Vertical distribution of dissolved CO_2 in lakes Nyos and Monoun (Cameroon) as estimated by sound speed in water

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Abstract: Vertical profiles of sound speed (SS) in lake water were measured at lakes Nyos and Monoun (Cameroon) in 2012 and 2014. A significant linear correlation with Pearson's r of 0.987 to 0.995 between total CO_2 concentration ($[CO_2] = [CO_{2(aq)}] + [HCO_3^-]$) and SS excess ($\Delta v$) in water was found. Based on this correlation, we propose the SS method as a new simple tool to monitor CO_2 concentration in lakes Nyos and Monoun. We applied this method to multi-point measurements of CO_2 in lakes Nyos and Monoun, and found horizontal differences between CO_2 distributions in the two lakes. The results indicated that Lake Nyos water was stably stratified, and the total CO_2 decreased from 2012 to 2014. At Lake Monoun, which consists of three basins (the western basin, the central basin, and the main basin), waters in the western and central basins exhibited a higher dissolved CO_2 content than those in the main basin at the same depth. An increase in CO_2 was also detected in the main basin from 2012 to 2014.

Limnic eruptions at Lake Nyos in 1986 and at Lake Monoun (Cameroon) in 1984 were caused by a sudden release of magmatic CO_2 dissolved in the lake waters. These disasters killed 1800 residents around the lakes (Kling *et al.* 1987; Sigurdsson *et al.* 1987). Measuring dissolved CO_2 over time appears to be the most suitable parameter to investigate the precursors of limnic eruptions and reduce the risks at these lakes. Some methods for determining CO_2 concentration have been proposed and applied. Kusakabe *et al.* (2000, 2008) used the MK method, which combines chemical analysis of sampled water, pH and electrical conductivity (EC). Yoshida *et al.* (2010) proposed the YY method, where volumes of dissolved CO_2 and water siphoned from deep water through a plastic tube were measured. Although these methods are very accurate, they require a great deal of time and effort. Thus, we propose an alternative method using sound speed (SS) in water. In this article, we present a simple and fast procedure for indirect calculation of the concentration of dissolved CO_2 by simplifying the SS–EC method proposed by Sanemasa *et al.* (2015). These authors estimated dissolved CO_2 using equation (1)

$$\Delta v = k_1[CO_{2(aq)}] + k_2[HCO_3^-], \qquad (1)$$

where Δv is an increment of SS due to dissolved ions, and k_1 and k_2 are empirical coefficients that can be determined experimentally. The $CO_{2(aq)}$ and HCO_3^- concentrations can be calculated using k_1, k_2, SS, temperature and electrical conductivity. The new method proposed in the present study is based on the SS–EC method; however, SS in water and temperature are the only two parameters that need to be measured to calculate the total concentration of dissolved CO_2. To demonstrate the suitability of our approach, this simplified SS method was used to estimate the vertical and horizontal distribution of dissolved CO_2 concentrations at lakes Nyos and Monoun in 2012 and 2014.

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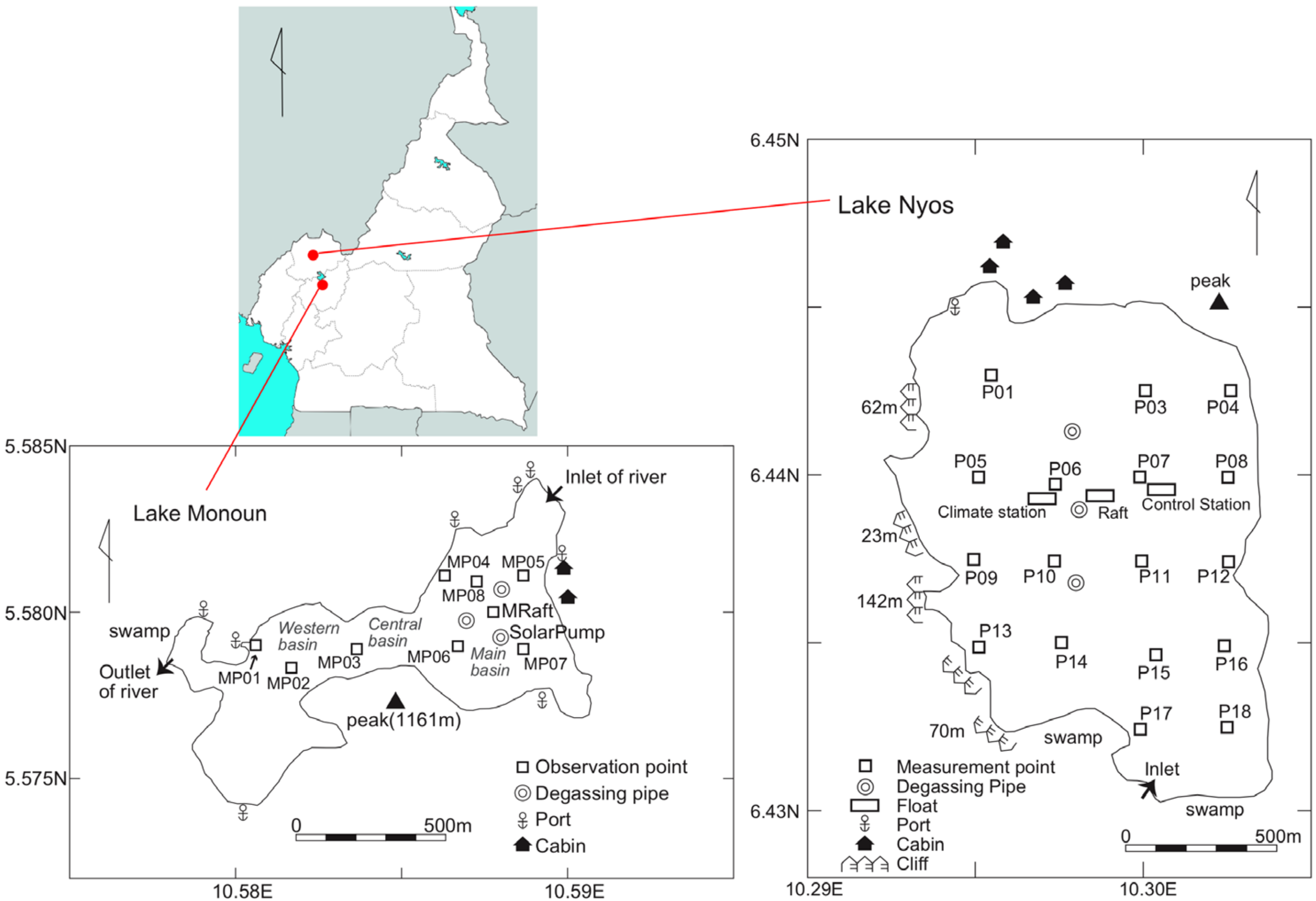

Fig. 1. Schematic maps of lakes Nyos and Monoun. The sites where the SS depth profiles were carried out are also indicated.

Table 1. *Measurement points for SS profile at Lake Nyos*

Point name	Latitude (°)	Longitude (°)	Date of measurements				
			5 March 2012	6 March 2012	7 March 2012	1 March 2014	2 March 2014
P01	6.44296	10.29549	✓	✓			
P03	6.44246	10.30011	✓		✓		
P04	6.44250	10.30262	✓				
P05	6.43992	10.29511		✓	✓	✓	✓
P06	6.43970	10.29739		✓	✓	✓	
P07	6.43991	10.29991		✓	✓	✓	
P08	6.43990	10.30253		✓			
P09	6.43746	10.29497		✓			
P10	6.43741	10.29737		✓	✓		✓
P11	6.43742	10.29997		✓	✓		
P12	6.43739	10.30255		✓			
P13	6.43485	10.29512		✓			✓
P14	6.43499	10.29758		✓			
P15	6.43462	10.30039		✓			
P16	6.43488	10.30243		✓			
P17	6.43240	10.29993		✓			
P18	6.43247	10.30252		✓			
Control station	6.43952	10.30053	✓	✓	✓		✓
Raft	6.43920	10.29842	✓	✓	✓		

Survey at lakes Nyos and Monoun

Two field surveys were conducted at both lakes from 5–10 March 2012 and from 1–5 March 2014. The data of the vertical profiles of SS, pressure and temperature were measured using an underwater data logger (Minos X, AML Oceanographic) at 19 stations at Lake Nyos and at 10 stations at Lake Monoun (Fig. 1). Latitude and longitude of sampling points (Tables 1 and 2) were determined using a portable GPS sensor; the accuracy of positions was 20 m, mainly because wind caused the boat to drift during measurement. The profiles of total CO_2 concentration were also determined using the MK method (Kusakabe *et al.* 2000) at the observation rafts of lakes Nyos (Raft) and Monoun (MRaft) (Fig. 1). The MK method and the total CO_2 measurement are described in detail by Ohba *et al.* (2015). The SS data and those of the total CO_2 acquired using the MK method were compared.

Results

The representative SS profiles obtained in the 2014 survey at lakes Nyos and Monoun are

Table 2. *Measurement points for SS profile at Lake Monoun*

Point name	Latitude (°)	Longitude (°)	Date of measurements			
			9 March 2012	10 March 2012	4 March 2014	5 March 2014
MP01	5.57901	10.58056	✓			
MP02	5.57833	10.58171	✓	✓	✓	
MP03	5.57890	10.58368	✓	✓	✓	
MP04	5.58110	10.58632	✓	✓	✓	
MP05	5.58110	10.58869	✓	✓		
MP06	5.57897	10.58670	✓	✓		
MP07	5.57888	10.58871	✓	✓		
MRaft	5.58002	10.58778	✓	✓	✓	
Solar pump	5.57925	10.58800			✓	
MP08	5.58093	10.58728				✓

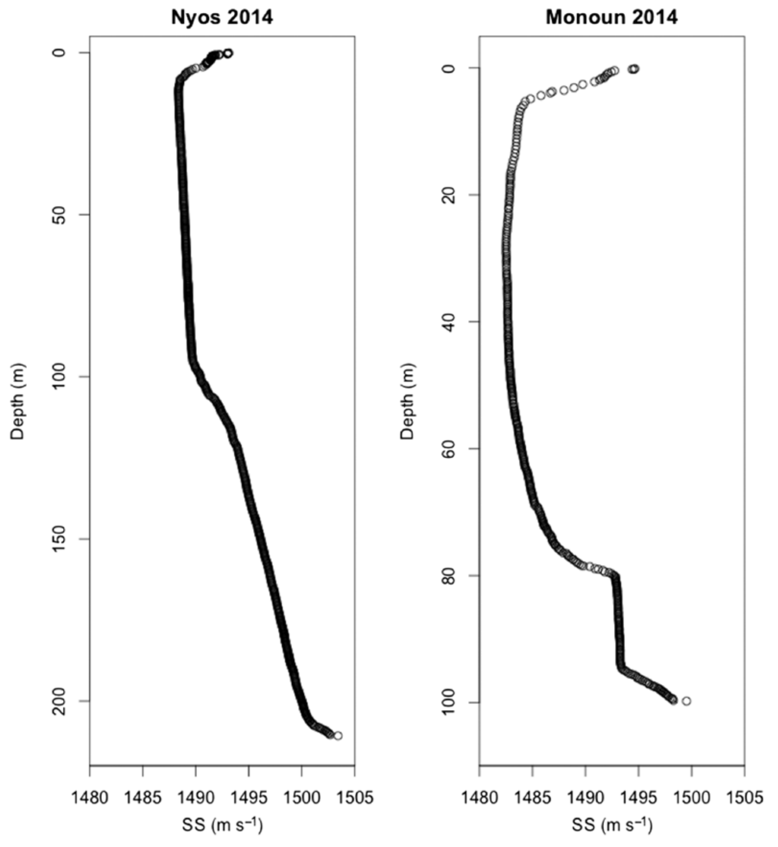

Fig. 2. Representative SS profile obtained in the 2014 survey at lakes Nyos (P06) and Monoun (MRaft).

presented in Figure 2. At both lakes, the SS profiles exhibit a steep decreasing trend around the surface. At Lake Nyos, SS decreased from 1493 m s^{-1} at 0.1 m depth to 1489 m s^{-1} at 7 m, and at Lake Monoun it decreased from 1495 m s^{-1} at 0.1 m to 1484 m s^{-1} at 6 m. It increased again to 1503 m s^{-1} at 209 m depth at Lake Nyos and to 1500 m s^{-1} at 99 m depth at Lake Monoun. To reduce the effect of SS dependence on temperature, we used the parameter Δv, which is the difference between the observed SS in the lake and the SS in pure water at the same temperature and pressure. We used the equation of Belogol'skii *et al.* (1999) to calculate the SS of pure water. Belogol'skii's equation is explained in detail in Sanemasa *et al.* (2015). The following equations were used to convert pressure data to depth.

$$\text{depth [m]} = \frac{209.0 \text{ [m]}}{203.5 \text{ [dbar]}} \times \text{pressure [dbar] at Lake Nyos} \quad (2)$$

$$\text{depth [m]} = \frac{99.0 \text{ [m]}}{95.63 \text{ [dbar]}} \times \text{pressure [dbar] at Lake Monoun} \quad (3)$$

These conversion factors were obtained at the bottom of both lakes from the bottom depth measured using a calibrated wire and the pressure measured using an underwater data logger. Lake Nyos is 209 m deep, corresponding to a pressure of 203.5 dbar; and Lake Monoun is 99.0 m deep, corresponding to a pressure of 95.63 dbar.

Figure 3 presents a diagram of the Δv versus total CO_2 concentration ($[CO_2] = [CO_{2(aq)}] + [HCO_3^-]$) for lakes Nyos and Monoun in 2012 and 2014. Good linear correlation between these two parameters is observed, since Pearson's *r* value is always >0.98. After reduction of the effect of temperature dependence, the decreasing trend of the SS profile near the surface disappeared. The concentration

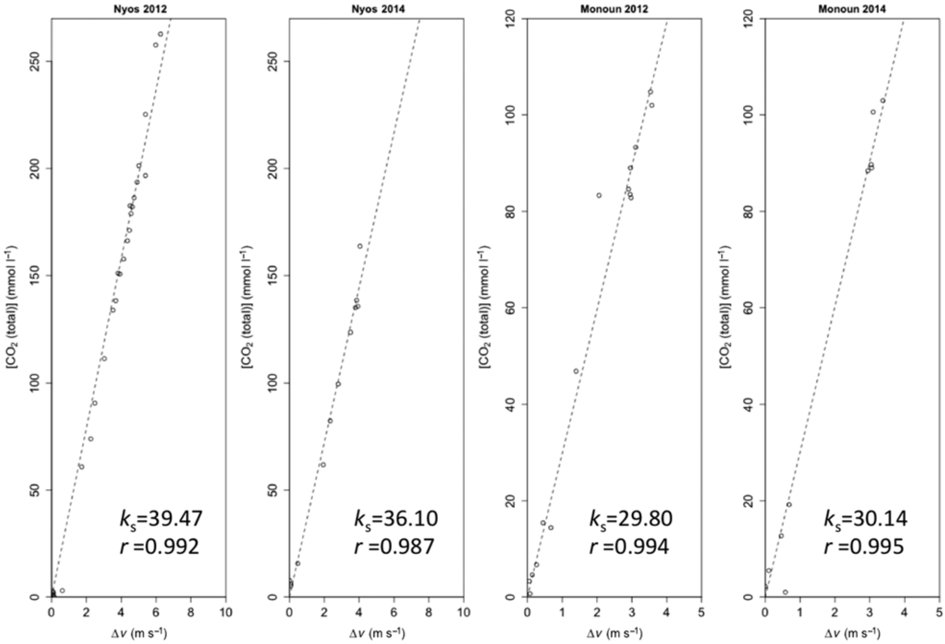

Fig. 3. Correlation between the change in SS and the total CO_2 concentration of lake water with Pearson's r at P06 of Lake Nyos and at MRaft of Lake Monoun. The value of k_s is the slope of the regression line.

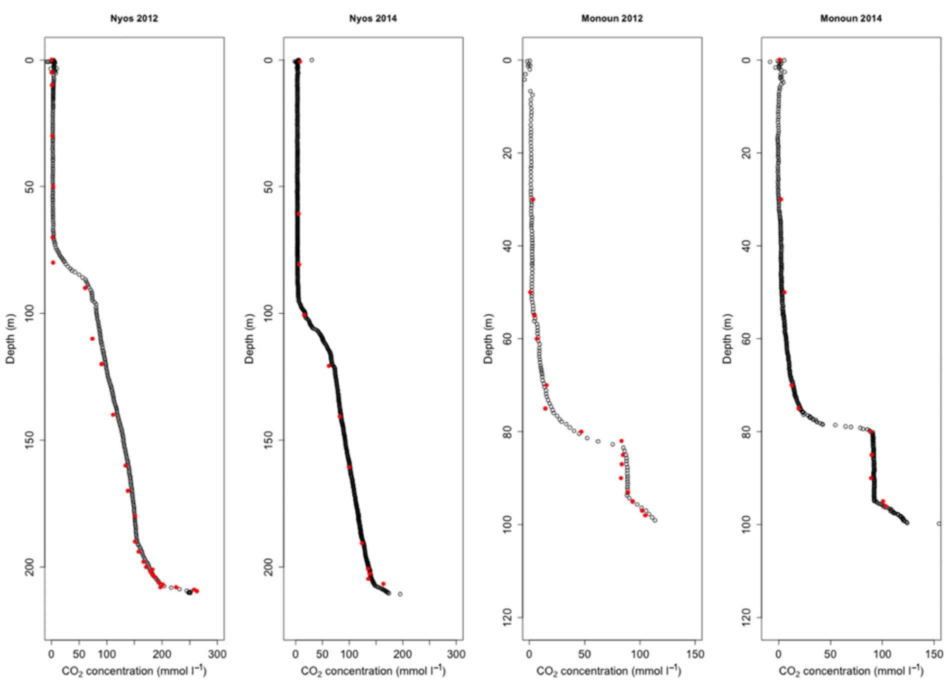

Fig. 4. Vertical profile of total CO_2 concentration estimated by SS change at Lake Nyos (P06) and at Lake Monoun (MRaft). The red dot denotes the results of the MK method.

of dissolved CO_2 was estimated by applying equation (4).

$$[CO_2] = k_s \Delta v \tag{4}$$

The value of k_s was determined as $k_s = 39.47$ from the slope of the regression line in Figure 3 (Lake Nyos) and $k_s = 29.80$ (Lake Monoun) in 2012, and $k_s = 36.10$ (Lake Nyos) and 30.14 (Lake Monoun) in 2014. Assuming a small variation of k_s within the same lake in the same year, we calculated the CO_2 profiles of all measured points. The obtained CO_2 profiles when compared with those obtained using the MK method indicated good agreement (Fig. 4), with differences within the range of ± 20 mmol l^{-1}.

Figure 5 plots all the results from Lake Nyos (Table 1), and Figure 6 plots those from Lake Monoun (Table 2). The multipoint depth profile data indicated the horizontal difference between the vertical profiles of CO_2 in lakes Nyos and Monoun.

Discussion

We found good correlation between SS and total CO_2 concentration at lakes Nyos and Monoun. The SS method is simple and accurate compared with the SS–EC method (Sanemasa *et al.* 2015). Using the SS–EC method, we assumed the formula $\Delta v = k_1[CO_{2(aq)}] + k_2[HCO_3^-]$, and k_1 and k_2 were determined through laboratory and field experiments. Although the k_1 values did not indicate any variations in either lake, in 2014 the k_2 value for Lake Nyos was 0.132 and that for Lake Monoun was 0.0930. However, with the SS method, we assumed the formula $[CO_2] = k_s\Delta v$; here, the k_s values for Lake Nyos are 39.47 (2012) and 36.10 (2014), and those for Lake Monoun are 29.80 (2012) and 30.14 (2014). The range of deviation of the coefficient k_s is smaller than that of k_2. Therefore, when there are no CO_2 data for calibration obtained in the same year, the SS method can be expected to provide more practical results using k_s of a different year. For example, we recalculated all the data with a single k_s value of 33.88, averaged from four k_s values in Figure 3. All results agreed well, with a maximum deviation of only 16%. The variation of k_2 value is due mainly to the change of the $CO_{2(aq)}/HCO_3^-$ ratio. Even if the ratio changes under the influence of other ion species, the total CO_2 does not change. That may be why k_s is relatively more stable than k_2. The difference

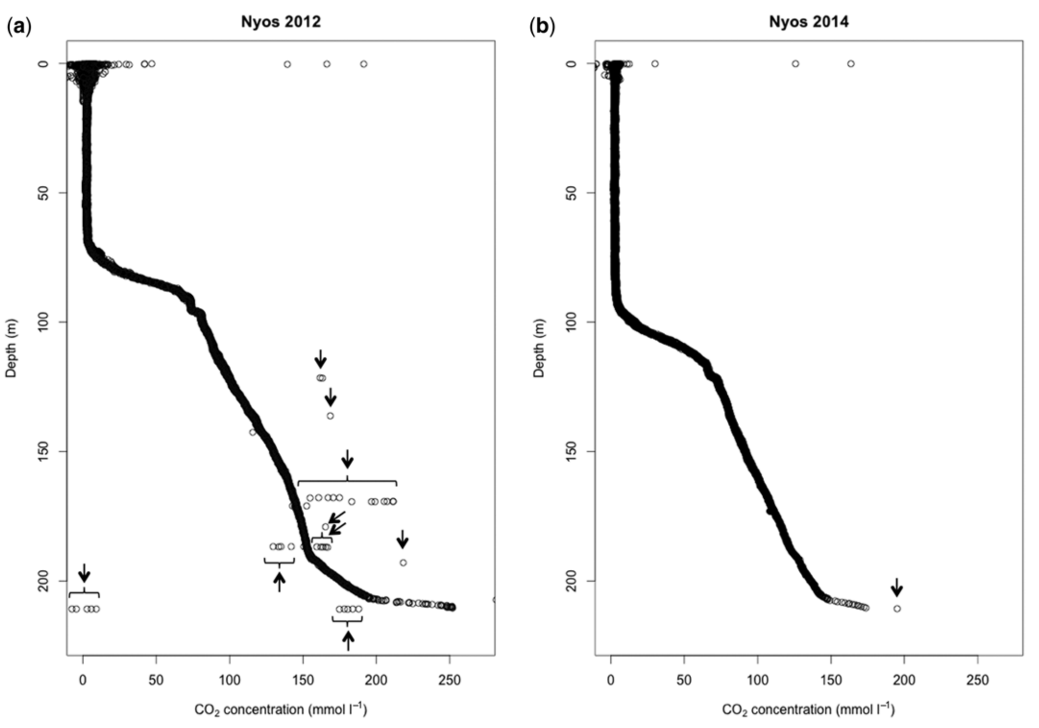

Fig. 5. Vertical profile of total CO_2 concentration estimated by SS change at Lake Nyos in (**a**) 2012 and (**b**) 2014. The twenty-nine measurements in 2012 and seven measurements in 2014 presented in Table 1 are plotted together. Some measurements have anomalous values, which are indicated by arrows at the bottom of each measurement.

in terms of k_s is caused by the variation of the cation species paired with carbonate ion, judging from the good correlation between [CO_2] and Δv. Although further verification is required, the accuracy of the SS method is sufficient for practical CO_2 monitoring. Furthermore, the shape of the profile, especially the layer thickness of high CO_2 concentration, is very useful for checking the effect of degassing and recharge of magmatic CO_2.

The fact that all the profiles at Lake Nyos were almost identical supports strong stratification of the lake observed by Kusakabe *et al.* (2008). The profiles at the same site on different dates for the same year were also identical, suggesting that there were no daily variations. In Figure 5, some points (indicated by arrows) fall outside the main profiles. They refer to the deepest portions of each measurement where anomalous values of dissolved CO_2 were recorded. In 2014, we measured only at deep points and conducted no shallow measurements. In some measurements from 2014 we observed a thin layer with extraordinarily high (sometimes low) SS just above the bottom. However, it is unclear whether the layer actually contains CO_2 as high as estimated by SS or not because the change of SS with depth is too abrupt there; and most of the measurements in 2012 and all measurements in 2014 do not indicate such anomalous values. Moreover, the depth resolution of MK data is too low to be compared with SS data. Possibilities include a chemically anomalous layer with some dense solution just above the bottom, or the effect of mud and other particles scattered by the falling SS sensor.

At Lake Nyos, we confirmed that no change exists in the profiles at all the measuring points in the same year. The thickness of the high CO_2 concentration layer from 100 m depth to the bottom exhibits a decreasing trend from 2012 to 2014. This result indicates the effectiveness of the artificial degassing equipment installed in Lake Nyos in 2001 (Halbwachs *et al.* 2004). However, the profiles at Lake Monoun suggest two patterns. Lake Monoun consists of three basins (Fig. 1). The main basin is the largest and deepest, and contains high gas concentrations (Kling *et al.* 2005). It has a depth of 100 m and is located on the east side. The western basin is 46 m deep, and the central basin is 56 m deep. While there is no distinct boundary between the western and the central basins, a ridge at 20 m depth separates the main basin and the central basin. Figure 6 plots the data at points MP01, MP02, and MP03 (within the western and central basins) in red and the data at the other points

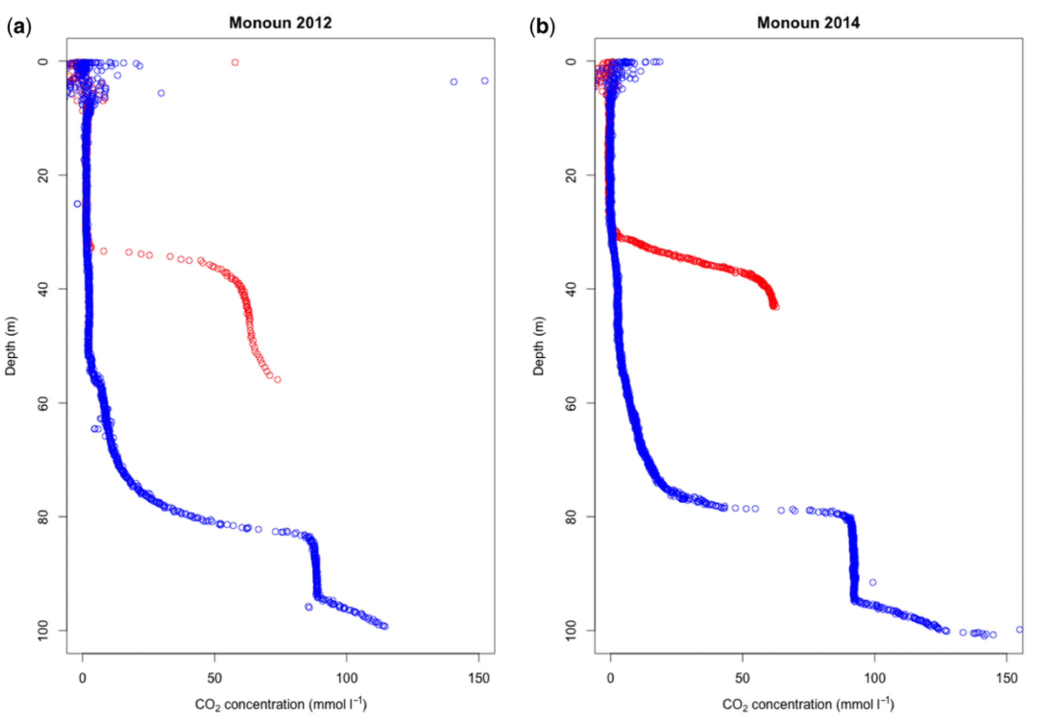

Fig. 6. Vertical profile of total CO_2 concentration estimated by SS change at Lake Monoun in (**a**) 2012 and (**b**) 2014. The fifteen measurements in 2012 and six measurements in 2014 presented in Table 2 are plotted together.

(within the main basin) in blue. The results suggest that lake water in the western and central basins has higher concentrations of CO_2 than those calculated in the waters from the main basin at depths ranging from 35–60 m.

The thickness of the high-CO_2 concentration layer around the bottom of the main basin increased from 2012 to 2014, suggesting a continuous supply of CO_2-rich waters from the bottom (Kusakabe 2015). However, the thickness of the high-CO_2 concentration layer at the bottom of the western and central basins apparently did not change. The depth where the profile branches into the main basin and the other basins is almost the same between 2012 (33 m) and 2014 (31 m). This depth is slightly greater than the depth of the ridge between the main and central basins. Therefore, the SS profiles at the western and central basins suggest that high CO_2 water overflowed from the main basin into the central and western basins when CO_2 concentration in the main basin was high and the high CO_2 layer could get over the ridge (before degassing began) and/or that there is another source of CO_2 in the western or central basin. Because high-density water with a high CO_2 content cannot go over the ridge between the main and central basins, it stays at the bottom.

Conclusions

A new method to determine the total CO_2 concentration using the SS in water was proposed. Multipoint measurement of the SS of lake water was carried out at lakes Nyos and Monoun. To estimate the CO_2 of lake water, we proposed the SS method assuming $[CO_2] = k_s \Delta v$, as there is good correlation between SS and $[CO_2]$. The value of k_s was determined from field data to be $k_s = 39.47$ (Lake Nyos) and 29.80 (Lake Monoun) in 2012, and $k_s = 36.10$ (Lake Nyos) and 30.14 (Lake Monoun) in 2014. The multipoint data indicated the 3D structure of CO_2 distribution in both lakes. All the SS profiles at Lake Nyos were almost identical, suggesting that the lake water was stably stratified even during degassing. The SS profiles at Lake Monoun differed between the main and central basins, indicating that CO_2 concentration in the central basin is higher than that in the main basin at the same depth. In addition, the increase or decrease of the volume of the high-CO_2 layer of each lake can easily be determined using this SS method.

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