Evolution of CO₂ in Lakes Monoun and Nyos, Cameroon, before and during controlled degassing

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Evolution of CO$_2$ in Lakes Monoun and Nyos (Cameroon) before and during controlled degassing is described using results of regular monitoring obtained during the last 21 years. The CO$_{2(aq)}$ profiles soon after the limnic eruptions were estimated for Lakes Monoun and Nyos using the CTD data obtained in October and November 1986, respectively. Based on the CO$_{2(aq)}$ profiles through time, the CO$_2$ content and its change over time were calculated for both lakes. The CO$_2$ accumulation rate calculated from the pre-degassing data, was constant after the limnic eruption at Lake Nyos (1986–2001), whereas the rate appeared initially high (1986–1996) but later slowed down (1996–2003) at Lake Monoun. The CO$_2$ concentration at 58 m depth in Lake Monoun in January 2003 was very close to saturation due to the CO$_2$ accumulation. This situation is suggestive of a mechanism for the limnic eruption , because it may take place spontaneously without receiving an external trigger.

The CO$_2$ content of the lakes decreased significantly after controlled degassing started in March 2001 at Lake Nyos and in February 2003 at Lake Monoun. The current content is lower than the content estimated soon after the limnic eruption at both lakes. At Monoun the degassing rate increased greatly after February 2006 due to an increase of the number of degassing pipes and deepening of the pipe intake depth. The current CO$_2$ content is ~40% of the maximum content attained just before the degassing started. At current degassing rates the lower chemocline will subside to the degassing pipe intake depth of 93 m in about one year. After this depth is reached, the gas removal rate will progressively decline because water of lower CO$_{2(aq)}$ concentration will be tapped by the pipes. To keep the CO$_2$ content of Lake Monoun as small as possible, it is recommended to set up a new, simple device that sends deep water to the surface since natural recharge of CO$_2$ will continue.

Controlled degassing at Lake Nyos since 2001 has also reduced the CO$_2$ content. It is currently slightly below the level estimated after the limnic eruption in 1986. However, the current CO$_2$ content still amounts to 80% of the maximum level of 14.8 giga moles observed in January 2001. The depth of the lower chemocline may reach the pipe intake depth of 203 m within a few years. After this situation is reached the degassing rate with the current system will progressively decline, and it would take decades to remove the majority of dissolved gases even if the degassing system keeps working continuously. Additional degassing pipes must be installed to speed up gas removal from Lake Nyos in order to make the area safer for local populations.

INTRODUCTION

Volatiles in the deep interior of the Earth are brought to the surface mainly by volcanic activity. In terms of the present-day global carbon cycle, CO$_2$ discharge from subaerial volcanism and passive CO$_2$ discharge from the craters or flanks of volcanoes are the major non-anthropogenic contributors to atmospheric CO$_2$ (e.g.,

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Kerrick, 2001). Lakes Nyos and Monoun in Cameroon, West Africa, are typical sites of passive degassing. They are volcanic crater lakes situated along the Cameroon Volcanic Line. Although no active volcanism is found near the lakes, magmatic CO_2 is continuously discharged from depth, and is trapped and accumulates in deep waters of the lakes (e.g., Kusakabe and Sano, 1992). This accumulation resulted in sudden outbursts of dissolved gases from Lakes Nyos and Monoun in 1986 and 1984, respectively, causing the gas disasters that claimed altogether close to 1800 lives (Sigurdsson et al., 1987; Sigvaldason, 1989). The term "limnic eruption" was coined by J.-C. Sabroux to describe gas outburst from a lake (Halbwachs et al., 2004), and will be used in this article. After the 1986 Lake Nyos gas disaster, many papers were published on the geological, geochemical, limnological, medical, and socio-anthropological aspects of the Nyos and Monoun events, some of which were included in the special issue of Journal of Volcanology and Geothermal Research edited by Sigvaldason (1989).

Follow-up studies of Lakes Nyos and Monoun indicated clearly that CO_2 content in the lakes was increasing at a high rate, unusually high as a geological phenomenon (Evans et al., 1993; Kusakabe et al., 2000). This situation induced scientists working on Lakes Nyos and Monoun to warn of the possible recurrence of a limnic eruption in the near future and to recommend artificial removal of dissolved gases from the lakes (Freeth et al., 1990; Tietze, 1992; Kling et al., 1994; Kusakabe et al., 2000). To achieve this goal, the Nyos-Monoun Degassing Program (NMDP) was set up. After experimental degassing at Lake Monoun (Halbwachs et al., 1993) and Lake Nyos (Halbwachs and Sabroux, 2001; Kusakabe, 2001), a permanent degassing apparatus was installed at Lake Nyos in 2001 and at Lake Monoun in 2003 under NMDP, funded by the U.S. Office of Foreign Disaster Assistance (USAID) and the Cameroonian and French Governments. The controlled degassing is continuing at both lakes. The degassing techniques and construction of the degassing system are described in Halbwachs et al. (2004). There was concern that artificial degassing might trigger another limnic eruption (e.g., Freeth, 1994). Numerical modeling of the evolution of CO_2 under different input conditions (Kantha and Freeth, 1996; McCord and Schladow, 1998; Kusakabe et al., 2000; Schmid et al., 2003, 2006) suggested that destabilization of the water column due to controlled degassing would not be a problem. In accordance with the results of the numerical modeling, the observed chemical structure of the lakes after initiation of the controlled degassing operation indicates that stable stratification has been maintained, and it remains basically the same as the pre-degassing situations at both lakes (Kling et al., 2005). This point will be confirmed by the present work.

Initially the controlled degassing system used a single pipe at each lake. The intake depth of degassing pipe was 203 m at Lake Nyos and 73 m at Lake Monoun. In January 2006, with funding from the French and Cameroonian Governments, two additional pipes were installed at Lake Monoun. At this occasion the intake depth was deepened from 73 m to 93 m (Michel Halbwachs, personal communication). The three pipes accelerated the rate of gas removal from the lake drastically, resulting in considerable deepening of the level of gas-rich water in a short period of time as shown in this paper. The gas removal rate by a single pipe at Lake Nyos, however, is low and insufficient to reduce the gas content to a safe level within several years (Kling et al., 2005). It is still important to keep monitoring the lakes' chemistry in order to know how much gas has been removed and to ascertain a stable stratification to avoid sudden releases of the remaining gas. This monitoring will serve to assess the safety of the lakes in the future. The purpose of the present paper is (1) to present the chemical compositions and CTD results of Lakes Monoun and Nyos obtained during the last 21 years, (2) to show chemical evolution of the lakes before and during controlled degassing, and (3) to assess the safety of these gassy lakes in the future. It is noted that only a limited number of papers have shown the chemical compositions of the lake water (e.g., Kling et al., 1987; Kusakabe et al., 1989; Evans et al., 1993, 1994; Nojiri et al., 1993). This paper compiles the chemical data obtained so far. The results before and during controlled degassing will be described using adjectives "pre-degassing" and "during degassing", respectively.

SAMPLING AND ANALYTICAL METHODS

Water was collected at the center of the lakes using a Niskin water sampler to which a plastic or aluminum gas bag was attached to prevent excessive pressure buildup by exsolving gases inside the sampler (Kusakabe et al., 1989) or by releasing the exsolved gases through a hole of the sampler when the sampler was retrieved to a depth of ca. 10 m. Immediately after water collection, sample water was filtered through a 0.45 μm membrane filter and divided into two fractions, one of which was acidified for cation analysis and another was untreated for anion and Na^+, K^+, NH_4^+ analyses. Mg^{2+}, Ca^{2+}, Fe^{2+}, Mn^{2+} and SiO_2 were analyzed with ICP (Inductively Coupled Plasma spectrometry), and Na^+, K^+, NH_4^+ were analyzed with IC (Ion Chromatography). Anions except HCO_3^- were analyzed with IC. Since the total number of moles of the sum of Cl^-, NO_3^- and SO_4^{2-} was less than 0.5% of that of total cations, essentially HCO_3^- electrically balances cations such as Na^+, K^+, NH_4^+, Mg^{2+}, Ca^{2+} and Fe^{2+}. Thus, the HCO_3^- concentration was calculated as a dif-

ference between the sum of equivalent concentration of cations and that of Cl^-, NO_3^- and SO_4^{2-}. Dissolved silica was assumed to exist as a neutral species.

CO_2 concentration was determined with two methods; the syringe method and the pH method (Kusakabe *et al.*, 2000). In the syringe method, the total dissolved carbonate (=H_2CO_3 + HCO_3^- + CO_3^{2-}) was fixed *in situ* in a plastic syringe containing concentrated solution of KOH and later determined in the laboratory using micro-diffusion analysis. The $CO_{2(aq)}$ (or H_2CO_3) concentration was obtained by subtracting HCO_3^- concentration from the total carbonate concentration. Analytical error of $CO_{2(aq)}$ determination by the syringe method is ±4.5 mmol/kg for $CO_{2(aq)}$ > 40 mmol/kg. For shallow waters which contain little $CO_{2(aq)}$, the syringe method gives increasingly inaccurate results because the titration difference between sample and blank becomes small. For this reason, $CO_{2(aq)}$ data from the pH method was used for waters containing $CO_{2(aq)}$ less than 40 mmol/kg after applying a small correction to the measured pH values to make the results consistent with the syringe CO_2 data. In the pH method, $CO_{2(aq)}$ concentration (or H_2CO_3 activity) was calculated using measured pH and temperature under the assumption that chemical equilibrium had been attained between dissolved carbonate species, i.e.,

$$\log(a_{H_2CO_3}) = -pH + \log(a_{HCO_3^-}) - \log K_1 \quad (1)$$

where K_1 stands for the first dissociation constant of carbonic acid. The activity coefficient of HCO_3^- was calculated using the Debye-Hückel equations. A CTD (Conductivity- Temperature-Depth profiler) provided continuous pH values from the surface to bottom, whereas the HCO_3^- values were obtained only for depths where chemical analysis was done after water sampling. Thus, a regressed relationship between HCO_3^- concentration and electric conductivity normalized to 25°C (abbreviated as C25 hereafter) was used to obtain HCO_3^- concentration at any depth. The temperature coefficient of electric conductivity was assumed to be 2%/°C. Since samples for total dissolved carbonate were collected using a syringe sampler (Kusakabe *et al.*, 2000) at depths slightly different from the depths where water was collected using a Niskin water sampler, the $CO_{2(aq)}$ concentration at a given depth in Table 1 was estimated to the appropriate depth from the relationship between C25 and $CO_{2(aq)}$.

In this paper the $CO_{2(aq)}$ concentrations by the syringe method were used preferentially, unless $CO_{2(aq)}$ data by the syringe method were not available (April 1996, June 2006 and January 2007). In the pH method, the results are influenced considerably by the accuracy of pH measurement. Measured pH values were corrected to align the pH-based $CO_{2(aq)}$ concentrations with the syringe-based
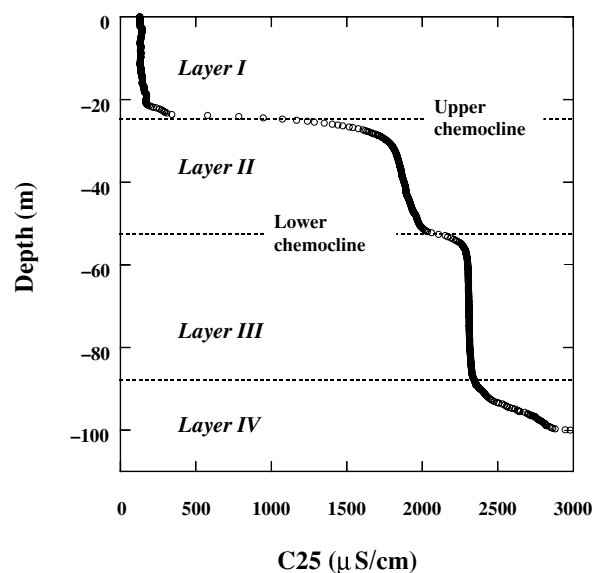

Fig. 1. Chemical structure of Lake Monoun as exemplified by C25 distribution measured in January 2003. Layers I, II and III are bordered by the upper and lower chemoclines. Layer IV is the deepest water characterized by increasing C25 toward the bottom.

$CO_{2(aq)}$ values when both were available. This correction may be justified because $CO_{2(aq)}$ concentrations of waters in mid-depth (80–140 m) of Lake Nyos show little change with time and those below the lower chemocline have become almost constant at Lake Monoun after November 1999. Such waters can be used to "calibrate" the measured pH values. The pH correction applied varied from year to year ranging from –0.13 to +0.18 pH unit. However, the precision of pH measurement is ±0.001, so the shape of $CO_{2(aq)}$ profiles from the corrected pH profiles is reliable. A change of ±0.01 pH unit resulted in a correction of ±0.3 mmol/kg of $CO_{2(aq)}$.

CTD MEASUREMENTS, CHEMICAL COMPOSITION AND CO_2 PROFILES OF LAKE MONOUN

For Lake Monoun we have CTD data in October 1986 (Kanari, 1989), March 1993, April 1996, November 1999, December 2001 and January 2003 as the pre-degassing casts, and in January 2004, January 2005, January 2006, June 2006 and January 2007 as the during-degassing casts. We used an Idronaut Model 316 CTD for most measurements. The CTD results were coupled with CO_2 determinations as stated below. There are too many CTD and CO_2 data to include in this paper, so they are presented only as figures. However, raw and processed data are accessible, for they are stored in a data archive operated by the Geochemical Society of Japan (Kusakabe,

Table 1. Chemical analysis of Lake Monoun, 1986–2006. Concentration is given in mmol/kg (abbreviated as mM/kg).

October 1986

Depth m	Temp. °C	C25 µS/cm	Na^+ mM/kg	K^+ mM/kg	NH_4^+ mM/kg	Mg^{2+} mM/kg	Ca^{2+} mM/kg	Fe^{2+} mM/kg	Mn^{2+} mM/kg	$SiO_{2(aq)}$ mM/kg	Cl^- mM/kg	NO_3^- mM/kg	SO_4^{2-} mM/kg	HCO_3^- mM/kg	$CO_{2(aq)}$ mM/kg	TDS mg/kg
0.0	22.75	109	0.48	0.08		0.17	0.11	0.03	0.00	0.61	0.01	—	0.00	1.17	—	113.2
−15.0	21.19	794	0.63	0.11	0.34	0.53	0.25	1.97	0.02	1.43	0.03	—	0.00	6.59	—	602.8
−25.0	21.18	1259	0.72	0.14	0.62	0.70	0.29	3.40	0.04	1.31	0.04	—	0.00	10.31	—	921.6
−50.0	21.70	1652	0.75	0.13	0.95	0.53	0.25	6.09	0.06	1.36	0.07	—	0.00	15.63	—	1400.7
−75.0	22.61	2199	0.92	0.18	1.41	0.90	0.44	9.67	0.08	1.51	0.07	—	0.00	24.63	—	2185.8
−95.0	22.64	2198	0.96	0.18	1.46	0.95	0.48	10.56	0.08	1.57	0.07	—	0.00	26.66	—	2365.8

Note 1. Data reproduced from Kusakabe et al. (1989).
Note 2. Temperature and C25 were reproduced from Kanari (1989). C25 has been corrected as described in text.

November 1993

Depth m	Temp. °C	C25 µS/cm	Na^+ mM/kg	K^+ mM/kg	NH_4^+ mM/kg	Mg^{2+} mM/kg	Ca^{2+} mM/kg	Fe^{2+} mM/kg	Mn^{2+} mM/kg	$SiO_{2(aq)}$ mM/kg	Cl^- mM/kg	NO_3^- mM/kg	SO_4^{2-} mM/kg	HCO_3^- mM/kg	$CO_{2(aq)}$ mM/kg	TDS mg/kg
0.0	24.796	124.9	0.30	0.07	0.00	0.18	0.19	0.04	0.00	0.29	0.02	0.00	0.00	1.18	0.1	105.4
−15.0	19.866	280.4	0.31	0.07	0.29	0.23	0.22	0.49	0.01	0.37	0.02	0.00	0.00	2.54	0.8	223.5
−40.0	21.660	1703.7	0.74	0.13	1.12	0.90	0.82	9.31	0.06	1.32	0.04	0.00	0.00	24.13	47.4	2131.8
−60.0	22.618	2184.3	0.88	0.14	1.43	1.06	1.07	11.23	0.08	1.62	0.05	0.00	0.00	29.26	129.1	2584.8
−65.0	22.934	2255.1	0.88	0.14	1.53	1.04	1.07	10.94	0.07	1.61	0.05	0.00	0.00	28.74	147.2	2537.7
−85.0	22.984	2263.5	0.93	0.14	1.69	1.11	1.11	11.59	0.08	1.67	0.06	0.00	0.00	30.46	149.3	2688.0
−95.0	23.093	2795.9	0.94	0.14	1.86	1.14	1.17	12.45	0.08	1.76	0.06	0.00	0.00	32.55	148.9	2873.0

Note 3. $CO_{2(aq)}$ concentrations were given as smoothed values of the CO_2 analysis by the syringe method.

December 2001

Depth m	Temp. °C	C25 µS/cm	Na⁺ mM/kg	K⁺ mM/kg	NH₄⁺ mM/kg	Mg²⁺ mM/kg	Ca²⁺ mM/kg	Fe²⁺ mM/kg	Mn²⁺ mM/kg	SiO₂(aq) mM/kg	Cl⁻ mM/kg	NO₃⁻ mM/kg	SO₄²⁻ mM/kg	HCO₃⁻ mM/kg	CO₂(aq) mM/kg	TDS mg/kg
−35	21.705	1846.4	0.81	0.10	0.97	1.06	0.92	9.51	0.07	1.51	0.04	<0.002	0.00	24.95	52.4	2228.6
−45	22.193	1918.3	0.80	0.10	0.95	1.00	0.96	9.36	0.07	1.49	0.05	<0.002	0.00	23.86	65.5	2196.9
−55	23.254	2300.0	1.04	0.12	1.53	1.25	1.33	12.57	0.09	1.93	0.07	0.00	<0.001	31.86	122.5	2945.0
−60	23.292	2305.8	1.05	0.13	1.55	1.25	1.33	12.48	0.09	1.92	0.07	0.00	<0.001	31.67	142.7	2929.3
−65	23.303	2303.5	1.04	0.13	1.54	1.25	1.34	12.44	0.09	1.90	0.07	<0.002	<0.001	31.61	151.8	2921.7
−70	23.307	2305.2	1.04	0.14	1.56	1.28	1.35	12.72	0.09	1.92	0.07	<0.002	<0.001	32.29	152.1	2981.7
−85	23.312	2305.7	1.06	0.15	1.60	1.26	1.34	12.31	0.09	1.92	0.07	<0.002	<0.001	31.47	152.2	2909.6
−95	24.274	3003.0	1.09	0.14	2.01	1.32	1.39	15.52	0.11	2.09	0.07	<0.002	<0.001	38.16	149.8	3532.3

Note 4. $CO_{2(aq)}$ concentrations were given as smoothed values of the CO_2 analysis by the syringe method. The value for 95 m was estimated.

January 2003

Depth m	Temp. °C	C25 µS/cm	Na⁺ mM/kg	K⁺ mM/kg	NH₄⁺ mM/kg	Mg²⁺ mM/kg	Ca²⁺ mM/kg	Fe²⁺ mM/kg	Mn²⁺ mM/kg	SiO₂(aq) mM/kg	Cl⁻ mM/kg	NO₃⁻ mM/kg	SO₄²⁻ mM/kg	HCO₃⁻ mM/kg	CO₂(aq) mM/kg	TDS mg/kg
−10.0	20.239	134.0	0.58	0.09	0.56	0.34	0.32	1.78	0.01	0.57	0.03	0.00	0.00	6.10	0.1	556.3
−20.0	20.052	169.9	0.67	0.11	0.68	0.62	0.51	4.64	0.03	0.96	0.04	0.01	0.00	13.03	0.4	1183.5
−30.0	21.374	1779.7	0.80	0.12	0.99	0.97	0.74	8.31	0.04	1.42	0.04	0.00	0.00	22.04	41.1	1994.1
−40.0	21.920	1883.6	0.83	0.13	1.03	0.97	0.82	9.04	0.04	1.52	0.05	0.00	0.00	23.75	51.6	2150.1
−50.0	22.543	1988.1	0.92	0.14	1.24	0.96	0.94	9.54	0.04	1.58	0.05	0.00	0.00	25.26	73.1	2285.0
−55.0	23.139	2272.8	1.02	0.14	1.56	1.09	1.14	11.48	0.05	1.83	0.07	0.00	0.00	30.22	153.6	2731.3
−75.0	23.333	2311.6	1.04	0.15	1.57	1.12	1.15	11.57	0.05	1.90	0.07	0.01	0.00	30.54	155.5	2762.4
−90.0	23.495	2409.0	1.03	0.14	1.64	1.14	1.20	12.21	0.05	1.90	0.07	0.00	0.00	32.02	154.8	2892.2
−95.0	23.983	2706.6	1.10	0.15	2.05	1.18	1.26	14.40	0.06	2.05	0.07	0.01	0.00	37.10	145.0	3346.4
−100.0	24.260	2935.8	1.11	0.19	2.18	1.18	1.27	16.44	0.07	2.15	0.08	0.01	0.00	41.38	137.6	3733.8

Note 5. $CO_{2(aq)}$ concentrations were given as smoothed values of the CO_2 analysis by the syringe method. The value for 100 m was estimated.

Table 1. (continued)

January 2004

Depth	Temp.	C25	Na⁺	K⁺	NH₄⁺	Mg²⁺	Ca²⁺	Fe²⁺	Mn²⁺	SiO₂(aq)	Cl⁻	NO₃⁻	SO₄²⁻	HCO₃⁻	CO₂(aq)	TDS
m	°C	μS/cm	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mg/kg
0.0	22.690	137.8	0.37	0.06	0.01	0.22	0.25	0.10	0.01	0.83	0.02	0.04	0.00	1.58	*0.3*	182.5
−7.0	20.337	155.9	0.35	0.06	0.01	0.22	0.26	0.18	0.01	0.83	0.02	0.03	0.00	1.73	*0.6*	193.9
−22.9	20.018	165.6	0.34	0.06	0.00	0.21	0.22	0.24	0.01	0.84	0.01	0.00	0.00	1.76	7.7	196.7
−30.0	20.787	878.2	0.65	0.10	0.53	0.84	0.64	5.83	0.05	1.56	0.04	0.00	0.00	15.99	20.7	1473.8
−45.7	21.933	1870.1	0.80	0.12	0.96	1.03	0.96	10.45	0.08	2.05	0.05	0.01	0.00	26.90	55.5	2458.3
−60.0	22.962	2136.2	0.93	0.14	1.28	1.15	1.21	12.08	0.09	2.33	0.06	0.00	0.00	31.40	138.5	2863.4
−80.0	23.472	2344.5	1.00	0.15	1.46	1.23	1.29	13.29	0.09	2.46	0.07	0.00	0.00	34.43	155.1	3135.9
−90.0	23.528	2364.5	0.99	0.15	1.47	1.22	1.32	13.89	0.09	2.48	0.09	0.00	0.00	35.66	156.7	3247.6
−95.4	24.054	2577.4	1.04	0.16	1.84	1.27	1.40	16.57	0.11	2.64	0.08	0.00	0.01	41.72	156.3	3789.7

Note 6. CO₂(aq) concentrations were given as smoothed values of the CO₂ analysis by the syringe method. Italicized numbers were estimated from the pH method in 2003 and 2005.

January 2005

Depth	Temp.	C25	Na⁺	K⁺	NH₄⁺	Mg²⁺	Ca²⁺	Fe²⁺	Mn²⁺	SiO₂(aq)	Cl⁻	NO₃⁻	SO₄²⁻	HCO₃⁻	CO₂(aq)	TDS
m	°C	μS/cm	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mM/kg	mg/kg
0.0	22.218	137.13	—	—	—	0.22	0.22	0.09	0.01	0.30	—	—	—	—	*0.4*	—
−7.0	19.897	140.90	0.37	0.07	0.15	0.22	0.23	0.03	0.01	0.31	0.03	0.00	0.00	1.51	*0.7*	144.7
−22.9	19.632	162.00	0.39	0.07	0.15	0.24	0.24	0.12	0.01	0.32	0.02	0.00	0.00	1.77	*1.3*	166.7
−30.0	19.563	185.65	0.38	0.07	0.18	0.24	0.24	0.22	0.01	0.33	0.04	0.01	0.01	1.97	*1.4*	188.7
−35.0	20.567	996.50	0.72	0.13	0.86	0.86	0.63	5.47	0.05	1.10	0.05	0.01	0.01	15.65	*2.7*	1421.3
−45.7	21.696	1883.10	0.89	0.14	1.19	1.04	0.85	9.54	0.07	1.51	0.07	0.01	0.02	25.12	43.9	2276.4
−50.0	21.881	1913.00	0.88	0.15	1.26	1.01	0.90	9.74	0.07	1.54	0.08	0.02	0.02	25.59	51.5	2323.0
−60.0	22.399	2009.05	0.89	0.15	1.33	1.02	1.00	10.32	0.08	1.57	0.07	0.00	0.00	27.13	76.0	2453.2
−80.0	23.477	2383.40	1.07	0.18	1.87	1.20	1.30	13.22	0.09	1.86	0.09	0.00	0.00	34.64	155.1	3124.2
−95.4	23.841	2645.00	1.12	0.20	2.33	1.27	1.41	17.30	0.11	2.13	0.09	0.00	0.00	43.72	154.1	3940.7

Note 7. CO₂(aq) concentrations were given as smoothed values of the CO₂ analysis by the syringe method. Italicized numbers were from the pH method.

January 2006

Depth m	Temp. °C	C25 µS/cm	Na$^+$ mM/kg	K$^+$ mM/kg	NH$_4^+$ mM/kg	Mg^{2+} mM/kg	Ca^{2+} mM/kg	Fe^{2+} mM/kg	Mn^{2+} mM/kg	SiO$_{2(aq)}$ mM/kg	Cl$^-$ mM/kg	NO$_3^-$ mM/kg	SO$_4^{2-}$ mM/kg	HCO$_3^-$ mM/kg	CO$_{2(aq)}$ mM/kg	TDS mg/kg
0.0	21.787	130.0	0.36	0.07	0.02	0.21	0.22	0.02	0.00	0.30	0.02	0.01	0.00	1.31	*0.4*	125.5
−3.0	20.518	125.7	0.34	0.06	0.02	0.20	0.21	0.01	0.00	0.29	0.02	0.01	0.01	1.24	*0.4*	120.1
−10.0	20.172	129.8	0.34	0.06	0.02	0.20	0.21	0.01	0.00	0.30	0.02	0.01	0.00	1.24	*0.7*	119.7
−15.0	20.058	138.2	0.35	0.06	0.04	0.21	0.21	0.05	0.01	0.31	0.02	0.01	0.01	1.35	*0.8*	130.2
−22.9	19.922	170.6	0.36	0.06	0.05	0.23	0.23	0.18	0.01	0.33	0.02	0.01	0.00	1.71	*1.2*	162.5
−35.0	20.088	441.6	0.46	0.08	0.19	0.44	0.36	1.58	0.02	0.53	0.02	0.01	0.00	5.50	*5.0*	500.7
−35.0	20.088	441.6	0.47	0.07	0.19	0.43	0.41	1.55	0.02	0.63	0.03	0.01	0.01	5.52	*5.0*	509.1
−45.7	21.638	1869.9	0.83	0.14	1.01	1.07	0.91	9.66	0.07	1.66	0.05	0.01	0.01	25.33	49.0	2297.5
−55.0	22.064	1950.6	0.85	0.15	1.06	1.10	1.01	10.51	0.08	1.60	0.06	0.01	0.01	27.36	62.0	2471.6
−61.0	22.474	2025.9	0.88	0.15	1.15	1.07	1.03	10.73	0.08	1.60	0.06	0.01	0.01	27.89	78.9	2518.5
−68.6	23.410	2366.3	1.04	0.18	1.59	1.21	1.27	13.08	0.09	1.94	0.08	0.01	0.01	34.02	155.2	3072.3
−79.6	23.442	2370.7	1.03	0.17	1.59	—	—	—	—	—	0.07	0.01	0.01	—	155.3	—
−90.0	23.451	2373.0	1.03	0.16	1.59	1.22	1.34	13.11	0.09	1.94	0.08	0.01	0.01	34.19	155.3	3085.6
−95.4	23.775	2628.5	1.11	0.19	2.08	1.27	1.40	17.26	0.11	2.39	0.08	0.01	0.01	43.33	147.6	3919.4

Note 8. $CO_{2(aq)}$ concentrations were given as smoothed values of the CO_2 analysis by the syringe method. Italicized numbers were from the pH method.

June 2006

Depth m	Temp. °C	C25 µS/cm	Na$^+$ mM/kg	K$^+$ mM/kg	NH$_4^+$ mM/kg	Mg^{2+} mM/kg	Ca^{2+} mM/kg	Fe^{2+} mM/kg	Mn^{2+} mM/kg	SiO$_{2(aq)}$ mM/kg	Cl$^-$ mM/kg	NO$_3^-$ mM/kg	SO$_4^{2-}$ mM/kg	HCO$_3^-$ mM/kg	CO$_{2(aq)}$ mM/kg	TDS mg/kg
0.0	23.643	174	0.39	0.06	0.01	0.26	0.27	0.39	0.01	0.35	0.02	0.10	0.01	2.21	*0.7*	215.3
−21.0	20.249	181.8	0.34	0.05	0.04	0.24	0.24	0.24	0.01	0.33	0.01	0.01	0.00	1.87	*0.9*	175.2
−26.0	20.105	187.4	0.35	0.05	0.05	0.23	0.23	0.27	0.01	0.32	0.02	0.00	0.00	1.92	*0.9*	179.4
−31.0	19.995	217.6	0.36	0.06	0.07	0.25	0.25	0.39	0.01	0.33	0.02	0.01	0.00	2.27	*1.2*	210.1
−36.0	20.001	285.5	0.40	0.06	0.11	0.31	0.29	0.75	0.02	0.39	0.02	0.01	0.00	3.27	*2.0*	300.2
−41.0	20.222	506.7	0.50	0.07	0.22	0.48	0.39	1.98	0.03	0.55	0.03	0.01	0.00	6.52	*6.6*	591.3
−46.0	20.832	1384.1	0.82	0.12	0.84	1.02	0.79	7.93	0.10	1.25	0.05	0.01	0.01	21.36	*31.7*	1924.6
−51.0	21.554	1841.7	0.85	0.13	1.00	1.10	0.92	9.73	0.11	1.49	0.05	0.01	0.01	25.64	*52.1*	2312.5
−86.0	23.421	2360.6	1.07	0.13	1.60	1.08	1.21	11.83	0.13	1.97	0.07	0.01	0.01	31.17	*154.6*	2825.0
−91.0	23.427	2361.8	1.06	0.15	1.56	1.12	1.23	12.32	0.13	2.00	0.07	0.01	0.01	32.26	*154.1*	2922.4
−96.5	23.981	2725.8	1.15	0.17	2.29	1.25	1.42	18.63	0.18	2.27	0.08	0.01	0.01	46.48	*150.7*	4189.2

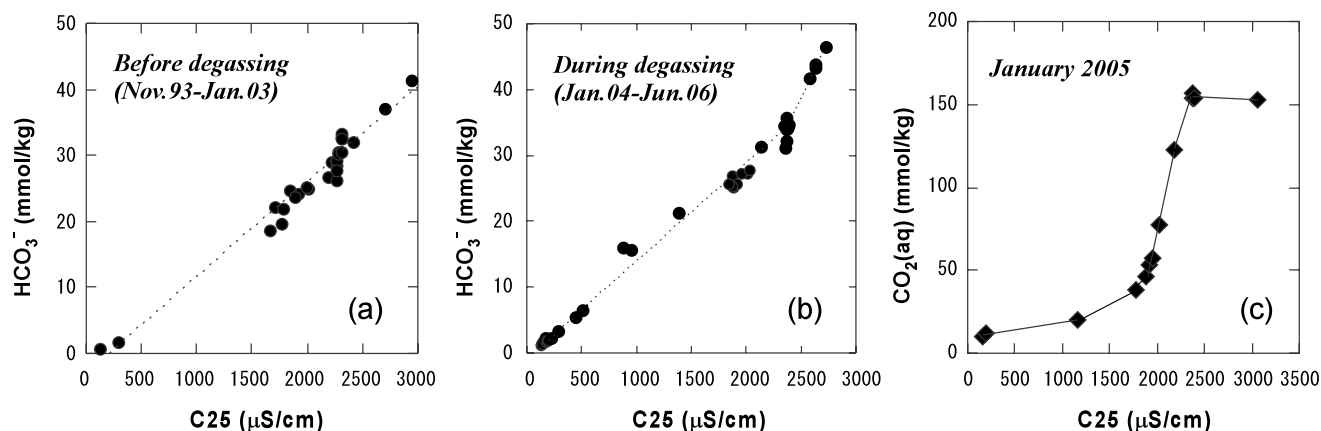Note 9. $CO_{2(aq)}$ concentrations were from the pH method.

Fig. 2. (a) Relationship between C25 and HCO$_3^-$ for waters collected before controlled degassing (November 1993, December 2001 and January 2003) at Lake Monoun. (b) Relationship between C25 and HCO$_3^-$ for during-degassing waters (January 2004, January 2005, January 2006 and June 2006). (c) A typical relationship between CO$_{2(aq)}$ and C25. The data points were connected from a segment to segment by appropriate linear or quadratic curves to smooth the data.

2007).

Table 1 shows the chemical composition of Lake Monoun collected at different depths at different times (1993–2006). Obviously CO$_{2(aq)}$ and HCO$_3^-$ show overwhelmingly the highest concentrations. The cation composition of Lake Monoun water is characterized by high concentrations of Fe^{2+} with subordinate concentrations of Mg^{2+} and Ca^{2+}. These metal elements derive from subsurface interaction of basaltic and peridotitic components with carbonic acid (Kusakabe *et al.*, 1989; Tuttle *et al.*, 1992). Unusually high Fe^{2+} concentration in deep waters has been attributed to reduction of laterite that was brought into the lake by the inflowing river and aeolian transport (Sigurdsson *et al.*, 1987).

The chemical structure of Lake Monoun is best shown by a depth-C25 relationship (Fig. 1, measured in January 2003). Lake Monoun can be divided into 4 layers. In January 2003 layer I is the shallowest, well-mixed, low conductivity water down to 23 m. It is separated by a sharp upper chemocline at 23 m, which is underlain by layer II that extends down to 51 m, where the lower chemocline develops. Below the lower chemocline, a well-mixed layer III continues down to *ca.* 85 m. Below this depth, conductivity (and temperature) increases steadily toward the bottom (layer IV). The C25 profile shown in Fig. 1 has changed with time, especially after initiation of controlled degassing. A general pattern, however, has remained similar as controlled degassing has proceeded.

Several steps were taken to obtain CO$_2$ profiles that form the basis for estimating the natural recharge and removal of CO$_2$ from the lake. Firstly, the measured HCO$_3^-$ concentrations were smoothed by correlating to C25. Figure 2a shows the pre-controlled degassing relationship between C25 and HCO$_3^-$ for waters collected in November 1993, December 2001 and January 2003. HCO$_3^-$ concentration is linearly correlated to C25. Although considerable scatter around the regression line can be seen, we believe the regression line represents the overall C25–HCO$_3^-$ relationship prior to the controlled degassing. The C25–HCO$_3^-$ relationship during-degassing for waters collected in January 2004, January 2005, January 2006 and June 2006 is shown in Fig. 2b. HCO$_3^-$ concentration is linearly correlated to C25, but the slope changes considerably to a higher value for waters with C25 > 2400 μS/cm. High C25 waters collected during controlled degassing tend to contain more Fe^{2+} than the other waters (Table 1). Calculations using a commercially available speciation program (REACT in Geochemist's Workbench 3.0, Bethke, 1996) indicate that up to 30% of total Fe exists as FeHCO$_3^-$, 20% of Ca as CaHCO$_3^-$, and 15% of Mg as MgHCO$_3^-$. Existence of these ion pairs would reduce C25 when compared to C25 of the solution in which Fe, Ca and Mg exist as free Fe^{2+}, Ca^{2+} and Mg^{2+} ions, since these ion pairs are monovalent and therefore would carry less electric conductivity per mole than unpaired ions, although molar ionic conductivity of the above ion pairs are not known. This effect may be partly responsible for a higher slope in the HCO$_3^-$–C25 relationship of deep waters having C25 > 2400 μS/cm. The slope of the regression for waters with C25 < 2400 μS/cm collected during controlled degassing is almost the same as that of the pre-degassing relationship. Enhanced Fe^{2+} concentrations of during-degassing deep waters were probably caused by dissolution of Fe(OH)$_3$ precipitates, which formed by oxidation of Fe^{2+} when Fe^{2+}-rich deep water from the degassing pipe was oxidized at the surface upon contact with the atmospheric oxygen through a reaction
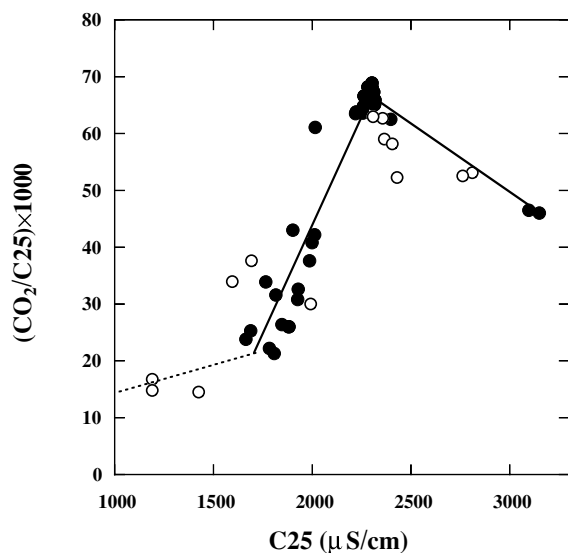
Fig. 3. Relationship between $CO_{2(aq)}/C25$ ratios and C25 values for pre-degassing waters (November 1993 to January 2003) at Lake Monoun. Two linear relationships, shown by solid lines, can be seen; one for waters with $1700 < C25 < 2300$ $\mu S/cm$, and another for waters with $C25 > 2300$ $\mu S/cm$. See text for regression equations. Open circles are for May 1987, December 1989 and April 1992. CO_2 concentration was measured with the cylinder method (Evans et al., 1993). Since no CTD data is available during these periods, C25 values were indirectly estimated using a $C25–SiO_{2(aq)}$ relationship observed during the pre-degassing period (November 1993 to January 2003). The $CO_{2(aq)}/C25–C25$ relationship for waters with $C25 < 1700$ was assumed as shown by a dotted line.

$$Fe^{2+} + 2HCO_3^- + \tfrac{1}{4}O_2 + \tfrac{1}{2}H_2O \rightarrow Fe(OH)_3 + 2CO_{2(g)}. \tag{2}$$
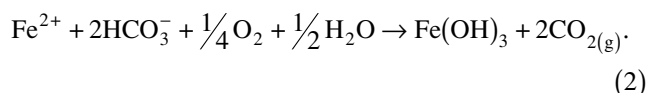
The precipitates sank to the deep anoxic zone (layers III and IV) where they were reduced to Fe^{2+} by reaction with organic matter. This interpretation may be supported by a constant Fe^{2+}/NH_4^+ ratio of waters in layers III and IV. Thus, we used the following regression equations for the pre- and during-degassing $HCO_3^-–C25$ relationships,

$$HCO_3^- = 0.01444*C25 - 2.86$$
$$\text{(pre-degassing, Fig. 2a)} \tag{3a}$$

$$HCO_3^- = 0.01449*C25 - 0.48 \ (100 < C25 < 2400)$$
$$\text{(during-degassing, Fig. 2b)} \tag{3b}$$

$$HCO_3^- = 0.03460*C25 - 47.61 \ (2400 < C25 < 2800)$$
$$\text{(during-degassing, Fig. 2c).} \tag{3c}$$

HCO_3^- concentration (in mmol/kg) calculated using Eqs. (3a) through (3c) was subtracted from the observed total

CO_2 concentration ($CO_{2(aq)} + HCO_3^-$ fixed in a syringe) to work out $CO_{2(aq)}$ concentration. Next, the $CO_{2(aq)}$ concentrations thus determined by the syringe method were correlated to C25 to obtain a smooth $CO_{2(aq)}$ profile. A typical correlation between $CO_{2(aq)}$ and C25 is shown in Fig. 2c. The data points were connected from segment to segment by appropriate linear or quadratic curves to smooth the data. This practice was applied to all observations of total CO_2 determinations by the syringe method from November 1993 to June 2006.

The earliest CTD measurement of Lakes Nyos and Monoun was made in October 1986 (Kanari, 1989). Comparison of the C25 data with those measured at Lake Nyos in November 1986 (Tietze, 1987) and later dates at Lake Nyos suggested that Kanari's C25 data appeared too low approximately by 25 $\mu S/cm$. After correcting the C25 data, the $CO_{2(aq)}$ profile in October 1986 was estimated as follows. If $CO_{2(aq)}/C25$ ratios obtained during the pre-degassing periods (November 1993 to January 2003) are plotted against C25, a linear correlation can be seen for waters having C25 between 1700 and 2300 $\mu S/cm$ (Fig. 3). A regression analysis gave the following equation for the waters with C25 between 1700 and 2300 $\mu S/cm$

$$Y = -111.9 \ (\pm 7.9) + 0.078 \ (\pm 0.004)*X \ (R^2 = 0.93) \tag{4a}$$

where Y stands for $CO_{2(aq)}/C25*1000$ and X for C25. Another linear relationship with a reversed slope exists for deep waters with C25 greater than 2260 $\mu S/cm$

$$Y = 123.59 - 0.0248*X. \tag{4b}$$

It was assumed that both $CO_{2(aq)}/C25*1000$ and C25 decrease linearly to the point of origin for layer II waters with C25 < 1700 $\mu S/cm$ (Fig. 3). These linear relationships were used to estimate a $CO_{2(aq)}$ profile for October 1986. Since no CTD data is available between 1987 and 1992 for Lake Monoun, we estimated C25 values of this period using a relationship between pre-degassing C25 and $SiO_{2(aq)}$ (Table 1) and $SiO_{2(aq)}$ analysis of pre-November 1993 waters (W. C. Evans, unpublished data). The estimated C25 combined with $CO_{2(aq)}$ values measured by the cylinder method (Evans et al., 1993, and unpublished data) confirmed the relationship in Fig. 3.

Temperature, C25, pH and CO_2 profiles of Lake Monoun before controlled degassing

Chemical evolution of Lake Monoun water before the controlled degassing is shown in Fig. 4. The temperature profiles (Fig. 4a) show minima at around 20 m, the depth of which changed with year and season when measurements were made. Below the minima, temperature increases gradually to *ca.* 23°C down to the lower chemocline at 50–60 m, remains constant down to 93 m,
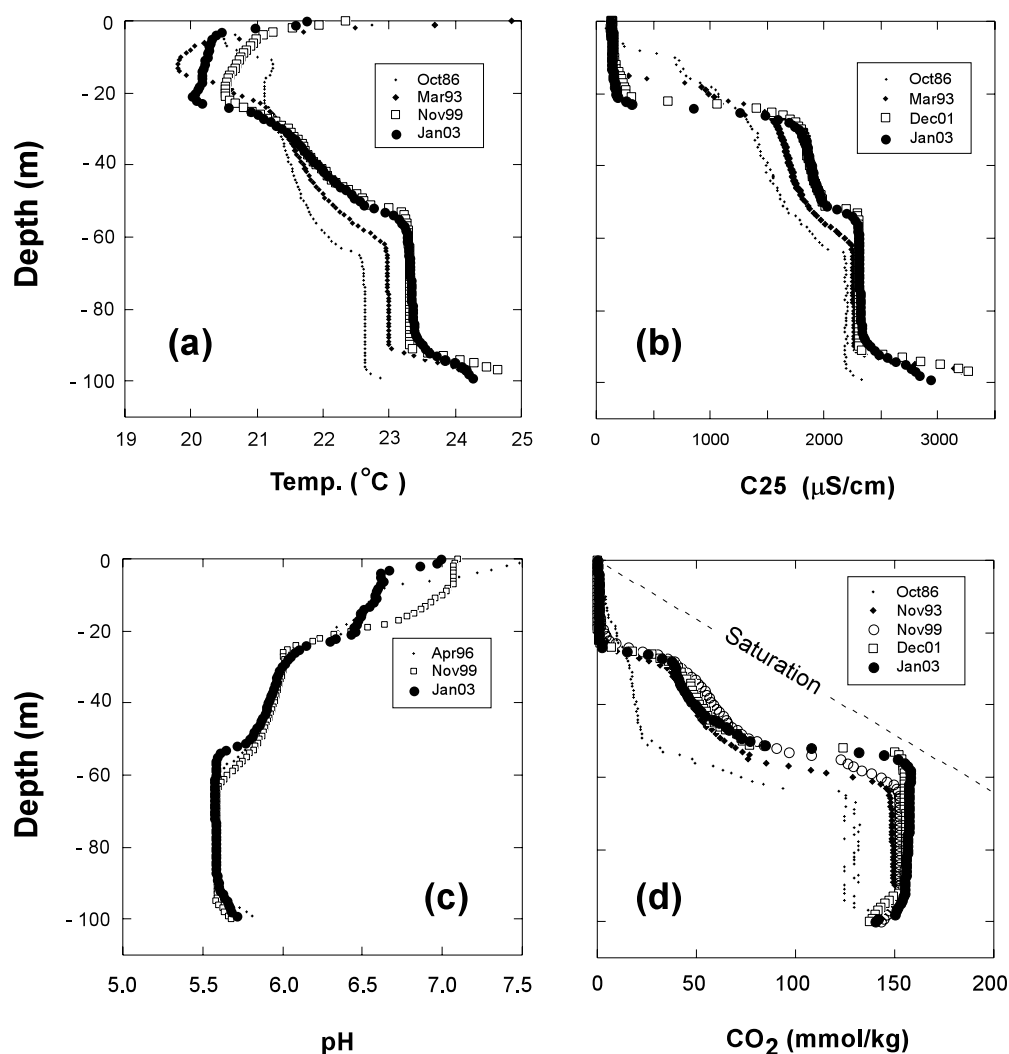
Fig. 4. Evolution of temperature (a), C25 (b), pH (c) and CO$_{2(aq)}$ (d) at Lake Monoun before controlled degassing (October 1986–January 2003). CO$_2$ saturation curve was calculated from Duan and Sun (2003).

and increases sharply to >24°C toward the bottom. It is noted that the temperature of water below layer II increased significantly between 1986 and 1999, and at the same time the layer III (thermally homogeneous zone) thickened forming a "shoulder" at a depth of 51 m by January 2003. Figure 4b shows the conductivity profiles of 1986, 1993, 2001 and 2003. An upper chemocline at 8 m in 1986 deepened to 23 m by 1999 (not shown) and remained unchanged until 2003. Below the upper chemocline, conductivity increased gradually until around 60 m depth, and stayed constant down to 90 m. Similar to the temperature profiles, a clear "shoulder" of conductivity formed at 51 m in 2003, shallower by 8 m than in 1993, which indicates thickening of homogeneous layer III having higher temperature and more salinity. In the very bottom water (layer IV, 90 m and below) conductiv-

ity increases sharply toward the bottom. The evolution of pH values is shown in Fig. 4c. Shallow water pH values varied with time, reflecting different rates of CO$_2$ consumption by algae in surface waters. Below the upper chemocline, pH decreases gradually until layer III is reached at around 60 m, and stays constant in layer III as is the case for temperature and conductivity. The pH values increase slightly toward the bottom in layer IV. CO$_{2(aq)}$ profiles obtained through the procedure previously described are collectively shown in Fig. 4d for the pre-controlled degassing period (October 1986–January 2003). Obviously the 1986 profile shows the lowest CO$_{2(aq)}$ concentrations in deep water (around 130 mmol/kg in layers III and IV) and the CO$_2$ shoulder at a depth of *ca*. 63 m. It is interesting to note that the CO$_{2(aq)}$ profiles evolved with time, but the greatest change appears
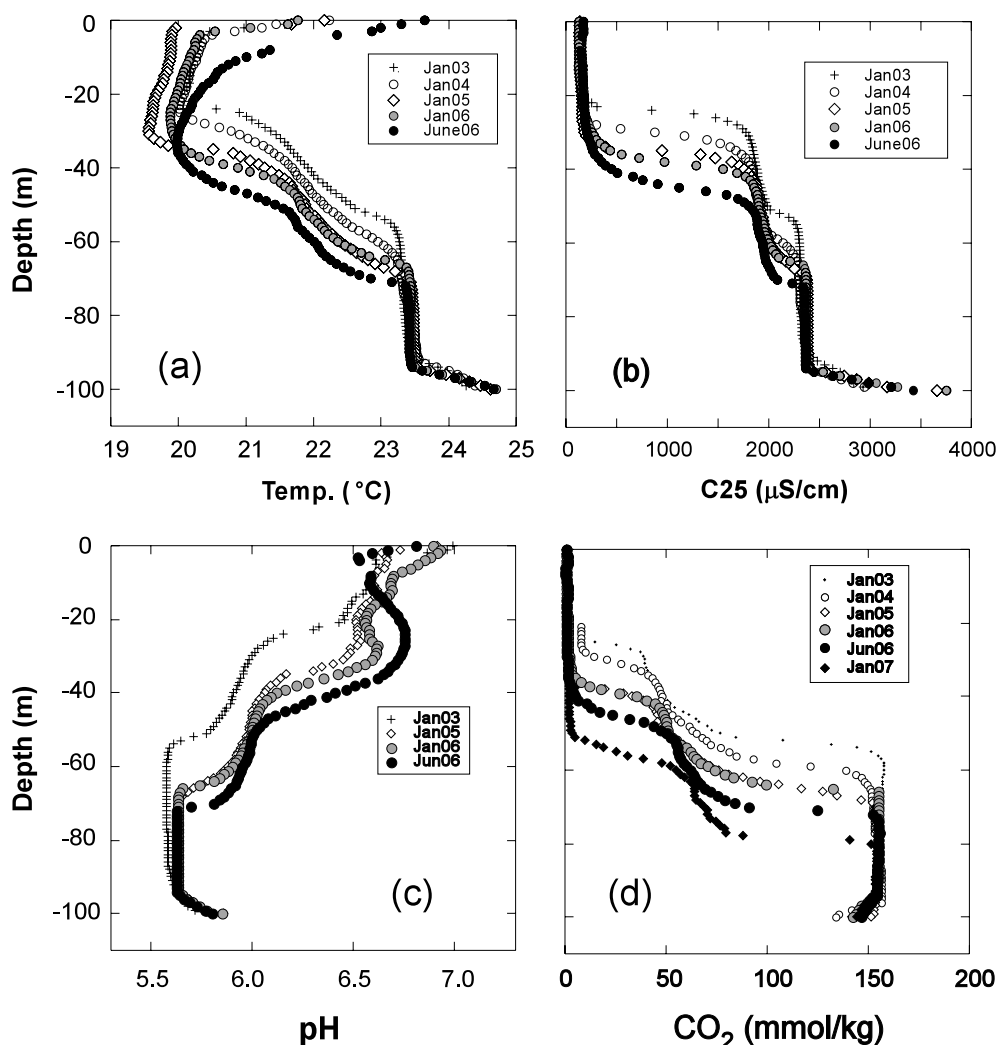
Fig. 5. Evolution of temperature (a), C25 (b), pH (c) and CO$_{2(aq)}$ (d) during controlled degassing (January 2003–January 2007) at Lake Monoun.

to have taken place between 1986 and 1993. Evolution after November 1999 seems slowed down. Layers III and IV expanded with time due to recharge of CO$_2$-rich water from the bottom, and the maximum CO$_{2(aq)}$ concentration reached 157 mmol/kg at 58 m depth in January 2003. A CO$_2$ profile close to the one of January 2003 was already attained in December 2001. The CO$_2$ shoulder at 58 m was very close to the CO$_2$ saturation curve at the depth. Since the depth of CO$_2$ saturation at 157 mmol/kg is 50 m (using the solubility equations in Duan and Sun, 2003), we had a saturation distance of only 8 m in January 2003. This was critical because spontaneous exsolution of CO$_2$ bubbles capable of triggering another limnic eruption could have occurred in a very short time if the degassing operation started later than 2003 (Kling *et al.*, 2005).

Temperature, C25, pH and CO$_2$ profiles of Lake Monoun during controlled degassing

Figure 5 shows evolution of temperature (a), conductivity (b), pH (c) and CO$_2$ (d) profiles at Lake Monoun during controlled degassing (January 2003–January 2007). Compared to the January 2003 profiles which are shown as the reference for the period just before controlled degassing started, all profiles maintained a similar shape but subsided considerably as degassing proceeded. Noticeable deepening of the upper and lower chemoclines with reduction of layer III thickness took place during the first 2 years (2003–2005) of degassing. However, little change was observed during 2005 and 2006, which indicates that performance of the degassing system declined during that period and that gas removal rate and recharge rate started balancing as predicted by a model
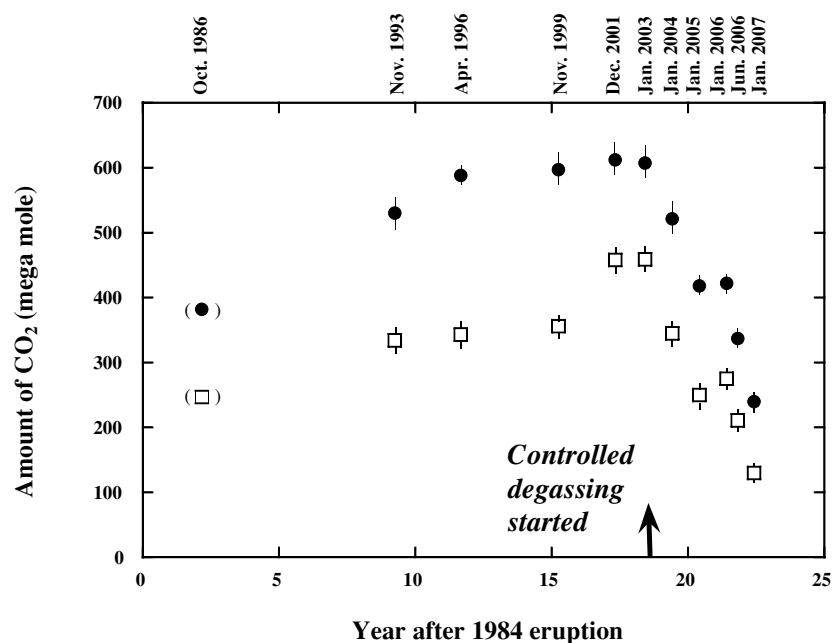
Fig. 6. Change with time in CO_2 content at Lake Monoun. Closed circles and open squares are for CO_2 content below the upper chemocline and below the lower chemocline, respectively. Error bars indicate the standard error associated with the estimation of each data point.

calculation (Kling *et al.*, 2005). After February 2006 when 2 additional degassing pipes were installed with intake depth deepened to 93 m (Halbwachs, personal communication), appreciable and rapid deepening of layer III resumed (Fig. 5, January 2006–January 2007).

A temperature minimum at 21 m in January 2003 deepened to 33 m in June 2006 resulting in expansion of layer I and lessening of sharpness of the upper thermocline (Fig. 5a). The lower chemocline deepened by 19 m from 51m in Jan 2003 to 70 m in June 2006 with associated deepening of the shoulders of temperature, conductivity, pH and CO_2. The meandering shape of shallow water pH profiles after 2005 reflects mainly changes in temperature, photosynthesis, and respiration that control CO_2 concentrations.

From the changes observed during 2003 and 2006 (Fig. 5), we can evaluate the change in CO_2 content over time and gas removal rate during controlled degassing as shown below. Note that the shape of each curve in Fig. 5 has remained similar during controlled degassing, indicating that layer III water has been carried up and added to the surface by the degassing pipe without changing a major chemical structure of the lake.

RECHARGE AND REMOVAL OF CO_2 AT LAKE MONOUN

The change in CO_2 content with time was quantitatively evaluated from the $CO_{2(aq)}$ profiles before control-

led degassing (Fig. 4d) and during controlled degassing (Fig. 5d). It is summarized in Table 2 and graphically shown in Fig. 6. The bathymetry used in Kling *et al.* (2005) was adopted to estimate the CO_2 content. In Table 2 the CO_2 contents of Lake Monoun were calculated for the main basin of the lake (total CO_2) and for waters below the surface chemocline (CO_2 below layer II) and below the deep chemocline (CO_2 below layer III). Since the water of Lake Monoun shallower than *ca.* 15 m is replenished by the Panke River, it is reasonable to discuss the change of CO_2 content below layer II in the main basin. The CO_2 content below layer II soon after the 1984 eruption is estimated to be approximately 330 mega moles (Fig. 6). It appears that the CO_2 content increased rapidly until 1996. Considering the fact that the CO_2 content in October 1986 was based on $CO_{2(aq)}$ concentrations estimated in an indirect way as mentioned previously, the overall rate of CO_2 accumulation below layer II is best calculated to be 8.4 ± 3.6 mega mole CO_2 per year for the pre-degassing period of 1993 to 2003 (Table 2). The CO_2 content below layer III also increased until January 2003 with the CO_2 accumulation rate of about 14 mega mole CO_2 per year. Kling *et al.* (2005) calculated a CO_2 recharge rate of 8.2 ± 1.5 mega mole CO_2 per year using data below layer II from 1992 to 2003. The value is consistent with the present value obtained using the syringe CO_2–C25 relationship. The recharge of CO_2 with this rate pushed the depth of lower chemocline gradually upward

Table 2. Change with time in CO_2 content of Lake Monoun

Date	Year after Aug. 1984	Total CO_2 mega mole	CO_2 below layer II mega mole	CO_2 below layer III mega mole	CO_2 accumulation rate mega-mole/yr	CO_2 removal rate mega-mole/yr
Pre-degassing						
October 1986	2.17	*383*	*382*	*288*	—	—
November 1993	9.25	531 ± 23	530 ± 23	334 ± 11	—	—
April 1996	11.67	*593 ± 9*	*588 ± 9*	*343 ± 9*	—	—
November 1999	15.25	603 ± 26	597 ± 26	356 ± 10	—	—
December 2001	17.33	612 ± 17	612 ± 17	458 ± 9	—	—
January 2003	18.42	608 ± 24	607 ± 24	459 ± 13	8.4 ± 3.6 (1993–2003)	—
Post-degassing						
January 2004	19.42	533 ± 23	521 ± 23	345 ± 10	—	—
January 2005	20.42	421 ± 17	418 ± 17	250 ± 7	—	
January 2006	21.42	430 ± 13	422 ± 12	275 ± 6	—	61.7 ± 8.9 (2003–2006)
June 2006	21.83	*351 ± 9*	*337 ± 8*	*211 ± 5*	—	—
January 2007	22.42	*254 ± 6*	*240 ± 5*	*130 ± 3*	—	182.0 ± 13.0 (2006–2007)

Note 1. The values for October 1986 were estimated using the CO_2/C25 vs. C25 relation ship (see text). Errors were not attached due to the assumptions involved.
Note 2. Italicized figures were obtained with the pH-CO_2 method.
Note 3. Errors associated with the figures were calculated for the uncertainties of ± 4.5 mmole $CO_{2(aq)}$/kg for the syringe method and that of ± 0.01 pH unit in the pH-CO_2 method.

from 63 m in October 1986 to 51 m in January 2003 (Fig. 4). As stated earlier the $CO_{2(aq)}$ concentration at 58 m depth in January 2003 was close to saturation. If the CO_2 profile before the 1984 event was similar to the profile in January 2003, natural recharge of CO_2 would have made the water at that depth saturated with CO_2, leading to spontaneous exsolution of gas bubbles and a limnic eruption. Although we have no data on the natural CO_2 recharge rate at that time, it could be higher than 8.4 mega moles CO_2 per year. In this scenario no external force is required to trigger a limnic eruption.

The effect of controlled degassing is remarkable (Fig. 6). The CO_2 content below layer II in January 2003, just before the degassing operation started, was 607 mega moles. It dropped to 521 mega moles in January 2004, and to 422 mega moles in January 2006, with a mean gas removal rate of 62 mega moles per year by a single pipe. This rate is slightly less than the initial gas removal rate of 86 mega moles per year calculated from the CO_2 contents below layer II in January 2003 and January 2004, but much greater than the recent natural recharge of ~8 mega moles per year. The observed pattern of the CO_2 content after 2003 is, generally, in good agreement with the prediction of model calculations by Kling *et al.* (2005). Between January 2005 and January 2006, the gas content below layer II remained unchanged. This observation is consistent with a model prediction (Kling *et al.*, 2005) that the rates of gas removal and recharge balance after 2 years of controlled degassing with the pipe intake at 73

m depth. The gas removal rate was also lowered by malfunctioning of the degassing pipe during that period. However, natural recharge of CO_2 continued as shown by the increase in the CO_2 content below layer III from 250 to 275 mega moles. After April 2006 when 3 pipes started extracting water from 93 m depth, CO_2 contents below layers II and III reduced drastically to 240 and 130 mega moles, respectively, as observed in January 2007. This reduction of the CO_2 contents translates into a gas removal rate of 182 mega moles per year. The accelerated reduction of the CO_2 content is similar to the model prediction which used two pipes. The observed pattern of reduction of the CO_2 content agrees with the prediction, although there was a delay of ~1 year in the initiation of actual degassing by three pipes compared to the model calculations.

If the current degassing systems keep working, gases dissolved in layer III (water between 80 to 92 m) will be removed by the end of 2007 since the amount of CO_2 there in January 2007 is 130 mega moles. After this, the $CO_{2(aq)}$ profile would be similar to the one in January 2007 (Fig. 5d) but with the lowest chemocline having deepened to 93 m. The gas content below 93 m will be unchanged at 24 mega moles unless the intake depth is deepened. $CO_{2(aq)}$ concentration coming into the pipes will gradually reduce from ~80 mmol/kg (Fig. 5d) to lower concentrations. This would result in a much smaller rate of gas removal leading to stoppage of self-siphoning from the pipe. After this stage has been reached, further deepen-

Table 3. Chemical composition of Lake Nyos, 1988–2006. Concentration is given in mmol/kg (abbreviated as mM/kg).

October 1986[*1]

Depth m	Temp.[*2] °C	C25[*2] μS/cm	Na+ mM/kg	K+ mM/kg	NH4+ mM/kg	Mg2+ mM/kg	Ca2+ mM/kg	Fe2+ mM/kg	Mn2+ mM/kg	SiO2(aq) mM/kg	Cl- mM/kg	NO3- mM/kg	SO4 2- mM/kg	HCO3- mM/kg	CO2(aq) mM/kg	TDS mg/kg
0.0	25.16	140	0.15	0.06	0.06	0.37	0.17	0.00	0.001	0.29	0.006	—	0.001	1.3	*3.0*	123.0
−7.0	23.34	341	0.18	0.08	0.12	0.58	0.26	0.03	0.012	0.39	0.006	—	0.001	2.1	*8.0*	189.3
−40.0	22.83	512	0.55	0.10	0.23	1.36	0.44	0.38	0.015	0.47	0.014	—	0.001	5.2	*36.0*	440.9
−80.0	22.92	647	0.58	0.12	0.28	1.60	0.55	0.54	0.018	0.50	0.016	—	0.000	6.4	*61.0*	534.6
−130.0	23.15	852	0.71	0.15	0.37	2.18	0.70	0.88	0.021	0.57	0.019	—	0.000	8.8	*106.0*	729.9
−200.0	—	—	0.77	0.17	0.44	2.63	0.77	0.98	0.025	0.66	0.017	—	0.000	10.2	*151.0*	845.4

[*1]Data copied from Kusakabe et al. (1989). CO2(aq) in italic was estimated using the relationship in Fig. 9.
[*2]Temperature and C25 values were taken from Kanari (1989). C25 values have been corrected as described in text.

December 1988[*3]

Depth m	Temp.[*2] °C	C25[*2] μS/cm	Na+ mM/kg	K+ mM/kg	NH4+ mM/kg	Mg2+ mM/kg	Ca2+ mM/kg	Fe2+ mM/kg	Mn2+ mM/kg	SiO2(aq) mM/kg	Cl- mM/kg	NO3- mM/kg	SO4 2- mM/kg	HCO3- mM/kg	CO2(aq) mM/kg	TDS mg/kg
0.0	24.519	87.7	0.15	0.03	0.00	0.25	0.15	0.00	0.000	0.24	0.005	0.000	0.002	1.0	*1.0*	89.8
−10.0	23.740	87.8	0.15	0.03	0.00	0.25	0.15	0.00	0.000	0.24	0.005	0.000	0.001	1.0	*1.0*	91.0
−20.0	22.900	139.0	0.19	0.04	0.00	0.36	0.20	0.00	0.001	0.27	0.005	0.022	0.001	1.3	*1.0*	121.9
−30.0	22.453	278.7	0.26	0.05	0.04	0.60	0.32	0.01	0.019	0.34	0.006	0.007	0.000	2.2	*7.0*	194.7
−50.0	22.776	604.1	0.48	0.10	0.07	1.43	0.71	0.89	0.020	0.57	0.006	0.001	0.000	6.7	64.3	576.3
−75.0	22.858	669.5	0.54	0.10	0.24	1.63	0.79	1.01	0.021	0.60	0.006	0.000	0.000	7.8	77.6	660.6
−100.0	22.974	762.4	0.61	0.11	0.27	1.87	0.90	1.18	0.023	0.66	0.007	0.000	0.000	8.9	97.1	756.4
−125.0	23.120	873.3	0.69	0.12	0.33	2.16	1.04	1.40	0.027	0.74	0.007	0.000	0.000	10.4	120.3	879.2
−150.0	23.238	934.4	0.75	0.13	0.37	2.34	1.12	1.52	0.028	0.78	0.007	0.000	0.000	11.2	132.0	950.3
−175.0	23.309	955.8	0.74	0.13	0.36	2.38	1.12	1.53	0.028	0.79	0.007	0.000	0.000	11.3	*140.0*	959.0
−190.0	23.677	1061.5	0.83	0.15	—	2.71	1.28	1.69	—	0.87	0.008	0.000	0.000	12.8	*173.0*	1078.1
−200.0	24.241	1342.5	1.10	0.20	0.44	3.73	1.73	2.28	0.031	1.09	0.007	0.000	0.000	17.3	259.0	1449.6

[*3]Data copied from Nojiri et al. (1993). CO2(aq) was measured with the syringe method. CO2(aq) in italic was estimated using the relationship in Fig. 9.

*Italic: November 1993[*4]*

Depth m	Temp.[*2] °C	C25[*2] μS/cm	Na+ mM/kg	K+ mM/kg	NH4+ mM/kg	Mg2+ mM/kg	Ca2+ mM/kg	Fe2+ mM/kg	Mn2+ mM/kg	SiO2(aq) mM/kg	Cl- mM/kg	NO3- mM/kg	SO4 2- mM/kg	HCO3- mM/kg	CO2(aq) mM/kg	TDS mg/kg
0.0	25.770	70.3	0.13	0.03	0.00	0.14	0.09	0.00	0.000	0.26	0.01	0.00	0.00	0.7	*0.0*	62.5
−30.0	22.046	96.9	0.16	0.04	0.00	0.24	0.13	0.00	0.001	0.30	0.01	0.00	0.00	1.1	*0.4*	93.3
−50.0	22.400	617.3	0.48	0.11	0.22	1.57	0.73	0.96	0.024	0.67	0.01	0.00	0.00	8.6	53.6	686.8
−100.0	22.997	783.7	0.65	0.12	0.33	2.10	0.95	1.21	0.024	0.79	0.02	0.00	0.00	11.3	91.2	896.4
−150.0	23.320	958.1	0.74	0.14	0.43	2.61	1.17	1.45	0.028	0.94	0.02	0.00	0.00	13.9	143.6	1096.8
−190.0	24.228	1208.2	0.82	0.16	0.47	3.26	1.35	1.83	0.030	1.17	0.02	0.00	0.00	16.8	222.5	1330.4
−205.0	24.986	1529.1	1.19	0.22	0.57	4.46	1.92	2.71	0.034	1.43	0.02	0.00	0.00	23.6	320.6	1866.7

[*4]*Data copied from Tanyileke (1994). CO2(aq) was measured with the syringe method. CO2(aq) in italic was estimated using the smoothed syringe CO2–C25 relationship.*

*December 2001[*5]*

Depth m	Temp.[*2] °C	C25[*2] μS/cm	Na+ mM/kg	K+ mM/kg	NH4+ mM/kg	Mg2+ mM/kg	Ca2+ mM/kg	Fe2+ mM/kg	Mn2+ mM/kg	SiO2(aq) mM/kg	Cl- mM/kg	NO3- mM/kg	SO4 2- mM/kg	HCO3- mM/kg	CO2(aq) mM/kg	TDS mg/kg
−165.0	23.604	1007.6	0.79	0.10	0.48	2.71	1.19	1.56	0.032	1.04	0.02	0.01	0.00	12.3	135.2	1047.6
−170.0	23.680	1024.6	0.77	0.11	0.46	2.76	1.19	1.64	0.032	1.05	0.02	0.00	0.00	12.6	*155.2*	1068.9
−175.0	23.765	1042.6	0.77	0.11	0.46	2.84	1.21	1.66	0.032	1.08	0.02	0.00	0.00	12.8	*159.3*	1089.3
−177.5	23.861	1064.7	0.80	0.13	0.46	2.94	1.23	1.67	0.032	1.09	0.02	0.00	0.00	13.1	*163.1*	1114.4
−180.0	23.986	1098.0	0.80	0.12	0.47	3.19	1.28	1.74	0.033	1.15	0.02	0.00	0.00	13.8	175.0	1176.4
−182.5	24.142	1133.7	0.83	0.14	0.47	3.24	1.31	1.75	0.033	1.17	0.02	0.00	0.00	14.1	180.5	1196.4
−185.0	24.345	1181.3	0.86	0.13	0.50	3.39	1.37	1.85	0.033	1.20	0.02	0.00	0.00	14.7	206.4	1251.7
−187.5	24.669	1298.9	0.93	0.15	0.53	3.67	1.49	2.08	0.034	1.25	0.02	0.00	0.00	16.1	250.6	1366.5
−190.0	25.036	1424.8	1.08	0.19	0.62	4.20	1.70	2.50	0.038	1.37	0.02	0.01	0.00	18.7	336.1	1585.7
−195.0	25.178	1444.2	1.06	0.16	0.59	4.26	1.72	2.58	0.039	1.39	0.02	0.00	0.00	19.0	342.7	1605.0
−200.0	25.200	1451.6	1.08	0.17	0.61	4.29	1.72	2.62	0.039	1.39	0.02	0.00	0.00	19.1	350.1	1619.3
−205.0	25.230	1474.5	1.08	0.16	0.58	4.29	1.75	2.65	0.039	1.36	0.02	0.00	0.00	19.2	335.9	1626.5
−208.0	25.377	1678.0	1.11	0.17	0.63	4.62	1.81	3.21	0.043	1.50	0.02	0.00	0.00	21.2	333.3	1801.1

[*5]*Italicized CO2(aq) values were estimated from the smoothed syringe CO2–C25 relationship.*

Table 3. (continued)

January 2003*6

Depth m	Temp.*2 °C	C25*2 µS/cm	Na+ mM/kg	K+ mM/kg	NH4+ mM/kg	Mg2+ mM/kg	Ca2+ mM/kg	Fe2+ mM/kg	Mn2+ mM/kg	SiO2(aq) mM/kg	Cl- mM/kg	NO3- mM/kg	SO4 2- mM/kg	HCO3- mM/kg	CO2(aq) mM/kg	TDS mg/kg
−30.0	21.868	98.5	0.22	0.04	0.01	0.23	0.13	0.02	0.003	0.25	0.01	0.01	0.00	1.0	0.7	96.4
−50.0	21.815	100.1	0.23	0.03	0.01	0.24	0.13	0.02	0.005	0.26	0.01	0.01	0.00	1.1	1.3	100.7
−80.0	22.740	724.1	0.59	0.12	0.34	1.70	0.78	1.16	0.024	0.69	0.02	0.00	0.00	8.3	*58.9*	713.8
−110.0	23.111	832.6	0.67	0.11	0.47	1.99	0.91	1.25	0.025	0.73	0.01	0.00	0.00	9.6	92.9	814.1
−130.0	23.305	932.0	0.71	0.13	0.45	2.24	1.01	1.41	0.029	0.83	0.01	0.00	0.00	10.7	105.1	906.6
−150.0	23.485	978.2	0.76	0.13	0.56	2.46	1.09	1.56	0.030	0.87	0.01	0.00	0.00	11.7	127.3	992.2
−160.0	23.569	999.0	0.76	0.13	0.58	2.43	1.09	1.53	0.029	0.90	0.01	0.00	0.00	11.6	130.2	986.6
−170.0	23.680	1026.0	0.77	0.13	0.54	2.48	1.12	1.61	0.029	0.98	0.01	0.00	0.00	11.9	*139.8*	1014.4
−180.0	23.959	1088.3	0.78	0.13	0.52	2.83	1.17	1.69	0.030	0.98	0.01	0.00	0.00	12.9	154.0	1089.5
−185.0	24.314	1159.0	0.87	0.17	0.61	3.16	1.27	1.77	0.031	1.05	0.02	0.00	0.00	14.1	196.7	1191.0
−190.0	25.042	1422.3	1.03	0.18	0.61	3.89	1.58	2.45	0.036	1.27	0.02	0.00	0.00	17.7	346.2	1496.7
−195.0	25.201	1443.9	1.08	0.19	0.61	3.91	1.62	2.47	0.036	1.31	0.02	0.00	0.00	18.0	355.4	1518.1
−200.0	25.234	1454.6	1.08	0.19	0.62	3.99	1.65	2.58	0.037	1.31	0.02	0.00	0.00	18.4	351.6	1553.8
−205.0	25.284	1492.0	1.14	0.20	0.73	4.15	1.70	3.26	0.039	1.45	0.02	0.00	0.00	20.4	349.0	1730.1
−208.0	25.425	1740.2	1.27	0.27	1.21	4.94	1.87	11.67	0.082	2.03	0.03	0.01	0.00	39.8	341.1	3468.0
−210.0	25.550	2744.5	1.34	0.32	1.31	5.02	1.89	13.11	0.087	2.06	0.02	0.01	0.00	43.1	—	3759.0

*6Italicized CO2(aq) values were estimated from the smoothed syringe CO2–C25 relationship.

January 2004*7

Depth m	Temp.*2 °C	C25*2 µS/cm	Na+ mM/kg	K+ mM/kg	NH4+ mM/kg	Mg2+ mM/kg	Ca2+ mM/kg	Fe2+ mM/kg	Mn2+ mM/kg	SiO2(aq) mM/kg	Cl- mM/kg	NO3- mM/kg	SO4 2- mM/kg	HCO3- mM/kg	CO2(aq) mM/kg	TDS mg/kg
0.0	22.216	108.2	0.16	0.04	0.01	0.24	0.13	0.01	0.002	0.28	0.008	0.000	0.000	1.0	*0.4*	94.5
−50.0	21.965	113.1	0.15	0.04	0.01	0.24	0.14	0.01	0.006	0.28	0.006	0.001	0.000	1.0	*11.8*	94.7
−55.0	22.143	413.9	0.31	0.07	0.11	0.88	0.43	0.50	0.052	0.51	0.008	0.000	0.000	4.2	47.5	369.0
−80.0	22.736	719.2	0.52	0.10	0.27	1.71	0.82	1.05	0.025	1.16	0.011	0.000	0.000	8.1	70.2	696.1
−120.5	23.207	887.1	0.55	0.10	0.31	2.12	0.98	1.36	0.027	1.33	0.010	0.000	0.000	9.9	95.5	849.3
−160.5	23.563	1002.1	0.73	0.13	0.42	2.56	1.15	1.47	0.030	1.47	0.028	0.002	0.005	11.7	129.0	995.6
−175.5	23.745	1047.0	0.76	0.14	0.45	2.74	1.19	1.60	0.031	1.52	0.017	0.000	0.001	12.5	135.0	1058.6
−186.5	24.560	1177.7	0.85	0.15	0.48	3.29	1.33	1.83	0.032	1.65	0.018	0.000	0.001	14.4	*202.6*	1218.2
−190.2	25.068	1426.0	0.97	0.17	0.53	3.99	1.64	2.40	0.037	1.85	0.018	0.000	0.000	17.8	337.9	1498.1
−197.8	25.215	1462.2	1.02	0.18	0.55	4.12	1.71	2.74	0.038	1.92	0.021	0.000	0.000	18.9	347.2	1596.8
−205.5	25.313	1489.5	1.00	0.17	0.53	4.40	1.77	2.73	0.040	1.97	0.023	0.000	0.002	19.6	357.8	1646.3

*7Italicized CO2(aq) values were estimated from the smoothed syringe CO2–C25 relationship.

January 2005[8]

Depth m	Temp.[2] °C	C25[2] µS/cm	Na⁺ mM/kg	K⁺ mM/kg	NH₄⁺ mM/kg	Mg²⁺ mM/kg	Ca²⁺ mM/kg	Fe²⁺ mM/kg	Mn²⁺ mM/kg	SiO₂(aq) mM/kg	Cl⁻ mM/kg	NO₃⁻ mM/kg	SO₄²⁻ mM/kg	HCO₃⁻ mM/kg	CO₂(aq) mM/kg	TDS mg/kg
0.0	22.076	109.2	0.18	0.04	0.12	0.28	0.15	0.00	0.002	0.26	0.028	0.002	0.001	1.2	*0.7*	111.2
−20.0	21.958	109.3	0.18	0.05	0.12	0.28	0.15	0.00	0.002	0.26	0.007	0.001	0.002	1.2	*0.5*	110.1
−50.0	21.953	109.6	0.18	0.05	0.14	0.28	0.15	0.00	0.002	—	0.020	0.002	0.002	1.2	*0.6*	—
−70.0	22.478	705.2	0.53	0.12	0.53	1.69	0.79	1.09	0.027	0.72	0.015	0.002	0.001	8.4	46.5	717.3
−98.8	22.877	767.1	0.62	0.13	0.60	1.91	0.91	1.22	0.026	0.75	0.022	0.000	0.000	9.4	74.0	806.2
−136.9	23.355	952.2	0.74	0.14	0.67	2.45	1.13	1.57	0.031	0.90	0.013	0.001	0.001	11.9	*117.8*	1010.0
−170.4	23.660	1029.4	0.82	0.17	0.89	2.71	1.20	1.70	0.032	0.99	0.021	0.003	0.000	13.1	131.8	1115.6
−180.0	23.848	1066.5	0.86	0.17	0.94	2.89	1.26	1.77	0.033	1.03	—	—	—	13.8	142.9	1173.5
−190.2	24.672	1240.8	0.97	0.20	1.00	3.62	1.47	2.14	0.035	1.24	0.021	0.005	0.004	16.7	314.6	1411.4
−197.8	25.232	1449.7	1.11	0.21	0.98	4.23	1.73	2.72	0.039	1.36	0.029	0.003	0.003	19.7	355.8	1665.3
−205.5	25.388	1543.9	1.20	0.21	1.04	4.45	1.79	3.12	0.042	1.45	0.027	0.003	0.005	21.2	355.8	1796.2

[8]*Italicized CO₂(aq) values were estimated from the smoothed syringe CO₂–C25 relationship.*

January 2006[9]

Depth m	Temp.[2] °C	C25[2] µS/cm	Na⁺ mM/kg	K⁺ mM/kg	NH₄⁺ mM/kg	Mg²⁺ mM/kg	Ca²⁺ mM/kg	Fe²⁺ mM/kg	Mn²⁺ mM/kg	SiO₂(aq) mM/kg	Cl⁻ mM/kg	NO₃⁻ mM/kg	SO₄²⁻ mM/kg	HCO₃⁻ mM/kg	CO₂(aq) mM/kg	TDS mg/kg
0.0	23.535	126.3	0.19	0.05	0.03	0.33	0.16	0.00	0.002	0.26	0.024	0.012	0.003	1.2	*0.1*	113.9
−20.0	22.457	117.0	0.17	0.04	0.02	—	—	—	—	—	0.008	0.009	0.001	1.2	*0.2*	—
−50.0	21.981	134.5	0.18	0.05	0.03	0.31	0.16	0.05	0.007	0.29	0.011	0.010	0.004	1.3	*1.7*	120.9
−75.0	22.522	718.0	0.54	0.11	0.32	1.66	0.80	1.10	0.027	0.74	0.022	0.022	0.031	8.0	*55.6*	698.2
−99.1	22.874	767.5	0.58	0.11	0.33	1.83	0.88	1.17	0.025	0.75	0.019	0.019	0.006	8.8	*69.5*	752.9
−100.0	22.887	769.8	0.59	0.11	0.33	1.84	0.89	1.18	0.025	0.75	0.021	0.013	0.005	8.8	71.3	757.8
−137.1	23.350	949.2	0.73	0.13	0.43	2.36	1.09	1.50	0.030	0.88	0.037	0.022	0.007	11.2	123.7	954.6
−170.7	23.658	1028.4	0.75	0.14	0.48	2.63	1.20	1.66	0.031	0.99	0.028	0.022	0.014	12.3	131.2	1053.2
−180.0	23.831	1063.2	0.77	0.12	0.48	2.77	1.24	1.75	0.032	1.05	0.025	0.022	0.010	12.9	139.8	1100.8
−185.0	23.981	1086.7	0.79	0.13	0.49	2.91	1.23	1.74	0.031	1.07	0.030	0.023	0.011	13.2	151.6	1122.9
−190.5	24.427	1158.4	0.84	0.15	0.53	3.19	1.32	1.86	0.032	1.14	0.029	0.026	0.015	14.2	183.1	1211.9
−195.0	24.902	1339.6	0.94	0.15	0.60	3.78	1.58	2.37	0.036	1.29	0.028	0.035	0.028	17.1	298.9	1455.6
−205.8	25.452	1635.5	1.12	0.19	0.66	4.45	1.81	3.45	0.042	1.55	0.029	0.038	0.016	21.4	366.6	1823.1

[9]*Italicized CO₂(aq) values were estimated from the smoothed syringe CO₂–C25 relationship.*

ing of the intake depth of the existing pipes is required, but the stoppage of self-siphoning will follow soon. Since natural recharge of CO_2 will continue, it is recommended to pump deep water continuously to the surface. For example, a screw pump driven by solar panels may be used, because it requires little maintenance. Another possibility may be the use of an air-lift system. Compressed air is sent to depth in the degassing pipe to facilitate pumping

water of that depth. A compressor powered by solar energy can be placed on shore. These additional pumping methods could make Lake Monoun completely free of dissolved gases and safe over the long-term. This attempt should be initiated as soon as most of dissolved gases have been removed from the lake by the current degassing system.

CHEMICAL COMPOSITION, CTD MEASUREMENTS AND CO_2 PROFILES OF LAKE NYOS

For Lake Nyos we have CTD data measured in October 1986 (Kanari, 1989), December 1988 (Nojiri *et al.*, 1993), November 1993, March 1995, April 1996, April 1998, November 1999, and January 2001 as the pre-controlled degassing casts, and in December 2001, January 2003, January 2004, January 2005, January 2006, June 2006, and January 2007 as the post-controlled degassing casts. As is the case for Lake Monoun, the CTD data are only partly shown by figures in this paper, but the raw and processed data can be found in a data archive operated by the Geochemical Society of Japan (Kusakabe, 2007). Table 3 shows the chemical composition of Lake Nyos waters sampled at different depths and at different times (1986–2006). For the sake of completeness, published data of October 1986 (Kusakabe *et al.*, 1989) and of December 1988 (Nojiri *et al.*, 1993) are included in Table 3, where $CO_{2(aq)}$ values were estimated using the relationship between $CO_{2(aq)}$/C25 and C25 as described below. $CO_{2(aq)}$ concentrations in Table 3 were basically obtained by the syringe method, but those for shallow waters were calculated by the pH method. The sampling and analytical methods are the same as those for Lake
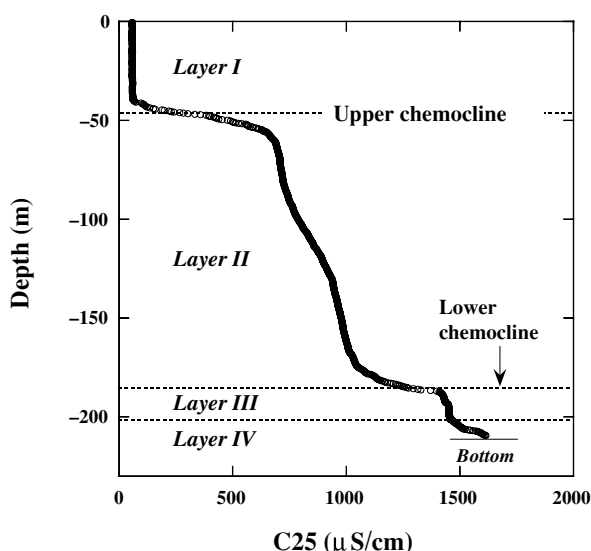
Fig. 7. Chemical structure of Lake Nyos as exemplified by C25 distribution measured in January 2001. Layers I, II and III are bordered by the upper and lower chemoclines. Layer IV is the deepest water characterized by increasing C25 toward the bottom.
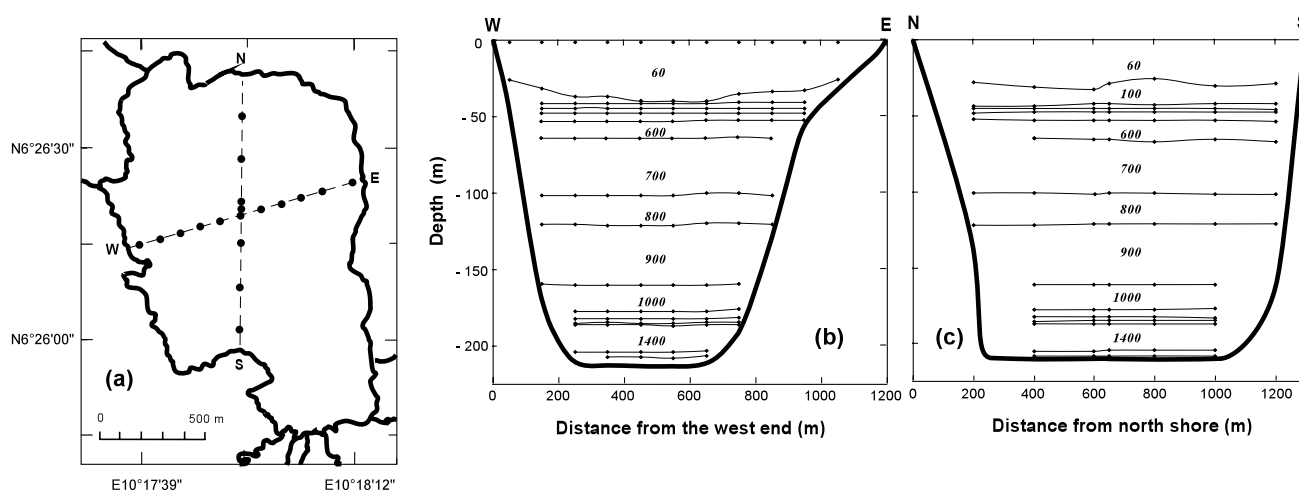
Fig. 8. Three-dimensional CTD measurements at Lake Nyos in January 2001. (a) Map of Lake Nyos. CTD cast was made at the points along the W-E and N-S transects. (b) C25 distribution along the W-E transect. (c) C25 distribution along the N-S transect. C25 values are given in μS/cm.

Monoun.

The chemical composition of Lake Nyos waters is, in general, similar to that of Lake Monoun (Table 1) with obvious predominance of $CO_{2(aq)}$ and HCO_3^- over the other dissolved chemical species. Ferrous ion concentrations in deep waters and salinity are lower than those of Lake Monoun as indicated by lower C25 values. Bicarbonate ion comprises more than 99.8% judging from the analyzed anion concentrations, again justifying the method of calculating HCO_3^- concentrations by the difference method as we did for Lake Monoun.

The chemical structure of Lake Nyos water is shown in Fig. 7 by the C25-depth relationship measured in January 2001. Similar to Lake Monoun, Lake Nyos can be divided into 4 layers. Layer I is the shallowest, well-mixed, low conductivity water, which is separated by a sharp upper chemocline at about 50 m. It is underlain by layer II that extends down to 180 m. A lower chemocline develops around this depth, below which a well-mixed layer III continues down to *ca.* 203 m. Below this depth, conductivity (and temperature) increases sharply toward the bottom (layer IV). Relatively speaking layer III thickness is much smaller than that of Lake Monoun.

Temperature, C25, pH and $CO_{2(aq)}$ profiles of Lake Nyos before controlled degassing

In January 2001 we made a CTD survey to determine the three-dimensional structure of the lake. Measurements were undertaken along the west-east and north-south transects of the lake following the mooring ropes for fixing the instrumental raft (Fig. 8a). Closely-spaced isopleths of C25 in Figs. 8b and 8c show the upper and lower chemoclines, respectively. Generally speaking the lake water was horizontally homogeneous and well-strati-
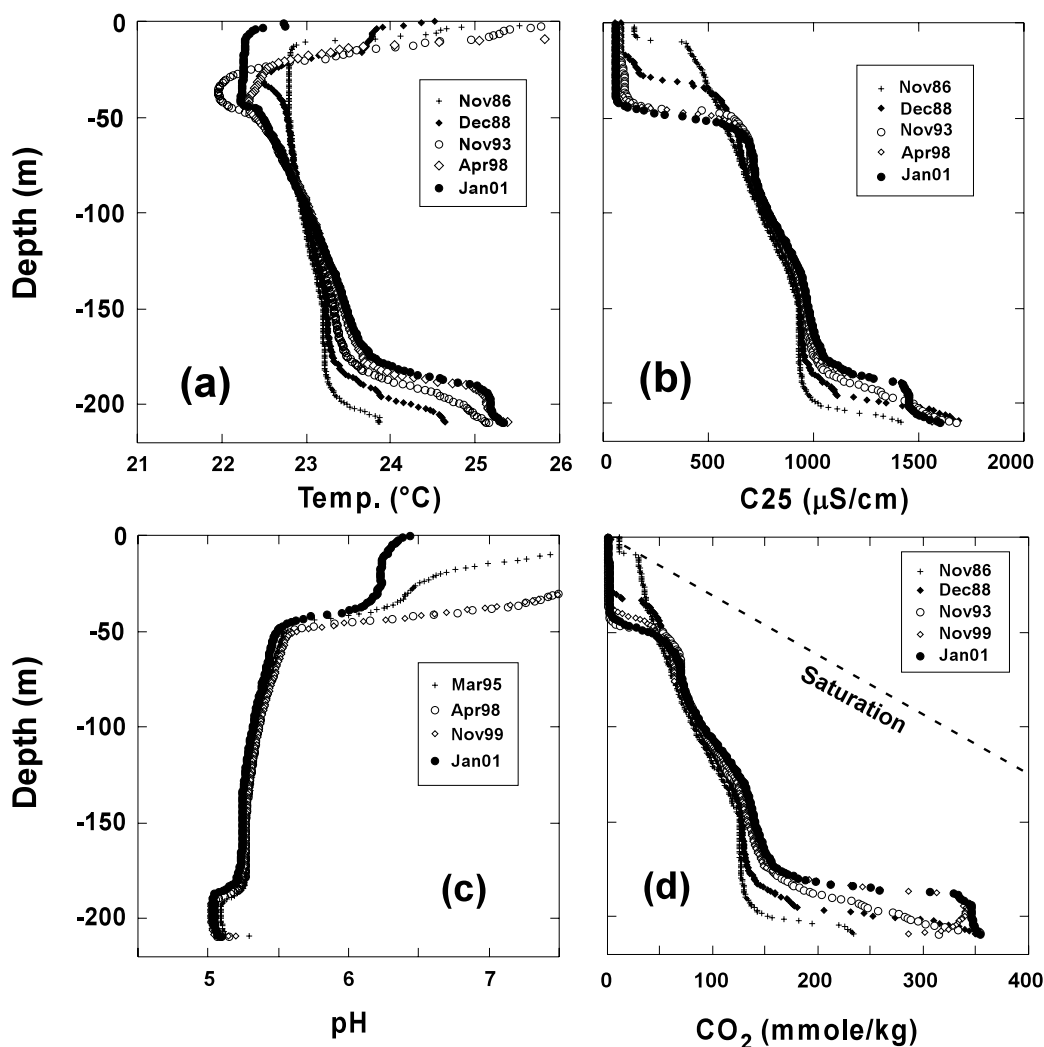

Fig. 9. Evolution of temperature (a), C25 (b), pH (c) and $CO_{2(aq)}$ (d) before controlled degassing (November 1986–January 2003) at Lake Nyos. CO_2 saturation curve was calculated from Duan and Sun (2003).
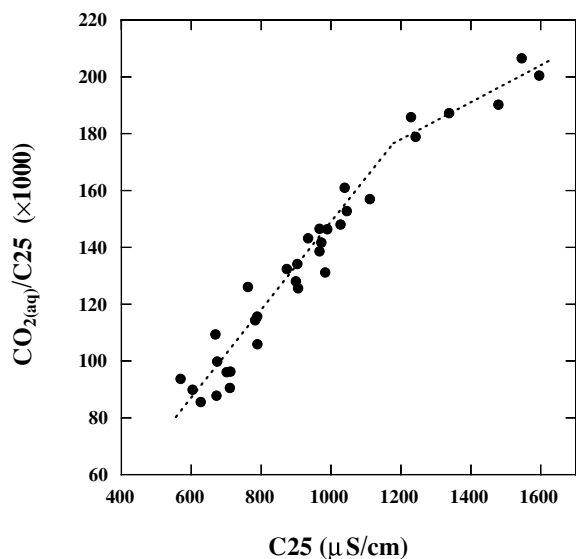
Fig. 10. Relationship between $CO_{2(aq)}$/C25 ratios and C25 values for waters collected before controlled degassing (December 1988 to January 2001) at Lake Nyos. Two linear relationships can be seen; one for waters with C25 < 1200 μS/cm, and another for waters with C25 > 1200 μS/cm. See text for regression equations.

fied below the upper chemocline. C25 values of the shallow water above the upper chemocline were slightly variable along the transects, reflecting diurnal effects, because measurements were made on different days and times.

Figure 9 shows the temperature, conductivity, pH and CO_2 profiles of Lake Nyos before controlled degassing started. The earliest temperature profiles in October 1986 (Kanari, 1989), measured 2 months after the limnic eruption in August 1986, showed monotonous increase below 10 m down to 145 m followed by constant temperature down to ca. 190 m. This pattern was confirmed by the CTD measurement in November 1986 (Fig. 9a) (Tietze, 1987). Below 190 m, a sharp rise of temperature was still observable even after the large scale gas explosion at that time. This may suggest that either the deepest water was disturbed only slightly during the explosion or the initial supply rate of warm recharge fluid from the bottom was very high. Surface temperatures changed considerably depending on the timing of measurements, but they decrease sharply towards the temperature minima just above the upper chemocline. The temperature variability in Layer I is affected by surface meteorological forcing such as solar irradiation, air temperature and rainfall, down to ~50 m (Kusakabe et al., 2000; Schmid et al., 2003, 2006; Kling et al., 2005). Temperatures below the upper chemocline increase gradually down to ca. 170 m. Below this depth, temperature increases sharply towards the bottom, exceeding 25°C at the deepest point after November 1993. The

temperature of deep water (170–210 m) increased noticeably with time as shown in Fig. 9a. There is a "temperature shoulder" in the January 2001 profile, but the shoulder appears to have already started to form back in 1998, suggesting initiation of mixing in layer III.

Similar to the temperature profiles, shallow waters in 1986 showed higher conductivity (Fig. 9b), indicating that deep, saline water was brought to the surface during the limnic eruption. The upper chemocline in October 1986 was at 7 m depth, but it deepened with time down to 47 m in 1993 and 50 m in 2001. Unfortunately the CTD measurement of October 1986 (Kanari, 1989) did not go deeper than 195 m. So we used the C25 data of November 1986 (Tietze, 1987) to estimate the $CO_{2(aq)}$ profile in 1986 as stated below. Conductivity profiles at mid-depths (70–160 m) stayed almost unchanged for 15 years after the eruption. The conductivity of the deep water (170–210 m) increased noticeably with time as shown in Fig. 9b. In January 2001 the conductivity profile between 185 m and 202 m became steep, with the associated reduction of earlier high conductivity in layer IV, indicating initiation of mixing in the deepest zone. From 205 m to the bottom, conductivity increased sharply towards a maximum value. Figure 9c shows that surface pH values are greater than 6.5 and decrease sharply toward the upper chemocline, converging at ca. 5.5. They then decrease gradually between 50 and 140 m, and stay almost constant between 140 and 185 m. They sharply decrease towards 195 m with minimum values found at around 200 m. The 2001 pH values are slightly lower than the 1993 and 1996 values when compared at the same depths, reflecting the CO_2 build-up in layers II and III.

Pre-controlled degassing $CO_{2(aq)}$ profiles obtained mainly by the syringe method are shown in Fig. 9d. We used $CO_{2(aq)}$ values estimated by the pH method for shallow waters that contain $CO_{2(aq)}$ less than 40 mmol/kg for the reasons described in the Lake Monoun section. We estimated the $CO_{2(aq)}$ profiles of November 1986 and part of December 1988 using the syringe-CO_2/C25 versus C25 relationship (Fig. 10). The relationship was obtained on the basis of measured $CO_{2(aq)}$ and C25 values during the pre-degassing period. It is approximated by equations

$$Y = 0.1476 \ (\pm 0.0076) * X - 1.76 \ (\pm 6.78)$$
$$(500 < C25 < 1200 \ \mu S/cm) \qquad (5a)$$

$$Y = 0.0572 \ (\pm 0.0076) * X + 111.19 \ (\pm 6.77)$$
$$(1200 < C25 < 1600 \ \mu S/cm) \qquad (5b)$$

where Y stands for $CO_{2(aq)}$/C25*1000 and X for C25. The uncertainties are for the standard error of regression. The $CO_{2(aq)}$ values for November 1986 and December 1988 thus estimated are included in Fig. 9d. It shows that $CO_{2(aq)}$ concentrations below the lower chemocline in-

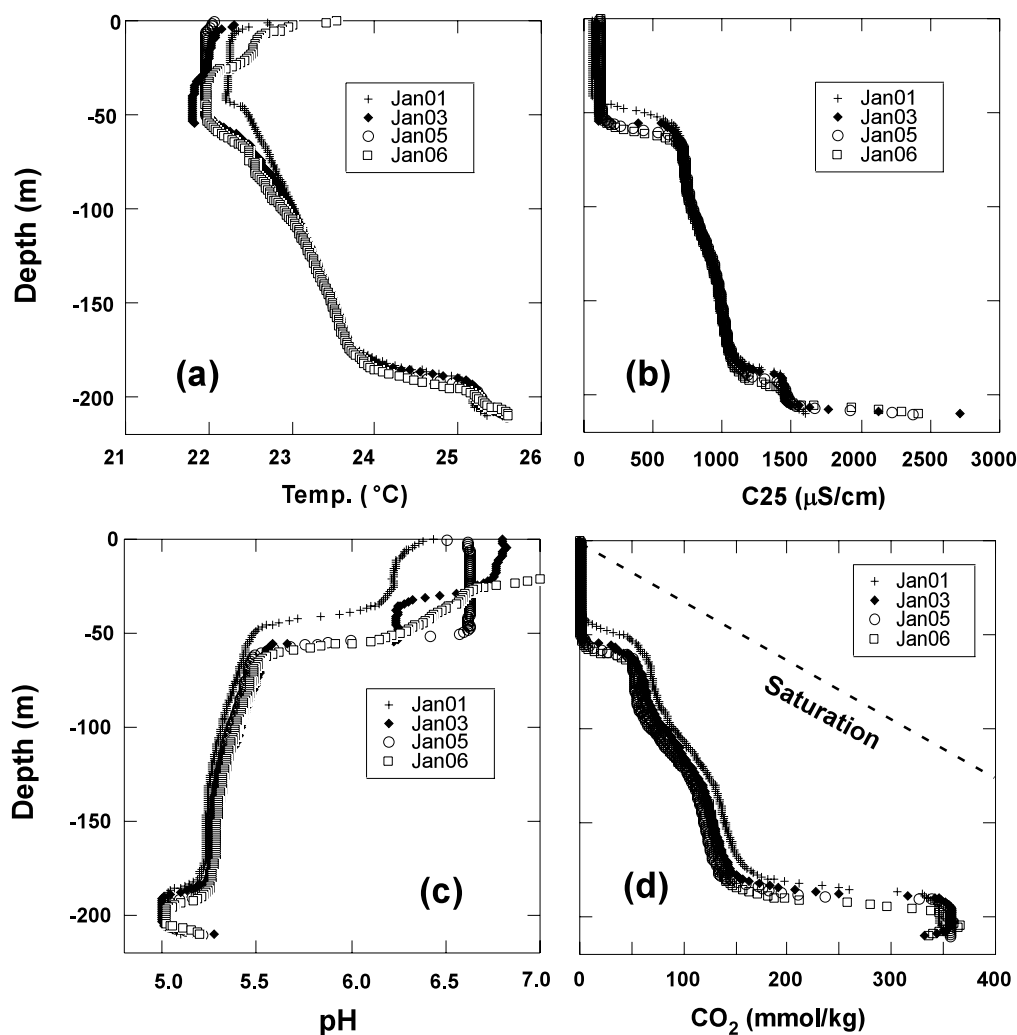*Fig. 11. Evolution of temperature (a), C25 (b), pH (c) and CO$_{2(aq)}$ (d) during controlled degassing (January 2001–January 2006) at Lake Nyos.*

creased considerably with time when compared at a given depth. This is especially conspicuous below 180 m, suggesting that CO$_{2(aq)}$ was added to water below 180 m (layers III and IV) during the pre-degassing period. CO$_{2(aq)}$ concentration at the bottom-most water is almost constant at a steady value of 360 mmol/kg since 1999.

Temperature, C25, pH and CO$_{2(aq)}$ profiles of Lake Nyos during controlled degassing

Following experimental degassing attempts, the first permanent degassing system was installed at Lake Nyos in January 2001 (Halbwachs *et al.*, 2004). A hardened polyethylene pipe of 203 m long carried water at that depth up to the surface. Since the initiation of controlled degassing, dissolved CO$_2$ has been released to the atmosphere while the water from 203 m depth has rained back to the surface of the lake. This has resulted in a small

increase in C25 values in layer I water and in gradual subsidence of temperature, pH and CO$_{2(aq)}$ profiles below the upper chemocline as shown in Fig. 11. It shows the during degassing evolution of temperature (a), conductivity (b), pH (c) and CO$_2$ (d) profiles for the period from January 2001 to January 2006. Compared to the January 2001 profiles which are shown as the reference for the last pre-degassing period, all the profiles maintained their general shape and structure but subsided noticeably as degassing proceeded. The upper chemocline deepened from the 2001 level of 47 m to 59 m in January 2006 (it deepened to 61 m in January 2007, not shown in Fig. 11b). The lower chemocline also subsided from the 2001 level of 188 m to 193 m in January 2006 (it deepened to 200 m in January 2007, not shown in Fig. 11b). The rate of subsidence of the lower chemocline is calculated to be ~1 m/year if the data between 2001 and 2007

Table 4. Change with time in CO₂ content of Lake Nyos

Date	Year after Aug. 1984	Total CO_2	CO_2 below layer II	CO_2 below layer II	CO_2 accumulation rate	CO_2 removal rate
		giga mole	giga mole	giga mole	giga-mole/yr	giga-mole/yr
Pre-degassing						
November 1986	0.17	*13.05 ± 0.07*	*12.88 ± 0.07*	*0.46 ± 0.02*	—	—
December 1988	2.33	*13.33 ± 0.05*	*13.29 ± 0.05*	*1.76 ± 0.02*	—	—
November 1993	7.25	13.57 ± 0.54	13.55 ± 0.54	2.57 ± 0.04	—	—
April 1998	11.67	14.06 ± 0.53	14.02 ± 0.54	3.48 ± 0.05	—	—
November 1999	13.25	14.41 ± 0.51	14.04 ± 0.52	3.39 ± 0.05	—	—
January 2001	14.42	14.78 ± 0.52	14.63 ± 0.62	3.52 ± 0.05	0.12 ± 0.04 (1986–2001)	—
Post-degassing						
December 2001	15.33	14.20 ± 0.49	14.13 ± 0.49	3.32 ± 0.04	—	—
January 2003	16.42	13.10 ± 0.48	13.05 ± 0.48	3.42 ± 0.04	—	—
January 2004	17.42	13.16 ± 0.49	12.96 ± 0.49	3.21 ± 0.04	—	—
January 2005	18.42	12.29 ± 0.45	12.17 ± 0.45	3.06 ± 0.04	—	—
January 2006	19.42	11.83 ± 0.43	11.70 ± 0.43	2.63 ± 0.04	—	0.59 ± 0.15 (2001–2006)

Note 1. Errors attached to the November 1986 and December 1988 figures were calculated for the standard error in the slope of regression equations. See text and Fig. 9 for the equations.
Note 2. Errors for the figures other than the November 1986 and December 1988 figures were calculated for the uncertainties of ±4.5 mmole $CO_{2(aq)}$ at each depth in smoothing the syringe and pH data.

are used. This implies that the lower chemocline will subside to 203 m, the current intake depth of the degassing pipe, in a few years, resulting in a reduced gas removal rate because of decreasing $CO_{2(aq)}$ concentration of incoming water to the pipe. It is recommended not only to increase the number of the degassing pipes but also to deepen the intake depth of the pipe at Lake Nyos in the near future in order to remove $CO_{2(aq)}$ and other dissolved gases in layer IV where the highest gas concentrations are observed.

A sudden rise in C25 of the deepest water was observed after the degassing operation was initiated (compare Figs. 9b and 11b). The deepest C25 values never exceeded ~1700 μS/cm in the pre-controlled degassing period, whereas those in the during-degassing period reached as high as 2700 μS/cm (January 2003, Fig. 11b). Ferrous iron concentration of the deepest water in January 2003 was unusually high (Table 3) while concentrations of the other dissolved ionic species including $SiO_{2(aq)}$ did not show such an abrupt rise. The increase in C25 is, therefore, likely due to dissolution of $Fe(OH)_3$ precipitate which formed when Fe^{2+}-rich deep water from the degassing pipe was exposed to the atmosphere at the surface; these particles then sank and redissolved under anoxic conditions in the deepest water.

RECHARGE AND REMOVAL OF CO₂ AT LAKE NYOS

The change in CO_2 content with time was quantitatively evaluated from the $CO_{2(aq)}$ profiles before control-

led degassing (Fig. 9d) and those during-degassing (Fig. 11d). It is summarized in Table 4 and graphically shown in Fig. 12. The bathymetry used by Kling *et al.* (2005) was adopted for quantification. In Table 4 the CO_2 content of Lake Nyos from 1986 to 2006 was calculated for the whole lake (total CO_2) and for waters below the upper chemocline (CO_2 below layer II) and below the lower chemocline (CO_2 below layer III). The first CTD measurement at Lake Nyos was made in October 1986 by Kanari (1989). However, his C25 data are consistently lower than the later measurements when compared at the same depth range, e.g., between 70 and 150 m, a zone of little change with time. For this reason, we used C25 values obtained in November 1986 by Tietze (1987) to make an estimate of the earliest CO_2 content after the limnic eruption. Using the C25 profile of November 1986 (Tietze, 1987) and the syringe-CO_2/C25 versus C25 relationship (Fig. 10), a CO_2 profile of November 1986 was estimated. It is consistent with measurements carried out in 1987 using the cylinder method (Evans *et al.*, 1994). Using the CO_2 profile, total CO_2 content in November 1986 was estimated to be 13.05 ± 0.07 giga-moles. CO_2 content below the upper chemocline was 12.88 ± 0.07 giga moles. The error was estimated only from the standard error of the regression equation used in Fig. 10, and the accuracy of the 1986 C25 measurement was not taken into account. This estimate of CO_2 content must be close to that of Lake Nyos soon after the August 1986 limnic eruption. CO_2 content below the upper chemocline kept steadily increasing and it reached 14.63 ± 0.62 giga moles
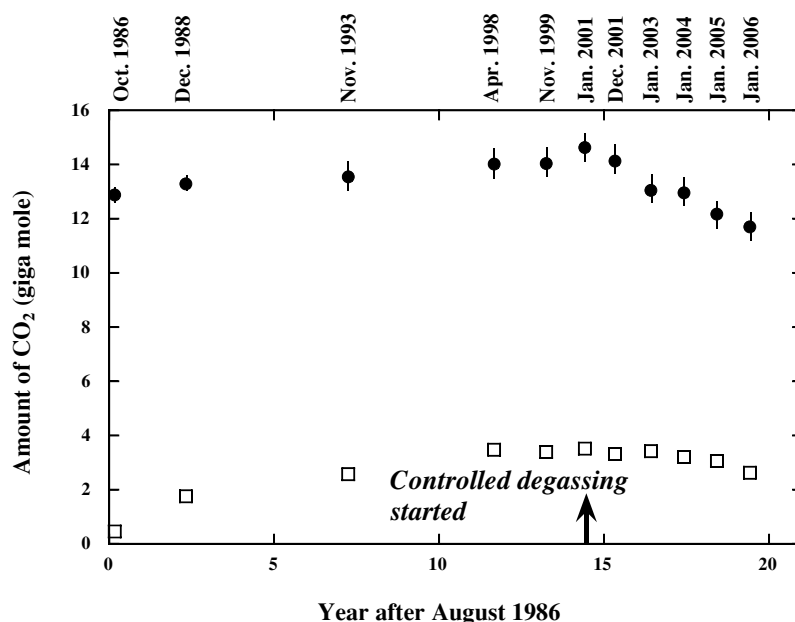
Fig. 12. Change with time in CO_2 content at Lake Nyos. Closed circles and open squares are for CO_2 content below the upper chemocline and below the lower chemocline, respectively. Error bars indicate the standard error associated with the estimation of each data point.

in January 2001, a maximum value in the pre-degassing period. The values in Table 4 are consistent with those reported in Kling *et al*. (2005) within error. From the change over time of the CO_2 content below the upper chemocline, we can evaluate the rate of CO_2 accumulation in Lake Nyos since its explosion in 1986 (Table 4). The mean rate of CO_2 accumulation below the upper chemocline is calculated as 0.12 ± 0.04 giga moles/year based on the data of November 1986 to January 2001. This agrees well with the CO_2 recharge rate of 0.126 ± 0.048 giga moles/year reported in Kling *et al*. (2005) who used the CO_2 contents of 1992 and 2001. Note that the CO_2 accumulation rate of 0.23 giga moles/year reported by Kusakabe *et al*. (2000) is considerably higher than 0.12 ± 0.04 giga moles/year stated above. The reason for the discrepancy is that Kusakabe *et al*. (2000) did not use CO_2 data below the upper chemocline but below 180 m only. The continued recharge of CO_2-rich water from the bottom resulted in the thickening of layer III (Fig. 9d). The increase in the CO_2 content in layer III since 1986 is 3.06 ± 0.01 giga moles (Table 4).

Evolution of the water column structure before controlled degassing (Fig. 9) indicates thickening of layers III and IV with time. This suggests that the CO_2 profile was developing in a similar way to the profiles observed at Lake Monoun. There is a possibility that layers III and IV of Lake Nyos may have been much thicker before the 1986 catastrophic event, and that the shape of pre-event CO_2 profile may have been similar to that observed at

Lake Monoun in 2003 (Fig. 4d), with the $CO_{2(aq)}$ concentration almost constant at 360 mmol/kg below the lower chemocline down to the bottom. If the lower chemocline was around 113 m (saturation depth for water containing 360 mmole-CO_2/kg) at that time, a small addition of CO_2 from natural recharge would have made the water at that depth saturated with CO_2, leading to spontaneous exsolution of gas bubbles and eventually to a limnic eruption. If this model is correct, the amount of CO_2 gas released to the atmosphere during the 1986 limnic eruption is calculated as ~14 giga moles or 0.31 km^3 at STP as a difference between the above assumed pre-event profile and the November 1986 profile multiplied by the lake's bathymetry. This value is greater than the estimate (0.14 km^3 STP) by Evans *et al*. (1994) by a factor of 2, but much smaller than an early estimate of ~1 km^3 at STP (Tuttle *et al*., 1987; Faivre Pierret *et al*., 1992). The estimated amount of CO_2 released obviously depends on the assumptions involved. As long as the lake receives natural recharge of CO_2, limnic eruptions can occur repetitively (Tietze, 1992; Kling *et al*., 1994) but may not be regular as described in a model calculation by Chau *et al*. (1996).

Controlled degassing started in March 2001 at Lake Nyos. The observed decrease of CO_2 content after 2001 (Fig. 12) is in general agreement with the prediction of the model calculations by Kling *et al*. (2005). The CO_2 content below the upper chemocline in January 2006 was 11.70 ± 0.43 giga moles, which is smaller than that soon

after the 1986 limnic eruption. The mean rate of gas removal in the during-degassing period is 0.59 ± 0.15 giga moles/year. The year-by-year rate of gas removal varies from 0.09 to 0.99 giga moles/year with the minimum rate observed between January 2003 and January 2004. During this period the degassing system was not functioning at its optimal rate and the gas recharge rate may have balanced the gas removal rate. Although the mean rate of gas removal of 0.59 ± 0.15 giga moles/year (calculated for the period 2001–2006) is greater than the mean rate of CO_2 recharge of 0.12 giga moles/year, the gas removal rate will reduce substantially when the lower chemocline subsides below the water tapping depth of 203 m. Currently the lower chemocline subsides approximately 1 m per year, and the lower chemocline was at 193 m depth in January 2006 and 200 m depth in January 2007. Thus it takes only a few years for the lower chemocline to reach the water tapping depth. After this is reached, CO_2 content of water tapped by the pipe will become lower and the gas removal will take several decades (Kling *et al.*, 2005; Schmid *et al.*, 2006). After this stage, the pipe should be lowered as close to the bottom as possible, and also a system to carry the bottom-most waters up to the surface to remove gas continuously (as proposed for Lake Monoun) must be installed, since natural recharge of CO_2 will continue.

The degassing operation since 2001 at Lake Nyos and 2003 at Lake Monoun has not changed the overall shape of each profile (Fig. 11), and has proven that the lakes have not been destabilized as initially suspected (e.g., Freeth, 1994). Since the effect of dissolved CO_2 on the density of lake water is far greater than that of the other dissolved chemical species at a given temperature (e.g., Kusakabe *et al.*, 1989), the CO_2 profiles essentially determine the density profiles. This implies that the overall stability of the lakes has become greater as degassing has proceeded, and therefore spontaneous overturn of the lake water is unlikely.

Conclusions

Regular monitoring of Lakes Monoun and Nyos since 1986 has shown that chemical evolution of the lakes is remarkably rapid as geological phenomena. Results of the monitoring obtained during the last 21 years including the earliest CTD measurement made in October/November 1986 indicated that the greatest changes of temperature, C25 and CO_2 concentration in the lake took place during the initial 10 years at Lake Monoun. Changes at Lake Nyos are fairly steady. The accumulation rate of CO_2 was estimated to be *ca*. 8 mega-moles/year at Lake Monoun before controlled degassing (November 1993 to January 2003). Similarly at Lake Nyos, the CO_2 accumulation rate was ~0.12 giga moles per year until January

2001 when controlled degassing started. If the pre-event CO_2 profile was close to saturation at a certain depth as observed at Lake Monoun in 2001, the lakes may have exploded spontaneously without receiving an external trigger.

The effect of degassing on CO_2 content is remarkable especially for Lake Monoun. By January 2007 the CO_2 content was lowered to ~40% of the maximum content attained just before controlled degassing started. The current CO_2 content is lower than the estimated initial content soon after the 1984 limnic eruption. The rate of gas removal will drastically decrease in a short period of time, because the lower chemocline will subside to the current pipe intake depth of 93 m within a year or so, assuming full performance of the degassing systems. After this stage is reached, the degassing pipe should be lowered as close to the bottom as possible to remove the remaining dissolved gases. The rate of CO_2 recharge seems to have declined in recent years at Lake Monoun, but it may change to a higher rate in the future. So, a system that carries the bottom water to the surface needs to be installed after degassing by the current system becomes ineffective. The new system should be simple, robust and maintenance-free, e.g., a small pump driven by solar power that pumps deep water to the surface.

Controlled degassing at Lake Nyos since 2001 has reduced the CO_2 content below the level found soon after the limnic eruption in 1986. However, the CO_2 content still remaining in the lake amounts to 80% of the maximum level of 14.8 giga moles observed in January 2001 and is thus still dangerous to people living around the lake. The depth of the lower chemocline may reach the water intake depth of 203 m within a few years. After this situation is reached the degassing rate with the current system will progressively be lowered, and it would take decades to remove a majority of dissolved gases even if the degassing system keeps working continuously for such a long time. Therefore additional gas removal systems need to be installed in order to speed up gas removal from Lake Nyos in a short period of time.

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