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Ultrasonic in situ measurements of density, adiabatic compressibility, and stability frequency

Abstract—An in situ density profile has been measured with an ultrasonic density probe in the mining lake Merseburg-Ost 1b. From the acquired measurements of sound speed and acoustic impedance, the important physical properties of in situ density and adiabatic compressibility could be calculated. It was also shown that these two properties sufficed to determine the stability frequency, which hence becomes a directly observable physical magnitude. Measurements of all magnitudes are presented. Currently the accuracy only suffices to provide new insight in the density structure of natural water bodies in cases of unusual composition of dissolved substances. However, as all required data are acquired in situ and density is measured directly, i.e., without need of empirical

formulations (“equations of state”), the ultrasonic approach has the potential to become a reliable reference for density measurements in limnic waters, when chemical conditions are complex or spatial or temporal variation of dissolved substances becomes relevant for the stability of the water column.

Owing to the variable composition of the dissolved substances, density measurements in inland waters are still hard to acquire at the required accuracy (Schimmele and Herzprung 2000). The oceanographic approach of calibrating temperature and electrical conductivity (via salinity) against

the potential density (UNESCO, Fofonoff and Millard 1983) claims an accuracy in the order of 10^{-6} for the open ocean; however, even for adjacent seas, e.g., the Baltic, this accuracy cannot be gained with the present approach.

The common procedure results in a separate calibration curve for fresh water (e.g., Bühner and Ambühl 1975; Chen and Millero 1986), which is modified for any closely investigated lake (for Lake Constance: Heinz 1990; Bäuerle et al. 1998). Additional density related physical properties can be included in the calibration curve for density measurements. The reference densities are commonly produced in the laboratory by frequency measurements of an oscillating pipe loop filled with the investigated water sample.

While for lakes with low salt content and small local and temporal changes of the composition of dissolved substances this is a viable way, for those lakes with a high concentration of dissolved substances and high gradients in the salt composition due to meromixis or strong groundwater inflows, this approach often can only provide a solution of restricted temporal validity (Gräfe 2000; Gräfe and Boehrer 2001). The accuracy that can be achieved under complex chemical conditions may be limited to 10^{-4} .

Especially in mining lakes, where dissolved substances show high variability in the vertical, e.g., due to meromixis (e.g., Boehrer 1998; Heidenreich et al. 1999; Boehrer et al. 1998, 2000a,b), a more direct measurement of density is required. On top of this, there are lakes with a considerable content of nonionic dissolved substances (e.g., Lake Vollert-Süd, Stottmeister et al. 1998, p. 113–121), which cannot be verified by measurements of electrical conductivity.

In addition to the above difficult field measurements, reference densities inherit a problem of reliability. Some lakes show a high concentration of dissolved gases, which can escape when pressure is relaxed (e.g., von Rohden 1998; von Rohden and Ilmberger 2001). Obviously, the contribution of those gases to water density is lost, but in addition some of those gases are pH relevant (e.g., CO_2 , H_2S), and the chemical conditions change with all their consequences for other dissolved substances before a good measurement of density can be attained. Additional difficulties, such as contact with oxygen when injecting anoxic water samples into the density measurement equipment, are also a major concern.

A direct measurement of density in situ would not inherit any of the above problems. However, until recently there was no sensor available that was suited for direct measurements, that worked at a required accuracy, and that was robust enough for field work. A recently developed ultrasonic sensor, capable of measuring sound speed and acoustic impedance, may be suited for in situ measurements in natural water bodies. The measurements of sound speed and acoustic impedance result not only in a direct measurement of density but also—until now a by-product—in direct measurements of adiabatic compressibility. The measurements of both physical properties allow a direct in situ measurement of the stability frequency, which commonly is regarded as the gradient of the potential density. This, on the other hand, might even be interesting for freshwater lake application, as adiabatic compressibility can be a crucial magnitude (Crawford and Collier 1997; Ravens et al. 2000; Wüest et al. 2000) for

deep water renewal at great depth, as well as for the open ocean, if the accuracy is good enough.

As a consequence, such a sensor originally designed for industrial application (Püttmer 1999; Püttmer et al. 2000) was attached to a pressure-resistant housing (Hoppe et al. 1999) to test the measuring principle and arrangement for measurements in lakes. For the first measurements, a lake with high vertical density gradients was chosen to confirm the proper function at the present accuracy.

Theoretical background—Stability: We consider a mass m of liquid that fills a (variable) volume V , and thus we define its in situ density as

$$\rho_l \equiv \frac{m}{V} \quad (1)$$

With the derivative $d\rho_l/dV = -m/V^2 = -\rho_l/V$, we can replace the volume dependence in the definition of adiabatic compressibility (compression by changing pressure p without heat exchange, i.e., $\delta Q = \text{const.}$) by a density dependence

$$\kappa \equiv -\frac{1}{V} \left(\frac{\partial V}{\partial p} \right)_{\delta Q} = -\frac{1}{V} \frac{1}{d\rho_l/dV} \left(\frac{\partial \rho_l}{\partial p} \right)_{\delta Q} = \frac{1}{\rho_l} \left(\frac{\partial \rho_l}{\partial p} \right)_{\delta Q} \quad (2)$$

For considerations of stability of a stratified water column, it is convenient to eliminate the pressure-dependent contribution from the total (=in situ) density gradient $\partial\rho_l/\partial z$ to gain the magnitude for the stability

$$(\partial\rho_l/\partial z)_p \equiv d\rho/dz \quad (3)$$

commonly referred to as potential density gradient. We are looking at processes (e.g., turbulent vertical exchange and waves) fast enough not to be affected by heat transport. In those cases, vertical transport, i.e., pressure-dependent change of density, can be considered to be adiabatic.

$$\frac{d\rho_l}{dz} = \left(\frac{\partial \rho_l}{\partial z} \right)_p + \left(\frac{\partial \rho_l}{\partial p} \right)_{\delta Q} \frac{dp}{dz} = \frac{d\rho}{dz} + \kappa \rho_l \frac{dp}{dz} \quad (4)$$

The change of (hydrostatic-pressure with depth results from the gravity g that acts on the mass of density ρ_l that lies above $dp/dz = g\rho_l$.

$$\frac{d\rho_l}{dz} = \frac{d\rho}{dz} + g\kappa\rho_l^2 \quad (5)$$

Thus the calculation of the stability becomes

$$N^2 \equiv \frac{g}{\rho_l} \frac{d\rho}{dz} = \frac{g}{\rho_l} \frac{d\rho_l}{dz} - \kappa g^2 \rho_l = g^2 \left(\frac{d\rho_l}{dp} - \kappa \rho_l \right) \quad (6)$$

Since most probes measure depth by acquiring the pressure, we have replaced the derivative after depth with the derivative after pressure. In conclusion, an instrument capable of measuring in situ density and adiabatic compressibility (in situ) can directly measure the stratification or potential density gradient.

Buoyancy frequency: On many occasions, listings for the potential density are readily available, but this might not be the case for the in situ density. Hence, in the aquatic field, it is commonly neglected that the denominator in Eq. 6 is

not the potential density but in situ density ρ_i , as shown in the following.

A fluid parcel of in situ density $\hat{\rho}$ experiences an acceleration from the surrounding in situ density field ρ_i .

$$\hat{\rho} \frac{d^2 z}{dt^2} = g(\rho_i - \hat{\rho}) \quad (7)$$

We consider infinitely small excursions of the fluid parcel, i.e., inclusion of first-order derivatives of the density field only, about its depth of neutral buoyancy z_0 . Hence $\hat{\rho} \approx \rho_i$.

$$\rho_i \frac{d^2(z - z_0)}{dt^2} = g \left(\frac{d(\rho_i - \hat{\rho})}{dz} \right)_p (z - z_0) \quad (8)$$

In the case of the fluid parcel, the derivative after z is solely due to the (hydrostatic) pressure, and thus $(d\hat{\rho}/dz)_p = 0$. By using the above definition of $d\rho/dz$ (Eq. 3), we find a differential equation

$$\rho_i \frac{d^2(z - z_0)}{dt^2} = g \frac{d\rho}{dz} (z - z_0) \quad (9)$$

which is solved by harmonic oscillations of z at the buoyancy frequency

$$N = \sqrt{\frac{g}{\rho_i} \frac{d\rho}{dz}} = \frac{2\pi}{T} \quad (10)$$

where T represents the period.

Speed of sound: The derivation of sound speed can be found in textbooks about mechanics (e.g., Gerthsen et al. 1982). For completeness, we included it here. In a compressible fluid, a pressure change dp results in a change of volume of a respective fluid parcel:

$$dV = -\kappa V dp \quad (11)$$

If the involved processes are fast enough, heat transport is not important and κ represents the adiabatic compressibility. On the other hand, a longitudinal compression is observed if the fluid shows gradients in the velocity dv/dx :

$$dV = V \frac{dv}{dx} dx \quad (12)$$

Replacing dV results in

$$\left(\frac{dp}{dt} \right)_{\delta Q} = -\frac{1}{\kappa} \frac{dv}{dx} \quad (13)$$

The fundamental equation connecting acceleration to a force acting on a fluid parcel reads

$$\frac{dv}{dt} = -\frac{1}{\rho_i} \frac{dp}{dx} \quad (14)$$

Now cross-differentiation of Eqs. 13 and 14 results in a wave equation for a pressure wave:

$$\left(\frac{d^2 p}{dt^2} \right)_{\delta Q} - \frac{1}{\kappa \rho_i} \left(\frac{d^2 p}{dx^2} \right)_{\delta Q} = 0 \quad (15)$$

This equation is solved by harmonic pressure waves in the liquid that travel at a speed

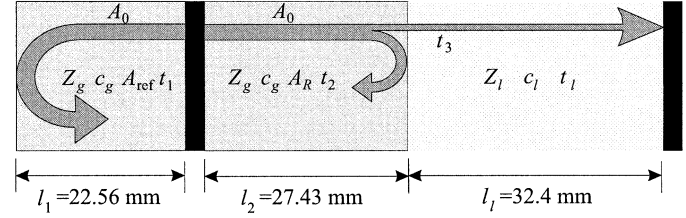


Fig. 1. Sketch of the sound paths through the ultrasonic sensor.

$$c_l^2 = \frac{1}{\kappa \rho_l} \quad (16)$$

As a conclusion, the last term of Eq. 6 can be determined from a sound speed measurement.

Transmission and reflection at an interface: To yield information about the physical properties of a liquid, we consider a wave that traverses a glass before hitting an interface to the liquid (see also Sutilov 1984; Püttmer 1999). The pressure wave will be partly transmitted into the liquid and be partly reflected back into the glass as sketched in Fig. 1.

The velocity at the interface is both the boundary condition of the propagating wave at the liquid side and the interference of incident and reflected wave in the glass:

$$v_l = v_g + v'_g \quad (17)$$

An index g refers to properties in the glass, an index l to properties in the liquid, and the prime indicates the reflected wave.

In addition, the kinetic energy of the wave has to be conserved, i.e., the energy flux of the incident wave must correspond with the energy flux of the emitted waves:

$$\frac{1}{2} \rho_g c_g v_g^2 = \frac{1}{2} \rho_g c_g v_g'^2 + \frac{1}{2} \rho_l c_l v_l^2 \quad (18)$$

As easily confirmed by inserting it above in Eqs. 17 and 18, the equation system for transmission and reflection of a pressure wave is solved by

$$\frac{v_l}{v_g} = \frac{2Z_g}{Z_g + Z_l} = T \quad (19)$$

$$\frac{v_g'}{v_g} = \frac{Z_g - Z_l}{Z_g + Z_l} = R \quad (20)$$

where we implemented the definition of acoustic impedance of the liquid (and correspondingly for glass) as:

$$Z_l = c_l \rho_l \quad (21)$$

$$Z_g = c_g \rho_g \quad (22)$$

T and R are called transmission or reflection coefficient, respectively. The reflection coefficient connects the acoustic impedance of the glass and the liquid (straight from Eq. 20) as

$$Z_l = \frac{1 + R}{1 - R} Z_g \quad (23)$$

Equipment—The sensor consisted of a piezo ceramic glued onto a quartz glass cylinder (see Fig. 1), through which the ultrasonic pulse of a frequency of 2 MHz was transmitted. The sound wave first traversed the quartz glass. Then at the interface to the investigated liquid, the sound wave was partially reflected and partially transmitted into the surrounding liquid, where the signal was recognized after a certain time (t_i) at a sensor a certain travel distance l_i away from the interface. The sound speed simply was evaluated from

$$c_i = l_i/t_i \quad (24)$$

While the time measurement does not pose any problems at the present requirements of accuracy, the determination of the l_i turned out to be more difficult and could only be achieved with calibration measurements using known liquids. In the end, we were confident to measure sound speed with an accuracy better than 10^{-4} . Hence the sound speed measurement does not noticeably affect the accuracy of the density measurement (see below).

For a measurement of the reflection coefficient R (Eq. 20), a second quartz glass cylinder was attached to the piezo ceramic. On this side of the sensor, the sound wave hit a glass–air interface, and thus was basically entirely reflected into the glass. The reflection coefficient was evaluated from the signal amplitudes $R = A_R/A_o$ at the piezo ceramic from both glass cylinders. A calibration measurement with air on both sides evaluated the asymmetry of the system.

The error involved in the measurement of the amplitude ratio R turned out to be the crucial point for the accuracy of the density measurements. In addition, $R \approx 0.75$ appears in the sum $1 - R$ of the denominator in Eq. 23 and thus is even amplified in its effect and limits the measurement of acoustic impedance to an accuracy of 10^{-3} .

With the reflection coefficient R , the ratio between acoustic impedance of the glass and the liquid is known (Eq. 23). A second calibration in a known liquid, e.g., aqua and NaCl solution, allowed measurement of the properties of the quartz glass: density $\rho_g = 2231.2 \text{ kg m}^{-3}$ and sound speed $c_g = 5888 \text{ m s}^{-1}$. The close investigation confirmed a temperature dependence. For this purpose a temperature sensor was included in the probe, and a slow temporal variation made an on-site calibration and the inclusion of temperature dependent calibration values inevitable. A detailed description of the procedure was included in Gräfe (2000).

The interesting physical properties of in situ density (Eq. 22) and adiabatic compressibility (Eq. 16) could be evaluated straightforwardly. The accuracy for both measurements was in the order of 10^{-3} . Empirically we found a response time of 150 s for the acoustic parameters, in the case of varying temperature, which complied with our anticipation for heat conduction through the quartz glass.

Results of in situ measurements—In situ measurements were conducted in the highly salinity stratified mining lake of Merseburg-Ost 1b in Central Germany. The density probe was attached to a probe for the measurement of electrical conductivity, temperature, and depth (CTD probe; Idronaut), so the density profile could be correlated to depth with the help of time and temperature. The range of observed tem-

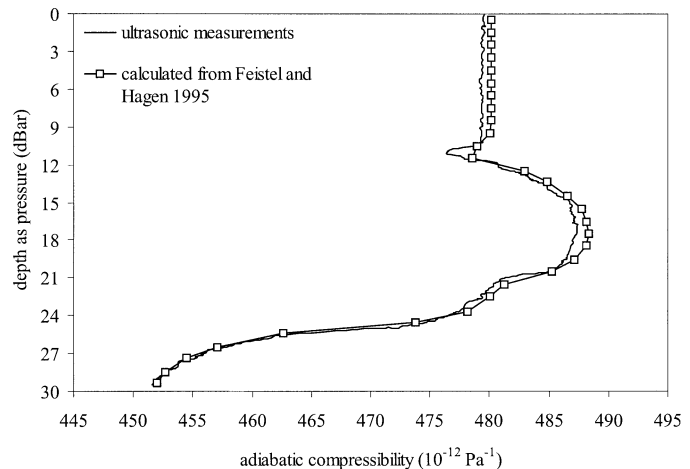


Fig. 2. Adiabatic compressibility in Lake Merseburg-Ost 1b against the pressure (depth) from ultrasonic data and calculated from the tables of Feistel and Hagen (1995) using the UNESCO-formula (Fofonoff and Millard 1983) for calculating salinity from electrical conductivity and temperature.

peratures was small (4.5 to 9°C). The location was chosen because the salinity varied between 5 and 25‰ and was dominated by NaCl. Under these conditions, density and salinity could be calculated the traditional way from electrical conductivity and temperature by application of the UNESCO formula (see Schimmele and Herzsprung 2000). Thus, a comparison of the present measurements could be related to tabulated values on the basis of measured temperature and calculated salinity (Feistel and Hagen 1995).

First we refer to the adiabatic compressibility evaluated from the measured data. They range from 4.5×10^{-10} to $4.9 \times 10^{-10} \text{ Pa}^{-1}$ (see Fig. 2). The values can be compared to the list in Feistel and Hagen (1995), on the basis of salinity calculated from measurements of electrical conductivity using the UNESCO formula. The agreement is within about 2×10^{-3} , though the range covers a 10% variation.

This is a very good agreement with literature data for in situ measurements of adiabatic compressibility. However, the evaluation of density from the same ultrasonic data (Z_i and c_p , see below) confirms an accuracy of about 10^{-3} . Equation 16 connects compressibility with density by a factor of c_i^2 . Sound speed (see above) can be acquired an order of magnitude more reliably. Hence, we would anticipate an accuracy of about 10^{-3} for the compressibility measurements. As a consequence, we were tempted to attribute part of the very consistent deviation between both approaches to the restricted validity of the UNESCO formula to the mixolimnion water of Merseburg-Ost 1b.

Figure 3 comprised the measurements of in situ density with the ultrasonic probe (jagged line), the calculated density from electrical conductivity and temperature by using the UNESCO formula, and three water samples collected at the respective depth and measured in the lab with the oscillator tube. The general step structure was confirmed by all techniques. The various approaches, however, differed by a factor of about 10^{-3} from each other.

The profiles of in situ density and adiabatic compressibil-

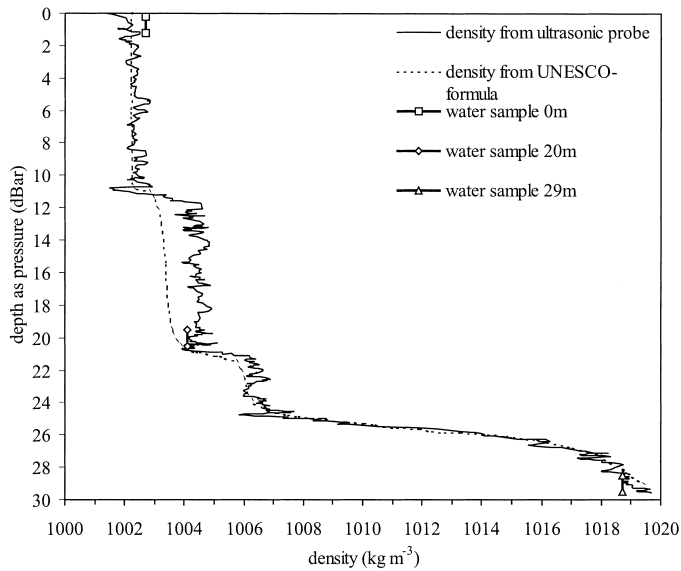


Fig. 3. Density versus pressure (depth): measured with the ultrasonic probe (solid, jagged line), calculated using the UNESCO-formula (Fofonoff and Millard 1983), and water samples measured at correct temperature under laboratory conditions with a high-accuracy oscillator tube instrument.

ity allowed the calculation of the stability frequency (*see* Eq. 6). Since the density measurements needed averaging before calculating differences, we could only provide one measurement per meter, as included in Fig. 4, based on in situ measurements only. The accuracy of N^2 measurement is limited to $2 \times 10^{-3} \text{ s}^{-2}$ by the accuracy of the in situ density measurements of $\pm 1 \text{ kg m}^{-3}$.

In conclusion, the measurement of in situ density and adiabatic compressibility with a submersible ultrasonic probe appears to be an interesting new approach to density and compressibility measurements in water bodies of the environment. The stability frequency can be measured directly in situ without the common assumption of the potential density concept, which is not an observable magnitude.

At present, the accuracy is of the order of 10^{-3} . As a consequence, new insight into the density structure of natural water bodies can only be provided in special cases where composition of dissolved substances is unknown or highly variable. For several reasons, the ultrasonic measurement of compressibility and density provides a promising way of investigating the density structure in natural water bodies: First, it is the only practicable and currently used way of a direct density measurement, since it does not require any empirical connection of acquired data and density, i.e., “equation of state.” Second, it provides the adiabatic compressibility as an independent measurement. Third, the method includes density contributions of nonionic dissolved substances.

Currently, implementation of the latest generation of analog-to-digital converters is being tackled, which could resolve the 2 MHz pressure wave pulse reasonably well in time. This will result in a much improved measurement of the reflection coefficient R (Eq. 20), which in the presented measurements is the limiting factor for the accuracy of the

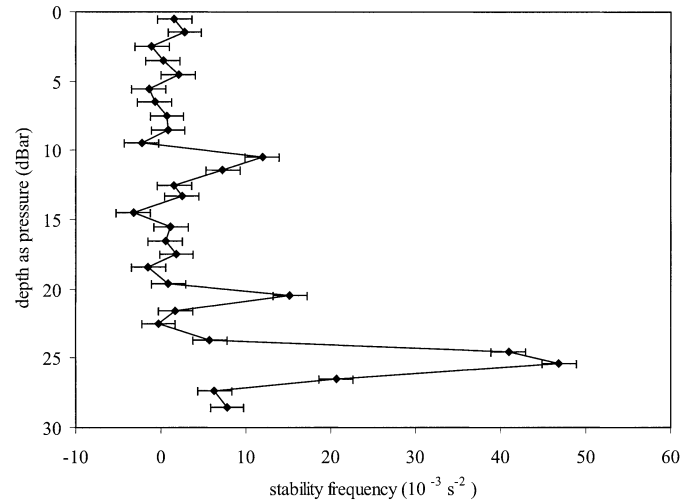


Fig. 4. Stability frequency (N^2) calculated from Eq. 6 using the direct ultrasonic measurements of compressibility and in situ density.

density data. Furthermore, we would like to point out the necessity for the development of the sensor from its current durable industrial design along limnophysical and oceanographic requirements to improve accuracy and time response, to proceed in the direction of a marketable product.

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