering is caused by the exponential degassing profile of CO₂ (see Fig. 1), whose release

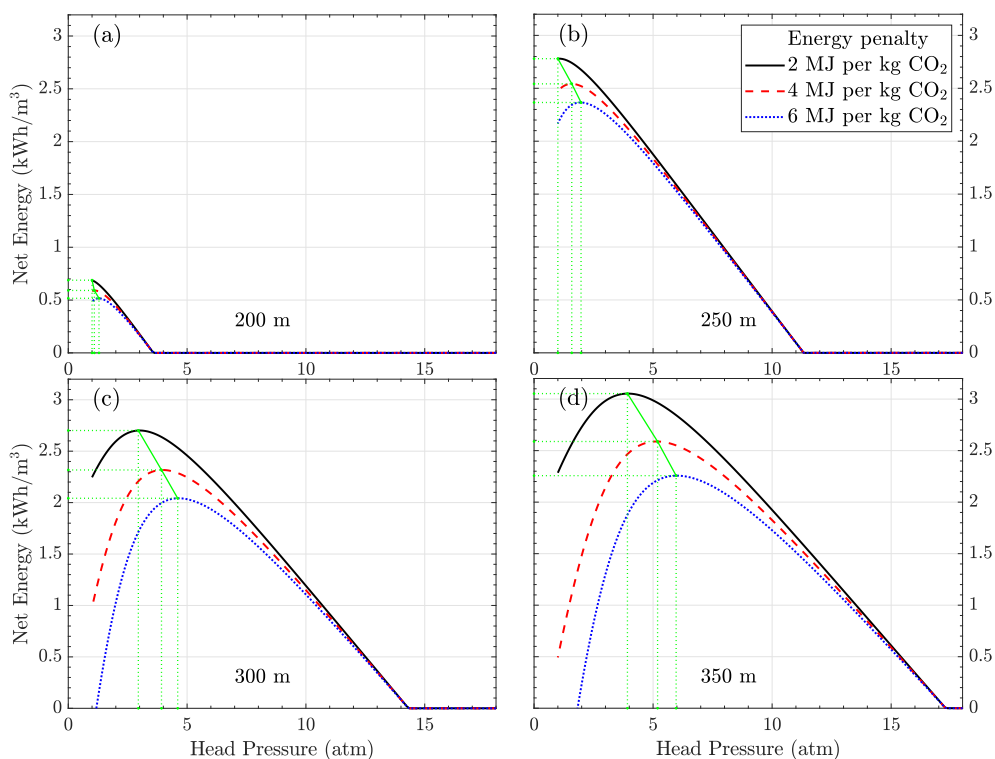


Fig. 2. Net energy recovered after subtracting energy cost of CO₂ separation. The green dotted line shows the point where the maximum energy is recovered, and the solid line is a trend line. The legend shows the energy costs considered in MJ kg⁻¹ of CO₂. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

greatly diminishes at higher head pressures.

The combustion process is strongly dependent on the gas mixture composition. When the volume content of CO₂ is above 73%, there is no combustion [26]. Currently, the KivuWatt gas extraction facility provides to the power plant a gas mixture with 85% of CH₄ [45,46]. Table S1 shows the fraction of each gas in the evolved gas mixture before any CO₂ separation. Operational conditions could provide parameters that allow to accommodate some CO₂ in the gas phase.

Because amine scrubbing is the most established separation technology, hereafter our analysis will be based on its energy cost of 4 MJ kg⁻¹ of CO₂ separated. Below we compare the following cases with our optimal strategy: 1) complete water degassing to 1 atm and 2) extraction of 90% of dissolved methane.

Fig. 3 summarizes the key operational and environmental outputs of the energy production process, assuming a value of 4 MJ kg⁻¹ of CO₂ for the gas separation process. The results are color coded for different head pressures. The depths of 200, 250, and 300 m are not in the resource zone and are shown here for comparison only. Methane must be recovered from the resource zone that starts at around 350 m of depth. Below this point, CH₄ and CO₂ concentrations are almost constant.

Fig. 3a plots the volume of water required to produce energy equivalent of 1 kWh of electricity (kWh_e) from the gas mixtures extracted at different depths. Fig. 3b shows the total CO₂ emissions. It includes the degassed CO₂, and the CO₂ emissions from the combusted methane. Fig. 3c shows the energy output as electricity. It accounts for the energy spent on gas separation, and we assume a power conversion efficiency of a single-cycle power plant of 40%. Fig. 3d shows the global energy efficiency of the system, considering all the above losses and the total energy available.

For the uptake at 350 m of depth, complete recovery of the gases is not practical, as almost all energy would be spent on gas separation. The 90% and our optimum recovery scenarios give similar results, and the net energy is 0.975 and 1.03 kWh_em⁻³, respectively, which implies a global efficiency of 27% for the 90% recovery and 31% at the optimum configuration.

It is interesting to note that at a the depth of 250 m, energy efficiency is better for the 90% recovery. However, energy output remains higher for the optimum process. This happens because more energy is consumed to separate CO₂ at optimum than at 90% recovery. In Fig. 3, we can discard the uptake at 200 m of depth, simply because it would cause a significant disturbance from the low energy output, implying that a higher volume of water would be pumped.

Supporting Table S2 summarizes the total gas fractions of CH₄ and CO₂ released from different depths for the optimum and 90% recoveries. Due to the dynamic nature of gas production in the lake, the optimum conditions may change with time. Therefore, continuous monitoring of methane content in the resource zone will be required.

3.3. Long-term exploitation strategy

For a long-term energy production strategy one must consider the following aspects: methane mass balance (current concentration, recharge rate, extraction rate, unrecoverable concentration), methane harvesting efficiency, and methane to electricity conversion efficiency.

Above, we have addressed harvesting efficiency of methane from the lake water. In this section, we discuss the limitations on methane recharge rate that refers to net accumulation of methane,

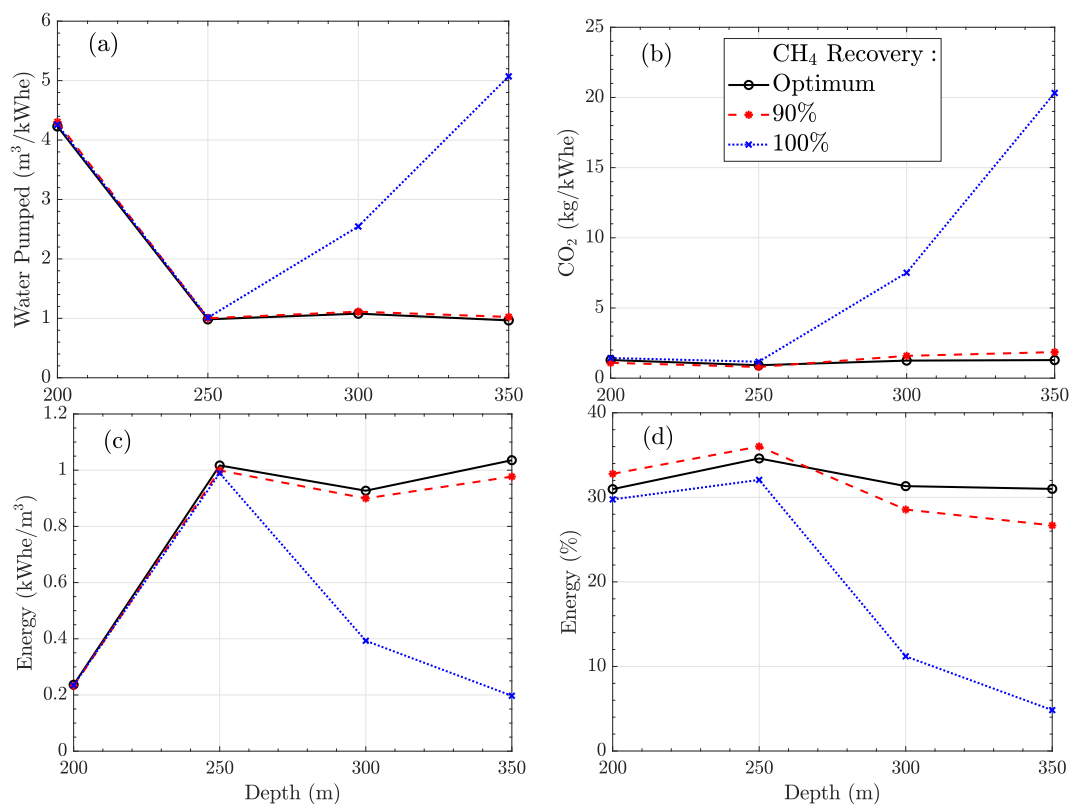


Fig. 3. Global efficiency and the environmental aspects of methane power production technologies in Lake Kivu. a) Water that should be pumped to provide 1 kWh_e; b) the amount of CO₂ that will be emitted is the sum of CO₂ degassed and that from the combustion process; c) Net energy produced per m³ of degassed water, considering the losses from gas separation and power conversion; d) Global energy efficiency of the system accounts for the energy used to separate the CO₂ and power conversion efficiency of 40%. This estimate is made considering the intermediate energy cost of gas separation equal to 4 MJ kg⁻¹ of CO₂. The x-axis is water uptake depth in the lake.

i.e., after natural losses. We consider two technologies for electricity generation: single cycle power plant and combined cycle power plant.

Current methane availability in the resource zone is estimated to be 40.9 km^3 [6]. The technological limit of CH_4 extraction is 5 mol m^{-3} of residual concentration [31], which implies that at the current concentration values, approximately 30% of produced CH_4 will be left behind.

CH_4 recharge rate is required to make accurate long-term forecasts. However, the literature estimates of this recharge rate are in disagreement. We estimate the recharge rate from the available literature sources starting in 1955. Our analysis results in a distinctly *lower value* of the methane recharge than previous estimates by Schmid et al. [4], Pasche et al. [12], Schmid et al. [6]. The details are presented in Supporting Information (Fig. 2S shows a robust linear fit of the data. The slope of the line in the $C_{\text{CH}_4} - t$ coordinates is the recharge rate of $0.012 \text{ mmol/L}_{\text{reservoir}}$ per year (an increase of 0.075% per year). Our fit accounts for the variation and uncertainty of all publicly available data points.) Because all the CH_4 samples were collected from the reservoir zone in the lake at a only a handful of locations, there is no systematic areal coverage, and apart from the method or measurement uncertainty, natural heterogeneity might be the cause of data variation. The increases and drops in CH_4 might be genuine, not caused by measurement error, but rather by natural lake cycles. We suggest that the regular areal CH_4 surveys of the lake will shed more light on the evolution of methane in the reservoir zone. Based on our analysis, and given the uncertainty, we conclude that the lake is in a steady-state, and the recharge rate is close to zero. This result is in agreement with the most recent estimate by 10. In the analysis below we consider two extremes of the recharge rate, 0 km^3 per year for the steady state based on our findings and 10, and 0.12 km^3 per year.

A stand-alone gas turbine has an efficiency of 35–40% [47]. Different techniques, like heat recuperation, reheating, inter-cooling and mass injection, can improve the efficiency of a pass-through gas turbine to 38–40% [48].

A higher efficiency alternative is a combined-cycle power plant that includes gas and steam turbines. The current designs are operating with efficiency of $\sim 60\%$, and there is potential to break this barrier [48,49].

GE Power estimates that a 1% increase in efficiency could save millions in fuel costs over a year [50]. The disadvantages of adopting the most efficient technology arise from the cost, which almost doubles for a combined-cycle power plant [51,52]. A more in-depth technical and economical analysis should be completed before any decision is taken. Worldwide, the average efficiency of gas power plants is 39% [53].

Below, we consider both power conversion cycles, assuming best case efficiencies of 40% for a single-cycle gas turbine, and 60% for the combined-cycle.

Fig. 4 shows the operational time for the considered nominal power, with the presence and absence of CH_4 recharge. The red dashed line designates 50 years of operational time – a target set by the Rwanda government.

Fig. 4a shows the operational time as a function of the nominal capacity of the power plant, assuming a fast methane recharge rate of 0.12 km^3 per year, which provides nearly $1.2 \times 10^6 \text{ MW h}$ of primary energy per year. This primary power is enough to run continuously approximately 50 MW of nominal power. The operational time shortens almost exponentially as nominal power drawn from the lake increases; it is a consequence of the recharge rate. A reduced extraction rate delays depletion of the reservoir, because over a longer period the recharge rate has a more significant contribution, similar to a compound interest effect.

Optimal harvesting with a combined cycle power plant is the

most appealing option that provides nearly 400 years of operational time at 100 MW of nominal power. However, because of the complexity of the combined cycle technology and the costs associated with the optimal harvesting approach (for example, the costs of continuous exploration to find methane concentration levels in the lake and adjust them in the models), a pass through-turbine with the 90% methane extraction scenario is a better base case. In this case, the lake can potentially support 50 years of operational time at 200 MW.

Although optimum methane production condition increases overall process efficiency by only 4%, it lengthens operational time by 20%. In comparison with the single cycle, the combined cycle increases the operational time by 55% on average, while the optimum condition can run 28% longer than the 90% recovery case.

Beyond the power conversion technology adopted, optimization of the process implies that for each 1% of improvement in the global efficiency, operational time extends by 5%.

A completely different scenario emerges when there is no recharge of CH_4 , see Fig. 4b. The high nominal power output will reduce the operational time by 15–23%. The most significant reduction occurs at 100 MW, with the best scenario operating for 150 years, while the single-cycle operates for close to 100 years. Because of uncertainties in CH_4 recharge rate, the best strategy is to start with low nominal power and initiate a monitoring program across the lake. As more data become available from the monitoring of Lake Kivu, the predictions and power generation can be adjusted accordingly.

3.4. CO_2 emissions and mitigation

In this section, we compare natural CO_2 and CH_4 emissions from the lake to the emissions from gas harvesting. Annually, the lake emits to the atmosphere $800 \times 10^3 \text{ t}$ of CO_2 [54], and $5.2 \times 10^3 \text{ t}$ of CH_4 [11].

Lake Kivu contains $564 \times 10^6 \text{ t}$ of CO_2 and $29.3 \times 10^6 \text{ t}$ of CH_4 [6]. CH_4 is 70 times more potent greenhouse gas than CO_2 by mass. A complete release of CH_4 from the lake into the atmosphere would be equivalent to releasing $2050 \times 10^6 \text{ t}$ of CO_2 . According to Brenbold et al. [10], the risk of such a release is not increasing with time, but is nonzero. Therefore, it is important to execute lake degassing activities at a rate faster than natural recharge.

For the harvesting scenarios considered above, emissions vary from 16.26×10^3 to $7.5 \times 10^3 \text{ t}$ of CO_2 per MWh_e per year. For 100 MW of installed power at 90% degassing, 0.1356 t of CO_2 will be generated per capita with the current population of 12 million people. These emissions could be reduced by a third if the optimum conditions were implemented.

The most recent World Bank data indicate that in 2014 Rwanda had the per capita CO_2 emissions of 0.076 t, while the average for low-income countries was 0.308 t. In comparison, a developed green country, like Sweden, emits 4.48 t of CO_2 per capita per year [55].

Adding the potential power plant emissions to the current emissions, Rwanda will be emitting approximately 0.2 t of CO_2 per capita, which is extremely low compared with other CO_2 producers. At the current economic status quo and level of development of the country, it does not make sense to enforce a mitigation plan for CO_2 emissions, because it will increase energy cost and hinder the development of Rwanda.

Rwanda's government reports that there is an unbalanced demand for wood that is heavily used as cooking fuel, and causes deforestation [21]. Thus, we estimate the feasibility of forestry and agroforestry as a CO_2 release and deforestation mitigation strategy in Rwanda.

Because of many benefits that go beyond carbon sequestration,

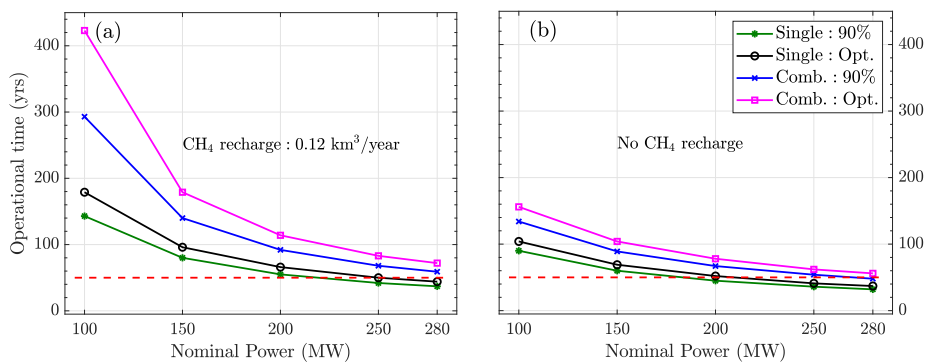


Fig. 4. Operational time with (a) CH₄ recharge, and (b) no recharge. The legend indicates the extraction method considered. Two power conversion cycles are considered: a pass-through cycle (power conversion efficiency of 40%) and combined cycle (power conversion efficiency of 60%). We compare two processes, 90% of methane recovery and the optimum process. Our calculations assume the intermediate value for gas separation of 4 MJ kg⁻¹ of CO₂, and water uptake at the main reservoir level (350 m depth). The red dashed line is the power threshold at 50 years. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

agroforestry seems to be a reasonable approach. It can reduce land pressure, enhance soil fertility, increase resilience to climate change, and provide fuelwood [56,57].

Carbon that could be stored in soil varies widely, depending on the approach and goals [58,59]. The values reported for carbon stored per hectare range from 5 to 300 t [56,59]. Considering that 30 t of carbon must be stored per hectare to mitigate emissions from 1 MWh_e per year, the specific requirement is approximately 100 ha of arable soil. In the case of a 100 MW power plant that operates for 100 years, the offset total requirement is 1 × 10⁶ ha. The required area corresponds to 38% of the territory of Rwanda, which is 26,338 km² [60]. Partitioning this area over one hundred years implies that each year 0.38% of Rwanda’s territory must be reforested. This area can change drastically depending on the assumptions, and a more detailed study should be performed.

Aligned with the goal to develop Rwanda is the possibility of aggregation of some of the CO₂ recovered. This aggregated CO₂ can be a raw material for different products, such as methanol, urea, and polyurethanes [61]. Assigning a price to CO₂ aligns with the effort to industrialize Rwanda; it should be considered and included in the development strategy for the country [21].

3.5. Degassed water disposal

Gas extraction and reinjection disturb the equilibrium of Lake Kivu. Table 1 shows the minimal volumetric water extraction rates (in m³ s⁻¹) for the different extraction scenarios proposed above (see Fig. 4). It serves as a baseline for the water reinjection rate needed to minimize perturbation of the stratified layers. However, the listed flow rates are based on the current gas concentration. As the reservoir depletes, the flow rates will have to increase to maintain the same rate of gas extraction. The most probable environmental impacts are well described by Kling et al. [62], and more recently by Hirslund and Morkel [63]. Significantly different extraction and reinjection scenarios were modeled by Wüest et al. [31], which – when complemented by this work – should provide a minimum understanding of the expected consequences.

Table 1
Volumetric flow rate for the nominal power of 1 MWh_e.

Cycle Depth (m)	Normal		Combined	
	90 (%)	Optimum	90 (%)	Optimum
Flow (m ³ s ⁻¹)	0.284	0.268	0.189	0.178

In order to estimate the effect of water reinjection, we consider the stratified lake structure and the physicochemical processes that define it. The role and interplay of double-diffusion convection, chemoclines, and subaquatic springs are still somewhat unclear, and the contradicting perspectives can lead to different descriptions of the lake [13,16,17,64]. Below we consider the consequences of disposal of the degassed water at the lake’s surface and its reinjection at various depths.

3.5.1. Water disposal at surface

Lake Kivu is oligotrophic [65], with reduced levels of nutrients and dissolved organic carbon (DOC) [35]. Availability of phosphorus is the key factor limiting primary production in the lake [33,65,66].

The degassed water from the deeper layers is anoxic but nutrient-rich. According to Morana et al. [35], deep waters of Lake Kivu have more than double of the DOC measured at the surface. Anoxic water can form an anoxic region in the lake, while excess nutrients can induce a eutrophic condition in the lake.

Total chemical oxygen demand could be used to estimate the impact of the anoxic condition. Assuming complete biological oxidation of ammonia to nitrate, hydrogen sulfide to sulfates, methane and organic matter to CO₂, approximately 500 mg of O₂ per liter of degassed water is required. With negligible aeration, 72 L of the surface water will provide the needed oxygen per liter of degassed water disposed at the surface. However, from the information available, it is impossible to estimate if surface disposal will induce an anoxic condition due to the fast reactions, or if it will only reduce the dissolved oxygen, not leading to a critical condition. The mitigation of this impact can be made through aeration systems and/or by avoiding fixed-point discharges.

Phosphorus is the limiting element of eutrophication. A minimum concentration of 30 µg L⁻¹ is required for the process [36]. The most recent measurements indicate the concentration of 18.6 µg of phosphorus per liter of water in the mixed layer [35]. In contrast, the degassed water has the mean phosphorus concentration of 5630 µg L⁻¹. To keep the phosphorus concentration under 30 µg L⁻¹, dilution by a factor of 500 is required. A 25 MW power plant that reinjects water at a rate of 7 m³ s⁻¹ will require mixing of the degassed water with 3500 m³ of surface water per second. Needless to say this requirement is unrealistic. Other potential problems with surface disposal are: evolution of unrecovered methane and hydrogen sulfide.

3.5.2. Water disposal at depth

The degassed water reinjection at a depth disturbs the meta-stable lake equilibrium and can lead to an alternation of current

stratified structure of the lake. According to Kling et al. [62], the two most significant barrier layers that must be preserved are located between 60 and 80 m of depth, which separates oxic water from the anoxic nutrient-rich layer, and at 250 m, the main chemocline which separates the gas-rich layer from the rest of the lake. The destruction of the first layer can cause lake eutrophication, and with the main chemocline largely gone, a gas outburst could occur. Also, there is a secondary chemocline at 190 m depth, which separates the transition zone from the potential resource zone.

In a typical reinjection scenario, degassed water extracted from the resource zone will be pumped back into the lake. We consider three scenarios: 1) the water is reinjected at 150 m in the transition zone; 2) the water is reinjected into a convection zone between 190 m and 240 m of depth; and 3) the water is reinjected into the upper part of the resource zone at 270 m.

Fig. 5 shows the buoyancy frequency depth profiles before and after the reinjection. The positive buoyancy frequency indicates stratification. The negative buoyancy frequency indicates convective overturning mixing.

For the current lake state, Fig. 5 shows two chemoclines in black. The main chemocline is at 250 m of depth (highlighted by a blue band, see Fig. 5), and the secondary chemocline at 190 m of depth (highlighted by a light blue band, see Fig. 5), where the buoyancy frequency is positive. All reinjection scenarios strengthen or initiate stratification above the reinjection point, and enhance mixing below the reinjection point.

In Fig. 5a the degassed water is reinjected at 150 m. It contains 18% more CH₄, 745% more CO₂ and is 120% more saline than the *in-situ* water at this depth. If a significant amount of degassed water is injected, mixing will be induced just above the chemocline at 190 m, and a new diffusion zone will form at 130 m, above the reinjection point.

Fig. 5b indicates the disturbance, induced when the degassed water is reinjected into a convection zone right above the main chemocline. In this case, the degassed water has 23% less CH₄, 200% more CO₂ and 80% more salinity than the water originally present in this region. A strong mixing region will then emerge right above the main chemocline, and the layer at 190 m of depth will strengthen, forming a new diffusion zone. Because of the limitations of the adopted model, it is hard to predict what is going to happen in the main chemocline (at 250 m of depth). However, the strong mixing likely indicates the thinning and weakening of this layer followed by its disappearance.

Fig. 5c portrays the disturbance of the stratified structure, when the degassed water is reinjected below the main chemocline. At this depth, the degassed water contains 77% less CH₄, 77% more CO₂, and is 15% more saline than the native water present at the

same depth. The main chemocline is strengthened and a new mixing front at 320 m forms, weakening the chemocline at 330 m of depth.

The last approach that involves reinjection at 250 m of depth is most appealing from the safety point of view, because it strengthens the main chemocline and prevents the dissolved gases from escaping the lake. However, the resource zone will be diluted by reinjection. Additionally, the reinjection will modify the upward flux of nutrients, most probably increasing the inflow of phosphates and nitrogen into the oxic layer.

In the case of reinjection at 190 m, the primary resistance will be displaced upwards by 60 m in depth, which weakens the resistance to a gas burst due to hydraulic pressure.

The four reinjection scenarios considered above cover all main options to tackle this challenge. These are the principal scenarios. All other alternatives will be small variations of the presented results, which could be refined for a more specific and targeted study.

3.6. Block harvesting approach

In this section we discuss a block exploitation strategy and the current limitations of gas mining or water reinjection. Currently, gas extraction is conducted from a barge floating on Lake Kivu [62,67,68]. Once the gas resource near the barge location depletes locally, the barge will have to move to another location. To optimize the harvesting time and reduce the number of relocations, we split the lake's surface into individual blocks with no or little cross-flow. Each block is 5 by 5 km². Fig. 6 shows in red the boundaries of the main gas reservoir, and the grid consists of 5 by 5 km² blocks. The lake is covered by nearly 30 such blocks.

Different factors affect lifetime of a block. For example, the time to extract all accessible methane (30% of CH₄ is not recovered) from a single block is 12 years at an intake of 7 m³ s⁻¹, which is required for 25 MW of electricity production. However, if the water is reinjected at the lake's surface, the layer will become anoxic in 35 days, and phosphate concentration will exceed the limit of eutrophication in 5 days.

The degassed water can be reinjected at a depth: into the transition region (150 m), or the potential resource zone (190 m), or the resource zone (270 m). The transition zone will be entirely replaced in 11 years in a single block. The potential resource zone will be entirely replaced in 7 years, and the time of reinjection into the resource zone, will be the same as the time to exploit the resource or 12 years.

This information complements the results shown in Fig. 5, where the impact of a full replacement of the layer with degassed water is discussed. The block approach allows one to estimate the

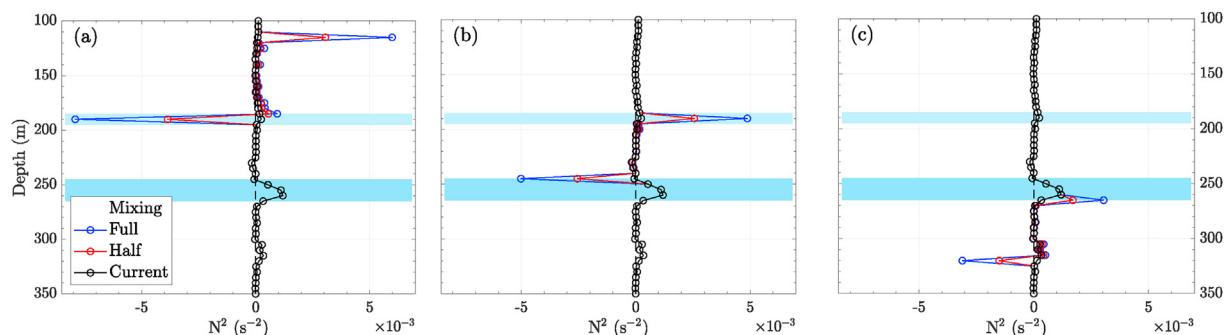


Fig. 5. Reinjection mixing. The black line is the actual lake profile before perturbations. The red line is when 50% of water volume in that layer is the reinjected fluid (assuming it is well-mixed). The blue line is when all the water is replaced with the reinjected fluid. (a) The reinjection depth is at 150 m, (b) the reinjection depth is at 190 m, and (c) the reinjection depth at 270 m. The areas shaded in blue indicate the chemoclines that should be preserved. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

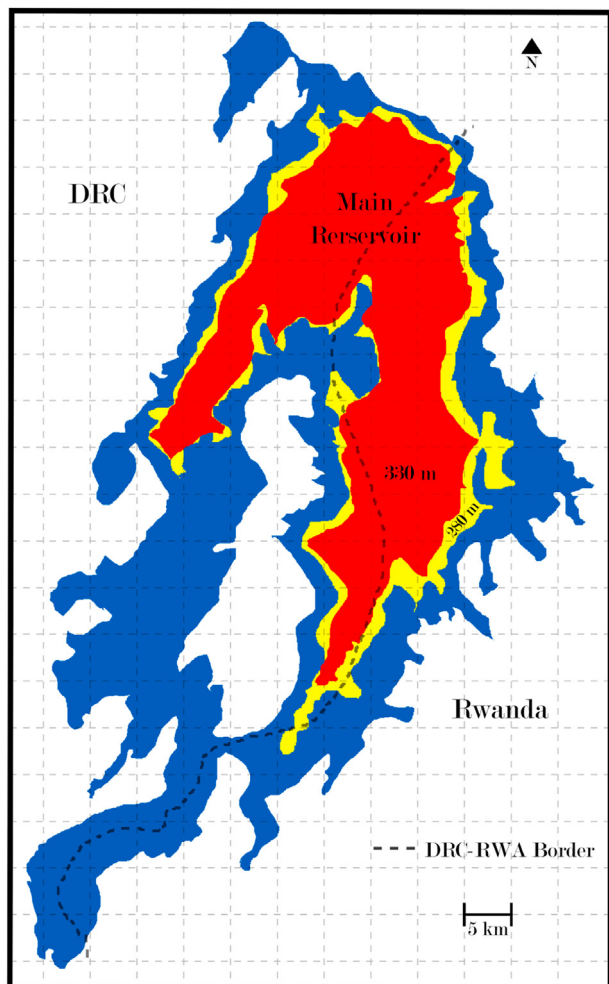


Fig. 6. Lake Kivu map. The grid shows the proposed “block” analysis. The red area is the lower reservoir of CH₄ with depths greater than 330 m, the yellow area is the upper part of the main reservoir. Adapted from Anonymous [69]. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

local impacts on the lake and quantify times for a move to another block. Alternatively, our block extraction model could be the basis of a more elaborate strategy to minimize environmental impacts on the lake.

Methane extraction and reinjection activities require proper monitoring to evaluate and mitigate their environmental effects. The current Lake Kivu Monitoring Program, aims to monitor on-plant, near the plant, and lake wide [70]. Activities related to the near plant monitoring that include the deep vertical profiles (pH, temperature, and salinity) should be emphasized for the reinjection control. For instance, it would be a good practice to monitor degassed water reinjection, mixing, and its effects on the lake stratification.

An important step is to make the data collected during these monitoring activities publicly available, so they could be widely used to understand the questions that remain open about Lake Kivu. An open data set will enable community contributions toward better understanding of the incredibly complex, interlinked, heterogeneous and dynamic open system that is Lake Kivu. This system is vital to the very survival and well-being of Rwanda’s people.

4. Conclusions

In this work, we have developed a numerical model of the physico-chemical equilibrium of Lake Kivu and confirmed it experimentally. Using this model, we quantified how pressure-dependent solubilities of CO₂ and CH₄ affect the efficiency of methane recovery and thus energy production. Based on the methane separation and conversion technology, siphoning effect, and water intake depth, we have proposed an energy-efficient methane harvesting strategy. The proposed strategy is 4–6% better than the alternatives. Although these improvements appear to be insignificant, they add up in the long term. Regardless of the power conversion technology selected, a 1% improvement in the global efficiency extends the operational time by ~5%.

By analyzing historical methane concentration data, we have shown that currently the lake is in a steady-state and net methane recharge rate is close to zero. This condition significantly affects the long term extraction strategy. By being extracted to deliver 200 MW, the lake could supply power for roughly one generation (40–50 years) without CH₄ recharge. At 100 MW, it could provide longer and perhaps more sustainable energy supply. We refrain from giving firm estimates for such long timescales because of the inevitable uncertainty surrounding them. We encourage the readers to make their own conclusions based on the analysis presented in this paper.

Gas emissions from methane extraction are negligible and their mitigation is unnecessary. Rwanda’s CO₂ emissions are insignificant, even when compared with average emissions from other low-income countries. Nevertheless, support for mitigation through forestry and agroforestry is desirable, given current dire problems with deforestation.

The impacts of methane extraction on the eutrophication of lake’s water indicate that water disposal at the surface is prohibited. It seems adequate to reinject the degassed water in the lower transition zone (150 m depth) or in the upper resource zone (270 m depth). These choices might allow full exploitation of the resource without interruptions due to environmental degradation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.energy.2021.120215>.

Credit author statement

Natanael Bolson: Experiment – Execution and Data Acquisition, Writing – original draft Preparation, Visualization and Software; Maxim Yutkin: Experiment – Experimental Setup and Data Interpretation, Writing – Editing and Reviewing, Software; Tadeusz Patzek: Supervision, Writing-Reviewing and Editing.

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