Aquat.sci.63 (2001) 417–431
1015-1621/01/040417-15 $ 1.50+0.20/0

Aquatic Sciences

Tracer experiment with sulfur hexafluoride to quantify the vertical transport in a meromictic pit lake

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Key words: Meromixis, mining lake, chemical stratification, vertical diffusion coefficients, SF_6 tracer experiment.

ABSTRACT

This paper describes a tracer experiment with sulfurhexafluoride (SF_6) in the monimolimnion of the meromictic mining lake Merseburg-Ost 1b. In October 1998, 1.1 mmol ($\simeq$160 mg) of the conservative gas SF_6 was released at the site of greatest depth 3.5 m above the sediment to observe its vertical spreading. An easy-to-use system to collect ~0.5 L water samples using evacuated flasks was developed. The headspace technique, gas chromatographic separation and ECD-detection were used to determine SF_6. The main objective is the evaluation of mean vertical diffusion coefficients for SF_6 and heat from periodically measured SF_6 and CTD profiles. During the study period of $1\,^1/_2$ years, the heat transport was estimated to be molecular in the stratified portion. A thermal flux from the sediments of 0.23 W/m^2 was found necessary to balance the heat. In the region of high stability ($N^2 \sim 10^{-2}\ s^{-2}$) diffusivities for SF_6 were $\sim 10^{-8}$ m^2/s, whereas in the lower part of the monimolimnion both tracers resulted in $K \sim 7 \cdot 10^{-6}$ m^2/s. We found K to be approximately proportional to $N^{-2.4\pm0.2}$.

Introduction

Vertical gradients of dissolved substances can cause a permanent density structure in the water column. Increasing density with depth in excess of the adiabatic compressibility reduces turbulence and therefore vertical mixing. Sources of these substances are remineralization in the sediments, chemical and biological processes in the water column or direct inflow of surrounding groundwater.

Experimental studies of vertical exchange processes mainly exist for the thermocline and for hypolimnia with comparatively weak stratification. Whereas temperature is the favored tracer used (Jassby and Powell, 1975; Heinz et al., 1990; Boehrer et al., 2000 (a)), measurements of parameters like the electrical conductivity, the concentration of dissolved oxygen and radionuclides as well as of several chemical and biological parameters (Melack and Jellison, 1998) have been performed. Quay et al. (1980) determined vertical diffusion rates by tritium tracer experiments. Structure und persistence of monimolimnia of meromictic lakes are described by Melack and Jellison (1998) and Stevens and Lawrence (1997, 1998). SF_6 was deployed as a tracer for horizontal and vertical mixing in Lake Constance (Maiss, 1992; Maiss et al., 1994) and in Lake Lucerne (Schlatter et al., 1997) and Lake Alpnach (Wüest et al., 1996). However, investigations of vertical transport in a strongly stratified water column with high stabilities ($N^2 > 10^{-2}$ s^{-2}) and small diffusivities ($K_z < 10^{-7}$ m^2/s) are sparse (von Rohden 1998; Walz, 1997; Maiss et al., 1997). In fact, mining lakes often tend to be meromictic because of saline groundwater inflows or chemical processes. For modeling and long-term prognosis of the development of such lakes (Sanderson et al., 1986; Boehrer et al., 1998) as well as for the assessment of fluxes of pollutants from the monimolimnion into the mixolimnion, experimental activities to quantify the vertical transport coefficients are essential.

Besides the thermal summer stratification suppressing direct input of turbulence below the thermocline, a constitutive peculiarity of lakes characterized by meromixis is the autumn and spring overturn reaching from the surface only to the chemocline. This has an eroding effect on the upper boundary layer of the monimolimnion. Thereby the chemocline is sharpened, because small gradients of salinity above the chemocline are removed. The interior of the monimolimnion is not directly affected by this mixing.

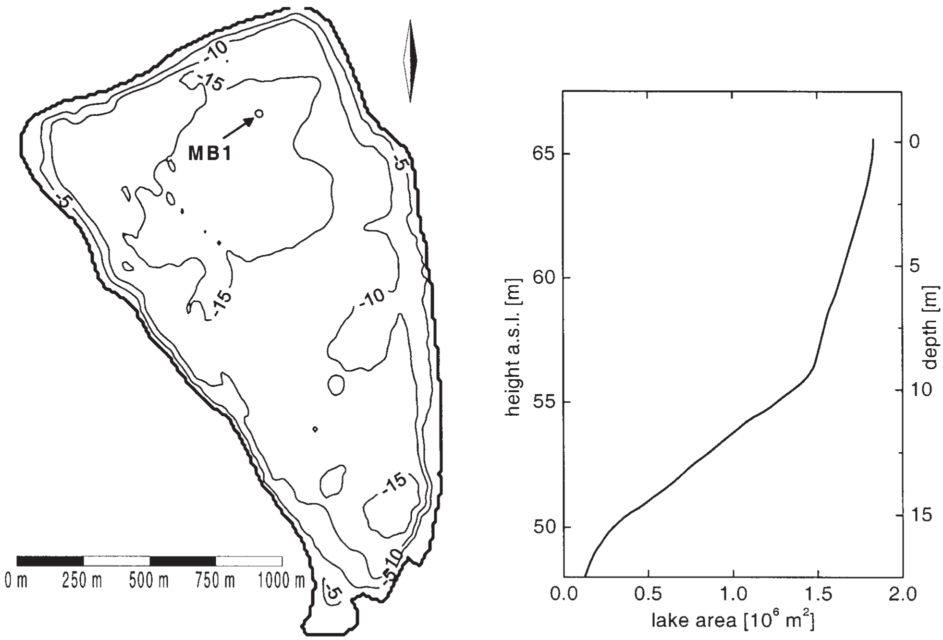

Figure 1. Depth contours and areas of the cross section of the mining lake complex Merseburg-Ost (near Leipzig, Central Germany), GPS- und echo-sound measured in April 1997 (UFZ Magdeburg). Since then, the water level rose continuously. The investigations took place at sampling site MB1

This study presents a tracer experiment to determine vertical diffusion coefficients in the monimolimnion of the pit lake Merseburg-Ost 1b. The tracer used is the mainly artificial gas sulfur hexafluoride (SF_6). The SF_6 diffusivity can be considered as operative to other solved solids when in a turbulent region. The SF_6 content of water samples can be measured with high sensitivity ($\sim 10^{-16}$ Mol/l) using gas chromatographic separation and an electron capture detector (ECD). SF_6 is a conservative tracer. Because of the geological surroundings at our sampling site and the amount added as tracer, natural sources as found by Busenberg and Plummer (2000) in some igneous, volcanic, and sedimentary rocks and in some hydrothermal fluids are of no pertinence for this study. The only relevant sources are the atmosphere and the low content in the infiltrating groundwater. The only known sink for SF_6 is photolysis in the stratosphere.

The experiment is based on the following conception: First, a spike solution is injected into the monimolimnion. After the horizontal homogenizing is completed, the vertical spreading of the tracer distribution is observed by measuring SF_6 concentration profiles every 1–2 months. The analysis relies on the assumption that horizontal exchange is significantly faster than vertical such that horizontal concentration gradients are smoothed out rapidly. Therefore one-dimensional considerations are sufficient. However, effects of changing layer volumes with depth are considered.

Site description

The system investigated is the pit Lake 1b in the opencast lignite mining Merseburg-Ost. It is located about 20 km west of Leipzig in Central Germany. Since cessation of the mining activities in 1991, the pit is filling from surrounding natural aquifers and precipitation. At the base of the lake there is a several meters thick anaerobic monimolimnion with a NaCl-dominated salt content of about 25 g/kg (Boehrer et al., 1998; Trettin et al., 1999). Since August 1998, controlled surface fresh water inflow from the nearby river Weiße Elster has raised the lake surface by ~10 m as of February 2000.

Temperature and electrical conductivity. Profiles of temperature and electrical conductivity were measured (hand probe TetraCon 96-T, WTW Weilheim, Germany) with a depth resolution of 10 cm in the monimolimnion.

Between March and April 1999 (Fig. 2a), (1) and (2)), the thermal stratification started, and the separation of the epilimnion and the hypolimnion by the thermocline began. Inflowing fresh water from the river Weiße Elster at the lake surface decreased salinity in the epilimnion until August 1999 (Fig. 2b), (1)–(4)), and a gradient of the electrical conductivity (κ_{25}) appeared in the thermocline region. From September 1999 (5), the autumn turnover began with increasing mixing depths manifested in the profiles from September 1999 to February 2000 ((5)–(8)). The density step associated with the conductivity gradient moved downwards. The hypolimnion mixed with the upper water column yielded an increase of the conductivity from August to December 1999 despite the fresh water inflow. The depth region below 62 m a.s.l (above sea level) was protected from direct wind during the

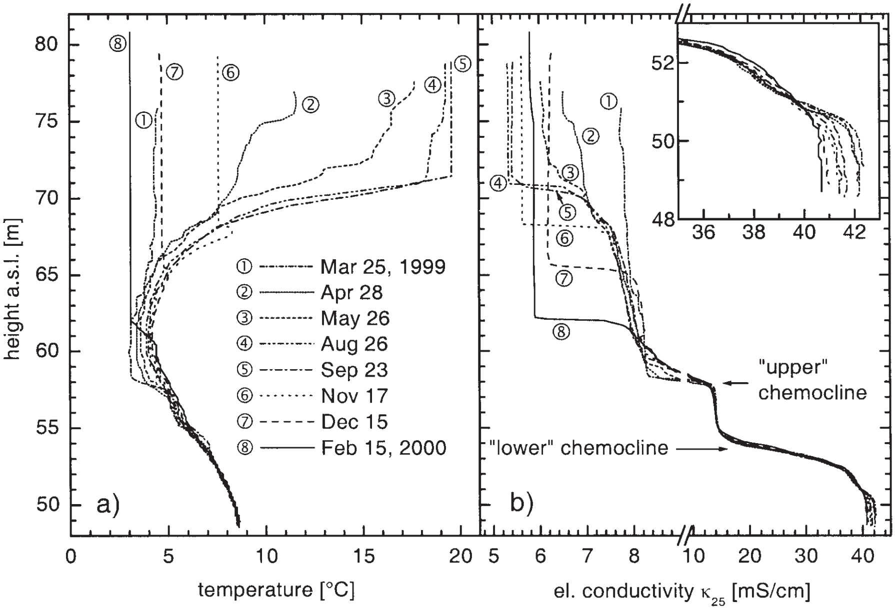

Figure 2. Temperature and electrical conductivity at the sampling site MB1 (Fig. 1) from March 1999 to February 2000

period from April 1999 to February 2000. In the deep monimolimnion, only small changes in the temperature profile occurred. The sediment temperature was found to be nearly constant at 8.6°C.

The overall accuracy of the measurements described and the NaCl-dominated (>85%, Boehrer et al., 2000 (b)) ion composition make the UNESCO expressions for ocean water (UNESCO 1983) acceptable to use for calculations of density and salt content from electrical conductivity and temperature measurements. The systematic error estimated for N^2 is smaller than a factor of two. Laboratory density measurements of anaerobic water samples from mining lakes are difficult and would have at least an error of ~1 kg/m^3 (Schimmele and Herzsprung, 2000; Gräfe and Boehrer, 2001) and are therefore not useful. In-situ density measurements with an ultrasonic method (Gräfe et al., 2001) reach at the moment an accuracy of ~0.1 kg/m^3, which is also less accurate than the UNESCO formula.

Figure 3 shows the Brunt-Väisälä-frequency calculated from the temperature and conductivity profiles from August 1999:

$$N^2 = -\frac{g}{\varrho_0}\frac{\partial \varrho}{\partial z} \equiv -\frac{g}{\varrho_0}\left\{\left(\frac{\partial \varrho}{\partial S}\right)\frac{\partial S}{\partial z} + \left(\frac{\partial \varrho}{\partial T}\right)\frac{\partial T}{\partial z}\right\} = N_S^2 + N_T^2 \qquad [s^{-2}], \tag{1}$$

where g is the acceleration of gravity, $\varrho = \varrho\,(\kappa_{25},T)$, ϱ_0 the density at the depth z, S the salinity and T the temperature. The contributions of temperature and salt content are plotted separately as N_S^2 for the salt gradient and as N_T^2 and $-N_T^2$, respec-

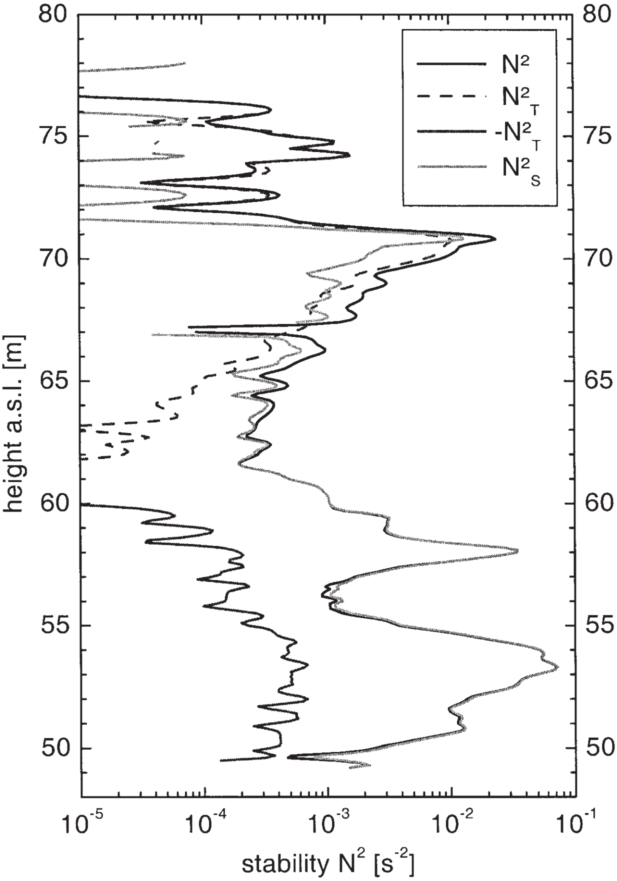

Figure 3. Stability in August 1999. N^2: stability from conductivity and temperature profiles, linearly interpolated at a 10 cm vertically spaced grid and smoothed, N^2_S: local stability from salt gradient, N^2_T: local stability due to temperature gradient. Below ~60 m a.s.l., where the temperature destabilizes ($N^2_T < 0$), the negative $-N^2_T$ is included

tively, for the temperature. The minus sign in $-N^2_T$ refers to the inverse temperature gradients, which cannot otherwise be included in the logarithmic scale. The increasing temperature with depth below 62 m a.s.l. has a slightly reducing effect on the stability. Below 50 m a.s.l., N^2 ($<10^{-3}$ s^{-2}) is one order of magnitude lower than the value of the summer stratification in the thermocline ($2 \cdot 10^{-2}$ s^{-2}).

Total salt content. The salt balance of the monimolimnion from bottom to ~58 m a.s.l. does not show a significant change over the period investigated. Due to the decreasing layer volumes towards the bottom, the temporal development of the salinity represented by κ_{25} especially in the deepest part in Fig. 2 is not reflected in the balance calculations within the error. Hence a significant inflow of saline groundwater into the monimolimnion cannot be stated at this time.

Measurements

The predominantly artificial gas sulfur hexafluoride is colorless, scentless and nonreactive. Its high chemical stability makes it suitable for electrical isolation and gas-shielded applications. Since mid of the 1950s, there is a rising level of SF_6 in the atmosphere by industrial emissions, well documented since the 1970s. The mixing ratio in the northern hemisphere was 4.78 pptv (pptv = 10^{-12} parts per volume) in January 2000, extrapolated from a quadratic fit based on atmospheric measurements (Maiss et al., 1998). The pre-industrial background level from natural sources was 0.054 pptv (Busenberg et al., 2000). The SF_6 concentration in the lake's surface layer develops according to the equilibrium solubility. The Ostwald solubility α, defined as the ratio of the concentrations ([fMol/l]) between water and atmosphere, can be specified from the Bunsen solubility β depending on temperature and salinity (Wanninkhof, 1992):

$$\beta(T, S) = \exp\left(75\,701 \cdot \ln(T) + 25\,060 \cdot \frac{1}{T} - 520\,606 - 0.0117 \cdot S\right)$$

$$\alpha = \beta \cdot \frac{T}{T_0}, \tag{2}$$

where T is the temperature [K], $T_0 = 273.15$ K and S is the salinity [g/kg]. Measured concentrations at the lake's surface vary seasonably from 1.3 to 2.8 fMol/l (1 fMol = 10^{-15} Mol). The molecular diffusivity for the temperature range of 5–10°C as found in the monimolimnion is $6–8 \cdot 10^{-10}$ m^2/s (King et al., 1995).

SF_6 tracer injection into the monimolimnion. In October 1998, the tracer was released from a rubber boat at the site of greatest depth of Lake 1b (MB1). To prepare the tracer, a 20 Liter tank with lake water was flushed with a gas composite of 10% SF_6 and 90% N_2 via a porous glass cap. Salt was added to this solution to attain the same density as the water at the depth of injection. Thereafter the spike was piped through a flexible hose and released horizontally ~3.5 m above the local lake bottom. To guarantee a sharp initial distribution of the tracer in the vertical, the ending of the hose was fixed horizontally at a rope, which was tied at the bottom with a weight. Sufficient SF_6 (1.1 mmol) was injected to achieve initial spike concentrations of around 1500 fMol/l after horizontal equalization, exceeding the background level by three orders of magnitude.

Sampling. The SF_6 sampling depths in the epilimnion and hypolimnion are selected on the basis of beforehand measured CTD profiles. At depth sections with rather strong density gradients (e.g. in the thermocline), a higher vertical sampling resolution was chosen. Within the monimolimnion, i.e. in the region of the tracer distribution, a 20 cm vertical sampling grid is attempted. Samples were taken monthly using a submersible battery-powered pump (Comet, Pfaffschwende, Germany) designed for groundwater wells. Water is pumped from the sampling depth to the surface through a 12 mm copper tube system at a rate of >5 l/min. While filling the sampling bottles (Fig. 4), an overpressure of up to 1 bar can be generated to prevent degassing in the copper pipe. The system provides efficient profiling of more than 10 samples per hour.

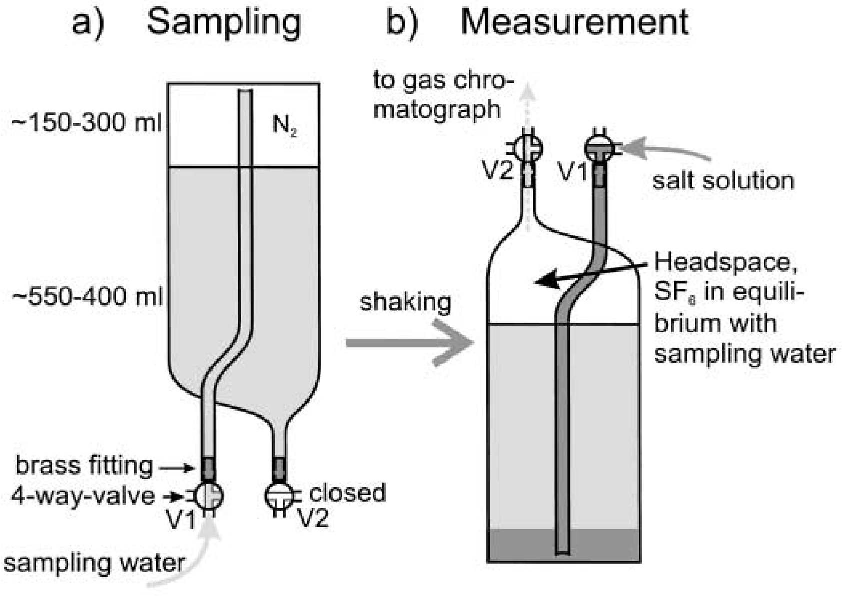

Figure 4. Sampling bottles

The sampling bottles (Fig. 4) were flushed and filled with ultrapure nitrogen in the laboratory and closed with some overpressure to prevent air contamination during transport. Just before taking the water sample, the bottles are evacuated to 0.2…0.5 atmospheric pressure, depending on the concentrations expected. After flushing the copper tubes, filling starts through valve V1, the bottle upside down, creating an absolute pressure of about 1100–1300 mbar in the remaining N_2-headspace. Due to the pressure drop within the bottle at the beginning of the filling, the strong degassing of the sample water helps to reach the equilibrium. A headlong transport of the bottles to the laboratory minimizes the risk of gas or water losses by possible leakages through the valves.

Laboratory measurements. SF_6 concentrations were measured using the method reported by Wanninkhof et al. (1991). In the closed bottle (Fig. 4), the water sample is shaken under controlled pressure and temperature conditions to achieve SF_6 equilibration between water and the nitrogen headspace. For the headspace measurements, a gas chromatographic system (Shimadzu GC-8AIE) specifically designed for SF_6 by Maiss (1992) with an ECD is used. After gravimetrical determination of the gas and sample water volumes, a SF_6 mass balance is applied to calculate from the measured headspace concentration c'_h in equilibrium with water to the original sample concentration c_w:

$$c_\mathrm{w} = c'_\mathrm{h}\left(\frac{V_\mathrm{h}}{V_\mathrm{w}} + \alpha\,(T, S)\right) \qquad \left[\frac{\mathrm{fMol}}{\mathrm{l}}\right]. \tag{3}$$

With the choice of the volume ratio of samples and headspace and different sample loops, we cover concentrations of 4 to 5 orders of magnitude from <1 fMol/l to several 1000 fMol/l. The most significant laboratory errors occur during the determination of headspace (V_h) and sample (V_w) volumes. Using comparable volumes of headspace and sample water, inaccuracies of the expression $\alpha\,(T, S)$ are dispensable. The error by the gas chromatographic system is ±1% for samples equilibrated with the atmosphere or for spike concentrations. The overall error of background lake measurements is at most ±10% and of spike measurements at most ±5%.

Results

The SF_6 profiles from November 1998 to February 2000 demonstrate the spreading of the initial distribution (Fig. 6). Maximum error bars of 5% are exemplary drawn for the September profile. For convenient calculations, they are linearly interpolated on a 10 cm grid. The temporal changes of the profile contours show the following characteristics: The maximum concentration decreases, while above and below the values increase. From the shape of the profiles we infer that SF_6 spreads faster towards the bottom than in the upward direction. The position of the SF_6 maximum moves upwards about 0.5 m. This can be addressed to the vertically varying mixing intensity which dominates the opposite effect due to the increasing layer volumes.

In Fig. 5 the SF_6 balance calculations are represented. The errorbars drawn result from the lake's surface level uncertainty of ±10 cm. This dominates the balance error vigorous. The nearly constant difference in the results of the two lower balance sections validates the less important role of other inaccuracies. The large

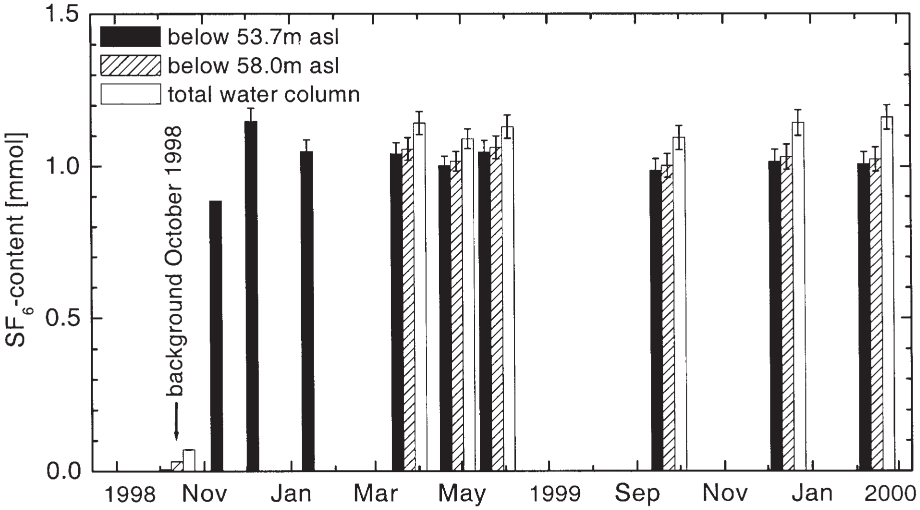

Figure 5. SF_6 content calculated from measured profiles (Fig. 6) for three vertical distances: bottom (48 m a.s.l.) to "lower" chemocline (53.7 m a.s.l.), bottom to "upper" chemocline (58 m a.s.l.) and total balance to the surface

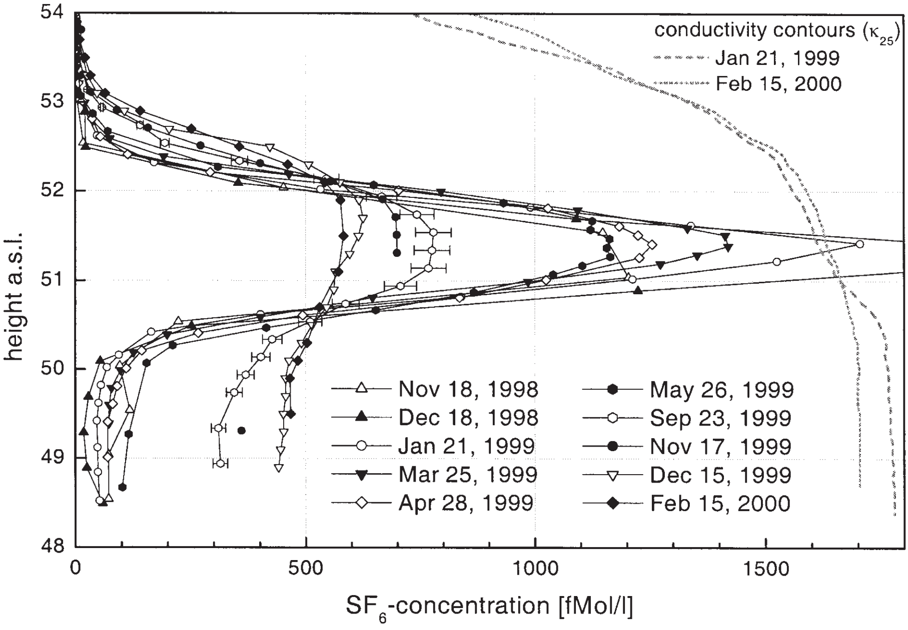

Figure 6. SF_6 profiles from November 1998 to February 2000. Exemplary, 5% errorbars for the September profile are plotted

variations before March 1999 are probably caused by the lower vertical sampling resolution and the still proceeding horizontal equalization. Relatively high SF_6 concentrations below 50 m a.s.l. in the November profile (Fig. 6) could possibly also be addressed to it. The ascending trend of SF_6 above the monimolimnion, i.e. the difference between total balance and balance below 58 m a.s.l. (Fig. 5, white column minus hatched column) is caused by the growing water column due to the surface inflow of water with background concentrations.

At March the horizontal settlement is assumed to be completed. After this time, the amount of SF_6 doesn't change, thus intense groundwater outflow or flushing of the monimolimnion can be excluded.

Vertical diffusion

As a first approach we can use the solution of Fick's 2nd law, starting with a δ-distribution, to estimate vertical coefficients of diffusion. Disregarding the effect of changing volume with depth, the decrease of the maximum concentration leads to a value of about 10^{-8} m^2/s. From the shape of the profiles close to the maximum (about ±1 m) the calculations for the measurements from December 1998 to May 1999 resulted in 10^{-8} m^2/s.

Eddy diffusivities for SF_6 were calculated with the flux-gradient method (Jassby and Powell, 1975; Heinz et al., 1990; Benoit and Hemond, 1996). It is based on the

First Fickian law $j = -K\ \partial c/\partial z$. The temporal change of heat or SF_6 content between the depth z and the maximum depth z_m yields the vertical flux through the lake area at this depth. Using the measured concentration gradients, K were calculated with

$$K_i = -\frac{\sum_{k=1}^{i} \frac{\Delta c_k}{\Delta t} \cdot V_k}{\frac{\Delta c_i}{\Delta z} \cdot A_i}. \tag{4}$$

where K_i are the diffusion coefficients at the surface of the layer i, c_k is the concentration in layer k, Δz is the layer thickness, V_i and A_i are the volume and the area of layer i; $i = 1$ refers to the bottom layer. As a first approach, the arithmetic mean of the vertical gradients from the chosen starting and ending profiles for each layer were used. The temporal change follows from the concentration difference between the starting and ending profiles at the depth z. The calculations showed that the K-profiles didn't change within the errors at chosen time intervals, i.e. up to now we don't see a dependence on the season. Therefore, the K-profile for the whole time step from March 1999 to February 2000 in the depth region between 49 m a.s.l. and 53 m a.s.l. was calculated as arithmetic mean of all feasible time intervals (Fig. 7) using the SF_6-profiles from March, April, May, September, December 1999 and February 2000. The errorbars represent the standard deviation of the results.

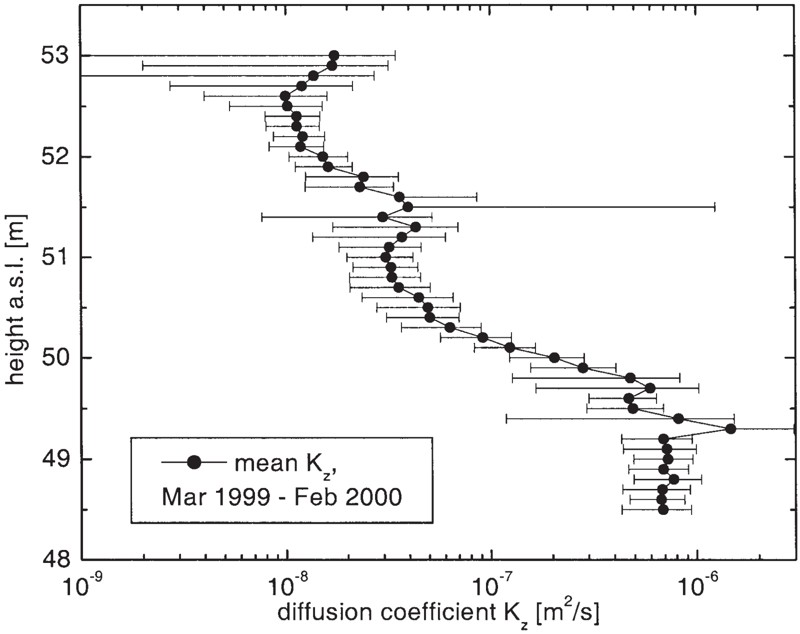

Figure 7. Mean vertical diffusion coefficients K, calculated by flux-gradient method from measured SF_6 profiles (Fig. 6)

The values are several times 10^{-8} m²/s (one order of magnitude lower than the molecular diffusivity of heat) slightly increasing from 52 m a.s.l. down to 50.5 m a.s.l. and showing a sharp increase at ~ 50 m a.s.l. by a factor of ~ 10 (~ $7 \cdot 10^{-7}$ m²/s) in the deep part of the monimolimnion about 2 m above the lake bottom. Because of the logarithmic scale, not all of the left side errorbars could be drawn, in particular for the depths around the SF_6-maximum. There (~ 51.5 m a.s.l.), the measured vertical concentration gradients tend to vanish (denominator in eq. 4). This entails large errors, but do not influences the calculations in the depth region above, because the mass balance (numerator in eq. 4) is not affected thereby. At depths above 53 m a.s.l., diffusion coefficients could not be evaluated as the concentration changes of SF_6 were not high enough.

In parallel, diffusion coefficients for heat transport can be evaluated from measured temperature profiles (Fig. 2) similarly to the SF_6. In the deep part of the lake, where the sediment area to volume layer ratios are comparatively large, a thermal flux Q from the sediment must be included as heat source, since it influences the heat balance significantly. Weighted with the sediment areas per layer volumes, Q [W/m²] is included in the expression for K (see eq. 4):

$$K_i = -\frac{\sum_{k=1}^{i} \left\{ \frac{\Delta T_k}{\Delta t} \cdot V_k - \frac{Q\,(A_k - A_{k-1})}{c_p\, \varrho} \right\}}{\frac{\Delta T_i}{\Delta z} \cdot A_i} . \tag{5}$$

where c_p and ϱ are the heat capacity and the density of the water. The value used for Q is 0.23 W/m². It is determined by the assumption that the heat diffusivity cannot exceed the molecular portion of ~10^{-7} m²/s, if the measured SF_6-diffusivity is definitely smaller in the same depth region (Fig. 8). Q is comparable with values found in the same manner in other Central German mining lakes (Walz, 1997; von Rohden, 1998). The uncertainty of Q (0.05 W/m²) is represented by the fluctuation of the Q used for all eligible time steps between March 1999 and February 2000. Below 50 m a.s.l., the mixing coefficients are higher than 10^{-7} m²/s and show the same depth dependence for both heat and SF_6 transports.

There are quite a few examples in the literature where a relationship of the type $K \propto (N^2)^{-q}$ is sought (e.g. Heinz et al., 1990; Quay, 1977). We expect the evaluated vertical diffusion coefficients to be a result of a combination of many processes, and regarding N^2 as the only relevant parameter would not hold for a closer investigation. But it seems reasonable to relate the diffusivity to the stability because the two influence each other. Figure 9 however confirms a connection between both magnitudes in a domain (N^2 ~ $10^{-3} \ldots 10^{-1}$ s^{-2}; K ~ $10^{-6} \ldots 10^{-8}$ m²/s, which previously has not been investigated. A fit to the data from 49.3 m a.s.l. to 52.5 m a.s.l. yields a slope of $q = 1.2 \pm 0.1$.

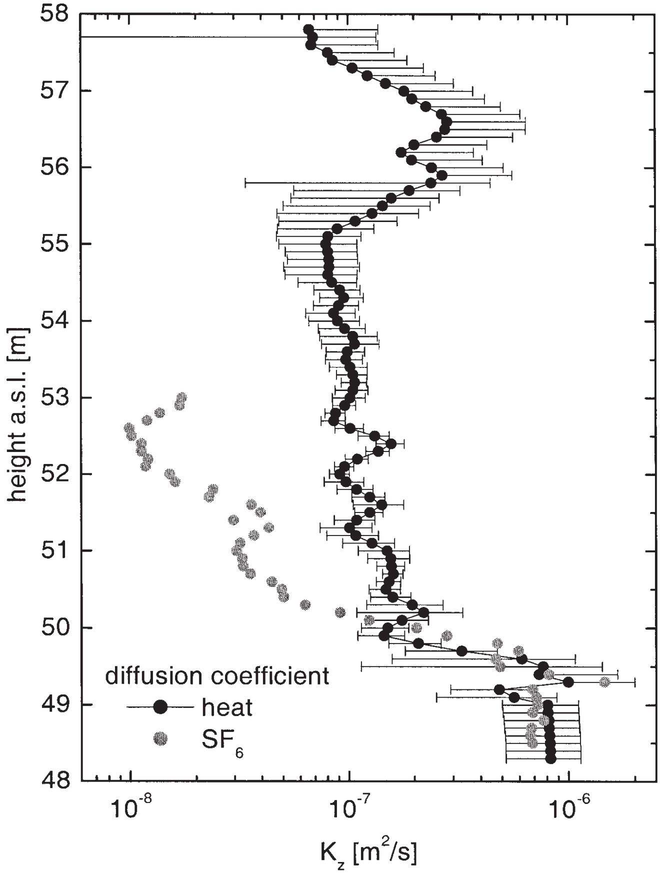

Figure 8. Mean vertical diffusion coefficients K, calculated by flux-gradient method from measured temperature profiles (Fig. 2) including a heat flux from the sediments of 0.23 W/m^2. The SF$_6$ diffusion coefficients are also plotted (faint symbols)

Conclusions

The main purpose of the investigations in the mining lake Merseburg-Ost 1b is the measurement of the vertical distribution of mean diffusivities in the stably stratified monimolimnion. At the stratification conditions established here, measured diffusivity and stability range over more than two orders of magnitude in a vertical range of a few meters with an overall slow characteristic of transport. The mixing patterns found are caused by various processes as wind forcing, internal waves and bottom boundary mixing (see Gloor et al., 2000; Goudsmit et al., 1997; Wüest et al., 1996 and Wüest et al., 2000). Considerations of the particular contributions of this processes to the production of turbulence require further substantial investigations which were not part of our analysis.

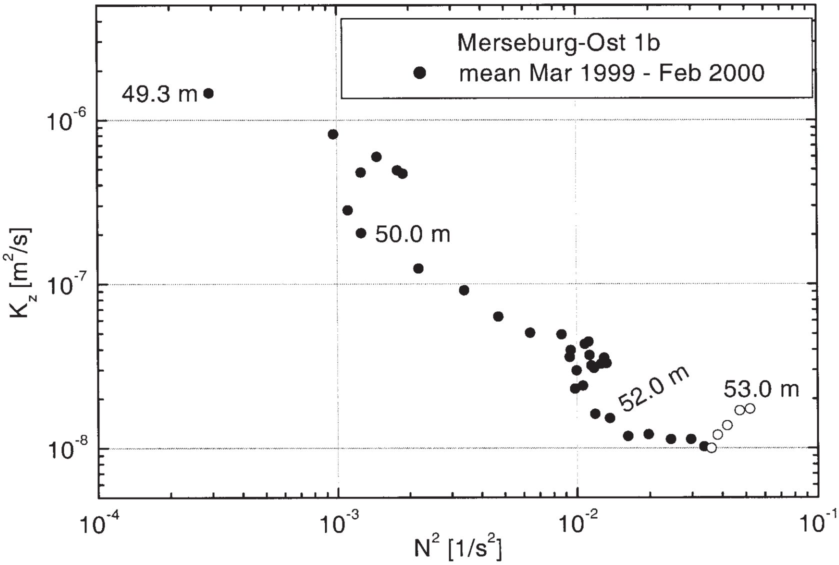

Figure 9. Vertical diffusion coefficients *K* (Fig. 7) versus stability N^2

The use of SF_6 in tracer experiments is a powerful method to quantify mean vertical diffusion coefficients under conditions described. It is even feasible to confirm diffusivities lower than the molecular diffusivity of heat. In the mining lake Merseburg-Ost 1b, diffusion coefficients of around 10^{-8} m^2/s were found in the monimolimnion between 50 and 53 m a.s.l. This is one order of magnitude above the molecular diffusivity of SF_6.

The SF_6-spike in lake 1b provides a good base for further experimental work. This includes measurements of the vertical transport from the monimolimnion into the mixolimnion across the chemocline and the estimation of possible exchange with the surrounding groundwater aquifers. The data collected so far will be used for model descriptions of the tracer development and its turbulent transport.

ACKNOWLEDGEMENTS

This work was funded by UFZ – Centre for Environmental Research, within research contract: "SF_6 als Tracer für Transport- und Mischungsvorgänge in Tagebaurestseen". The authors thank their colleagues of UFZ-Limnophysik for great support with field trips and useful discussions.

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Received 13 February 2001;
revised manuscript accepted 18 June 2001.