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**GHG emissions from  
alpine reservoirs**

T. Diem et al.

# Greenhouse gas emissions (CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O) from perialpine and alpine hydropower reservoirs

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## Abstract

In eleven reservoirs located at different altitudes in Switzerland depth profiles of greenhouse gas ( $\text{CO}_2$ ,  $\text{CH}_4$ , and  $\text{N}_2\text{O}$ ) concentrations were measured several times during spring and summer. Trace gas emissions were calculated using surface concentrations, wind speeds and transfer velocities. Additionally we assessed methane loss at the turbine and the methane input by inflowing water. All reservoirs were net emitters of  $\text{CO}_2$  with an average of  $1030 \pm 780 \text{ mg m}^{-2} \text{ d}^{-1}$  and of methane with an average of  $0.20 \pm 0.15 \text{ mg m}^{-2} \text{ d}^{-1}$ . One reservoir (Lake Wohlen) emitted methane at a much higher rate ( $160 \pm 110 \text{ mg m}^{-2} \text{ d}^{-1}$ ), most of which (>98%) was due to ebullition. Only lowland reservoirs were sources for  $\text{N}_2\text{O}$  ( $72 \pm 22 \text{ } \mu\text{g m}^{-2} \text{ d}^{-1}$ ), while the subalpine and alpine reservoirs seem to be in equilibrium with atmospheric concentrations. Methane loss at the turbine was as large as the diffusive flux from the surface for two subalpine reservoirs and around five times smaller for a lowland reservoir. The available data suggests greenhouse gas emissions from reservoirs in the Alps are minor contributors to the global greenhouse gas emissions.

## 1 Introduction

In the early 1990s artificial lakes and reservoirs were discovered as potential greenhouse gas emitters (Kelly et al., 1994; Rudd et al., 1993). The question was put forward, whether hydroelectric reservoirs, especially in the tropics could still be considered cleaner energy sources, compared to fossil alternatives (Fearnside, 1997, 2002). There is however, a high variety of trace gas emissions between different reservoirs, which leads to large uncertainties in the quantification of global emissions. Another problem is the small number of available data. Several studies were done in tropical (e.g. Galy-Lacaux et al., 1997, 1999; Keller and Stallard, 1994; Rosa et al., 2003, 2004) and boreal reservoirs (e.g. Duchemin et al., 1995; Rudd et al., 1993; Tremblay et al., 2005), but so far there is limited information about emissions from reservoirs

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in the temperate climate zone (Soumis et al., 2004). Both in the tropics and in boreal/temperate reservoirs CO<sub>2</sub> is the most emitted trace gas (St. Louis et al., 2000). Taking the Global Warming Potential (GWP) into account, methane release in the tropics contributes more to global warming than CO<sub>2</sub> since the GWP of CH<sub>4</sub> is 25 times that of CO<sub>2</sub> on a 100 year basis (Forster et al., 2007).

In this study we try to increase the knowledge of greenhouse gas emissions from hydropower reservoirs across an altitude gradient in the Alps, Central Europe. We calculated diffusive fluxes of CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O from the surface concentrations of several Swiss reservoirs at different times of the year. Eleven reservoirs at different altitudes allowed a comparison of greenhouse gas emissions under different climatic conditions and possible changes in the importance of CO<sub>2</sub> compared to CH<sub>4</sub> release.

Nitrous oxide emissions from freshwater bodies published so far are very small and often linked to the littoral zones of the lakes (Huttunen et al., 2003; Wang et al., 2007). N<sub>2</sub>O production in lakes is not limited by low temperatures, as Huttunen et al. (2002) reported N<sub>2</sub>O accumulation during winter in two boreal reservoirs. Swiss alpine reservoirs have little or no littoral zones, as the water depth increases very fast along the steep mountain slopes and the reservoirs are usually deeper than 50 m. By measuring N<sub>2</sub>O concentrations in reservoirs at different elevations the relevance of littoral zones for N<sub>2</sub>O production in cold water reservoirs and consequently N<sub>2</sub>O emissions is investigated.

Besides methane supersaturation in lakes and reservoirs, enhanced methane concentrations have been found in rivers (Abril et al., 2006; de Angelis and Lilley, 1987) and below the outlet of reservoirs (Kemenes et al., 2007). The importance of these additional sources and sinks of methane in hydropower reservoirs is assessed by methane concentration and carbon isotopic composition measurements. The isotopic composition of methane can be used as a tracer to distinguish between different sources of methane. Biogenic methane usually has a more negative  $\delta^{13}\text{C}$  than thermogenic methane (Whiticar et al., 1986). The only process relevant in lakes having an essential effect on the isotopic signal is methane oxidation, which can result in a significant

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increase in  $\delta^{13}\text{C}$  (Barker and Fritz, 1981; Whiticar, 1999). Turbulent diffusion does not affect the isotopic composition of methane and the change caused by methane emission is small (Knox et al., 1992). The use of carbon isotopic composition also helps to distinguish between internal and external methane sources in the reservoirs

## 2 Study sites

From September 2003 to August 2006 eleven Swiss reservoirs from different areas and elevations were sampled for greenhouse gases (Table 1 and Fig. 1). The reservoirs are distributed along an elevation gradient from 481 to 2368 m a.s.l. Hence, climate varies greatly between the different reservoirs. Average yearly temperatures range from about  $8^\circ\text{C}$  at Lake Wohlen to nearly  $0^\circ\text{C}$  at Lake Oberaar. Precipitation in the reservoirs of the central part of Switzerland is up to four times higher than for the reservoirs in the southwestern parts and eastern parts of the country.

Electricity production for most Swiss storage reservoirs is higher in winter than in summer, while the majority of the water filling the reservoirs is available from spring to autumn. This leads to a large decline in reservoir water level. In some cases the remaining water in early spring is less than 10% of the maximum water volume.

Two of the reservoirs (Lakes Oberaar and Sihl) are pump-storage reservoirs, which receive water from a lower lying reservoir (Lake Grimsel for Lake Oberaar) or lake (Lake Zurich for Lake Sihl). While the water volume of Lake Oberaar gets replaced up to 10 times every year, pumping only contributes a minor part to Lake Sihl.

Seven of the reservoirs (Lake Wohlen, Lake Gruyère, Lake Sihl, Lake Luzzzone, Lake Santa Maria, Lake Oberaar and Lake Lungern) were sampled several times during the campaign to investigate changes in the temporal development of the greenhouse-gas concentrations. The first six reservoirs were sampled three times from May 2005 to August 2005. Due to snow conditions, measurements at higher elevations started later than ones in lower elevations. Lake Lungern was sampled once in October 2005 and three times between August and September 2006.

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### 3 Methods

#### 3.1 Hydrographic data

A SBE 19 CTD probe (Sea Bird Electronics) equipped with oxygen and pH sensors was used to collect hydrographic data (conductivity, temperature, depth, light transmission, pH and dissolved oxygen). The water column was sampled with a 5l Niskin bottle and aliquots were immediately transferred into sample bottles with plastic tubing (Winkler bottles for oxygen, 600 ml glass bottles for methane and nitrous oxide concentration and 200 ml plastic bottles for alkalinity). Winkler samples were used to correct the offset in the oxygen sensor. Alkalinity was titrated with 0.1 M HCl. Samples for dissolved gas analysis were flushed with 2–4 times the bottle volume before the samples were preserved with NaOH (pH > 12) or Cu(I)Cl, closed with butyl septa, while carefully avoiding air bubbles in the bottles. To calibrate the pH sensor, solutions of known pH (pH=4, 7 and 9) were used before each sampling date.

#### 3.2 Dissolved gases

##### 3.2.1 CO<sub>2</sub>

Dissolved CO<sub>2</sub> was calculated using the measured alkalinity, temperature, pH, and the dissociation constants of H<sub>2</sub>CO<sub>3</sub> and HCO<sub>3</sub><sup>-</sup> (Plummer and Busenberg, 1982). Samples for alkalinity were only taken at the surface and at the bottom of the water column.

According to Neal et al. (1998) the error of CO<sub>2</sub> concentration calculations using temperature and pH is less than 2% for pH values < 9. The error of pH measurement is more important and an uncertainty of 0.05 units results in an error of 12% for CO<sub>2</sub> concentrations.

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### 3.2.2 CH<sub>4</sub> and N<sub>2</sub>O

Concentrations of dissolved methane and nitrous oxide were measured by the headspace technique similar to McAuliffe (1971). A sample volume of 50 ml was replaced by an inert gas (helium or nitrogen) and equilibrated in an ultra-sonic bath for about 30 min. Nitrous oxide was measured with a Dani 86.10 HT gas chromatograph (GC) with a Porapak Q column (Supelco) and an electron capture detector (ECD). The oven temperature was kept constant at 70°C and the detector temperature was 340°C.

Methane concentrations were measured on a HRGC 5160 Mega Series (Carlos Erba Instruments) with a flame ionization detector (FID), a GS-Q P/N 115-3432 column (J and W Scientific) and hydrogen as a carrier gas. Temperatures were 40°C for the oven and 200°C for the detector. Gas volumes of 2 ml for N<sub>2</sub>O and 200 µl for CH<sub>4</sub> were injected. Replicate measurements yielded an accuracy of ±5% for methane and ±10% for nitrous oxide.

From 2006 on, the measurements were made on an Agilent GC using a GS-Carbonplot column (Agilent) for nitrous oxide and a Carboxen 1010 Plot column (Supelco) for methane. Again an ECD was used for N<sub>2</sub>O and a FID for CH<sub>4</sub>. The temperature was kept constant at 40°C for 5 minutes and then raised to 120°C at the rate of 10°C/min. The GC had a 1-ml sample loop for nitrous oxide and a 500 µl sample loop for methane. Accuracy on this GC improved to ±3% for methane and ±5% for nitrous oxide.

Standards used for calibration were supplied from Scotty Specialty Gases. Concentrations were 15 ppm, 1000 ppm and 1% for methane and 1 ppm and 10 ppm for nitrous oxide.

Dissolved gas concentrations were calculated using solubility data from Wiesenburg and Guinasso (1979) for methane, from Weiss and Price (1980) for nitrous oxide and from Weiss (1974) for carbon dioxide.

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### 3.3 Stable isotopes

The isotopic signature of methane was determined similar to the method described by (Sansone et al., 1997). Measurements were done with an IsoPrime mass spectrometer connected to a TraceGas preconcentrator (GV Instruments, UK). The amount of injected gas depended on the sample concentration, ranging from a few  $\mu\text{l}$  to several ml. Results are noted in the standard  $\delta$ -notation relative to Vienna PeeDee Belemnite (VPDB).

$$\delta^{13}\text{C} = \left( \frac{R_{\text{sample}}}{R_{\text{Reference}}} - 1 \right) \times 1000 \quad (1)$$

Here  $R_{\text{sample}}$  is the ratio of  $^{13}\text{C}/^{12}\text{C}$  of the sample and  $R_{\text{reference}}$  the ratio of the reference material and  $\delta^{13}\text{C}$  the isotopic signature of methane in ‰ vs. VPDB. A Standard (1%  $\text{CH}_4$  in argon) of known isotopic composition was injected between every two or three sample runs. The accuracy of the method was  $\pm 0.7$  ‰.

### 3.4 Gas fluxes

Greenhouse-gas fluxes were calculated using the boundary layer model as described by Liss and Slater (1974).

$$F = k \cdot c \cdot (C_w - C_{eq}) \quad (2)$$

The model estimates the air-water flux  $F$  [ $\text{mg m}^{-2} \text{d}^{-1}$ ] using the water saturation concentration  $C_{eq}$  [M], the measured water concentration  $C_w$  [M] of the greenhouse-gas, the transfer velocity  $k$  [ $\text{cm h}^{-1}$ ] and a unit conversion factor  $c$ . For the calculation of the transfer velocity  $k$  we used the bi-linear relationship given by (Crusius and Wanninkhof, 2003):

$$\text{for } U_{10} < 3.7 \text{ m s}^{-1} \quad k_{600} = 0.72 U_{10}$$

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$$\text{for } U_{10} > 3.7 \text{ m s}^{-1} \quad k_{600} = 4.33U_{10} - 13.3$$

where  $k_{600}$  is the transfer velocity for the Schmidt number  $Sc=600$ , and  $U_{10}$  the wind speed ten metres above the ground. To convert  $k_{600}$  to the actual transfer velocity  $k$  of the gas we used

$$k = k_{600}(Sc/600)^c \quad (3)$$

where  $Sc$  is the Schmidt number of the greenhouse gas ( $\text{CH}_4$ ,  $\text{CO}_2$  and  $\text{N}_2\text{O}$ ) at water surface temperature and  $c$  is  $-2/3$  for  $U_{10} < 3.7 \text{ m s}^{-1}$  and  $-1/2$  for higher wind speeds (Liss and Merlivat, 1986).

Equilibrium concentrations were determined using an air concentration of 1.77 ppm  $\text{CH}_4$ , 379 ppm  $\text{CO}_2$  and 319 ppb  $\text{N}_2\text{O}$  (Forster et al. 2007), corrected to the reduced pressure of the lake elevation, and measured water temperatures. Schmidt numbers were calculated for the measured water temperatures according to Wanninkhof (1992) and the authors cited therein. Wind data was supplied by MeteoSwiss from the ANETZ- or ENET-Station closest to the lake in question.

### 3.5 Methane mass balance studies

Several of the following methods were used to allow a simple mass balance of methane fluxes in and out of the reservoirs. Table 1 gives an overview what measurements were done in which reservoirs.

#### 3.5.1 Sediment cores

Cores were taken with a gravity corer. Samples were collected every centimetre from holes in the side of the core using a plastic syringe. 2 ml of sediment and 5 ml of  $\text{NaOH}$  (5%) were filled into 25 ml glass bottles and closed with butyl septa. Methane concentration and isotopic composition were measured in the headspace as described above.

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### 3.5.2 Ebullition

To estimate methane flux from the sediments via bubbles, funnels were deployed around one meter above the sediment and left for several hours in one spot. Gas bubbles were sampled in a cylinder on top of the funnel. The volume of gas was estimated from the area of the cylinder no longer filled with water. Samples were collected from the cylinder through a butyl septum and filled into pre-evacuated, sealed 25 ml glass bottles. These bottles were taken to the lab and analyzed for concentrations and isotopic signature as described above.

### 3.5.3 Inflows, outflows

Methane flowing in and out of the reservoirs was sampled in six reservoirs. If possible the CTD probe was used, but if depth of the river was not sufficient, temperature and conductivity were measured with a WTW LF 330 conductivity meter, pH with a Metrohm 704 pH-meter and oxygen with a WTW Multi 340i multi probe. Samples for methane measurements were sampled as described above. Only major inflows were sampled, as well as spill-water from the reservoir and water after passage of the turbine.

## 4 Results

### 4.1 CO<sub>2</sub> concentrations and emissions

Surface concentrations of CO<sub>2</sub> were supersaturated in all five reservoirs for which data are available (Table 2), with concentrations ranging from 40–280 μM. This is in accordance with findings by Cole et al. (1994), who analyzed lakes world-wide, most of which (87%) were supersaturated with an average of about three times the equilibrium concentration (~15 μM). In a tropical reservoir water CO<sub>2</sub> concentrations were in the same range (50–400 μM) as our data (Abril et al., 2006).

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The fluxes we calculated were on average  $1030 \pm 780 \text{ mg m}^{-2} \text{ d}^{-1}$  (Median  $710 \text{ mg m}^{-2} \text{ d}^{-1}$ ). Emissions at the first sampling dates in May are nearly four times higher ( $1820 \pm 490 \text{ mg m}^{-2} \text{ d}^{-1}$ ) than at the remaining dates ( $520 \pm 290 \text{ mg m}^{-2} \text{ d}^{-1}$ ; Table 2).

In low conductivity lakes we had a problem with the pH sensor we used. The accuracy of the sensor was not sufficient for these conditions and concentrations and fluxes for reservoirs with conductivities below  $100 \mu\text{S cm}^{-1}$  were not calculated.

## 4.2 Nitrous oxide concentrations

Concentration ranges and profiles were similar to open water concentrations found in other studies on lakes and reservoirs (Huttunen et al., 2003; Mengis et al., 1996). Minimum concentration was 6 nM (55% saturation) in Lake Zeuzier and 41 nM (260% saturation) at the bottom of Lake Lungern. Figure 2a and b show a typical profile for an alpine reservoir (Lake Grimsel) and for a lowland reservoir (Lake Lungern). The remaining profiles are documented in the electronic supplement (<http://www.biogeosciences-discuss.net/5/3699/2008/bgd-5-3699-2008-supplement.pdf>, Figs. B1–B3).

Concentrations in the three alpine reservoirs were close to the atmospheric equilibrium concentration in the water column. Although they differed from equilibrium concentrations at the surface we assumed these alpine reservoirs to be neither a source nor a sink. Both lowland reservoirs were supersaturated with  $\text{N}_2\text{O}$  throughout the water column, being small nitrous oxide sources of  $72 \pm 22 \mu\text{g m}^{-2} \text{ d}^{-1}$  (Lake Wohlen) and  $50 \pm 13 \mu\text{g m}^{-2} \text{ d}^{-1}$  (Lake Lungern, all sampling campaigns).

## 4.3 Methane concentrations, $\delta^{13}\text{C}$ isotopic composition and emissions

In the following, we will group the reservoirs into three categories and illustrate the different categories with one example profile. The profiles of the remaining reservoirs are documented in the electronic supplement (<http://www.biogeosciences-discuss.net/>

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The categories are:

- (i) uniform methane profile (Lakes Oberaar, Dix, Bianco, Grimsel and Wohlen),
- (ii) increasing methane concentrations towards the sediment (Lakes Sihl, Zeuzier, and Santa Maria),
- (iii) profiles with methane maxima in the water column (Lakes Luzzone, Lungern, and Gruyère).

(i) uniform methane profiles

To greater or lesser extent constant methane concentrations in the water column were found in these five reservoirs. In Lake Dix, concentrations were around 20 nM at most depths, and up to three times higher at certain depths (Fig. 3a). These depths coincided with small temperature perturbations and were most likely the result of inflowing water, stratifying in these layers (Fig. 3b). The water was warmer than the remaining lake water because it was pumped from lower altitude reservoirs or neighbouring valleys into Lake Dix. While the  $\delta^{13}\text{C}$  values for most of the reservoir were between  $-40\text{‰}$  and  $-37\text{‰}$ , at the depths with enhanced concentrations, the  $\delta^{13}\text{C}$  also differed from the remainder of the reservoir, which supported the hypothesis of an external  $\text{CH}_4$  source. Sediments did not seem to contribute to the methane content of the reservoir.

Concentrations in Lake Bianco were at saturation level ( $\sim 3\text{ nM}$ ), hence the methane emissions were negligible (see Table 2). The three other alpine reservoirs Lake Dixence, Oberaar and Grimsel emitted methane at  $0.05\text{ mg m}^{-2}\text{ d}^{-1}$ ,  $0.28\pm 0.03\text{ mg m}^{-2}\text{ d}^{-1}$ , and  $0.37\pm 0.16\text{ mg m}^{-2}\text{ d}^{-1}$ , respectively.

*Lake Wohlen*

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Methane concentrations nearly doubled from the entrance of the reservoir towards the dam, where concentrations at the surface were 880 nM in May 2005 and around 650 nM for all other dates we sampled (Fig. 4). The profiles in the basin in front of the dam barrage were not uniform, in July 2005 and September 2006 concentrations increased towards the bottom and reached values of 1470 nM and 1000 nM respectively. Changes in concentrations within the year 2005 were marginal except for the barrage basin, where concentrations were about 300 nM higher.

The carbon isotopic composition changed little over the measurement period, but changes occurred with distance from the dam. Values at the entrance of the reservoir were around  $-50\text{‰}$ , decreasing to  $-60\text{‰}$  in the middle of the reservoir and at the dam.

Diffusive fluxes in Lake Wohlen were one order of magnitude higher than in the other reservoirs at  $6.3 \pm 3.6 \text{ mg m}^{-2} \text{ d}^{-1}$ . As we noticed intense bubble flux in some areas of the lake we also measured the ebullition flux from this reservoir with funnels. From these funnel measurements we found ebullition to account for an additional  $900 \pm 700 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$  (Range 200–2000  $\text{mg m}^{-2} \text{ d}^{-1}$ , Median 700  $\text{mg m}^{-2} \text{ d}^{-1}$ ) (Electronic supplement Table C1). Assuming the area of intense bubble flux to be approximately one third of the reservoir, preliminary ebullition fluxes for Lake Wohlen would be  $300 \pm 230 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ . This is in the range of results Keller et al. (1994) and Galy-Lacaux et al. (1997) found for two tropical reservoirs, but more than three times higher, than what dos Santos et al. (2006) found for reservoirs in Brazil or Duchemin et al. (1995) and Huttunen et al. (2002) for several boreal reservoirs.

(ii) increasing methane concentrations from the surface to the sediment

The reservoirs in this category had a more or less steady increase of methane concentrations from the water surface to the sediment surface in common. Concentrations increased with a gradient close to  $1 \text{ nmol m}^{-1}$  (range 0.4–1.4) in Lakes Zeuzier and Santa Maria, and about ten times faster in the shallow Lake Sihl (range 5.4–12.8  $\text{nmol m}^{-1}$ ). Methane diffusing from the sediment seems to be the cause for

the higher concentrations at the lake bed.

In Lake Santa Maria, methane concentrations on all three sampling dates (June, July, and August) increased towards the bottom (Fig. 5a). In June surface concentrations were 55 nM, while on the other two dates concentrations were about 15 nM.

Concentrations right above the sediment decreased from June to August, from 100 nM to 63 nM. Meanwhile the carbon isotopic signal of methane in June was  $-34\text{‰}$  from the surface down to 40 m and the decreased to  $-40\text{‰}$ . On the other two dates the value at the surface was  $-40\text{‰}$  and values decreased to  $-54\text{‰}$  in July and  $-51\text{‰}$  in August. The rapid change in the isotopic composition is reflected in temperature (Fig. 5b) and other hydrographic parameters (data not shown) as well.

The emission flux was lowest in Lake Zeuzier at  $0.07 \text{ mg m}^{-2} \text{ d}^{-1}$ , three times higher in Lake Sihl at  $0.21 \pm 0.08 \text{ mg m}^{-2} \text{ d}^{-1}$  and still higher in Lake Santa Maria at  $0.32 \pm 0.29 \text{ mg m}^{-2} \text{ d}^{-1}$ . Emissions in Lake Santa Maria were highest in June 2005 with  $0.65 \text{ mg m}^{-2} \text{ d}^{-1}$ , while in July and August values were similar to the ones in Lake Luzzone for the same time span ( $0.15 \pm 0.06 \text{ mg m}^{-2} \text{ d}^{-1}$ ).

(iii) enhanced methane concentrations in an intermediate layer

Profile shapes in these reservoirs did not increase steadily from the water surface to the sediment, but a (local) maximum in methane concentrations was detected in intermediate water layers. From the lower methane minimum downwards, concentrations increased again towards the sediment. We suggest methane entering the reservoir with inflowing water stratifying at intermediary depth to be the reason for this profile shape.

Lake Luzzone was sampled twice in July and August 2005 (Fig. 6). Both times, methane concentrations in the enhanced intermediary layer were about twice as much as the upper and lower minimum. Concentrations increase again towards the sediment and reach the highest concentrations above the sediment at 100 nM in August. The carbon isotopic compositions followed the concentration profile with values between

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–38 and –40‰ at the minima and –50 to –52‰ at higher concentrations.

Methane emissions from each lake individually were  $0.15 \pm 0.02 \text{ mg m}^{-2} \text{ d}^{-1}$ ,  $0.13 \pm 0.12 \text{ mg m}^{-2} \text{ d}^{-1}$  and  $0.13 \pm 0.01 \text{ mg m}^{-2} \text{ d}^{-1}$  for Lake Gruyère, Lake Lungern and Lake Luzzone, respectively. Changes during the sampling period were small in Lake Gruyère and Lake Luzzone, while in Lake Lungern emissions decreased in the year 2006 from  $0.34 \pm 0.08 \text{ mg m}^{-2} \text{ d}^{-1}$  in early August to  $0.07 \pm 0.06 \text{ mg m}^{-2} \text{ d}^{-1}$  for four sampling dates in September and October. The situation in Lake Lungern was special however, as due to a defect in that year large amounts of water supersaturated with air flowed into the lake. This resulted in air saturations of more than 150% and caused a mixing of the water body down to 40 m, while in normal years the epilimnion is only 10 m deep in summer (Jaun et al., 2006). However, emissions in October 2006 were not much lower than in October 2005, when no malfunction occurred.

#### 4.4 Inflows and outflows

Methane concentrations of inflows and outflows are given in Table 2. Concentrations were supersaturated with methane for all sampling dates, with a range of 10 to 420 nM. Methane loss at the turbine was calculated by using the concentration of the closest sampling point to the depth of the outlet. Concentrations did not differ before and after the turbine in Lakes Wohlen and Gruyère, while at Lakes Sihl, Luzzone and Grimsel concentrations are between 16 and 73% lower after the water passed the turbine (Table 2).

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## 5 Discussion

### 5.1 Diffusive surface emissions

#### 5.1.1 Carbon dioxide

Similar to other boreal and temperate reservoirs, CO<sub>2</sub> is the most important (in regard to volume and GWP) greenhouse gas emitted from Swiss reservoirs. The emissions are in the same range as emissions from other temperate and boreal reservoirs (Table 3).

In their analysis Tremblay et al. (2005) found among others correlations between CO<sub>2</sub> flux and the parameters pH and month of sampling. In our data (except for Lake Luzzone), there is as well a correlation between CO<sub>2</sub> flux and pH ( $R^2=0.741$ ,  $p<0.0001$ ), which can easily be explained by our calculation of the CO<sub>2</sub> concentration from alkalinity and pH. Another correlation existed between CO<sub>2</sub> flux and date ( $R^2=0.6137$ ,  $p<0.0001$ ). There is, however, no significant correlation between time and elevation for the three Lakes Wohlen, Gruyère and Sihl (ANOVA,  $F=6.6387$ ,  $p=0.057$ ). Both correlations are done with a low sample number ( $n=11$ ) and have to be treated with caution. The agreement with the findings of Tremblay et al. (2005), however, suggests the effect of season on CO<sub>2</sub> emissions to be real.

The date of separation was set somewhat arbitrarily, but for all lakes (except Lake Luzzone) emissions decreased from spring (May, early June) to summer, while pH increased during the same time (data not shown). This suggests a reduced CO<sub>2</sub> emission in summer, not from decreased DIC concentrations, but from a shift to bicarbonate and lower dissolved CO<sub>2</sub> concentrations.

#### 5.1.2 Nitrous oxide

Similar to previous findings fluxes of N<sub>2</sub>O in lakes and reservoirs are small in open water (Huttunen et al., 2002) and are slightly higher in littoral zones (Huttunen et al.,

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2003). While emissions from the two lowland reservoirs are in the same range as previous results (Table 3), in the alpine reservoirs concentrations throughout the water column are very close to atmospheric equilibrium. Only N<sub>2</sub>O concentrations at the surface deviate from the equilibrium concentrations. There is no production of N<sub>2</sub>O in the deep alpine reservoirs we sampled and we assume calculated fluxes are probably overestimated. However, the amount of data from alpine reservoirs is limited and alpine reservoirs might be small sources and sinks for N<sub>2</sub>O as well, depending on the continuity of surface concentrations differing from the atmospheric equilibrium.

Overall, if we take the “Global Warming Potential” into account (for a 100 year time period the impact of N<sub>2</sub>O is about 12 times higher than the one of CH<sub>4</sub>; Forster et al., 2007), the fluxes of N<sub>2</sub>O in the lowland lakes have the same warming potential as CH<sub>4</sub>.

### 5.1.3 Methane

All lakes except Lake Bianco were supersaturated with methane. Lake Bianco was saturated within measurement accuracy and does not emit methane to the atmosphere (Table 2).

If we separate the lakes into alpine, subalpine and lowland lakes and exclude Lake Wohlen, we get emissions of  $0.25 \pm 0.17 \text{ mg m}^{-2} \text{ d}^{-1}$ ,  $0.21 \pm 0.21 \text{ mg m}^{-2} \text{ d}^{-1}$  and  $0.16 \pm 0.09 \text{ mg m}^{-2} \text{ d}^{-1}$ , respectively. There are no significant differences between the different elevation levels (ANOVA,  $F=1.651$ ,  $p=0.21$ ) and none between emissions and date.

In comparison to reservoirs from other regions (Table 3), emissions from the eleven Swiss reservoirs we sampled are more than one order of magnitude lower than emissions from temperate and boreal reservoirs and three orders of magnitude lower than methane emissions from tropical reservoirs. Only the diffusive flux from Lake Wohlen is in the same range as the ones from the temperate and boreal reservoirs, while the ebullition flux from this reservoir is in the range of emissions from tropical reservoirs.

There are several possible reasons, why the diffusive methane fluxes in our study are much lower, than the ones in other studies. The most important ones are

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- (i) the use of the thin boundary layer model (TBL) to calculate emissions
- (ii) age of the reservoirs and
- (iii) shape of the reservoirs

#### 5 (i) Thin Boundary Layer method

It has been shown (Duchemin et al., 1999), that the TBL tends to underestimate fluxes as opposed to emissions measured by static chambers, especially in shallow water. In a comparison of the two methods, Matthews et al. (2003) found differences of more than an order of magnitude for very low wind speeds. They suggested additional turbulences caused by the chambers to be the cause for this discrepancy. However, Soumis et al. (2004) found good agreement between the two methods for wind speeds under  $3 \text{ m s}^{-1}$ , as did Gu erin et al. (2007) who let their chambers float free with the river to avoid additional turbulence. An underestimation in our fluxes can not be excluded; however, as both methods in most cases are within the same order of magnitude the low fluxes we found seem reasonable.

The choice, which linear relationship is used to calculate the fluxes can also influence the results. We used the bi-linear relationship from Crusius and Wanninkhof (2003), and not the constant-linear one, since a comparison of these two showed only minor differences in emissions (data not shown). However, when we use the relationship of Cole and Caraco (1998) differences between the lowland and the subalpine/alpine reservoirs are significant (ANOVA,  $F=6.415$ ,  $p=0.0061$ ), leading to higher emissions from lowland reservoirs.

#### 25 (ii) Age

Studies of newly impounded reservoirs reported a peak of greenhouse gas emissions shortly after the flooding, caused by the degradation of the submerged organic material

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that was present in the area. Emissions decline after two to three years (Galy-Lacaux et al., 1999) and reach a steady state at a much lower level after 10 years (Abril et al., 2005).

All reservoirs we investigated were older than 35 years at the date of the sampling (Table 1), thus emissions should have levelled off and reached their “base level” more than twenty years ago. For reservoirs, that flooded areas rich in organic material the “base level” is maintained by the degradation of carbon stored in the flooded soil. The rather low emissions of the Swiss reservoirs could be an indication of low productivity of the reservoirs and a used up carbon stock in the sediments.

### (iii) Reservoir shape

Another important factor in emissions from lakes and reservoirs is the shape of the basin. Duchemin et al. (1995) found emissions from deep parts of LaGrande-2 reservoir to be less than half of the emissions from shallow areas. Additionally littoral zones from lakes and reservoirs have been identified as important areas for greenhouse gas emissions (Huttunen et al., 2003; Wang et al., 2007). The subalpine and alpine reservoirs in Switzerland were created in narrow gorges with steep slopes, thus the ratio of shallow to deep areas is very small in most cases. As a result littoral zones are small and of minor importance and low flux areas determine the overall emissions.

## 5.2 Other emission pathways

### 5.2.1 Loss at the turbines

Another important emission pathway for reservoirs is the loss caused by the turbulence caused during turbine passage. Of the five lakes we sampled for investigating methane loss at the turbine, two (Lake Wohlen and Lake Gruyère) did not lose methane (Table 2). The average loss for the three other lakes (Lake Sihl, Lake Luzzone

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and Lake Grimsel) was  $46 \pm 18\%$  (Range 16–73%), matching the findings of Kemenes et al. (2007). Whereas at Lake Wohlen and Lake Gruyère the water drops only a few meters down to the river, for the other three reservoirs, the height difference between the dam and the river is several hundreds of meters. While this drop seems to create enough turbulence for the water to degas on its way down the pipe, the short drop through the air does not seem to be sufficient for a measurable loss of gas.

If we use the average loss at the turbine to calculate the importance of this emission path for the total methane flux, we receive  $51 \pm 13\%$  for the subalpine and alpine reservoirs, while for Lake Sihl gas loss at the turbine accounts for  $14 \pm 7\%$  of the total emissions at the time of the measurement.

### 5.2.2 Ebullition

Ebullition in Lake Wohlen is nearly two orders of magnitude higher than the diffusive surface flux and is thus the most important emission pathway of methane in this reservoir. We believe the high input of organic matter caused by the short water residence time of this run-of-the-river reservoir and subsequent anaerobic degradation is the source of the high bubble flux and the high methane emissions compared to the other Swiss reservoirs. Given the low methane concentrations, especially in alpine reservoirs we do not expect ebullition in these reservoirs.

### 5.3 Methane sources

Generally, the carbon cycle in oxic lakes and reservoirs assumes methane production in the sediments and methane oxidation, while methane is diffusing from the sediments into the water column (Kuivila et al., 1988). This oxidation can be traced with the increasing  $\delta^{13}\text{C}$  of methane (Barker and Fritz, 1981; Whiticar, 1999). Concentration and isotopic composition profiles in the lowland reservoirs agree well with this trend. Alpine reservoirs, on the other hand, did not show this behaviour and there seems to be no methane flux from these sediments into the water column.

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We, therefore, assume external sources to be responsible for the methane content in the alpine reservoirs. Most alpine reservoirs collect additional water from neighbouring valleys, which is transported there via tubes and in some cases pumped up from lower altitude. Methane concentration measurements in the retention basin Carassina of Lake Luzzone for example were 120 nM below the surface and 270 nM at the bottom of the basin in August. Enhanced methane concentrations (Fig. 6) and enhanced conductivity (data not shown) are characteristic for the intermediate layer in Lake Luzzone originating in a mixture of different inflows (among them water from the Basin Carrasina).

The only high alpine reservoir we have inflow data for is Lake Oberaar. Here, methane concentrations of the water flowing from the glacier into the reservoir reach 30 nM in August with values in June and July are lower (around 15 nM), possibly due to dilution by snow melt water. Concentrations and isotopic composition of the inflowing water (Table 2) are very similar to the values in the lake (Electronic supplement Fig. A7). It has been shown, that methane can be produced in glaciers and low temperature environments (e.g. Price, 2007; Wadham et al., 2008) The  $\delta^{13}\text{C}$  value of  $-35\text{‰}$  suggests that, methane produced in the glacier has already been partially oxidized by the time it left the glacier snout.

Not only in alpine reservoirs are inflows important for the methane content in and the emissions from the reservoirs. Estimating methane inflow from the river Jogne and Sarine into Lake Gruyère and comparing it to methane emissions shows that 4.8 and 1.5 times more methane enters the reservoir in May and June, respectively than leaves the reservoir via surface diffusion. Only in August was diffusion five times higher than methane inflow. It seems that during late spring and early summer methane diffusing from the lake surface originates mostly from reservoir inflows, while in autumn, when the water mixes and in spring, before the reservoir is stratified methane from the sediment is more important.

On the other hand methane diffusing from the sediments is only a minor source for the water column and a minor source of methane to the atmosphere as most of the

methane is already oxidized before it reaches the water column. This is reflected in the isotopic composition with values from  $-55\text{‰}$  to  $-20\text{‰}$ . Methane flux from the sediments in the middle basin of Lake Lungern, calculated from the methane concentration profile (Electronic supplement Fig. A2b), was  $410 \mu\text{mol m}^{-2} \text{d}^{-1}$ , similar to the flux found in aerobic Lake Constance (Frenzel et al., 1990). Methane oxidation consumed 93% of the methane diffusing upwards in the sediment at Lake Constance and a similar rate is to be expected in the reservoirs we sampled, as at Lake Lungern methane concentrations in the water above the sediment were always lower than 80 nM.

#### 5.4 Origin of methane in the inflows

To determine where the methane in the inflow originates from, we measured concentrations upstream and downstream of a wastewater treatment of the Sarine, an inflow of Lake Gruyère, and the river Minster, an inflow of Lake Sihl. While concentrations in the Sarine did not change or decreased slightly after the inlet of the cleaned wastewater, concentrations in the Minster increased 2–5 times together with a decrease of  $\delta^{13}\text{C}$  of up to  $20\text{‰}$ , indicating biologically produced methane in the outflow of the wastewater treatment plant.

One of the major inflows of Lake Sihl, the river Minster, crosses a plain of agriculturally used land. At the sampling date in August we measured methane concentrations at the beginning of the plain and after the plain at the inflow to the lake. Methane concentration rose more than 10-fold to 367 nM after the passage of the plain and the carbon isotope signal fell from  $-39\text{‰}$  to  $-56\text{‰}$ . Thus the main source of methane in the river Minster lies within this agricultural influenced plain.

## 6 Conclusions

The most important greenhouse gas emitted from perialpine and alpine reservoirs in Switzerland is  $\text{CO}_2$ . On average, reservoir emissions are  $860 \pm 700 \text{mg m}^{-2} \text{d}^{-1}$  and

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therefore slightly smaller than emissions from boreal and temperate reservoirs in other parts of the world. Emissions in the spring time were higher than in summer and autumn ( $p < 0.0001$ ), while no elevation effect could be found.

Alpine reservoirs were in equilibrium with atmospheric  $\text{N}_2\text{O}$  concentrations, whereas two lowland reservoirs emitted small amounts of  $\text{N}_2\text{O}$  at  $72 \pm 22 \mu\text{g m}^{-2} \text{d}^{-1}$ .

Methane emissions were an order of magnitude smaller than values published for reservoirs in temperate and boreal climates. Average emissions were  $0.2 \pm 0.15 \text{ mg m}^{-2} \text{d}^{-1}$  for all reservoirs, except Lake Wohlen, which emitted  $1.5 \pm 0.4 \text{ mg m}^{-2} \text{d}^{-1}$  via surface diffusion and  $300 \pm 230 \text{ mg m}^{-2} \text{d}^{-1}$  via bubble flux. There was no significant difference between different elevations, as higher wind speeds at higher elevations compensated the lower methane concentrations. The amount of external methane entering via inflows is sufficient to explain the emission rates found in most reservoirs. Methane input from sediments is only of minor importance for open water sites. Only during times of mixing can accumulated methane in the water layers above the sediment contribute significantly to methane emission. The isotopic composition of sedimentary methane suggests a biogenic source and partial oxidation.

Methane loss at the turbine accounted for around 50% of total emissions (diffusive surface flux + gas loss at the turbine) in subalpine and alpine reservoirs. This type of emissions was less important in lowland reservoirs such as Lake Sihl, where it contributed only 14% of the total  $\text{CH}_4$  flux to the atmosphere.

In two lowland reservoirs with a small height difference (smaller than 10m) between reservoir and receiving river, no additional emissions at the turbine were found.

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**Table 1.** Properties of the sampled reservoirs.

Lake	Location (Latitude/Longitude)	Elevation (m)	Year of construction	Volume (10 <sup>6</sup> m <sup>3</sup> )	Surface (km <sup>2</sup> )	Greatest Depth (m)	Average Depth (m)	Retention time (d)	Meteo station	Additional measurements
1. Lake Wohlen	46° 58' N/7° 19' E	481	1920	25	3.65	20	7	2–3	Mühleberg	In- and Outflows, Ebullition
2. Lake Gruyère	46° 39' N/7° 06' E	677	1947	200	9.6	75	21	75	Payerne	In- and Outflows
3. Lake Lungern	46° 48' N/8° 10' E	689	1920	65	2.01	68	32	100–200	Giswil	Sediment core
4. Lake Sihl	47° 08' N/8° 48' E	889	1936	96.5	10.85	23	9	140 <sup>a</sup>	Oberaegeri	In- and Outflows
5. Lake Luzzzone	46° 34' N/8° 58' E	1591	1963	88	1.44	181	61	230	Hinterrhein	In- and Outflows
6. Lake Zeuzier	46° 21' N/7° 26' E	1777	1957	51	0.85	140	60	120	Montana	
7. Lake Grimsel	46° 34' N/8° 20' E	1908	1932	101	2.72	100	37	20–50 <sup>b</sup>	Grimsel-Hospiz	
8. Lake Santa Maria	46° 34' N/8° 48' E	1908	1968	67	1.17	86	57	100–200 <sup>c</sup>	Piotta	In- and Outflows
9. Lago Bianco	46° 24' N/10° 01' E	2234	1912	21	1.5	53	14	100–200 <sup>d</sup>	Robbia	
10. Lake Oberaar	46° 33' N/8° 16' E	2303	1953	61	1.46	90	42	30–60 <sup>b</sup>	Grimsel-Hospiz	In- and Outflows
11. Lake Dix	46° 04' N/7° 24' E	2368	1961	401	4.3	227	93	30–50	Evolène	

<sup>a</sup>about 10% of the water in the Lake are pumped from Lake Zurich

<sup>b</sup>water from Lake Grimsel is pumped into Lake Oberaar at night and released back to Lake Grimsel during the day for energy production; this way the volume of Lake Oberaar gets replaced about ten times every year

<sup>c</sup>is connected with two other reservoirs to one power station

<sup>d</sup>is a storage reservoir for Lake Palü, no direct energy production

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**Table 2.** Diffusive fluxes of CH<sub>4</sub>, CO<sub>2</sub> and N<sub>2</sub>O in the studied reservoirs and average wind speed on the sampling dates.

Reservoir	Date	CH <sub>4</sub>				CO <sub>2</sub>			N <sub>2</sub> O		Wind speed average (m s <sup>-1</sup> )
		surface concentration (nmol l <sup>-1</sup> )	diffusive flux (μg m <sup>-2</sup> d <sup>-1</sup> )	inflow (nmol l <sup>-1</sup> )	outflow <sup>a</sup> (nmol l <sup>-1</sup> )	loss at turbine <sup>b</sup> (%)	surface concentration (μmol l <sup>-1</sup> )	diffusive flux (mg m <sup>-2</sup> d <sup>-1</sup> )	surface concentration (nmol l <sup>-1</sup> )	diffusive flux (μg m <sup>-2</sup> d <sup>-1</sup> )	
Lake Wohlen	24.05.2005	516±527	1239±1341	97	870	not observed	279±98	1558±613			1.0
	14.06.2005	572±333	1672±666	61±21	1412	not observed	140±10	1053±72			1.2
	10.08.2005	498±256	1183±605	102	653	not observed	52±6	276±57			1.0
	28.09.2006	1039±302	2025±569						23±4	72±22	0.9
Lake Gruyère	24.05.2005	48	142	42; 189	102	not observed	185.0	1518			1.4
	15.06.2005	42±16	135±55	15; 84	23	not observed	88±17	707±170			1.4
	10.08.2005	49±16	179±63	12; 27	15	not observed	80±18	712±182			1.4
Lake Lungern	25.10.2005	104	163								0.7 <sup>c</sup>
	01.08.2006	101±23	340±80				50±14	353±136			1.3
	01.09.2006	48±11	86±20				40±0	132±2	18±3	47±16	0.8
	14.09.2006	62±13	147±32						19±0	69±1	1.0
	21.09.2006	22±9	35±15						17±1	38±4	0.8
Lake Sihl	19.10.2006	22±7	30±10						20±5	46±24	0.7
	14.05.2005	67±1	286±11	416; 172	53	16	203±23	2391±362			1.7
	17.06.2005	51±10	133±29	128; 102	23	73	74±14	460±960			1.1
Lake Luzzzone	09.08.2005	72±9	205±25	367; 102	32	46	68±30	451±249			1.2
	21.07.2005	12	122	26.5;36.8;23.8	13	46	53	1313			2.7
Lake Zeuzier	22.08.2005	27	140	26±4; 197±105	33	45	72	2516			2.4
	10.09.2003	15	65						6	-27	1.1
	07.06.2005	55	650	29±7							4.0
Lake Santa Maria	06.07.2005	16	189	22±4							3.5
	23.08.2005	15	108	11±1							3.4
	09.10.2003	34	212	46	20	57			11	-11	2.8
Lake Grimsel	05.07.2005	18	355								5.5
	28.07.2005	13	532								7.3
Lago Bianco	28.09.2004	3	-1								0.9
Lake Oberaar	05.07.2005	13	254	19±2	18						5.5
	28.07.2005	9	313	18	15						7.3
	24.08.2005	25	262	31	31						3.9
Lake Dix	10.09.2003	31	48						17	65	2.6

<sup>a</sup>measured after the water passed the turbine<sup>b</sup>calculated from the difference of the methane concentrations closest to the depth of the outlet and the outflow concentration<sup>c</sup>wind speed data from 2006

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**Table 3.** Greenhouse gas emissions from reservoirs in different climates.

CO <sub>2</sub> (mg m <sup>-2</sup> d <sup>-1</sup> )	CH <sub>4</sub> (mg m <sup>-2</sup> d <sup>-1</sup> )	N <sub>2</sub> O (mg m <sup>-2</sup> d <sup>-1</sup> )	Remarks	Reference
boreal zone				
1508±1771 ≈ 1200	8.8±12 5 to 10	-1.38 to 3.10		Tremblay et al. (2005) Duchemin et al. (1995)
504–3192	2.5 to 133	-0.089 to 0.270	two Finnish lakes	Huttunen et al. (2002)
temperate zone				
1137±156	3.2 to 9.5 21±6		2 out of 6 reservoirs sampled	Soumis et al. (2004) Smith and Lewis (1996)
1400	26±19 20 (10–80)		natural lakes	St.Louis et al. (2000)
tropical zone				
5800 1600±1300 3500	80±60 50 300 (20–1500)		open water flooded forest	Abril et al. (2005) St.Louis et al. (2000)
	13±22 71±107		deep reservoir shallow reservoir	Lima (2005)
2400 to 18800	15 to 3240 117		early filling stage	Galy-Lacaux et al. (1997) Fearnside (2002)

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**Fig. 1.** Locations of the sampled reservoir (for numbers see Table 1).

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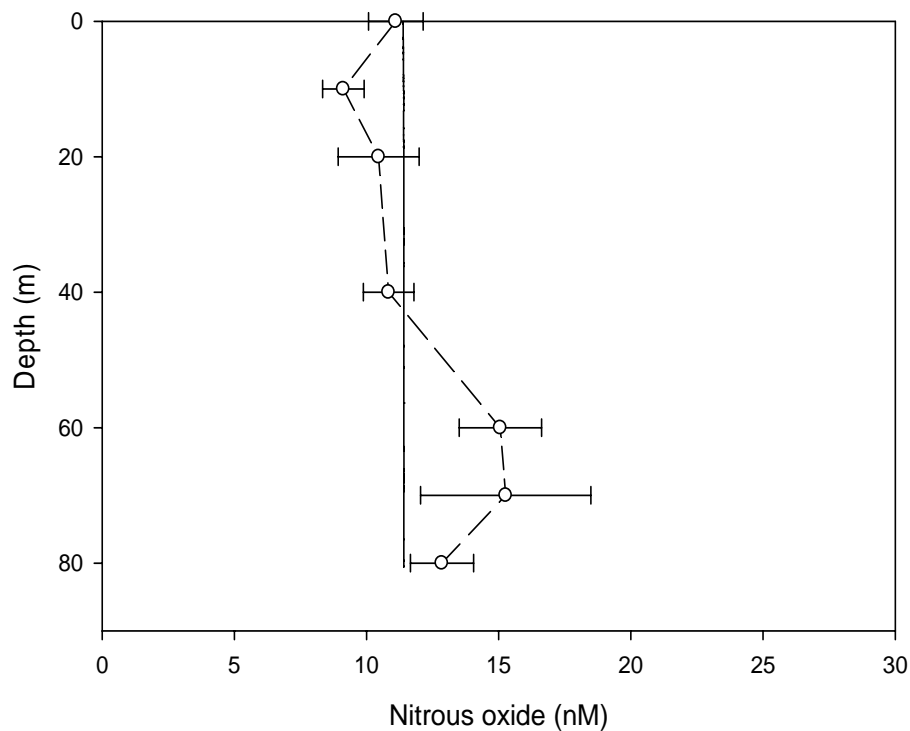
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**Fig. 2a.** N<sub>2</sub>O concentrations (circles) and atmospheric equilibrium concentration (solid line) of Lake Grimsel on 9 October 2003.

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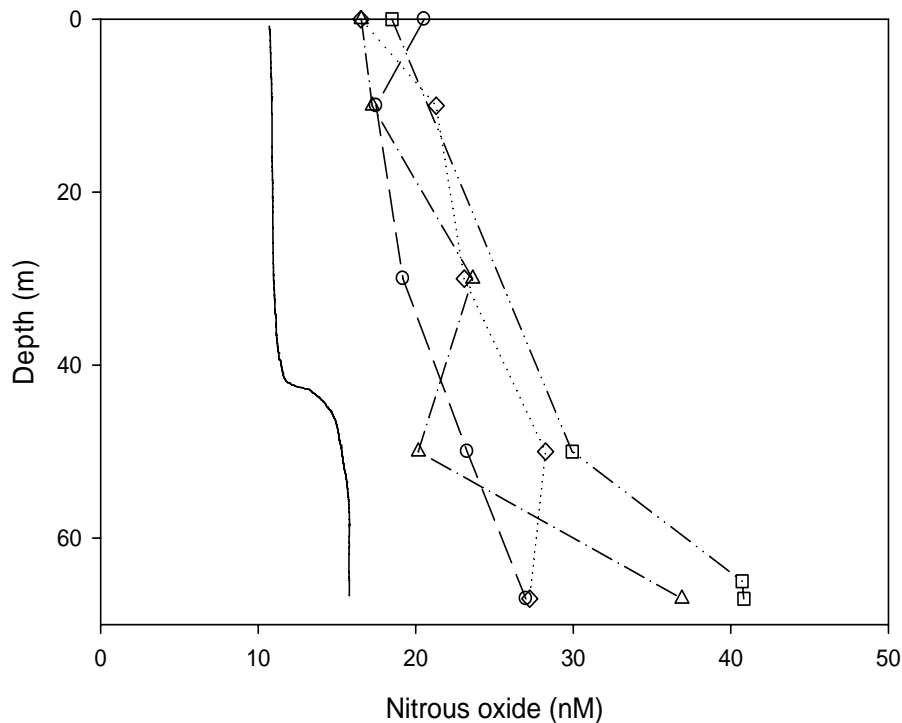
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**Fig. 2b.** N<sub>2</sub>O concentrations in Lake Lungern on 19 October 2005 (diamonds), 1 September (circles), 14 September (squares), and 21 September 2006 (triangles) and atmospheric equilibrium concentration (solid line) of Lake Lungern.

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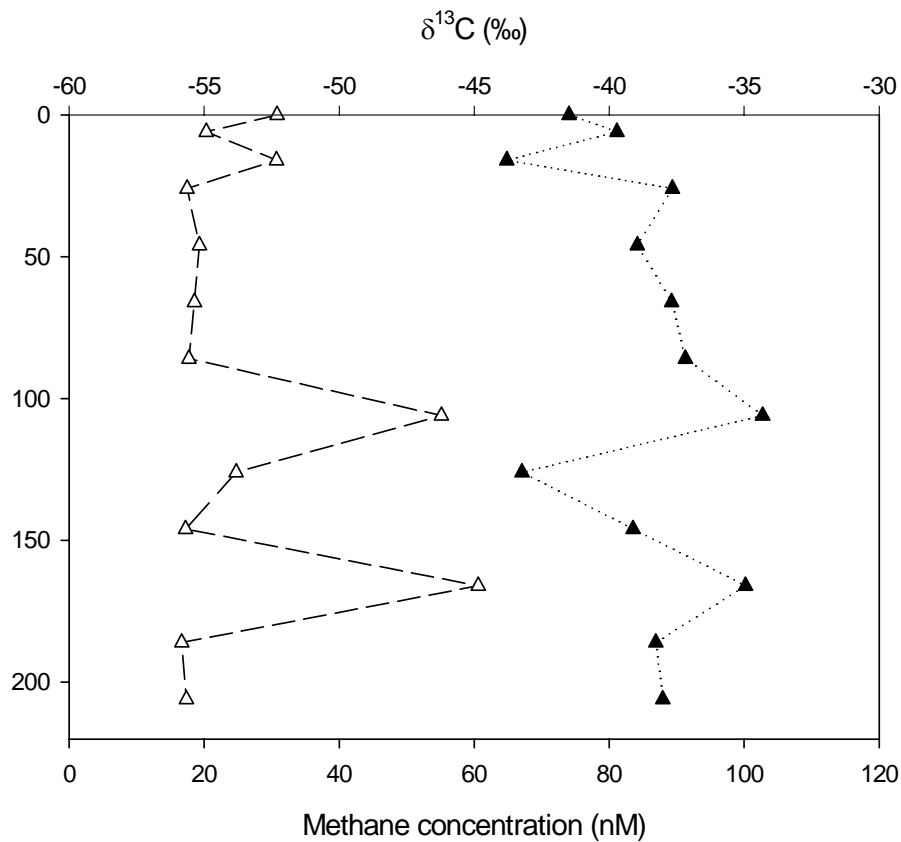
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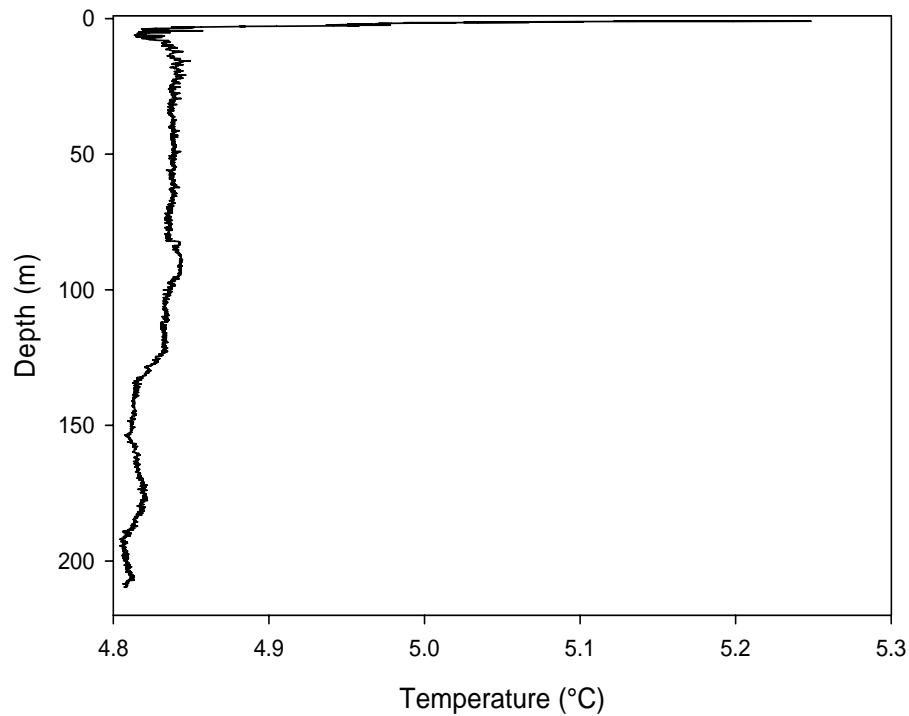
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**Fig. 3a.** Methane concentration (triangles) and isotopic composition (black squares) in Lake Dix.

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**Fig. 3b.** Temperature profile of Lake Dix.

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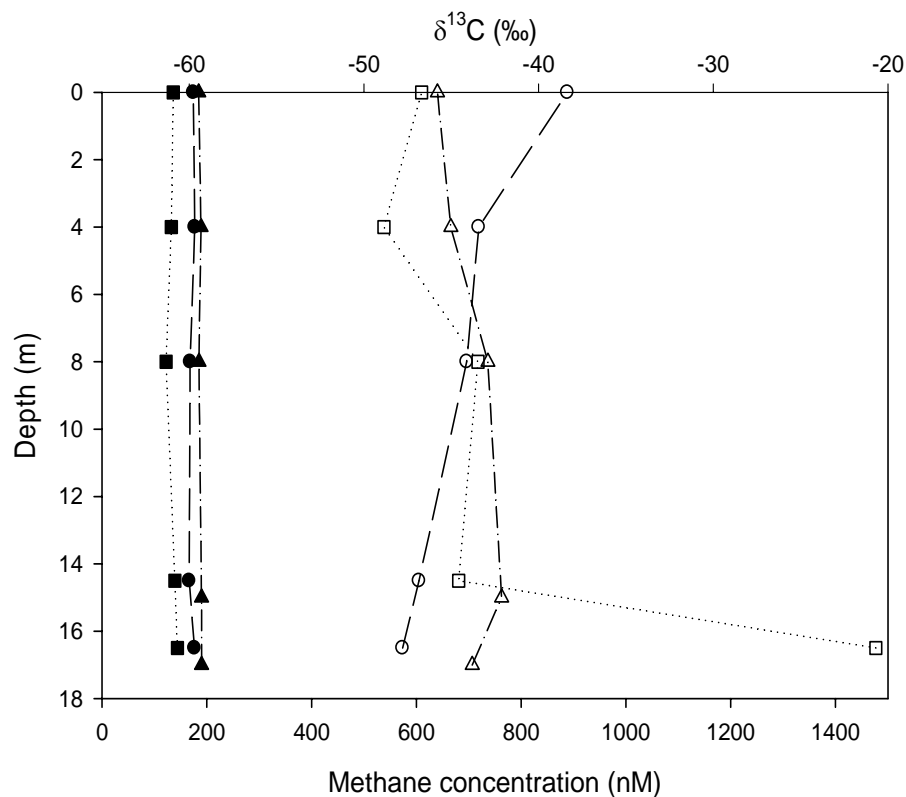
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**Fig. 4.** Methane concentrations (open symbols) and isotopic composition (full symbols) in Lake Wohlen on 24 May (circles), 14 June (squares) and 10 August 2005 (triangles).

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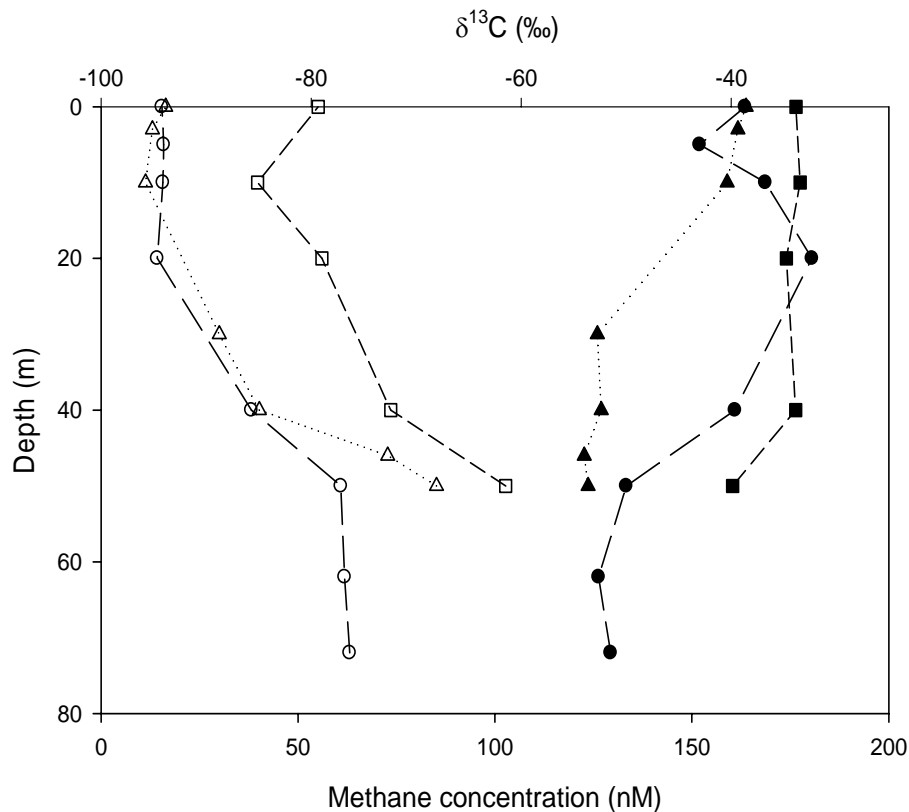


Fig. 5a. Methane concentrations (open symbols) and isotopic composition (full symbols) in Lake Santa Maria on 7 June (squares), 6 July (triangles) and 23 August 2005 (circles).

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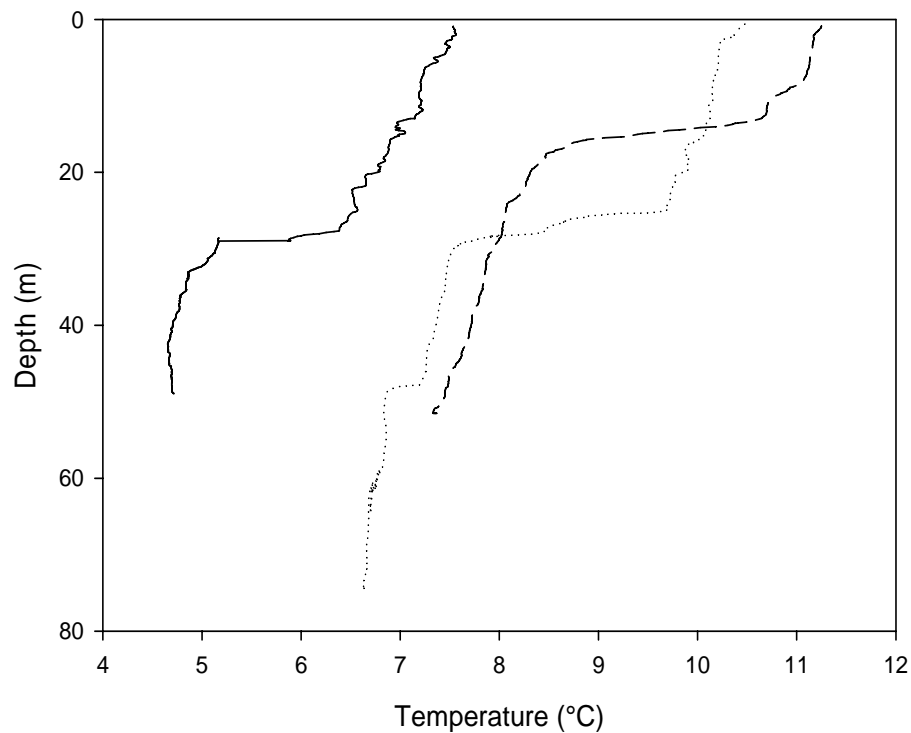
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**Fig. 5b.** Temperature profiles of Lake Santa Maria on 7 June (solid line), 6 July (dashed line) and 23 August 2005 (dotted line).

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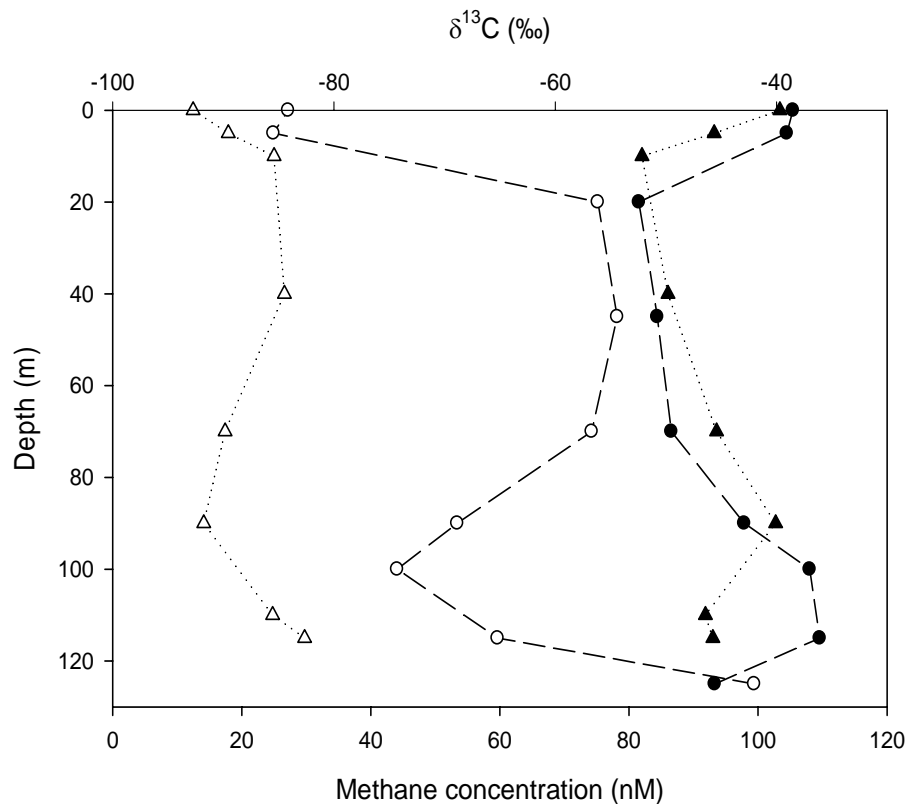
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**Fig. 6.** Methane concentrations (open symbols) and isotopic composition (full symbols) in Lake Luzzzone on 7 June (rectangles), 6 July (triangles) and 23 August 2005 (circles).

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