



Diluting a salty soup: Impact of long-lasting salt pollution on a deep Alpine lake (Traunsee, Austria) and the downside of recent recovery from salinization

Harald Ficker¹ · Martin Luger¹ · Barbara Pamminger-Lahnsteiner¹ · Daniela Achleitner¹ · Albert Jagsch¹ · Hubert Gassner¹

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Abstract

Saline emissions can elevate water densities and have the potential to alter stratification and mixing dynamics in lakes. In the light of rising anthropogenic salinization of temperate freshwater lakes, it is highly relevant to understand how saline emissions, as well as the rehabilitation from salt pollution might affect lake circulation patterns. In this study, we present the impact of industrial pollution with chloride salts on the deep Alpine lake Traunsee in Austria from 1930 to 2005 and the observed limnological changes during the recovery from salinization in the following 12 years. We assembled and analysed a unique dataset of monthly sampling profiles covering chloride, conductivity, temperature and dissolved oxygen over a 87-year period. We quantified the impact of saline emissions on the lake's total chloride content, water densities, stability indices and deepwater oxygen concentrations. Time-series of water density profiles and stability indices revealed a significant impact of salinization on seasonal stratification and mixing. Higher dilution and shallower release of saline wastes and the short water retention in Traunsee helped to prevent a long-lasting density stratification. Nevertheless, two periods of salt-induced meromixis occurred in the lake. The first was caused directly by the disposal of saline wastes into the deepwater in the 1930s and 1940s, whereas the second was induced by the recovery from salinization in recent years. The naturally faster wash-out of salts from shallower water layers reinforced the density gradient and impeded vertical water circulation after salt pollution ceased. The rehabilitation from salinization had a stronger effect on stratification intensity and deepwater oxygen concentration than the continuous salt pollution over the last century.

Keywords Meromixis · Salinization · Soda production · Temperate lake · Oxygen depletion · Water density

Introduction

Salinization of freshwater lakes by various anthropogenic sources of chloride salts is an emerging issue and has the potential to threaten lake ecosystems especially in temperate regions (e.g. Kaushal et al. 2005; Müller and Gächter 2011; Rogora et al. 2015; Dugan et al. 2017a, b). High chloride concentrations can have multiple adverse effects on freshwater organisms such as chronic toxicity or changes in species composition (e.g. EPA 1988; Evans and Frick 2001; CCME

2011). Effects on the whole lake ecosystem are also possible through changes in density stratification and seasonal mixing dynamics. The impact of salts on the density stratification of lakes is well-known from coastal basins where sea water was trapped by a freshwater layer or from alkaline lakes where evaporation and geochemical processes caused the development of a saline gradient (e.g. Anderson 1958; Sanderson et al. 1986; Miller et al. 1993; Hakala 2004; Wüest et al. 2012). Due to the salt-induced density gradient, such lakes can remain meromictic over several thousand years with only partial mixing and anoxic conditions in the saline deepwater (Wüest et al. 2012; Boehrer et al. 2017).

How seasonal mixing can be affected by salts is an interesting and insufficiently investigated aspect in the light of rising man-made salinization of temperate inland waters (Dugan et al. 2017b). A salty effluent usually has a higher density compared to lake water and flows to the

✉ Harald Ficker
harald.ficker@baw.at

¹ Institute for Freshwater Ecology, Fisheries Biology and Lake Research, Federal Agency for Water Management, Scharfling 18, 5310 Mondsee, Austria

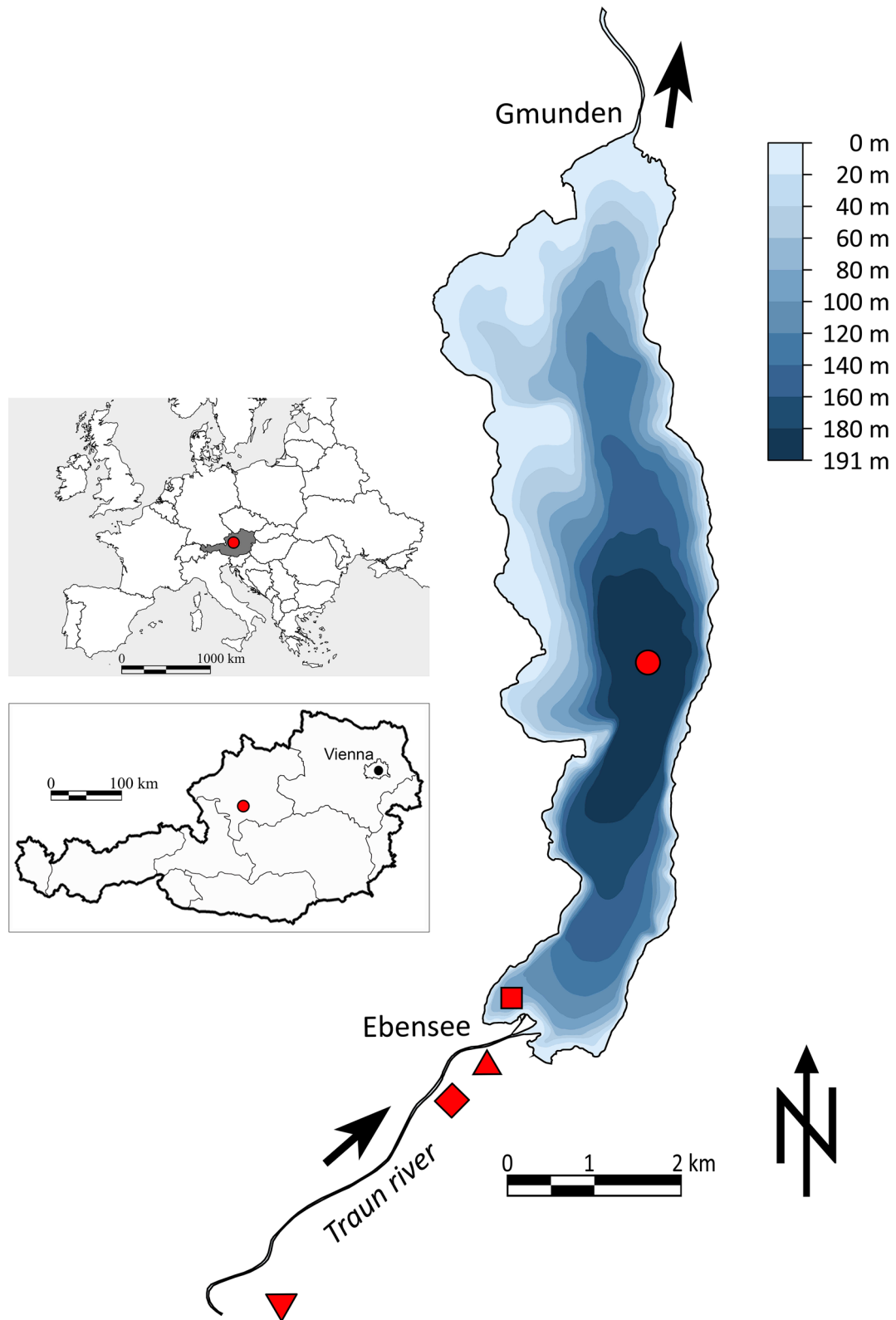


Fig. 1 Location and morphometry of Traunsee. Circles indicate the location of Traunsee in Europe and Austria and the location of the sampling site. The diamond symbol marks the position of the soda work in Ebensee. Triangle symbols give the positions of the old salt

work (upward triangle) which was shut down in 1979 when the new salt work (downward triangle) started its production. The square marks the position of the floating pier for the disposal of industrial wastes in the bay of Ebensee

depth where it is in equilibrium with ambient water densities. This can boost vertical mixing by the induced convection and lead to the dispersion of discharged salts within the lake. When saline emissions are highly concentrated or are released into the deepwater it is more likely that salty waste waters accumulate in the hypolimnion, reinforce the density gradient and induce meromixis of the lake (e.g. Effler and Owens 1996; Ficker et al. 2011). In cases of strongly reduced or ceased salt pollution one would expect a continuous decline of the lakes' salt content until the natural geochemical state is reached. However, water renewal and consequently the wash-out of dissolved salts is a function of depth and time in seasonally stratified lakes (e.g. Varekamp 2002; Ambrosetti et al. 2003; Pilotti et al. 2014). More precisely, the salt wash-out from the hypolimnion can only be effective during the mixing period in the cold season, whereas it is a perennial process in the epilimnion. Higher precipitation and runoff from the catchment may further amplify the wash-out from the epilimnion in summer (Imboden and Wüest 1995; Rimmer et al. 2005; Pilotti et al. 2014). Hence, a saline gradient could appear which reinforces the density stratification and impairs or hinders complete vertical mixing in consequence of the recovery from salinization.

In this study, we present the exceptionally well documented case of salt pollution in Traunsee, a deep Alpine lake in Austria. Saline emissions from soda and salt production were continuously released into the lake over the last century until the shutdown of the soda work in the year 2005. Accordingly, the Traunsee case represents a perfect opportunity to investigate both, the impact of continuous salt pollution on seasonal stratification and mixing and the potential reinforcement of the density gradient due to the recovery from salinization. We assembled and analysed a unique data set comprising 87 years of monthly lake sampling profiles of Traunsee between 1930 and 2016. Our research aims were to (1) assess the contribution of industrial salt disposal to the lake's chloride content, (2) quantify changes in profiles of chloride concentrations and electrical conductivities, (3) investigate if saline emissions had an impact on seasonal mixing and stratification through analyses of dissolved oxygen profiles, water density profiles and calculated stability indices and (4) test if density stratification was reinforced by the recovery from salinization from 2005 onwards.

Materials and methods

Study site

Traunsee is situated in the "Salzkammergut" lake district and is the deepest and one of the largest lakes of Austria.

The geologic surrounding of the lake is formed by the Northern Calcareous Alps in the southern part and by flysch and glacial deposits in the northern part (Schneider et al. 1986). The name "Salzkammergut" originates from the large salt deposits and the long salt mining tradition in the region. The river Traun represents the main inflow and the main outflow of Traunsee (Fig. 1). This tributary has high annual discharging rates resulting in a short theoretical water renewal time of about 1 year (Table 1).

Industrial disposal of chloride salts

Minor effects of salinization of Traunsee date back to 1607 when the first salt extraction plant started its operations at the southern lake shore (Fig. 1). A new era of industrial salt pollution began in 1885 with the launch of soda production in Ebensee (Fig. 1; Pechlaner and Sossau 1982; Steinhauser 2008). The by-products of the production process, dissolved saline waste and calcium-rich solid waste, were discharged initially into the main tributary, the river Traun. Dissolved saline waste from the soda work mainly consisted of calcium chloride (CaCl_2), sodium chloride (NaCl) and to a much lesser degree of potassium chloride (KCl), calcium hydroxide ($\text{Ca}(\text{OH})_2$), calcium sulfate (CaSO_4) and ammonia (NH_3). The total amount of disposed chloride was on average 98.4 kg m^{-3} waste (see Table 2).

In 1927, the soda company started to discharge all wastes directly into the lake together with saline waste from the salt industry. The disposal facility was situated on a floating pier in the southern part of Traunsee (Fig. 1). In the 1930s and early 1940s, liquid waste emissions had densities between 1106 and 1116 kg m^{-3} and were released into Traunsee at a depth of 30 m below the floating pier (unpublished report by W. Einsele 1940; Sossau 1982). Between 1941 and 1948, the outflow was repeatedly lifted to the final depth of about $3\text{--}5 \text{ m}$ below the surface and equipped with several tubes and nozzles to improve the dispersion in the epilimnion. During that time, also the dilution of effluents was improved by technical modifications of mixing pumps, leading to a density reduction to 1023 kg m^{-3} until 1951. In 1969, the density of saline wastes was reduced once again to 1018 kg m^{-3} .

The major part of saline emissions entering Traunsee originated from soda production, whereas on average only 6% of industrial salt disposal originated from salt production over the past decades (Fig. 2). In the late 1920s, annual waste disposal amounted to about $62,302 \text{ t}$ of dissolved saline waste from soda production (i.e. $39,250 \text{ t year}^{-1}$ chloride; Fig. 2). In 1940, legal authorities limited the annual disposal to $170,090 \text{ t}$ (i.e. $107,157 \text{ t year}^{-1}$ chloride; Fig. 2). The legal limit was

Table 1 Limnological characteristics of Traunsee

Geographic coordinates (deepest site, WGS 84)	47°51'15"N, 13°48'10"E
Elevation (m a.s.l.)	422
Surface area (km ²)	24.35
Max. length (km)	12.20
Max. width (km)	2.94
Max. depth (m)	191
Mean depth (m)	94.54
Volume (million m ³)	2302
Catchment area (km ²)	1417
Theoretical water renewal (years)	1.04
Trophic status	Oligotrophic

Table 2 Average compositions of dissolved wastes from soda ash production

Composition of saline waste	kg m ⁻³	Cl ⁻ fraction (kg m ⁻³)
CaCl ₂	107.4	68.6
NaCl	46.6	28.3
KCl	3.1	1.5
Ca(OH) ₂	1.4	–
CaSO ₄	0.9	–
NH ₃	0.006	–
	159.4	98.4

further elevated in 1951 to 211,700 t (i.e. 133,371 t year⁻¹ chloride; Fig. 2) and once again in 1970 to 240,900 t (i.e. 151,767 t year⁻¹ chloride; Fig. 2). In contrast, the saline input by the salt industry increased only slightly until the 1970s. In 1979, the old salt work at the lake shore was closed and a new salt work started its production about 5 km south of the lake (see Fig. 1). From that time, waste

from salt production (i.e. “mother liquor”) was re-used in the soda work which led to a reduction of saline emissions by the salt industry (Fig. 2). In summer 2005, the soda work was shut down and the vast majority of chloride pollution was terminated. The remaining minor emissions from salt production stopped in 2007 due to the backhaul of saline waste to salt caverns.

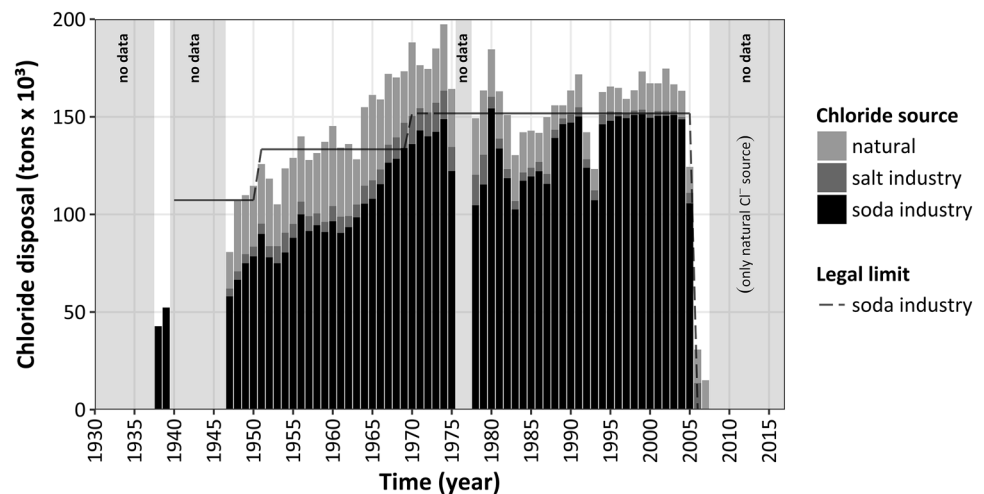
Lake monitoring data

Vertical sampling at the deepest site of Traunsee started in June 1930. Since then, water samples were collected with a 5-L container in multiple depths between surface and lake bottom on a regular basis. Water temperatures were determined at the sampling site with a mercury thermometer. Electrical conductivities of water samples were determined in the laboratory with an electrode at reference temperatures of 18 °C (1930–1974), 20 °C (1975–1985) and 25 °C (1986–2016). Dissolved oxygen profiles were additionally recorded since 1941. Dissolved oxygen concentrations were quantified in the laboratory with the Winkler method (Winkler 1889). The total available measurements amount to 17,991 temperature records, 16,956 conductivity records, 16,785 chloride records and 15,785 oxygen records collected on 960 days during the last 87 years (1930–2016). Further details about temporal resolution of sampling data are presented in Figs. 10, 11, 12 and 13 in Appendix.

Data preparation

First, we made a quality control of all available sampling data. Review and quality check followed the general requirements for the competence of testing and calibration laboratories (ISO/IEC 17025:2005 + Cor

Fig. 2 Annual quantities of chloride disposal into Traunsee between 1930 and 2016. Chloride disposals are grouped by natural sources and emissions from the local salt and soda industry. Periods with missing data are shaded in light grey



1:2006). Second, we corrected electrical conductivities (K) recorded between 1930 and 1985 from the reference temperatures of 18 °C and 20 °C to the reference temperature of 25 °C with $K_{25} = K_{18} \times 1.168$, respectively, $K_{25} = K_{20} \times 1.116$ according to the non-linear temperature correction described in ISO 7888:1985. Third, we applied a two-stage interpolation procedure to fill spatial and temporal data gaps (see also North and Livingstone 2013; North et al. 2014; Ficker et al. 2017). We used linear interpolation over depth to generate vertical profiles with 1-m resolution per sampling date. Subsequently, we filled temporal data gaps between sampling dates with linear interpolation for conductivity and chloride data and cubic spline interpolation for temperature and oxygen data. The resulting time-series data contained daily vertical lake profiles in 1-m resolution between surface and the deepest part of Traunsee (i.e. 191 m) from June 1930 to December 2016 with 5,966,076 individual values of water temperature, conductivity and chloride concentration and 5,232,063 values of dissolved oxygen concentration. Data of 1945 were excluded in all calculations due to the low sampling frequency during that year (see Appendix).

Chloride content

We multiplied interpolated chloride concentrations by the respective volume of each 1-m stratum to determine the total chloride masses in Traunsee between 1930 and 2016. Since annual chloride contents are probably linked to the annual chloride disposal by the industry (Fig. 2), we made a non-parametric correlation analyses on the two parameters (i.e. Spearman's rank correlation).

Recovery from salt pollution

Wash-out of a chloride salts from periodically stratified lake systems generally appears as a negative exponential model (Varekamp 2002; Ficker et al. 2011; Pilotti et al. 2014) with

$$C_t = C_0 e^{-Rt},$$

where C_t stands for the chloride mass at time t , C_0 is the initial mass and R is the washout rate that describes the reduction per unit time. To examine the recovery of Traunsee from salt pollution, we fitted the reduction model to monthly chloride masses and mean conductivities between surface and the deepest part of the lake from 2005 to 2016. We additionally calculated mean dissolved oxygen concentrations in water layers > 185 m depth to investigate the development of dissolved oxygen in the deepest part of the lake in recent years.

Water density and stratification

Calculation of water densities is crucial to investigate stratification and mixing dynamics of lakes. The majority of reliable formulas, that consider the contribution of dissolved salts in addition to the contribution of temperature to water density, requires the estimation of salinity. These approaches originate from oceanography and are not perfectly suited for freshwater lakes. A more appropriate and simple method is the inclusion of conductivity in density calculations to account for the contribution of dissolved salts (Moreira et al. 2016). Therefore, we chose the method RHO_LAMBDA (ρ_λ) proposed by Moreira et al. (2016) which requires temperature (T), conductivity (K_{25°) and two coefficients (λ_0 and λ_1) to calculate water density with

$$\rho_\lambda(T, K_{25^\circ}) = \rho_w(T) + K_{25^\circ} [\lambda_0 + \lambda_1 \times (T - 25^\circ \text{C})].$$

We obtained the two λ -coefficients from density calculations of Lake Constance, which has a comparable chemical composition to Traunsee ($\lambda_0 = 0.62$ and $\lambda_1 = -0.0014$; see Moreira et al. 2016; Pechlaner and Sossau 1982). Calculation of density as a function of temperature ($\rho_w(T)$) refers to the International Temperature Scale of 1990 (ITS-90) and the density formulation of Jones and Harris (1992) with

$$\rho_w(T) = 999.85308 + 6.32693 \times 10^{-2}T - 8.523829 \times 10^{-3}T^2 + 6.943248 \times 10^{-5}T^3 - 3.821216 \times 10^{-7}T^4.$$

We calculated $\rho_w(T)$ and $\rho_\lambda(T, K_{25^\circ})$ with the interpolated time-series data of Traunsee for each depth and day between June 1930 and December 2016.

With the calculated density profiles we evaluated daily water column stability N^2 for every depth (i.e. squared Brunt-Väisälä frequency) as indicator of stratification and mixing dynamics within the water column and daily Schmidt stability (S) as indicator of stratification intensity of the whole water column. The buoyancy frequency quantifies the oscillation of vertical displacement in relation to density differences between specific water layers, whereas the Schmidt stability expresses the amount of work that is required to mix the entire water column to a uniform density. We calculated two types of stability indices N^2 and S to evaluate the contribution from dissolved salts, one based solely on water temperatures (i.e. N_t^2 and S_t) and the other based on temperatures and conductivities (i.e. N_{tk}^2 and S_{tk}). For density and stability computations we utilized the R-package "rLakeAnalyzer" (Read et al. 2011). We adapted density equations in the R-package with $\rho_w(T)$ and $\rho_\lambda(T, K_{25^\circ})$ in the calculations of N_t^2 and S_t , respectively, N_{tk}^2 and S_{tk} . Subsequently, we compared the development of daily profiles of N_t^2 and N_{tk}^2 to investigate the impact of saline emissions on stratification and mixing in the water column and annual mean values of S_t and S_{tk} to examine salt-induced changes

in the overall stratification intensity of Traunsee. We also contrasted minimum S_t and S_{tk} of each year to inspect the effects of salinization on water column stabilities during the mixing period in the cold season.

Without salinization effects, the intensity of seasonal thermal stratifications are usually controlled by epilimnic water temperatures which in turn are affected by prevailing air temperatures during the warm season. Climatic changes considerably affected temperate lakes over the past decades (e.g. Adrian et al. 2009; O'Reilly et al. 2015; Ficker et al. 2017). Therefore, we also tested if summer temperatures had an impact on stratification intensity of Traunsee. We used available monthly air temperature data from two meteorological stations near Traunsee (i.e. station Bad Ischl and Feuerkogel; see HISTALP station map and data available at: <http://www.zamg.ac.at/histalp> and Auer et al. 2007) and conducted a Pearson product-moment correlation analyses with annual mean S_t and mean air temperatures between April and September.

Results

Chloride content

The total chloride content of Traunsee was considerably elevated by industrial salt pollution over the last century. We identified a significant statistical correlation between annual chloride disposal and the lake's annual chloride masses (Spearman's $\rho=0.83$ with $p<0.0001$ and $n=58$; see also Figs. 2 and 3). Starting in 1930, the lake's chloride content increased until 1943 followed by a slight reduction in the years after the Second World War (Fig. 3). From 1950 onwards, the chloride mass in Traunsee increased steadily until its maximum in 1973. A longer period of decreasing chloride masses set in afterwards. In 1989, the chloride content began to rise again and stabilized in the mid-1990s till

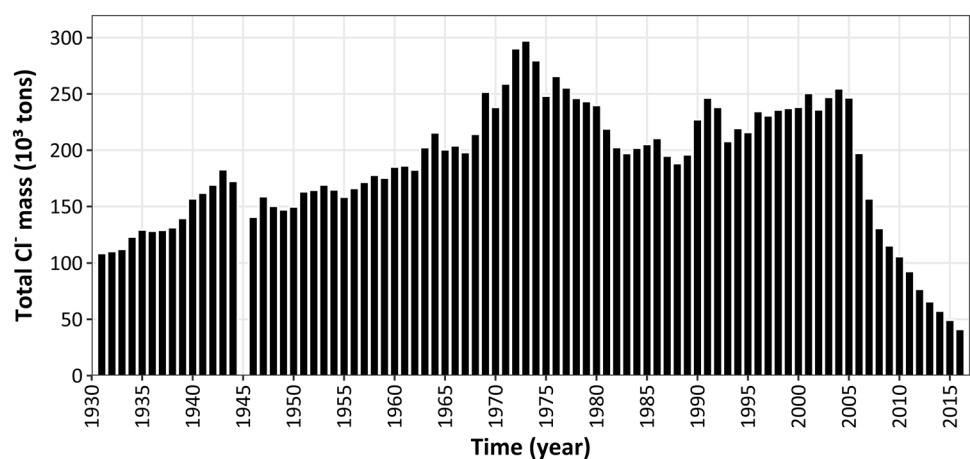
the mid-2000s. From 2005 to 2016, total chloride amounts in Traunsee declined substantially, leading to the lowest chloride content in the lake since the beginning of the monitoring program (Fig. 3).

Profiles of chloride, conductivity and oxygen

Chloride concentrations in Traunsee showed seasonal patterns in water layers between surface and about 50 m depth. We observed lower chloride levels in the epilimnion during summer than during winter (Fig. 4a). Chloride concentrations in water layers below 50 m were free from seasonal variations. During the Second World War (1939–1945) chloride levels showed a peak in the hypolimnion with concentrations >100 mg L⁻¹. After slightly decreasing chloride levels in the post-war years, concentrations increased in the hypolimnion almost constantly in the 1950s, 1960s and 1970s. The maximum chloride concentrations were recorded in early spring 1973 with a peak of 170 mg L⁻¹ at depths from 180 to 191 m. At the same time, very high chloride concentrations with a maximum of 166 mg L⁻¹ appeared also in shallow water layers. In the following years, chloride concentration decreased in the hypolimnion below 120 mg L⁻¹ and started to increase again from 1990 onwards. A short period of chloride reduction in the deep-water occurred in 1992 and 1993 in response to the lower chloride disposal in these years (see Fig. 2). Between 1996 and 2005, chloride levels increased steadily with a final peak in the years 2004 and 2005. After industrial salt input ceased in 2005, chloride concentrations declined more rapidly in shallower water layers than in deepwater layers. The chloride gradient was still noticeable in 2016. The difference between surface and the deepest part of the lake was about 24 mg L⁻¹ the smallest chloride gradient since the beginning of the monitoring program in 1930.

During the last 87 years the development of conductivities was similar to the course of chloride concentrations

Fig. 3 Development of annual chloride contents of Traunsee derived from sampling data



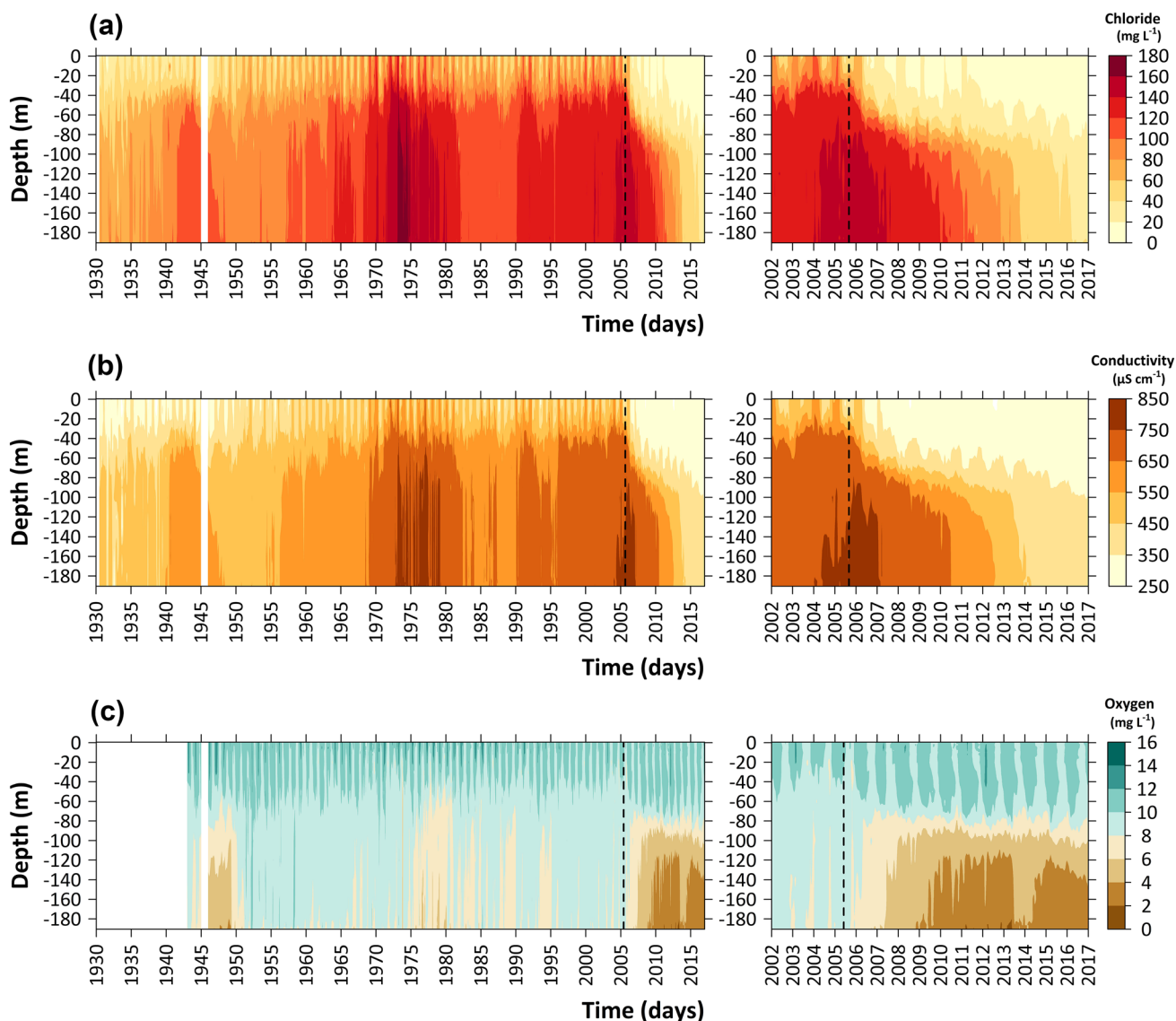


Fig. 4 Isopleth diagrams of interpolated daily vertical profiles of **a** chloride concentration, **b** conductivity and **c** oxygen concentration between June 1930 and December 2016 (left panel) and between

January 2002 and December 2016 (right panel). Vertical dashed lines indicate the shutdown of the soda production in summer 2005

(Fig. 4b). A statistical comparison of daily vertical profiles of conductivities and chloride concentrations resulted in a highly significant correlation of the two parameters (Spearman's $\rho = 0.98$ with $p < 0.0001$; see Figure S4). We observed three distinct periods with exceptionally high conductivities in the hypolimnion, comparable to the development of chloride concentrations. The first period with deepwater conductivities up to $623 \mu\text{S cm}^{-1}$ during the Second World War, the second with values up to $820 \mu\text{S cm}^{-1}$ in the mid-1970s and the third with conductivities up to $766 \mu\text{S cm}^{-1}$ in the mid-2000s.

Dissolved oxygen concentrations, recorded since 1941, revealed periods of stronger oxygen depletion in the

hypolimnion (Fig. 4c). We identified two meromictic periods indicated by continually declining dissolved oxygen in the deepwater over several years. The first meromictic period appeared between 1944 and 1949 and the second in the time after salt disposal ceased in 2005 until present. During meromixis, dissolved oxygen declined to minimum concentrations of 3.4 mg L^{-1} in December 1948 and 1.2 mg L^{-1} in March 2013, respectively. Distinct reductions in dissolved oxygen concentrations in the hypolimnion appeared as well from 1975 to 1980, from 1982 to 1984, from 1988 to 1990 and from 1993 to 1995. However, the depletion of oxygen was less pronounced as during the 1940s and late 2000s and repeatedly interrupted by rising oxygen concentrations.

Anoxic conditions never occurred in Traunsee between 1930 and 2016.

Recovery from salt pollution

The total chloride content of Traunsee and mean conductivity in the water column declined quickly after salt pollution ceased in summer 2005. Reductions in chloride concentrations and conductivities were clearly faster in the epilimnion than in the hypolimnion which raised the chloride and conductivity gradient. Until 2016, the overall annual chloride mass in Traunsee decreased from 246,660 to 41,183 t (see Fig. 3), while annual mean conductivities decreased from 700 to 344 $\mu\text{S cm}^{-1}$. Reductions in monthly mean chloride content and conductivity showed some seasonal oscillations and appeared as exponential decays (Fig. 5a, b). During the recovery of Traunsee dissolved oxygen concentrations in water layers > 185 m depth declined exponentially from about 9 mg L^{-1} in 2005 to below 2 mg L^{-1} in 2013. An abrupt increase in deep oxygen concentrations to roughly 4 mg L^{-1} appeared in June 2013 (Fig. 5c).

Water densities, stratification and mixing

Water densities in Traunsee derived solely from temperature data (i.e. $\rho_w(T)$) showed a seasonal pattern. Lower densities appeared in the epilimnion during summer due to warmer temperatures (Fig. 6, upper panel). Densities $\rho_w(T)$ in the hypolimnion remained constant between 999.9 kg m^{-3} and 1000.0 kg m^{-3} . Water densities derived from temperature and conductivity data of Traunsee (i.e. $\rho_\lambda(T, K_{25})$) additionally revealed elevating effects of salinization on water densities in the hypolimnion (Fig. 6, lower panel). Salt pollution raised water densities in deeper water layers by 0.1–0.5 kg m^{-3} . This resulted in higher density gradients in the water column, especially during the two meromictic periods in the 1940s and 2000s when distinct oxygen reductions occurred in the hypolimnion.

Comparison of daily buoyancy frequencies N_{ik}^2 and N_i^2 , with and without inclusion of measured conductivities, demonstrate the impact of saline emissions on stratification and mixing dynamics in Traunsee from 1930 to 2016 (Fig. 7). Calculated buoyancy frequencies in Fig. 7 indicate locally stable water layers when $N^2 > 0$, local neutral stability when $N^2 = 0$ and the appearance of convection when $N^2 < 0$. A closer look on the development of N_{ik}^2 confirmed only partial mixing during the two observed meromictic periods, while N_i^2 values suggested complete vertical mixing in the cold seasons of every single year. Very interestingly, the termination of salt pollution in 2005 was followed by strongly increasing local stabilities from the epilimnion down to a depth of about 100 m (see N_{ik}^2 in Fig. 7). This process was induced by the faster wash-out of salts from

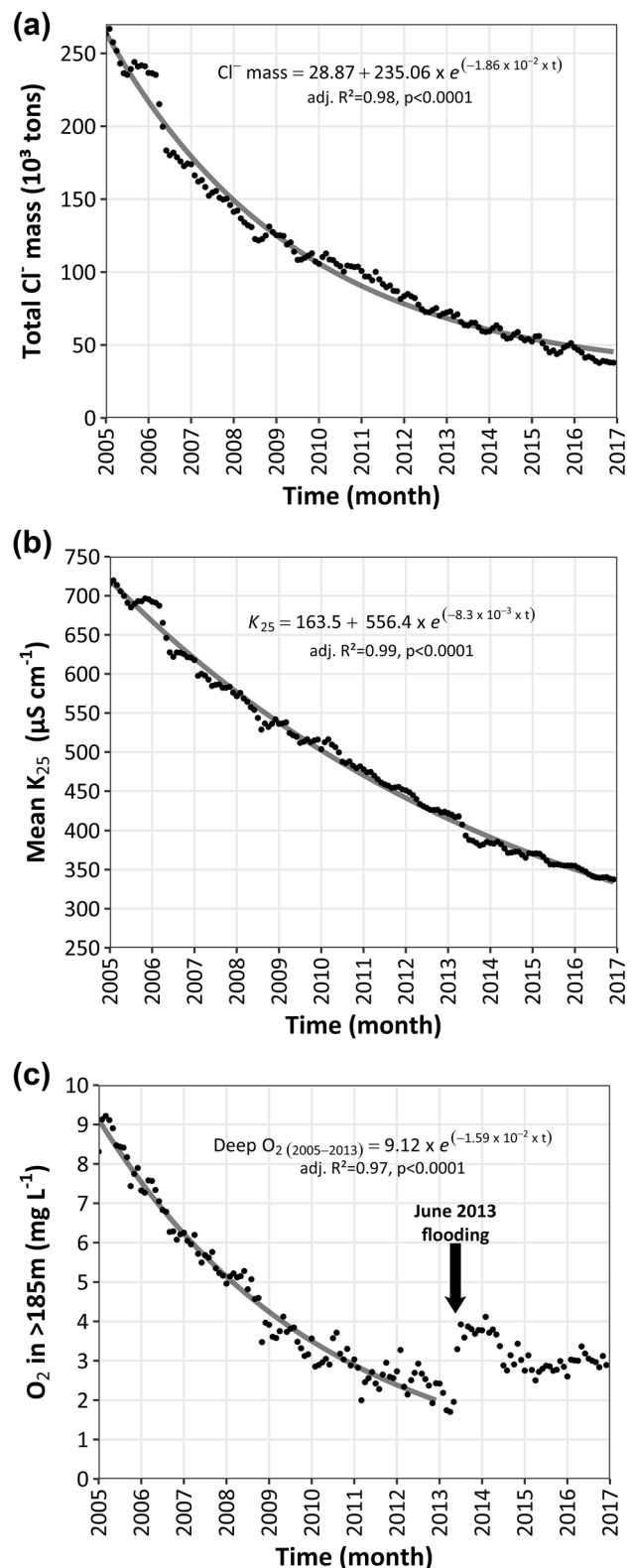


Fig. 5 Monthly mean chloride contents (a), mean conductivities in the water column (b) and deepwater oxygen concentrations (c) from January 2005 to December 2016 with fitted exponential reduction models. The arrow marks the appearance of a huge flooding event in June 2013 affecting deepwater oxygen concentrations

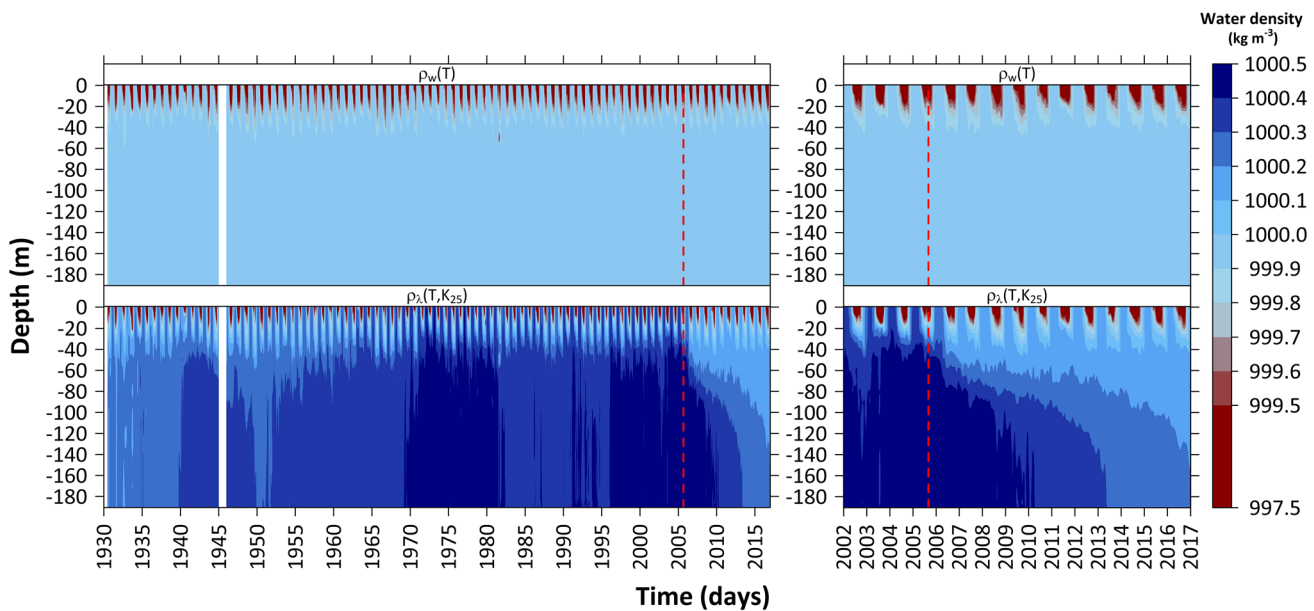


Fig. 6 Calculated water densities derived from daily temperature data ($\rho_w(T)$, upper panel) and from daily temperature and conductivity ($\rho_\lambda(T, K_{25^\circ})$, lower panel)

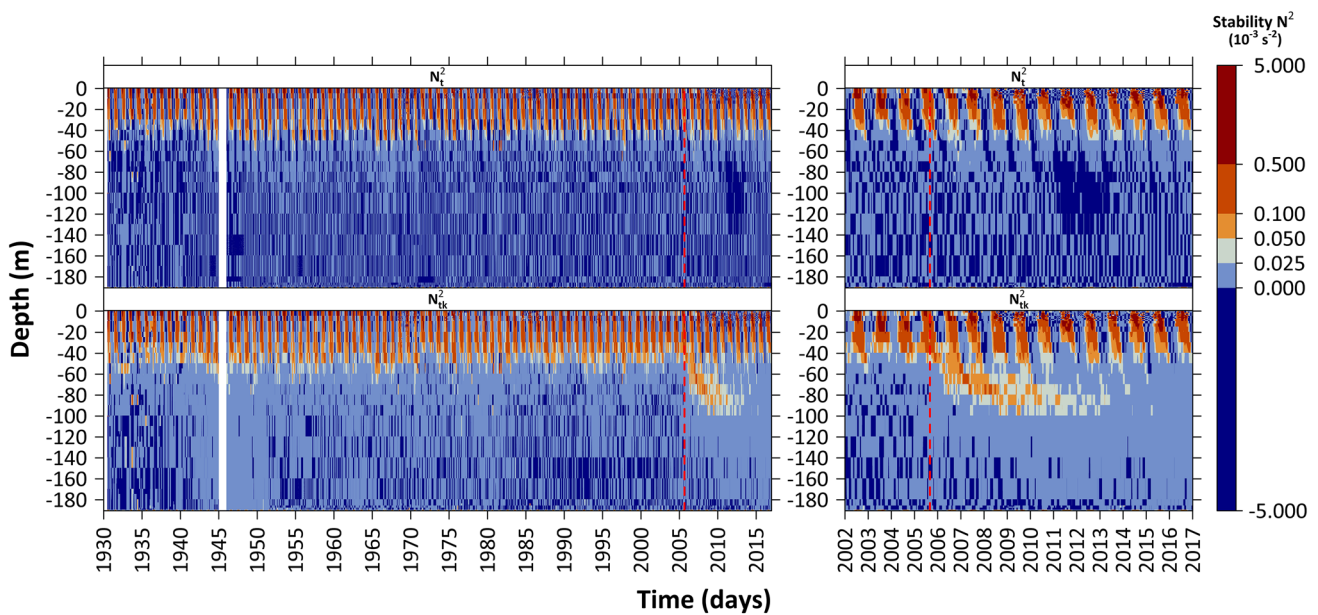


Fig. 7 Squared buoyancy frequencies N_t^2 and N_k^2 derived from Traunsee water densities based on temperatures $\rho_w(T)$ and on temperatures and conductivities $\rho_\lambda(T, K_{25^\circ})$ from 1930 to 2016. Dark blue areas illustrate $N^2 < 0$ and indicate convection of the specific water

layer, whereas $N^2 > 0$ indicate stable conditions and stratified water layers with increasing intensity. Vertical dashed lines mark the shut-down of the soda work in 2005

the upper water column that reinforced the density gradient and decoupled vertical circulation of Traunsee. Also the comparison of annual mean stability indices S_{tk} and S_t underline a severe impact of salt pollution on stratification intensity of Traunsee (Fig. 8a). Annual mean S_{tk} values were on average 60% higher than S_t over the last 87 years

and more than twice as high as S_t during the recovery from salinization and the salt-induced meromixis. During the cold season, annual minimum values of S_t were close to or below 0 J m^{-2} , whereas minimum S_{tk} remained always higher than 200 J m^{-2} (Fig. 8b). We detected exceptionally high annual

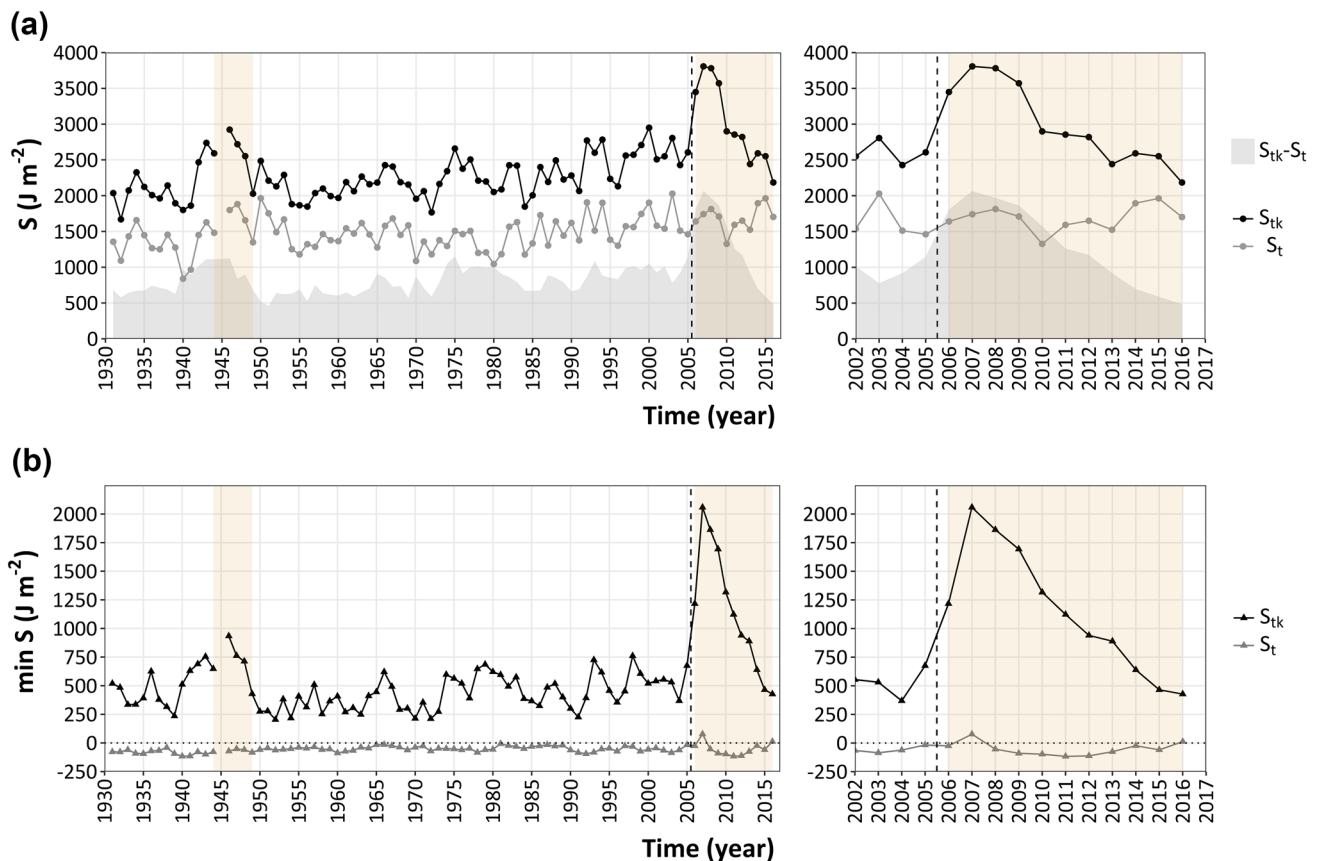


Fig. 8 Development of **a** annual mean Schmidt stabilities and **b** annual minimum stabilities derived from temperature data (S_t) and temperature and conductivity data (S_{tk}). The grey area in **a** shows differences between calculated stabilities ($S_{tk}-S_t$) and illustrate the con-

tribution of chloride pollution to stratification indices of Traunsee. The brown shaded areas indicate the two meromictic periods with continuous oxygen depletion in the hypolimnion (see also Fig. 3c). Vertical dashed lines mark the shutdown of the soda work in 2005

mean and minimum Schmidt stabilities at the beginning of the two meromictic periods (Fig. 8a, b).

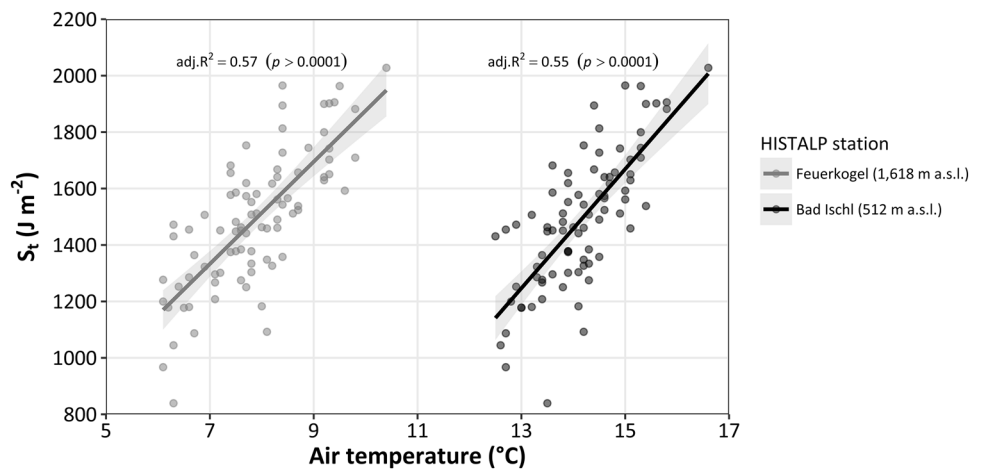
In general, the annual course of water densities and all computed stability indices followed a seasonal pattern with maximum values between August and October and minimum values between December and March (Figs. 7, 8; see also Fig. 14 in Appendix). When neglecting the effect of salt pollution, seasonal stratifications were obviously controlled by the temperature gradient in Traunsee. We found that warmer summers were related to higher mean stability S_t of Traunsee indicated by positive linear regressions between mean annual S_t and mean air temperatures measured at two locations near Traunsee (Fig. 9). Statistical correlation analyses of S_t and mean air temperatures between April and September resulted in Pearson's $r=0.74$, $p<0.0001$ for the Bad Ischl station and $r=0.76$, $p<0.0001$ for Feuerkogel station.

Discussion

Our analyses of almost nine decades of Traunsee sampling data exemplifies that pollution with chloride salts can severely affect density stratification and mixing dynamics of a temperate freshwater lake. Nonetheless, the constant input of very large amounts of salts into Traunsee did not cause a long-lasting meromixis with anoxic conditions in the hypolimnion. This was very likely due to the high inflow of the main tributary, the short theoretical water renewal of the lake and several technical adaptations in the disposal of saline emissions. Most notably, the recent recovery from salinization of Traunsee had a more serious impact on stratification and deepwater oxygen depletion than the input of large quantities of dissolved salts over the last century.

The Traunsee data confirmed that a faster wash-out of dissolved salts from the epilimnion can strongly reinforce a lake's density gradient and induce meromixis during the recovery from salinization in a seasonal stratified lake. After industrial pollution ceased in summer 2005, Traunsee quickly became meromictic and dissolved oxygen in the

Fig. 9 Positive correlations between mean air temperatures from April to September and mean stabilities (S_t) of Traunsee derived from water temperatures only



hypolimnion substantially decreased in the following years. A remarkable abrupt increase in deepwater oxygen concentrations gave the impression that the salt-induced stratification disappeared in 2013. However, the replenishment of hypolimnic oxygen was coincident with a huge flooding event of the river Traun in June 2013 (Blöschl et al. 2013; see Fig. 5c). The flood probably caused plunging turbid water that carried oxygen into the hypolimnion although meromixis still remained.

A period of recovery from salinization, indicated by declining total chloride masses in Traunsee, occurred also in the 1970s. However, the ongoing input of industrial saline waste into the epilimnion compensated the development of a saline gradient by the wash-out process and prevented Traunsee from becoming meromictic (Sossau and Pechlaner 1988). The stronger oxygen depletion in the hypolimnion between 1975 and 1980 was more likely the result of eutrophication and not due to salt-induced meromixis (Jagsch et al. 2002).

The only other meromixis that we observed in Traunsee apart from the meromixis in recent years happened in the 1940s. In contrast to the recent situation, the persistent stratification between 1944 and 1949 was caused directly by the disposal of saline waste and not by the wash-out process of dissolved salts. Saline emissions had densities between 1106 and 1116 $kg m^{-3}$ and were released below the epilimnion at a depth of 30 m into Traunsee during the 1930s and early 1940s (Sossau 1982). Because of the deep discharge and the high density of effluents, dissolved saline wastes accumulated in the deeper water and provoked stratification and meromixis. Several technical modifications of the disposal facility were carried out between 1941 and 1948 to counteract the salt-induced stratification (Sossau 1982). The outflow of dissolved waste was modified and repeatedly lifted into the shallow water of the epilimnion. Furthermore, the installation of mixing pumps on the floating pier led to a fourfold dilution of saline emissions with lake water and reduced

densities of effluents to 1023 $kg m^{-3}$ before they entered the lake. These adaptations helped to erode the salt-induced stratification and re-established complete vertical mixing of the lake in the early 1950s (Sossau and Pechlaner 1988). Such modifications of the disposal facility also helped to avoid meromictic conditions during the late 1960s and early 1970s, even though they could not prevent the accumulation of salts in the lake (Sossau and Pechlaner 1988). Only the reduction in the disposal of salts was effective to lower the total chloride mass in Traunsee.

The observed exponential reduction in chloride masses represents a typical development of a lake's salt content after the input of salt has been stopped (e.g. Effler et al. 1989; Doerr et al. 1994). Moreover, the reduction in chloride masses is comparable to the recovery from salt water intrusion in Hallstätter See, a neighbouring lake situated in the catchment of Traunsee (Ficker et al. 2011). Hallstätter See was contaminated by brine spills from salt mining that induced meromixis, analogous to the situation of Traunsee. Chloride reduction models of both lakes have similar exponential constants with $R=0.18$ $year^{-1}$ of Hallstätter See in Ficker et al. (2011) compared to $R=0.16$ $year^{-1}$ of Traunsee in this study (i.e. derived from monthly R in figure). The slightly higher reduction rate of Hallstätter See may be due to the shorter theoretical water renewal of 0.5 years and smaller maximum depth of 125 m compared to about 1 year, respectively, 191 m in Traunsee.

Declining chloride loads generally behave like hydrological tracers that are used to evaluate the actual water residence time (e.g. Cole and Pace 1998; Thies et al. 2002; Rimmer et al. 2005; Pilotti et al. 2014). In the case of Traunsee, the effects of salinization on water densities limit the use of chloride as an inert tracer of the lake's hydrology. Nevertheless, the overall chloride reduction in Traunsee reflected the actual water renewal time of the lake that was obviously prolonged by the reinforced density gradient during the recovery from salinization. According to the exponential

decline in chloride loads of Traunsee and to the derivations of water renewal time described in Pilotti et al. (2014) and Thies et al. (2002) the complete water exchange took about 6 years after chloride pollution stopped. The actual water renewal of Traunsee was therefore six times longer than the theoretical water renewal estimated by the ratio of lake volume to tributary discharge. Taking into account the recent development of chloride concentrations, conductivities, densities gradients and stability indices, a complete vertical mixing of Traunsee and oxygenation of the hypolimnion can be expected in the next few years.

Comparable datasets of temperate freshwater lakes that tracked severe salt pollution and subsequent recovery from salinization are rare. Dugan et al. (2017b) recently assembled a global database on long-term lake chloride concentrations but none of the 529 lake datasets comprised such long time-series and high temporal resolution as the Traunsee sampling data. A noteworthy example that can be related to the Traunsee case besides Hallstätter See is the salinization of Onondaga Lake in central New York in the United States (Effler and Matthews 2003). Onondaga Lake's density stratification was also considerably impacted by external loading with saline emissions from soda ash production (Effler and Driscoll 1986; Effler and Owens 1996). In contrast to the Traunsee case, Onondaga Lake did not become meromictic after the closure of the soda work. This was maybe due to the shallower character of the lake with a maximum depth of only 20 m.

As our study revealed, the downside of reduced or stopped salt pollution is the deterioration of seasonal mixing conditions and the depletion of oxygen in the hypolimnion in deep temperate lakes. Similar effects, albeit to a lesser extent, can arise from climate warming (e.g. Sahoo et al. 2015; Schwefel et al. 2016; Ficker et al. 2017). Longer persistence of thermal summer stratification due to higher water temperatures could additionally intensify the wash-out of dissolved salts from the epilimnion and reduce the upward mixing and wash-out from the hypolimnion. This would consequently lead to a slower recovery from salt pollution. Salinization and climate change might therefore have synergistic effects that weaken vertical mixing dynamics and seasonal replenishment of deepwater oxygen.

In addition to the salt-induced changes in mixing and oxygen conditions, chronic exposure to high chloride concentrations are suspected to cause negative effects on aquatic life. Chronic thresholds for aquatic life have been rarely proposed so far. According to EPA (1988) chronic effects on freshwater organisms could appear at chloride concentrations $> 230 \text{ mg L}^{-1}$. More recently, the Canadian Water Quality Guidelines suggested that long-term exposure to concentration $> 120 \text{ mg L}^{-1}$ chloride can impair aquatic life of freshwater lakes (CCME 2011). Chloride concentrations $> 120 \text{ mg L}^{-1}$ occurred in Traunsee in the 1970s, 1990s

and 2000s. However, negative effects of dissolved salts on aquatic life of Traunsee have not been detected, whereas the disposal of calcium-rich solid wastes clearly affected benthic organisms (e.g. Löffler 1983; Griebler et al. 2002; Sonntag et al. 2002; Wanzenböck et al. 2002). Besides a direct impact of industrial wastes, the appearance of meromixis had probably indirect negative effects on aquatic life of Traunsee. The development of hypoxia in the deepwater perhaps reduced the available habitat for fish and invertebrates in the hypolimnion (e.g. Zadereev et al. 2017).

Conclusions

The Traunsee case clearly shows that the recovery from long-term salinization can deteriorate vertical mixing and hypolimnic oxygen concentrations of temperate freshwater lakes. This effect may be more serious in lakes with long water retention times leading to a slow rehabilitation of the deepwater from salinization and the threat of long lasting meromixis. Warming surface temperatures due to climatic changes could exacerbate this situation. Mitigation of salinization effects on mixing dynamics can be achieved by reducing densities of saline effluents, improving their dispersal in the epilimnion and avoid the release of saline emissions into the deepwater. However, to prevent the accumulation of chloride salts in lakes only the reduction of saline emissions is effective. Even though salt pollution from a point-source is a rare situation of salinization, this study exemplifies that even low saline gradients can alter density stratifications and mixing of freshwater lakes. Hence, the prevalent and increasing salinization of many lakes by non-point salt pollution, such as road salts, fertilizers and wastewaters, could cause similar consequences.

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Appendix

Temporal and spatial dimensions of sampling profiles

The temporal and spatial resolution of sampling increased steadily over the first ten years of the monitoring program conducted from 1930 to 2016. The long-term records of water temperature, conductivity and chloride concentration predominantly comprise monthly vertical sampling profiles in at least 17 depth layers between surface and lake bottom from the 1950s onwards.

See Figs. 10, 11, 12 and 13.

Fig. 10 **a** Number of temperature profiles per year and **b** mean number of individual temperature samples per profile. Error bars indicate varying numbers of samples in the profiles during a year

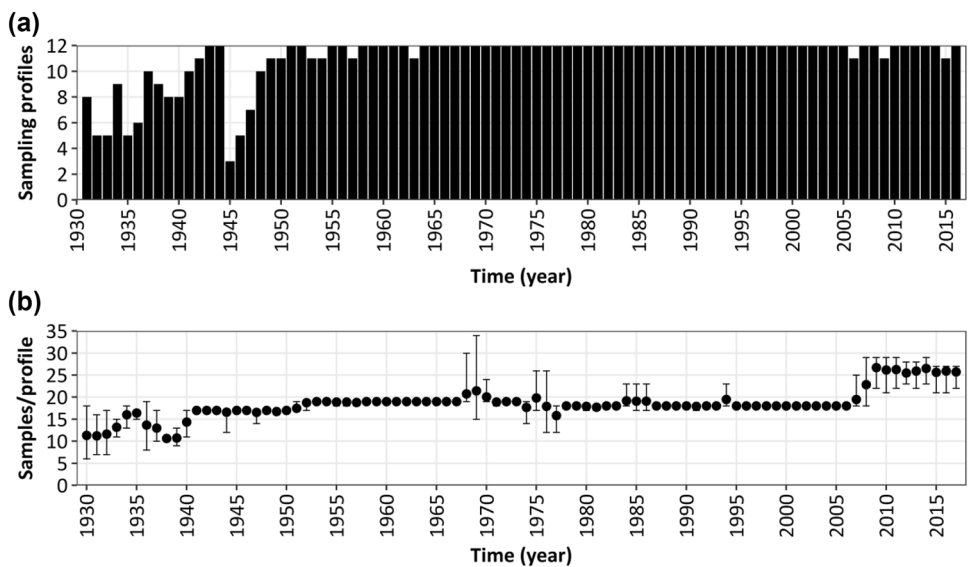


Fig. 11 **a** Number of conductivity profiles per year and **b** mean number of individual conductivity samples per profile. Error bars indicate varying numbers of samples in the profiles during a year

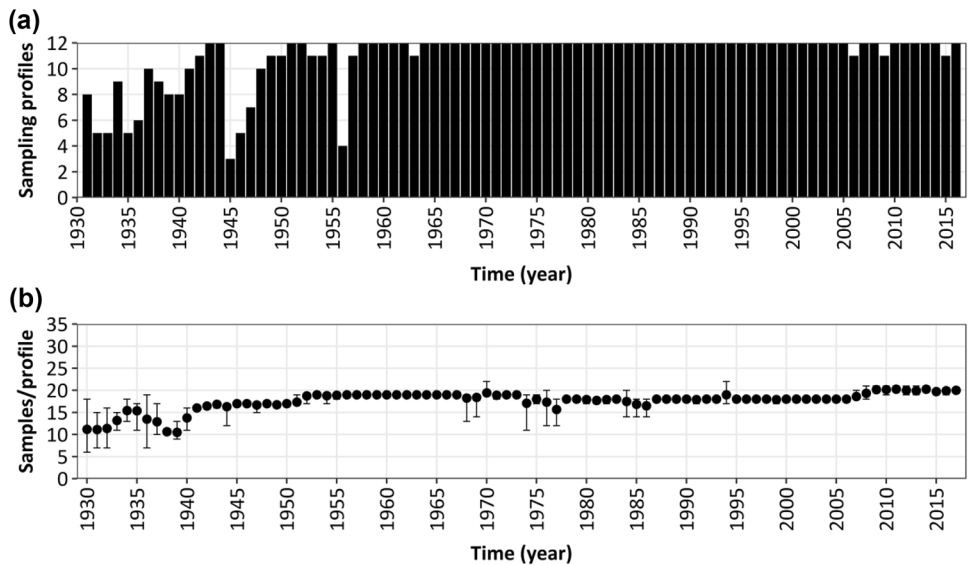


Fig. 12 **a** Number of chloride profiles per year and **b** mean number of individual chloride-samples per profile. Error bars indicate varying numbers of samples in the profiles during a year

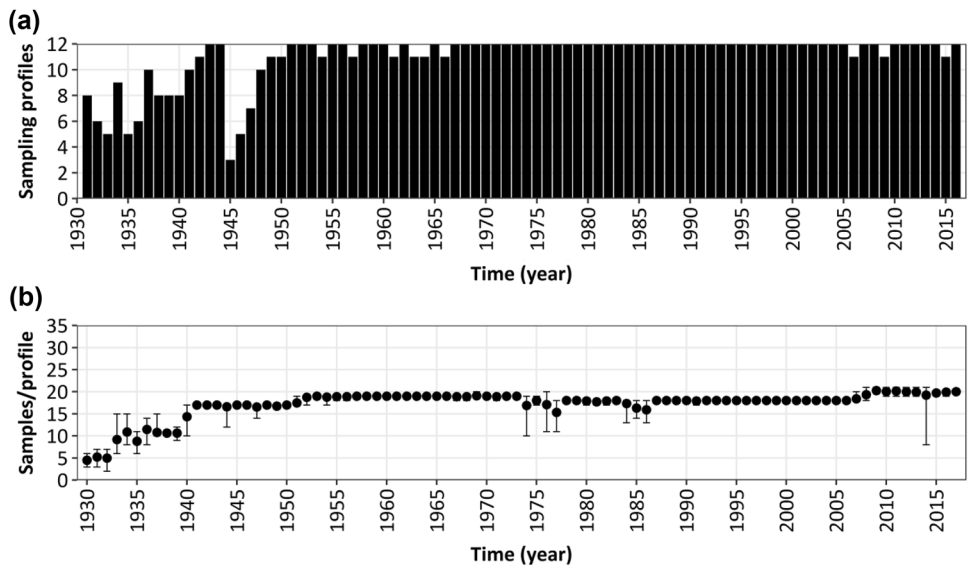
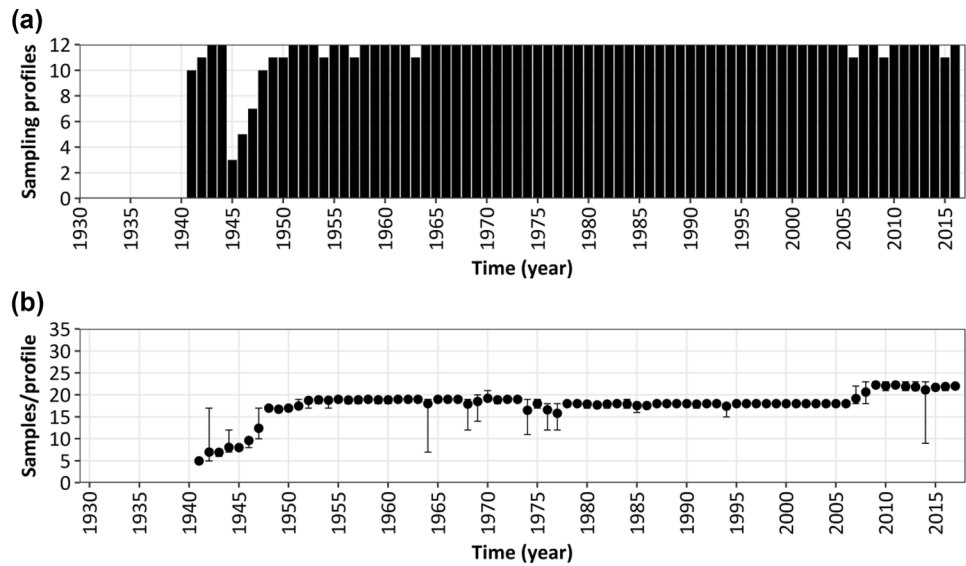


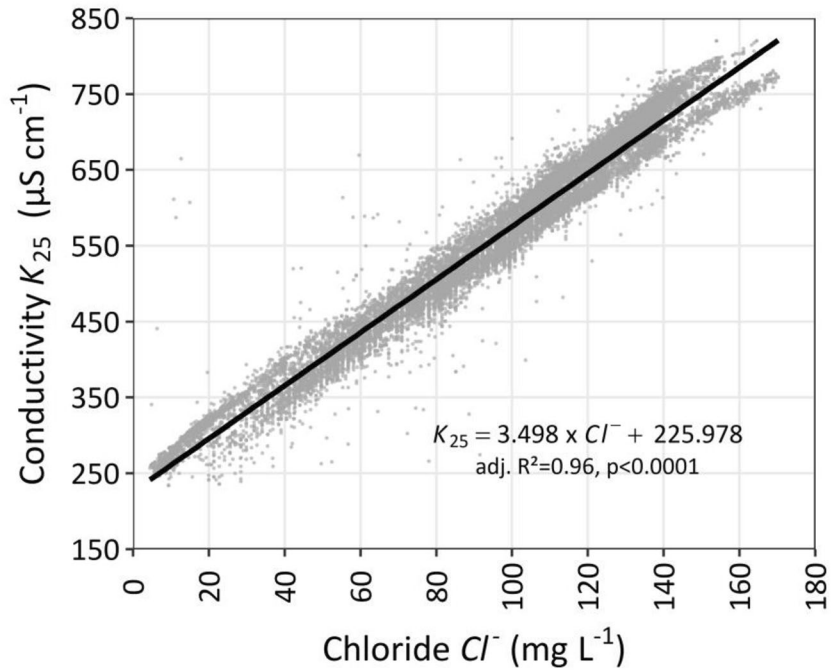
Fig. 13 **a** Number of oxygen profiles per year and **b** mean number of individual oxygen-samples per profile. Error bars indicate varying numbers of samples in the profiles during a year



Linear regression of chloride vs. conductivity measurements

See Fig. 14.

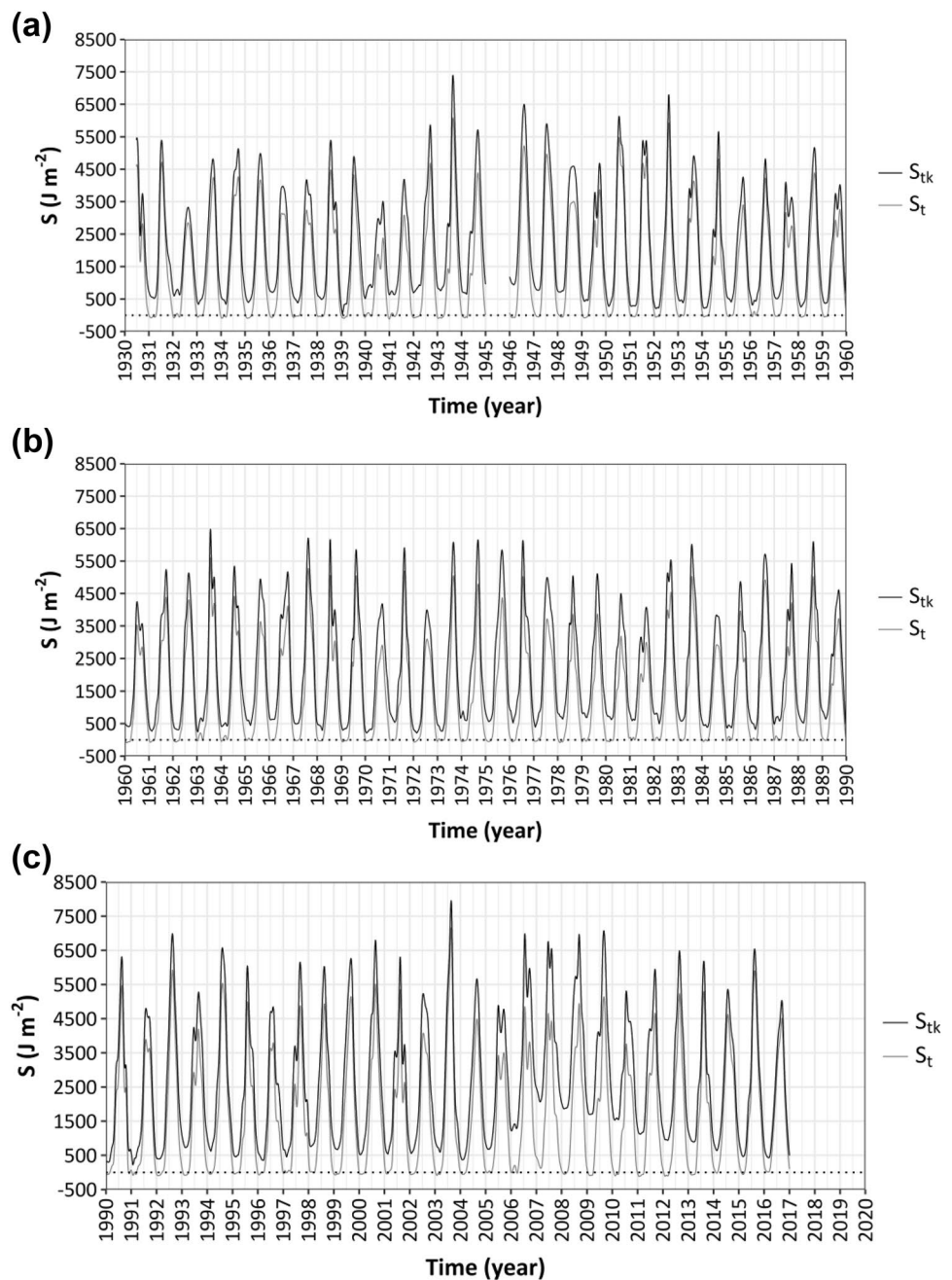
Fig. 14 Linear relationship between measured chloride concentrations and measured electrical conductivities in Traunsee between June 1930 and December 2016



Development of daily stability indices of Traunsee between 1930 and 2016

See Fig. 15.

Fig. 15 Daily time-series of Schmidt stability indices calculated with temperature data (S_t) and temperature and conductivity data (S_{tk}) from **a** June 1930 to December 1959, **b** January 1960 to December 1989 and **c** January 1990 to December 2016



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