

require changing  $k$ ,  $k'$  and  $D$  from constants to operators that are non-local in space and time. The delta function of the source and sink terms in equation (6) localizes  $\mathbf{r}$  to within a volume  $a^3$ , where  $a$  is the range of lengths over which a hydrogen bond might exist. The quantity  $\rho(\mathbf{0}, t)$  is thus  $n(t)/a^3$ . This connection, with equations (5) and (6) gives three equations for three unknowns, and these comprise a diffusion model for hydrogen-bond kinetics. The model can be solved straightforwardly for  $k(t)$  and  $k_{in}(t)$ . The solution depends on the length  $a$  as it appears in the time  $\tau = a^2/D(6\pi^2)^{2/3}$ . Diffusion constants of water are of the order of  $10^{-5} \text{ cm}^2 \text{ s}^{-1}$ . The radius of a water molecule is  $\sim 1.5 \text{ \AA}$ . Thus,  $\tau$  is expected to be in the range of 0.1–1 ps, neither short nor long compared to  $1/k$  and  $1/k'$ .

Figure 1 shows a comparison of  $k(t)$  obtained from our computer simulation with that obtained from the diffusion model with  $\tau = 0.4 \text{ ps}$  and with  $k$  and  $k'$  fixed at the values found from the correlation plot, Fig. 2. Less favourable comparisons are found by varying  $\tau$  by 25% from this value. A similar comparison using the same values of  $k$ ,  $k'$  and  $\tau$  is made in Fig. 3 for  $k_{in}(t)$ . The diffusion model is seen to provide an explanation of most of what is found from the computer simulation calculations, including the fact that  $k_{in}(t)$  becomes negative after a period of somewhat less than 1 ps. This behaviour is associated with trajectories that initially have a  $\dot{h}(0) > 0$ , and thus initially enter into a hydrogen bond. These trajectories are trapped, re-emerging only later into the unbonded region of configuration space.

Quantitative improvements to the diffusion model could be made by refining its resolution in space and time. Independent of such improvements, our analysis establishes the existence of well defined rate constants  $k$  and  $k'$  and their connection to microscopic correlation functions. This connection can be used in general to analyse the dynamics of hydrogen-bonded liquids. Our phenomenological model can be applied to interpret inco-

herent neutron scattering<sup>21</sup> and perhaps transient vibrational spectroscopy<sup>22</sup>. Experimental verification of the model will require coordinated application of two different techniques because  $c(t)$  and  $n(t)$  are independent functions that vary on the same timescale. □

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## Dynamics of CO<sub>2</sub>-driven lake eruptions

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ON 21 August 1986, a massive release of carbon dioxide from Lake Nyos in Cameroon killed about 1,700 people. A similar event occurred on 15 August 1984 at Lake Monoun, also in Cameroon. It was suggested<sup>1–5</sup> that the CO<sub>2</sub> released was initially dissolved in the hypolimnion (dense lower layer) of the lake, and was released by eruptive outgassing. Because of its violence, the Nyos outburst was at first thought<sup>6</sup> to have been volcanic, but undisturbed sediments and other evidence indicate that no large volcanic eruption occurred<sup>7–9</sup>. Recent experiments<sup>10,11</sup> have shown that decompression of CO<sub>2</sub>-saturated water is able to power explosive eruptions. Here I analyse the dynamics of CO<sub>2</sub>-driven lake-water eruptions by deriving an equation of state for gas–liquid mixtures and using it to integrate the Bernoulli equation, which describes the dynamics of the bubbly flow. I find that under certain conditions these eruptions can be violent: the lake-surface exit velocity of an initially gas-saturated water parcel may reach 89 m s<sup>-1</sup> for Lake Nyos and 51 m s<sup>-1</sup> for Lake Monoun. The dynamics are similar to those of water-driven volcanic eruptions, which are also powered by gas exsolution from a liquid.

The dissolved CO<sub>2</sub> content of the water of Lake Nyos, and hence the density of that water, increases with depth owing to slow leakage of magmatic CO<sub>2</sub> into the lake<sup>8,9</sup>. But as the partial pressure of CO<sub>2</sub> ( $p_{\text{CO}_2}$ ) in the water increases, the hydrostatically

stable water at the lake bottom becomes increasingly dynamically unstable because a small perturbation due to a landslide, sinking of cold rain water, an internal wave, a small volcanic injection of CO<sub>2</sub>, or a heavy flood of water into the lake might move the water up sufficiently to reach saturation ( $p_{\text{CO}_2} = P_{\text{total}}$ ), allowing bubbles to form and grow. Being less dense than the surrounding water, the bubbly water ascends with increasing speed, resulting in an “overturn” of the lake or a limnic eruption<sup>1</sup>, which releases the lethal CO<sub>2</sub> (ref. 9). The dynamics of CO<sub>2</sub>-driven limnic eruptions have not been fully investigated<sup>12</sup>.

Rise of CO<sub>2</sub>-saturated water may be explosive because of strong positive feedback between bubble formation and growth, volume expansion and buoyancy rise. Once triggered, uprising of bottom water may form an erupting “conduit”<sup>13,14</sup> into which more bottom water may be drawn (Fig. 1), allowing it to sustain itself throughout the eruption. In the Lake Nyos eruption, an estimated  $(1–3) \times 10^8 \text{ kg}$  of CO<sub>2</sub> (refs 5, 9) was released during an eruption that possibly lasted as long as 4.5 hours (ref. 12). Hence the process probably lasted much longer than the time needed for a water parcel to ascend through the erupting conduit ( $\leq 10 \text{ s}$ , see later discussion on ascent velocity). That is, the process may be roughly at steady-state. Steady state also requires that the pressure in the conduit at each depth be independent of time. Because the medium through which limnic eruptions occur is fluid, the pressure  $P$  at each depth in the erupting conduit is roughly hydrostatic (denoted as  $P_h$ ) and hence independent of time. Otherwise, water surrounding the conduit would flow into, compress and constrict the conduit so as to maintain hydrostatic equilibrium if the pressure in the conduit at each depth is less than  $P_h$ ; surrounding water would flow away to let the conduit expand if the pressure inside the conduit is greater than  $P_h$ . The approximate independence of pressure with time shows that the

a

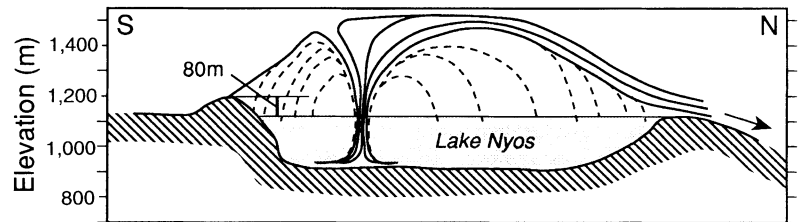
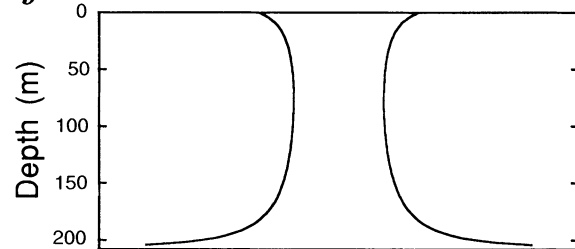


FIG. 1 a, Schematic south-north cross-section of Lake Nyos showing probable movement of gas cloud (solid curves) and water droplets (dashed curves) during a limnic eruption driven by  $\text{CO}_2$  dissolved in the water. The trajectory of water droplets depends on their size; larger droplets rain down closer to the conduit. The vertical and the horizontal scales are the same. Turbidity of the flow is not shown. The position of the conduit is not important to the model presented and could as well be towards the north end of the lake<sup>1</sup>. The flow is shown as non-symmetrical because of possible wind. b, Close-up view of idealized erupting conduit shape, calculated by assuming that the mass flux is constant; that is,  $A\rho u = \text{constant} = (A\rho u)_{\text{exit}}$  where  $A$  is the cross-section area. The conduit diameter depends on the mass flux into it (hence no scale is given for the horizontal axis) and reaches a minimum at a depth of 77 m.

b



flow can be steady once initiated and when there is enough supply of  $\text{CO}_2$ -saturated (or nearly saturated) water. Therefore, the flow is assumed to be roughly at steady-state in the model presented below.

Denoting the depth below water surface as  $h$ , the total lake depth as  $h_0$ , and the height above the bottom as  $z$ , then  $z = h_0 - h$ . In an erupting conduit, the pressure increases with depth as  $dP = \rho(g + a)dh = -\rho(g + a)dz$ , where  $\rho$  is the density of the gas-liquid mixture and varies with pressure (compressible flow),  $g$  is acceleration due to gravity, and  $a$  is the upward acceleration of the flow and varies with  $z$ . Thus  $d(u^2/2)/dz = a = -g - (1/\rho)dP/dz$  where  $u$  is the ascent velocity depending on  $z$ . Furthermore,  $P \approx P_h = P_0 - \rho_0gz = P_{\text{atm}} + \rho_0gh$ , where  $\rho_0$  is the density of bubble-free water and is assumed to be constant, and  $P_{\text{atm}}$  is the atmospheric pressure on the surface of the lake. If the flow is also barotropic (that is, local  $\rho$  is a function of  $P$  only), integration of  $d(u^2/2) = -gdz - (1/\rho)dP$  from  $z = 0$  (where  $P = P_0$  and  $u = 0$ ) to an arbitrary  $z$  (where pressure is  $P$ ) leads to the Bernoulli equation:

$$\int_{P_0}^P \frac{dP}{\rho} + \frac{1}{2}u^2 + gz = 0 \quad (1)$$

If the saturation depth is above lake bottom, the pressure at that depth should be substituted for  $P_0$  in equation (1) and the equations given below. To integrate equation (1), the relationship between  $\rho$  and  $P$ , that is, the equation of state for the gas-liquid mixture, must also be known. The liquid density is roughly constant ( $\rho_0$ ), and the liquid mass is also roughly constant. Hence,  $\rho_0/\rho \approx 1 + V_{\text{gas}}/V_{\text{liq}}$  where  $V_{\text{gas}}/V_{\text{liq}}$  is the ratio of the local gas-phase volume to liquid volume. The decompression process is roughly adiabatic with respect to the gas-liquid mixture. Because of the high heat capacity of water, the temperature decrease due to gas exsolution and expansion is small (the temperature decrease is estimated as  $\sim 4^\circ\text{C}$  if  $P_0$  is 2.0 MPa and final  $P$  is 0.1 MPa). Therefore the effect of temperature variation on density is ignored. When the pressure is less than 2 MPa, the  $\text{CO}_2$  gas is roughly ideal. Experiments show that in a  $\text{CO}_2$ -water system bubble growth is rapid (millimetre-sized bubbles can form in 0.02 s) and nucleation of bubbles is not difficult even with distilled water<sup>10,11</sup>. Hence, details of nucleation and bubble growth are not considered and bubble growth is assumed to be rapid enough to keep equilibrium between the gas and liquid. This assumption is different from that of Wilson<sup>15</sup> who assumed that for volcanic eruptions the mass of the gas phase is constant after fragmentation. The difference is due to the much greater diffusivity of  $\text{CO}_2$  in water at room temperature<sup>16</sup> than that of  $\text{H}_2\text{O}$  in

rhyolitic magma at  $850^\circ\text{C}$  (ref. 17). For example, the diffusion distance of  $\text{CO}_2$  in water is  $45 \mu\text{m}$  in one second whereas that of  $\text{H}_2\text{O}$  in magma is  $\sim 3 \mu\text{m}$  in one second. Fragmented water droplets are expected to be small and diffusion may be rapid enough to maintain rough chemical equilibrium.

For  $\text{CO}_2$  in water below 2 MPa, the Ostwald solubility coefficient  $\lambda$  is roughly independent of pressure. With the above assumptions and approximations,  $\lambda \equiv C_{\text{liq}}/C_{\text{gas}} = (n_{\text{liq}}/V_{\text{liq}})/[(n_0 - n_{\text{liq}})/V_{\text{gas}}] \approx (V_{\text{gas}}/V_{\text{liq}})/(P_0/P - 1)$  where  $n_{\text{liq}}$  is the number of moles of the gas in the liquid,  $C_{\text{liq}}$  is the gas concentration in the liquid in  $\text{mol m}^{-3}$  and  $C_{\text{gas}}$  is that in the gas. Therefore  $V_{\text{gas}}/V_{\text{liq}}$  can be replaced by  $\lambda(P_0/P - 1)$  and the equation of state for a  $\text{CO}_2$ -water mixture becomes:

$$\rho_0/\rho \approx 1 + \lambda P_0/P - \lambda \quad (2)$$

Using the above equation to integrate  $\int (1/\rho)dP$  in the Bernoulli equation yields:

$$\frac{1}{2}u^2 \approx \lambda \frac{P_0}{\rho_{\text{liq}}} \left( \ln \frac{P_0}{P} - 1 + \frac{P}{P_0} \right) \quad (3)$$

The acceleration of the upward flow can be found to be  $\lambda g(P_0/P - 1)$  in which  $P$  is decreasing. That is, the ascending flow has an increasing acceleration owing to continuous pressure decrease, unlike the constant acceleration in experiments in which  $\text{CO}_2$ -saturated water is decompressed instantaneously to a constant pressure<sup>10,11</sup>. The velocity  $u$ , calculated using equation (3), is a maximum estimate because the effects of turbulence, shallow water entrainment, and oversaturation of  $\text{CO}_2$  in water have been ignored. Hence, a more accurate account of the non-ideality of  $\text{CO}_2$ , the variation of  $\lambda$  with pressure, and the density variation of  $\text{CO}_2$ -saturated water is not warranted at present.

Figure 2a shows  $u$  as a function of depth for Lake Nyos assuming an initially saturated water parcel (with 3 wt% of dissolved  $\text{CO}_2$ ) starts at lake bottom. As expected, the velocity increases as the gas-liquid mixture rises. With decreasing depth, the velocity first increases slowly and then rapidly. As the column leaves the lake surface at 0.1 MPa pressure, the maximum exit velocity of the erupting column is  $89 \text{ m s}^{-1}$ . At this maximum exit velocity and ignoring entrainment of air, the erupting column would rise to a height of 400 m ( $0.5u_{\text{exit}}^2/g$ ). The minimum column height was estimated to be 120 m for the 1986 Lake Nyos eruption based on the distribution of dead animals<sup>5</sup>.

Figure 2b shows the calculated maximum exit velocity as a function of saturation depth. For Lake Monoun (depth 96 m), the calculated maximum exit velocity is  $\sim 51 \text{ m s}^{-1}$ . Ignoring entrain-

ment of air, an erupting column with such velocity would rise to a height of  $\sim 130$  m. If Lake Nyos erupted from a saturation depth of 150 m (instead of 208 m), the maximum exit velocity would be  $70 \text{ m s}^{-1}$  and the column would rise to a height of  $\sim 250$  m. The above results, with modifications to account for different initial and boundary conditions, may also be applied to analyse the dynamics of eruption through pipes designed to degas the gas-saturated deep water in these lakes<sup>18</sup>.

Magmatic  $\text{CO}_2$  leaks into the bottom of lakes Nyos and Monoun at a rate that could cause saturation of water at lake bottom on a timescale of years to decades<sup>18</sup>. It has been suggested that the 1986 eruption of Lake Nyos began at the base of a shallow chemocline<sup>13,14</sup>. Although eruption triggering is stochastic and could occur at shallower depths, a more disastrous, more easily triggered, and possibly more likely event would be the eruption of a significant amount of  $\text{CO}_2$ -rich bottom water if a substantial thickness of the bottom water is near saturation. Therefore, I present below and in Fig. 1 a model for a limnic eruption sustained by saturated bottom water. Because of some triggering mechanism, saturated bottom water is displaced upward. An erupting conduit is formed and sustained by drawing more bottom water into the rapidly rising column. By analogy with experimental results<sup>10,11</sup>, the rising flow is initially roughly a uniform bubbly flow and then fragments into a mist<sup>9</sup> when porosity reaches some

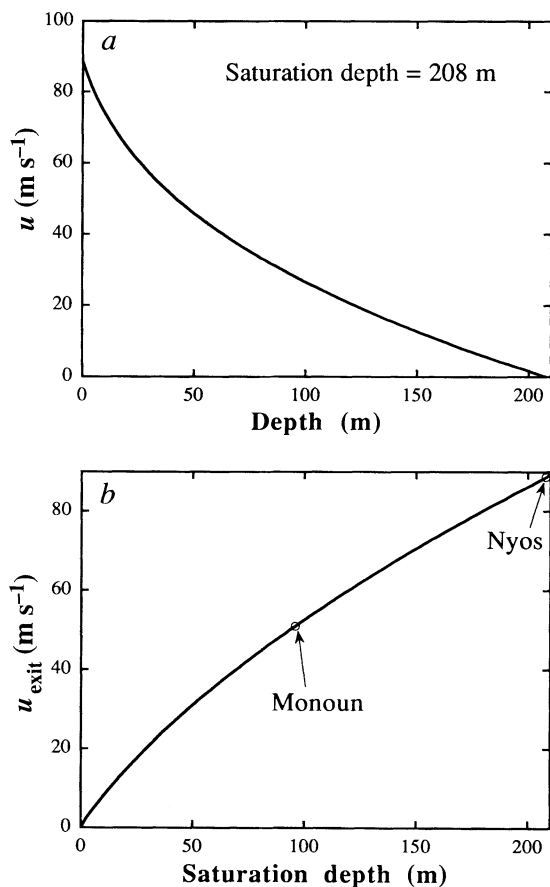


FIG. 2 a, Calculated maximum ascent velocity ( $u$ ) of an erupting  $\text{CO}_2$ -water mixture as a function of depth for Lake Nyos. The calculation uses equation (3) with  $\lambda = 0.87$  ( $T = 23^\circ\text{C}$ ),  $P_0 = 2.14 \text{ MPa}$ ,  $\rho_0 = 1,000 \text{ kg m}^{-3}$ , and pressure  $P$  in the conduit being hydrostatic. The  $\lambda$  value is calculated using the equation  $\ln \lambda = 3.12272 - 4042/T + 911269/T^2$  (where  $T$  is in kelvin), derived from data in refs 20–23. The value of  $u$  is not strongly dependent on small variations of the atmospheric pressure. For example, changing the atmospheric pressure from 0.1 to 0.088 MPa (for an elevation of 1,100 m) changes  $u_{\text{exit}}$  from 89 to 91  $\text{m s}^{-1}$ . b, Calculated maximum exit velocity ( $u_{\text{exit}}$ ) for  $\text{CO}_2$ -driven water eruption as a function of initial saturation depth using equation (3) with  $\lambda = 0.87$  and  $P = 0.1 \text{ MPa}$ . The  $u_{\text{exit}}$  for the eruption of bottom water of lakes Nyos and Monoun is indicated by arrows.

critical value (70–80%). Water droplets are carried up by the ascending gas and are roughly in thermal and chemical equilibrium with the gas, depending on the size of the droplets. Therefore, not only is  $\text{CO}_2$  gas lost from the lake, but water would rise above the surface level of the lake. Most of the water probably rains back into the lake. Because  $\text{CO}_2$ -rich gas is denser than air, the erupting column eventually collapses to form a  $\text{CO}_2$  flow down the external slope (a gravity current), carrying fine water droplets with it. By analogy to 'pyroclastic' flow, the  $\text{CO}_2$  flow carrying water droplets at ambient temperature may be termed an ambioructic (combination of 'ambient' and 'eruct') flow. At the exit velocity of  $89 \text{ m s}^{-1}$ , a conduit  $\sim 10$  m in diameter could erupt  $\text{CO}_2$  at a rate ( $4 \times 10^7 \text{ kg h}^{-1}$ ) comparable to that estimated for the 1986 Lake Nyos eruption<sup>5,9,12</sup>.

Tazieff<sup>6</sup>, assuming that a lake overturn or a limnic eruption would be slow, used the violence and localized nature of the process to argue against the limnic hypothesis and to suggest the volcanic hypothesis. My calculations and arguments above show that  $\text{CO}_2$ -driven water eruptions can be violent (as also shown by experiments<sup>10,11</sup>) and localized. Therefore, the Lake Nyos and Lake Monoun events are consistent with  $\text{CO}_2$ -driven water eruption. Because of the large amount of  $\text{CO}_2$  dissolved in the deep water of Lake Nyos, any volcanic-gas eruption through the lake would probably trigger a  $\text{CO}_2$ -driven limnic eruption and would be amplified, and probably dominated (depending on the magnitude of the volcanic eruption), by contribution from the limnic eruption.  $\text{CO}_2$ -driven water volcanism has also been proposed to play a role in the resurfacing of the jovian moon Europa<sup>19</sup>.

$\text{CO}_2$ -driven limnic eruptions are similar to  $\text{H}_2\text{O}$ -driven violent volcanic eruptions in that they are both powered by exsolution of gas from a liquid, though there may be small differences regarding the role of buoyancy and turbulence. Buoyancy helps limnic eruptions (as the eruption is through fluid) but not violent volcanic eruptions. On the other hand, turbulence plays a greater role in reducing the erupting velocity in limnic eruptions (owing to the low viscosity of water and the instability of a foam) than in violent volcanic eruptions. Ignoring these differences, the violence of a gas-driven eruption is largely determined by the solubility coefficient ( $\lambda$ ) and by the total decompression ratio ( $P_0/P$ ), based on equation (3). Therefore, both  $\text{CO}_2$ -water systems at room temperature and  $\text{H}_2\text{O}$ -magma systems at magmatic temperatures can power violent eruptions. On the other hand,  $\text{CO}_2$ -magma (except for  $\text{CO}_2$ -kimberlitic magma) systems may not be able to power violent eruptions owing to the low solubilities of  $\text{CO}_2$  in rhyolitic to basaltic magma. □

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