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Nitrous oxide emissions to the atmosphere from an artificially oxygenated lake

Abstract—Nitrous oxide (N_2O) production at the sediment surface of eutrophic Lake Baldegg was quantified with three independent methods: pore-water samplers, benthic chambers, and mass balances of the aerated-oxygenated hypolimnion. N_2O production at the sediment surface was the most important source in this lake and led to an accumulation in the hypolimnion during summer stratification. Highest rates of N_2O emission to the atmosphere ($24 \mu\text{mol m}^{-2} \text{d}^{-1}$) were observed after the onset of winter overturn, when hypolimnetic water enriched in N_2O came in contact with the atmosphere. During summer stratification N_2O emissions to the atmosphere decreased to $\sim 4 \mu\text{mol m}^{-2} \text{d}^{-1}$. The winter fluxes are close to the highest reported N_2O emissions from marine systems.

Nitrous oxide (N_2O) contributes to global warming and catalyzes the decomposition of stratospheric ozone. Its time-integrated warming effect per unit mass (global warming potential) is 200–300 times larger than that for CO_2 (IPCC 1990). Anthropogenic N_2O emissions contributed an estimated 6% to the total change in radiative forcing in the period 1980–1990. The major sink for N_2O is photolysis in the stratosphere—a process that provides active chemical species for the decomposition of ozone.

At present, N_2O concentration in the atmosphere (310 ppb) is increasing by $0.25\% \text{yr}^{-1}$ (IPCC 1990). Because the driving processes for this global increase are poorly understood, measurements of N_2O fluxes from soils and oceans have been intensified in recent years.

N_2O is produced by several microbiological processes. In natural waters nitrification and denitrification are considered to be the major processes producing N_2O . Law and Owens (1990) and Codispoti et al. (1993) reported significant N_2O emissions from eutrophic marine systems, indicating that global N_2O fluxes from aquatic systems to the atmosphere could be higher than the value of $1.4\text{--}2.8 \times 10^{12} \text{g N yr}^{-1}$ proposed by Butler et al. (1989). Little is known about the significance of N_2O emissions from freshwater systems. Lakes with N_2O supersaturated surface waters (Yoh et al. 1988; Downes 1991), as well as eutrophic lakes that act as sinks for atmospheric N_2O (Knowles et al. 1981; Lemon and Lemon 1981), have been described in the literature.

Law et al. (1992) suggested that N_2O emissions from eutrophic estuaries should be considered in global N_2O budgets and that N_2O supersaturation in the water column resulted mainly from sedimentary sources. In our study, we chose a highly eutrophic lake with an artificially

Table 1. Benthic fluxes ($\mu\text{mol m}^{-2} \text{d}^{-1}$) of N_2O , NO_3^- , and NH_4^+ calculated either from pore-water profiles (peeper) or from flux chamber experiments.

Method	Sam- pling site (m)	N_2O^*	$\text{N}_2\text{O}^\dagger$	NO_3^-	NH_4^+	Comment
Peeper						
Dec 93	66	1.13	-1.73	-230	2,430	
Feb 94	66	0.18	-0.19	-210	4,450	
May 94	66	1.46	-0.66	-350	2,580	
Jul 94	30	4.02	-3.78	-380	2,350	
Jul 94	66	1.68	-1.26	-250	2,360	
Sep 94	30	1.89	-0.86	-140	1,890	
Sep 94	66			-350	5,250	
Flux chamber‡						
Feb 94	66	13.5		-2,920	2,420	
Feb 94	66	11.0		-3,630	3,230	
May 94	66	11.8		-5,650	4,410	
May 94	66	13.8		-5,460	4,760	
Aug 94	66	19.3		-4,540	2,680	
Aug 94	66	17.2		-2,880		+ ATU
Oct 94	66	80.3		-3,900	4,350	+ NaNO_3
Oct 94	66	113.0		-2,830		+ ATU, + NaNO_3

* Flux to hypolimnion.

† Flux to deeper sediment layers.

‡ Replicate data of both flux chambers are shown.

oxygenated hypolimnion. Because N_2O is nearly inert in the presence of oxygen (Broecker and Peng 1982), this lake allows us to determine benthic N_2O fluxes from the N_2O accumulation in the hypolimnion. We compared flux estimates from hypolimnetic mass balances with N_2O fluxes obtained from pore-water profiles and from benthic chamber studies.

Lake Baldegg, a eutrophic lake in central Switzerland, has a surface area of 5.2 km², a volume of 0.173 km³, and a maximum depth of 66 m. The mean water residence time is 5.5 yr. The actual dissolved phosphorus concentration at overturn is 80 $\mu\text{g P liter}^{-1}$ but was as high as 460 $\mu\text{g P liter}^{-1}$ in 1975. The annual productivity was determined in 1974 and 1982–1983 as 420 and 350 g C m⁻² yr⁻¹, respectively (Stadelmann 1984). During summer stratification the hypolimnion is enriched with pure oxygen; during winter, water circulation is enhanced by pumping air to deeper layers of the lake. Due to these measures the oxygen concentration always exceeded 2 mg liter⁻¹ in the entire water column during our study.

The water column of Lake Baldegg was sampled at 4-week intervals between February 1994 and March 1995. N_2O concentrations were analyzed by headspace gas chromatography and an electron capture detector. The sediment pore water was sampled six times at two different depths with dialysis pore-water samplers ("peepers"). Design and deployment of peepers has been described by Hesslein (1976). We modified the peeper in such a way that it could be covered gastight in situ at the lake bottom. Two stainless steel lids were released by a burning wire after a 3-week exposure. Magnets kept the lids closed. To sample the peeper, we injected helium through butyl septa and transferred the displaced water into gastight syringes

within 1 h after retrieval. This procedure allows sampling of the peepers without substantial loss of N_2O and without air contamination.

Fluxes of N_2O across the sediment–water interface were measured four times with the aid of an in situ flux chamber (Devol 1987). The instrument contains two stainless steel flux chambers that cover sediment areas of 400 cm² and typically enclose 3–6 liters of water. After lowering the instrument to the sediment surface, an electronic system triggers up to 15 mechanical functions. From each chamber, 10 samples of ~50 ml were drawn at preprogrammed time intervals. During the experiment the enclosed water in the chambers was mixed by a rotating stirrer. We used stirring velocities of ~1 rpm, resulting in a diffusive boundary-layer thickness of ~1 mm (Wehrli et al. in prep.). At the end of the experiment the grab shovels at the bottom of the chambers were closed and two box sediment cores of about 30 cm were retrieved.

The flux chamber experiments lasted 40–60 h, during which time 10 samples were taken in parallel from each of the two flux chambers at time intervals of 2–8 h. At the end of all four experiments, oxygen was depleted in the chambers. Because the oxygen concentration is a key variable of the production and consumption of N_2O by nitrification and denitrification (Downes 1988), the N_2O flux across the sediment surface is likely to change during the experiment. To minimize this change, we considered only samples that were drawn during the first 8 h of the experiment for flux calculations. Dates and sampling sites of all flux chamber experiments and peeper deployments are summarized in Table 1. In two flux chamber experiments we injected allylthiourea (20 mg liter⁻¹ in the flux chamber), a known nitrification inhibitor (Hall 1984),

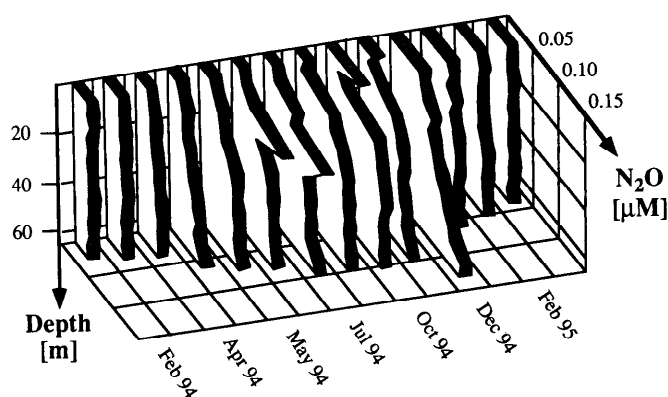


Fig. 1. N_2O in the water column of Lake Baldegg, February 1994–March 1995.

into one of the flux chambers. In another experiment we injected $NaNO_3$ into both flux chambers to increase the ambient NO_3^- concentration from 120 to 240 μM .

Profiles of N_2O concentration in the water column are presented in Fig. 1. Hypolimnetic concentrations increased as soon as stratification developed in spring. Highest concentrations ($\sim 0.13 \mu M$) were observed in December. Because the saturation value for water in equilibrium with atmospheric N_2O is $14.3 \times 10^{-3} \mu M$ at $6.2^\circ C$ (calculated from solubility constants, Weiss and Price 1980, and an assumed atmospheric N_2O concentration of 310 ppb), this value exceeded saturation more than 9-fold. Many profiles show increasing concentrations toward the sediment surface, indicating that the sediment-water interface represented a source for N_2O . In June and July, we observed peak concentrations in the region of the thermocline, which points toward a second N_2O source in the metalimnion. These peak concentrations coincided with oxygen minima. N_2O concentrations at the lake surface were always supersaturated. N_2O concentrations in the surface water were higher during winter overturn than during summer stagnation. The highest concentrations were measured in October. After the onset of winter overturn in December, the N_2O content of the lake decreased.

Figure 2 shows typical pore-water profiles. The vertical resolution for N_2O was 1 cm; for NO_3^- and NH_4^+ , it was 0.5 cm. All pore-water profiles (four at 66-m, two at 30-m water depth) exhibited a N_2O peak at the sediment surface. Concentrations ($1.4 \mu M$) were highest in summer, although no obvious seasonality was observed. Profiles at 30- and 66-m depth did not differ significantly. The NO_3^- profile suggested NO_3^- consumption in the region of the sediment-water interface. NO_3^- is consumed at the sediment surface or in the top sediment layer by denitrification and NO_3^- ammonification. NH_4^+ diffuses from deeper sediments to the sediment surface and eventually to the overlying water where it likely would be oxidized to NO_3^- by nitrifiers. Because nitrification, as well as denitrification, produces N_2O as an intermediate product or by-product (Downes 1988), the dominant process producing N_2O cannot be identified from pore-water profiles.

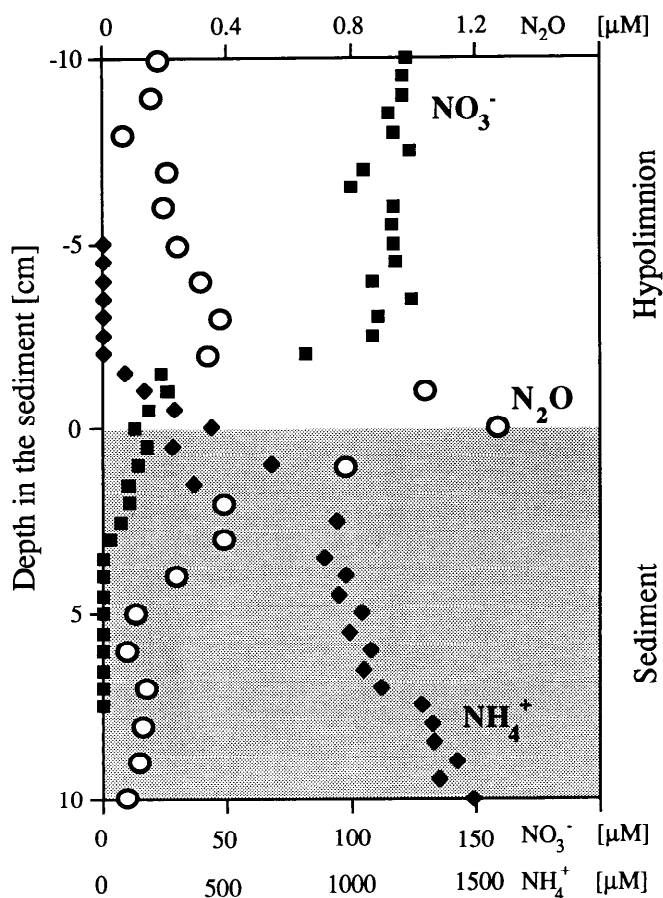


Fig. 2. N_2O , NO_3^- , and NH_4^+ at the sediment surface of Lake Baldegg, 30-m depth, July 1994.

Figure 3 shows a typical result from a flux chamber experiment. N_2O concentrations in the flux chamber increased in all four experiments during the first 8 h. The increase in concentration, however, varied significantly between experiments. Allylthiourea, known to inhibit nitrification, did not influence N_2O production in the flux chamber. In the experiment with artificially increased NO_3^- concentrations (from 120 to 240 μM), the N_2O flux ($100 \mu mol m^{-2} d^{-1}$) was five times larger than the average value of the other three flux chamber experiments ($19 \mu mol m^{-2} d^{-1}$). Because allylthiourea showed no effect, but NO_3^- addition increased N_2O production, denitrification rather than nitrification is likely the dominant source of N_2O .

Applying Fick's first law, $F = \phi \times D \times dC/dz$ (where ϕ is porosity, D is the diffusion coefficient, and dC/dz the concentration gradient), to the pore-water profiles we calculated the flux (F) of N_2O across the sediment surface. For ϕ we used a value of 0.92 and for D the molecular diffusion coefficient $1.08 \times 10^{-5} cm^2 s^{-1}$ ($5^\circ C$, Lerman 1979). Fluxes based on the flux chamber experiments were calculated directly from the change in concentration in the overlying water during the first 8 h, taking into account the enclosed water volume and the surface area of the sediment in contact with it.

Fluxes of N_2O , NO_3^- , and NH_4^+ estimated either from

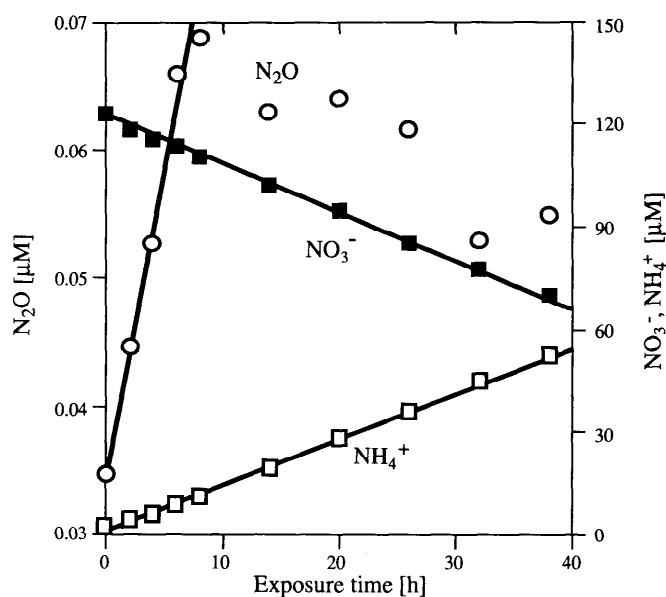


Fig. 3. N_2O , NO_3^- , and NH_4^+ in the flux chamber, Lake Baldegg, 66-m depth, March 1994.

pore-water profiles or from flux chamber experiments are summarized in Table 1. Fluxes for NH_4^+ calculated from the two methods agreed well, whereas those for N_2O and NO_3^- differed more than 10-fold. In the case of N_2O and NO_3^- , fluxes calculated from pore-water profiles may be underestimated for two reasons. First, due to insufficient vertical resolution, fluxes based on pore-water profiles often fail to reflect redox processes occurring in the top few millimeters of the sediment. Second, we considered the molecular diffusion coefficient to estimate fluxes, although the concentration gradients of N_2O and NO_3^- extended from the sediment into the overlying water where turbulent mixing should be taken into account. In the case of NH_4^+ however, the gradients are mainly in the top layer of the sediments where only molecular diffusion must be considered. Fluxes based on flux chamber experiments may overestimate in situ fluxes due to the mixing of the overlying water by a mechanical stirrer. The real N_2O fluxes across the sediment-water interface are constrained by the results of the two methods. The peepers yield a lower limit (avg value, $2 \mu\text{mol m}^{-2} \text{d}^{-1}$) and the flux chamber data represent an upper boundary (avg value, $19 \mu\text{mol m}^{-2} \text{d}^{-1}$).

Because N_2O is rather inert in oxic lake and seawater, most of the N_2O diffusing from the sediment surface into the lake accumulates in the hypolimnetic water during summer stratification. From May to October, the average increase of the hypolimnetic N_2O content yielded 60 mol d^{-1} . From the concentration gradients in the thermocline ($5 \times 10^{-3} \mu\text{M m}^{-1}$) and the depth-dependent vertical diffusion coefficients of $\sim 1\text{--}3 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$ (calculated with the heat budget method, Powell and Jassby 1974) we calculated a flux of $1.7\text{--}5.3 \text{ mol N}_2\text{O d}^{-1}$ from the hypolimnion into the epilimnion. Therefore, total production in the hypolimnion ranged from 60 to 65 mol d^{-1} , or 11 to $13 \mu\text{mol m}^{-2} \text{d}^{-1}$. This value lies well within the

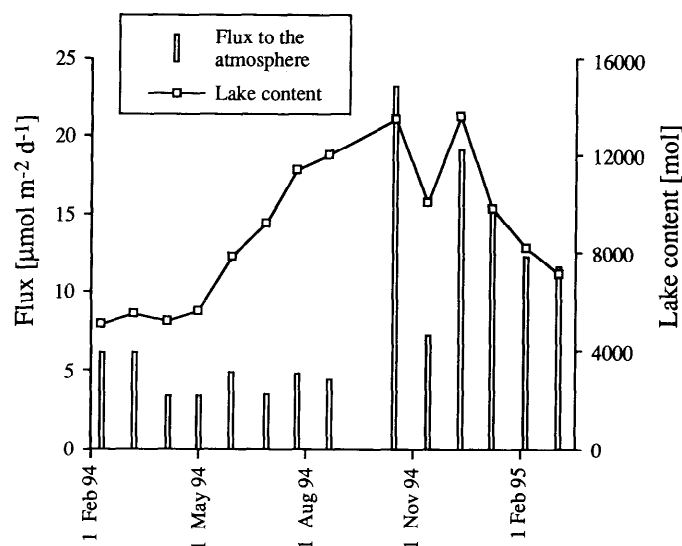


Fig. 4. N_2O content and flux to the atmosphere of Lake Baldegg, February 1994–March 1995.

range of our flux measurements with the peeper and flux chamber method and indicates that the sediment surface is the dominant source of N_2O in the hypolimnion. A sharp negative N_2O concentration gradient was always observed below the sediment-water interface, suggesting a sink for N_2O ($1.4 \mu\text{mol m}^{-2} \text{d}^{-1}$) in these anoxic deeper sediment layers. N_2O probably is reduced to N_2 by denitrifiers.

The surface waters of the lake were always supersaturated with respect to atmospheric N_2O concentrations during our 1-yr study. The N_2O fluxes from the lake into the atmosphere were calculated with

$$F = v_{\text{tot}} \times (c_w - c_s) \text{ (mol m}^{-2} \text{d}^{-1}\text{)},$$

based on the stagnant boundary theory. v_{tot} represents the transfer velocity, c_w the surface concentration, and c_s the concentration in equilibrium with the atmosphere, calculated with solubility constants from Weiss and Price (1980) and an assumed atmospheric N_2O concentration of 310 ppb.

For Lake Baldegg an average v_{tot} (0.4 m d^{-1}) for O_2 was determined from oxygen budget calculations during winter overturn. Because the gas exchange of both N_2O and O_2 is dominated by transfer through the water boundary layer, we calculated the transfer velocity for N_2O with the formula (Schwarzenbach et al. 1993)

$$v_{\text{tot}}^{\text{N}_2\text{O}} = v_{\text{tot}}^{\text{O}_2} \times (D_w^{\text{N}_2\text{O}}/D_w^{\text{O}_2})^{0.5} \approx 0.4 \text{ m d}^{-1}.$$

D_w is the molecular diffusion coefficient in water ($\text{N}_2\text{O} = 1.08 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$, $\text{O}_2 = 1.26 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$, 5°C , Lerman 1979). The resulting N_2O emission rates are presented in Fig. 4. Emissions were larger from October 1994 to March 1995 than from February 1994 to September 1994. These larger emissions are likely due to increased mixing that transported hypolimnetic water enriched with N_2O to the epilimnion during overturn and thus increased the N_2O concentration at the lake surface. As a conse-

Table 2. N₂O emissions to the atmosphere from aquatic systems.

Region	N ₂ O emission ($\mu\text{mol m}^{-2} \text{d}^{-1}$)	Reference
Marine systems		
Average marine emission	0.6	Butler et al. 1989
NW Indian Ocean (upwelling region)	9–16	Law and Owens 1990
E Pacific (Peruvian upwelling region)	39	Codispoti et al. 1993
Tamar Estuary, U.K.	7–12	Law et al. 1992
Lakes		
Laurentian Great Lakes	± 77	Lemon and Lemon 1981
Lake Okaro, N.Z.	0.6	Downes 1991
Lake Baldegg, Switzerland	8–16	This study

quence of increased emissions, the N₂O content of the lake decreased from 13.6×10^3 mol in December 1994 to 7.1×10^3 mol in March 1995. However, the lake as a whole remained supersaturated because at 5.2°C (lake temperature in March 1995) the equilibrium concentration would be $\sim 15 \times 10^{-3}$ μM , which corresponds to a lake content of 2.6×10^3 mol N₂O. N₂O production at the sediment surface during winter and the slow gas exchange accounts for this nonequilibrium even at the end of spring overturn. The aeration of this lake does not significantly enhance the gas exchange (Wüest et al. 1992). The N₂O fluxes into the atmosphere are smaller during summer stratification with an average value of $4 \mu\text{mol m}^{-2} \text{d}^{-1}$.

The N₂O profile in October suggests either a source of N₂O close to the lake surface or a zone of N₂O consumption at a depth of ~ 8 m. Law et al. (1993) showed with incubation experiments that denitrification coupled with N₂O production in aerobic surface waters can enhance the sediment N₂O flux by 150–500% in eutrophic estuaries. The concentration peaks at a depth of 10–20 m in June and July suggest a second source of N₂O in the water column. Such N₂O maxima that coincide with oxygen minima are often observed in oceans and usually are attributed to N₂O production through nitrification in the water column (Butler et al. 1989). Knowles et al. (1981) also observed such N₂O maxima at the thermocline in Lake Erie and suggested nitrification to be the source of this N₂O.

Combining our various observations, we conclude that the sediment surface is the major source of N₂O ($11\text{--}13 \mu\text{mol m}^{-2} \text{d}^{-1}$) in Lake Baldegg. On a yearly average, the lake exports $8\text{--}16 \mu\text{mol N}_2\text{O m}^{-2} \text{d}^{-1}$ into the atmosphere and $1\text{--}2 \mu\text{mol N}_2\text{O m}^{-2} \text{d}^{-1}$ to the deeper sediments. These emissions to the atmosphere are of the same order of magnitude as the highest N₂O emissions reported that were found in estuaries and marine upwelling zones (Table 2). With respect to the nitrogen cycle of Lake Baldegg, the N₂O emissions are unimportant because they represent only 0.2% of the total nitrogen input into the lake and $\sim 0.3\%$ of the estimated N₂ fluxes from the lake to the atmosphere (Höhener and Gächter 1993).

Lake Baldegg is a highly eutrophic lake and is artificially oxygenated, so our results cannot be extrapolated to the global surface area of lakes. The impact of artificial oxygenation on fluxes of different nitrogen species across

the sediment–water interface have been studied in detail in Lake Sempach (Höhener and Gächter 1994). Lake Sempach, a eutrophic 87-m-deep lake in central Switzerland, is ~ 8 km southwest of Lake Baldegg. Its trophic state is similar to that of Baldegg and, since 1984, the hypolimnion of Sempach has been oxygenated and artificially mixed by the same technique as applied in Baldegg. Höhener and Gächter (1994) found that artificial oxygenation of the hypolimnion increased benthic denitrification rates. They speculated that this was due to the presence of *Beggiatoa* at the sediment surface. This bacterium, known to oxidize H₂S, was not present in the pelagic sediment of Baldegg and Sempach before artificial oxygenation began. By oxidizing H₂S, *Beggiatoa* detoxifies the environment for nitrifying bacteria at the sediment surface. As a consequence, the distance between the sites of NO₃⁻ generation through nitrification and NO₃⁻ reduction through denitrification decreased. The resulting steeper NO₃⁻ gradients allowed an enhanced overall denitrification rate.

We postulate higher denitrification rates in Lake Baldegg as a consequence of artificial mixing and oxygenation, so N₂O production by benthic denitrification might also be enhanced in Baldegg compared to other eutrophic lakes. In eutrophic marine or freshwater systems, anoxic deep waters mostly act as a sink for N₂O produced either by nitrification or denitrification at the transient zone between oxic and anoxic water layers (Broecker and Peng 1982; Codispoti et al. 1993; Downes 1991; Knowles et al. 1981). Therefore, artificial oxygenation of the hypolimnion of Lake Baldegg not only may have increased N₂O production at the sediment surface but also eliminated anoxic conditions in the hypolimnion that formerly favored N₂O reduction. As a consequence, artificial oxygenation might increase net production of N₂O and therefore increase N₂O emissions to the atmosphere.

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Pelagic responses to changes in dissolved organic carbon following division of a seepage lake

Abstract—Within 2 yr of dividing a multibasin lake into discrete lakes for experimentation, dissolved organic carbon (DOC) concentrations and water color (absorption coefficient g at 440 nm, g_{440}) increased in the east basin and decreased slightly in the west basin. These changes were not explainable by watershed vegetation or groundwater chemical composition. However, g_{440} increased from 0.7 to 4.2 m^{-1} for water moving through the sediment-water interface in the east basin. In the west basin, g_{440} of groundwater (0.6 m^{-1}), in-seeping water (0.7 m^{-1}), and lake water (0.7 m^{-1}) were all similar. Patterns of DOC distribution matched time trends in the surface waters. In the east basin, DOC concentration doubled and g_{440} increased 3-fold from 1990 to 1993. Trends in the west basin were more complex, but in general, there was a decrease in DOC and g_{440} over the same period. Changes in the light regime affected phytoplankton vertical distribution, but total areal chlorophyll

and epilimnetic chlorophyll concentrations were not altered. The depth of oxygenation was changed by altered mixing characteristics and phytoplankton distribution. Changes in light-attenuating DOC affected pelagic responses to nutrient inputs.

Dissolved organic carbon (DOC) plays multiple and pivotal roles in lake ecosystems (Wetzel 1992; Schindler et al. 1992). Allochthonous DOC is rich in colored, relatively refractory components (Thurman 1985) that have significant effects on physical and chemical processes and ecosystem structure (Jones 1992). This refractory DOC alters light penetration, pH, and trace metal and nutrient availability (Jackson and Hecky 1980; Jones 1992). Staining by DOC decreases phytoplankton production (Jack-