

The influence of the content of phosphates in water on the propagation speed of ultrasonic waves

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The phosphate content of a test sample is one of the indicators of the trophic status of the test water. In this work, an attempt was made to use a non-destructive ultrasonic technique to determine this parameter. For this purpose, a specially prepared measuring station was used to test distilled water samples with different phosphate contents. Specially prepared samples contained 0, 20, 40, 60, 80, and 100 kg/m³ of phosphates. In addition, tests were carried out on the effect of sample temperature on the values of the characteristic parameter of the wave, in the range from 12 to 30°C. All tests were carried out using two ultrasonic heads with a wave frequency of 2 MHz. The ultrasonic wave parameter analysed in the study was the propagation speed of the ultrasonic wave. The results obtained indicate that the ultrasonic method is useful for non-destructive evaluation of phosphate content in the sample. Additionally, they show a large influence of the sample temperature on the results read.

Key words: *non-destructive ultrasound technique, phosphates, temperature, ultrasonic wave, propagation speed*

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1. Introduction

Due to the increasing anthropopressure on the natural environment, their conditions are deteriorating due to the increasing pollution of the surrounding nature. This process applies to all aspects of the natural environment – air, land, water, and the broadly understood biosphere.

In the case of lakes, the influence of anthropopressure is particularly visible. Firstly, because of their relatively small dimensions – the concentration of pollutants is much higher than, for example, in the case of air or seas. Secondly, because of the very diverse purpose of lakes' use. They can be used as a breeding place, and a source of fish for fish farms as tourist attractions – a place for water sports, and their shores are attractive places for the construction of housing estates or hotels. Finally, lakes can serve as drinking water reservoirs. Of course, in practice, all these uses coexist to varying degrees.

The anthropopression exerted on lakes accelerates the natural process of eutrophication (aging of lakes). Excessive inflow of pollutants (first of all – nutrients – nitrogen and phosphorus) causes a number of unfavorable changes in water ecosystems [20]. These changes consist in a decrease in water transparency as a result of the intensification of primary production processes (excessive development of phytoplankton and also bacterioplankton – cyanobacteria), changes in water salinity (increase in electrolytic conductivity), changes in the biological structure of the ecosystem (decrease in biodiversity, disappearance of macrophytes, replacement of valuable fish species by the less valuable) [16, 20]. Excessive production of organic matter causes the consumption of oxygen for the processes of its decomposition, which takes place in the deeper layers of water and causes the development and strengthening of oxygen deficits in the bottom zone.

It may cause an increase in water pollution in lakes and, consequently, the loss of some of their qualities – a ban on bathing due to pollution, or a decline in the fish population.

Surface water pollution can be of biological, chemical or physical character. In particular, it may be contamination of water with bacteria, e.g., *E. coli*, the influx of toxic chemicals into the water, or pollution with heavy metals. Eutrophication is a slow increase in lake pollution caused by the nutrients export from the catchment area, increasing water fertility. The rate of this nutrient alimentation in natural conditions is slow, but human activity has significantly accelerated this process. In the first stage, this leads to the flourishing of life, and after some time – due to the consumption of oxygen resources in the lower layers of water – to a series of unfavorable changes mentioned above.

There are various methods of examining the state of water in terms of physical (temperature, pH, color, smell, Secchi disc visibility, electrolytic conductivity), chemical (determination of the content of various elements and chemical compounds by analytical chemistry methods) [18] or biological (e.g. analysis of species composition, number of organisms, biomass of organisms) [20]. Optical methods are used to measure the phytoplankton content in waters [3].

As a result of the research, it can be proved, that there is a specific relationship between a specific deviation of the parameters of the sound emitted in the water of the tested reservoir depending on its chemical and physical state.

It is known from physics that the speed of sound in liquids depends on the modulus of volumetric elasticity and the density of the liquid – according to the equation (1)

$$v = \sqrt{K \cdot \rho^{-1}} \quad (1)$$

where: K – modulus of volumetric elasticity, ρ – liquid density.

The density of the liquid is primarily influenced by temperature and salinity. It is known from research on physical phenomena that the speed of sound waves in water depends on the temperature and salinity of the water. In

turn, the water elasticity coefficient is influenced by pressure, temperature and viscosity [15, 34]. These are relatively old studies – more recent studies in connection with the effect of solution concentration are shown e.g. in the works by Koszela-Marek [22], Koszela et al. [21]. This type of research was conducted especially for sea waters – the basic description of the acoustic properties of sea water is given in the works of Dera [8, 9].

It is known from the literature, that the speed of sound in water is described by the formula (2):

$$c(S,T,P) = 1449.14 + \Delta c_S + \Delta c_T + \Delta c_P + \Delta c_{S,T,P} \quad (2)$$

where 1449.14 m/s = $c^\circ(35, 0, 0)$ is the speed of sound under standard conditions, adopted for ocean water with a salinity of 35‰ at 0°C and atmospheric pressure. The remaining components of this formula expressing corrections for other conditions are given in formulas (2a) [40]:

$$\begin{aligned} \Delta c_S &= 1.3980(S - 35) + 1.692 \cdot 10^{-3}(S - 35)^2 \\ \Delta c_T &= 4.5721T - 4.4532 \cdot 10^{-2}T^2 - 2.6045 \cdot 10^{-4}T^3 \\ &\quad + 7.985 \cdot 10^{-6}T^4 \\ \Delta c_P &= 1.60272 \cdot 10^{-1}P + 1.0268 \cdot 10^{-5}P^2 + \\ &\quad + 3.5216 \cdot 10^{-9}P^3 - 3.3603 \cdot 10^{-12}P^4 \end{aligned} \quad (2a)$$

$$\begin{aligned} \Delta c_{S,T,P} &= (S - 35)(-1.1244 \cdot 10^{-2}T + 7.7711 \cdot 10^{-7}T^2 + \\ &\quad + 7.7016 \cdot 10^{-5}P - 1.2943 \cdot 10^{-7}P^2 + 3.1580 \cdot 10^{-8}PT + \\ &\quad + 1.5790 \cdot 10^{-9}PT^2) + P(-1.8607 \cdot 10^{-4}T + 7.4812 \cdot 10^{-6}T^2 + \\ &\quad + 4.5283 \cdot 10^{-8}T^3) + P^2(-2.5294 \cdot 10^{-7}T + 1.8563 \cdot 10^{-9}T^2) + \\ &\quad + P^3(-1.9646 \cdot 10^{-10})T \end{aligned}$$

where: T – water temperature, P – water pressure, S – water salinity.

Since this formula does not take into account other factors – e.g. the influence of gas bubbles or the content of organic substances – simpler formulas are used in practice, but also in the form of empirical regression formulas – see, for example, Hamilton [17], Mackenzie [29] or Fine et al. [11].

These formulas describe the relationship between the basic parameters of water (salinity, temperature, pressure) and the speed of sound wave propagation in water.

The phenomenon of absorption of sound waves in water has three main causes: thermal conductivity, molecular viscosity, and molecular relaxation processes. They cause an observable reduction in the acoustic energy carried by the sound wave. For the research hypothesis, the third important reason is relaxing molecular processes [6, 25].

It relies on transforming particles into new structures, caused by an increase in pressure (resulting from the passage of a sound wave). It has been experimentally confirmed that the greatest energy loss (absorption) occurs when the period of the sound wave is equal to the time needed for the particle to transform into a new structure. At present, this phenomenon has been found for three frequencies of the sound wave [12, 30]:

- 1 kHz for boric acid
- 100 and 200 kHz for transformation of $MgSO_4$
- 105 MHz for structural transformation of water.

These phenomena are still being investigated – for example, studies for poly-methylacrylics are described in the work of Ceccorulli and Pizzoli [4].

It implies, that depending on the frequency of the sound wave (ratio of wavelength and particle diameter), it will be possible to notice increased wave scattering in the frequency band corresponding to specific particle diameters [32]. For given phytoplankton components, their diameter is determined by species and stage of development. This should make it possible to find the relationship between the number of organisms (and other particles suspended in water), their type or number of gas particles, and the length (frequency) of the scattered wave [28, 38].

In the literature research, references can be found showing the use of various models of sound wave propagation in water depending on its physical or geometric properties. Primarily for marine waters [1, 24].

In the description of sound propagation phenomena, various mathematical models are used to describe the phenomena occurring in water [7, 26, 27]. The models presented in those works, their mathematical description, and various methods of solving problems related to modeling phenomena occurring during the propagation of the sound wave in the water show that these phenomena are still poorly understood and ambiguous from the point of view of their understanding or mathematical description.

In the case of surface waters – PAM (Passive Acoustic Monitoring) is currently used. In the study by Desjonquères et al. [10], the authors describe the potential application of the PAM method for the assessment of biological phenomena and ecological assessment of surface waters. Putland and Mesinger [36] clearly emphasized that freshwater acoustic monitoring is a field of knowledge that is still more unknown than known. In their research, they focused on monitoring lakes in Minnesota and the impact of sonic anthropopressure on lake waters. However, they also used the PAM method. Similar conclusions can be drawn from the work by Proulx et al. [35].

Also, the work by Rountree et al. [37] focused on "new" sources of sounds in surface waters – from artificial sources (cars, air transport, and others), while confirming that living organisms are the source of a huge sound background in lakes – in practice mostly unrecognized yet.

In turn, Geay et al. [13] analyzed the method of propagation of sounds in shallow mountain streams in their work. In their research, they focused on the use of sound to assess the intensity of gravel and stone erosion in the bottoms of mountain streams. The use of acoustic monitoring methods is not limited to natural reservoirs.

However, there is relatively a lot of work on the propagation of sound waves in sea waters. In these works, the authors discuss the influence of physical parameters (temperature, density) and chemical parameters – salinity) on the propagation speed of the sound wave – pro. For example, works contained in post-conference materials published by Murali et al. [31] or specifically the work by Annalakshmi, and Murugan [2], where the authors devote an entire chapter to measuring the speed of sound in sea waters using the CTD data analysis (conductivity, temperature, depth – pressure). Similar studies are also carried out in

Poland – for example, the work by Opaliński [33] describes a study of the impact of salinity and persistent thermal stratification on the accuracy of bathymetric measurements of water reservoirs with the use of echolocation methods.

To summarise the review, the description of physical phenomena related to the propagation of sound waves in water used in the measurement of the position of objects in the water (e.g. submarines, fish schools, underwater obstacles, etc.), i.e., it is well scientifically described, e.g. by Hodges [19]. The available sonic techniques can observe zooplankton clusters [14] – but there is very little research on sound wave propagation and showing the effects of pollution and temperature on sound wave propagation in lake waters [5].

The development of the above-described relationships between the propagation of a sound wave in water and its physicochemical properties will allow for a better understanding and more accurate description of the phenomena occurring in water during the propagation of a sound wave for different frequencies and water conditions. The development of new – more accurate regression formulas for the above-mentioned dependence will not only contribute to the development of science, but may be the basis – in the next step – for the development of new methods of analysis of the state of the water. Currently, such analyses require either measurements made with the use of multi-parameter probes or detailed laboratory analyses (physical, chemical analyses, microscopic analyses of phytoplankton and zooplankton), which are time-consuming and require the work of a team of specialists. These analyses – often tedious and time-consuming from the point of view of current standards, are often based on chemical methods.

2. Research hypothesis

As it was written in paragraph 1 – there are no articles available in the literature that illustrate the relationship between the trophic state of surface waters and physical phenomena related to the propagation of sound waves in water. Conducting the planned research will allow to find mathematical relationships (regression equations) between the mentioned above phenomena of sound wave propagation in water and its chemical, and physical state. Consequently, the scientific goal of the study is to find the relationship between the speed of propagation of sound and the physico-chemical state of surface waters.

The research hypothesis is as follows: a specific water status described by physical-chemical parameters will give characteristic parameters of sound wave propagation.

As a consequence, it is possible to build dependencies that will clearly define the trophic state of water based on the measurements of sound wave propagation.

3. Measuring stand

The ultrasonic measuring stand used in the study consisted of a Panametrics 5800PR ultrasonic wave generator, a Tektronix TDS 1012B digital oscilloscope, a set of M02 2L0o20C INCO measuring heads, and a measuring module that guaranteed a fixed, coaxial position of the heads opposite each other. An Adwa AD1020 laboratory meter with an accuracy of 0.1°C was used for temperature control (Fig. 1).

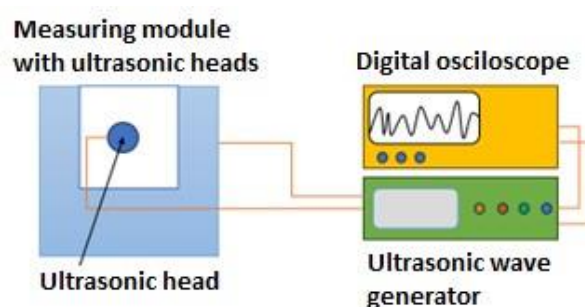


Fig. 1. Measuring stand

Knowing the distance of the heads in the measuring device ($s = 48 \pm 0.05$ mm) and measuring the ultrasonic wave transition time between the measuring heads (± 0.01 μ s), the propagation speed of the ultrasonic wave at a frequency of 2 MHz was calculated based on the relation (3) assuming a constant propagation speed of the ultrasonic waves at a short distance.

$$v = s \cdot t^{-1} \text{ [ms}^{-1}\text{]} \quad (3)$$

where: v – ultrasonic wave propagation speed [ms^{-1}], s – distance between measuring heads [m], t – time of ultrasound wave transition through the test medium [s].

4. Material and methods

The research was organised into two stages, the first was to test the effect of phosphates in the sample on the ultrasonic wave propagation velocity, while the second consisted of testing one sample at selected temperatures to determine the effect of temperature on the ultrasonic wave propagation velocity.

In the first stage, laboratory samples based on distilled water with a phosphate content of 0, 20, 40, 60, 80 and 100 $\text{kg} \cdot \text{m}^{-3}$ were obtained by adding an appropriate amount of di-Potassium hydrogen phosphate 3 ($\text{K}_2\text{HPO}_4 \times 3\text{H}_2\text{O} - 228.23 \text{ g} \cdot \text{mol}^{-1}$) to water were used. The substance was then mixed with a mechanical stirrer until the components were dissolved in distilled water. The test consisted in determining changes in the speed of propagation of the ultrasonic wave depending on the phosphate content of the sample. For this purpose, each sample was tested 10 times using a specialised measuring stand described in more detail in section 4, and then the results were compared with each other. Additionally, in order to eliminate the effect of temperature on the measurement results, each of the tested samples was kept at $20 \pm 0.5^\circ\text{C}$.

The second stage consisted of using a tap water sample with unknown chemical content. The test was carried out using the same sample at temperatures ranging from 12 to 30°C , changing it every 1°C . Due to technical limitations of the measuring stand, 3 repetitions of the ultrasound propagation time were performed for each temperature value of the sample.

5. Results and discussion

The ultrasonic velocity calculated from the laboratory samples is shown graphically in Fig. 2.

The propagation velocity of the ultrasonic wave for the reference sample with zero phosphate content ranged from

1476.0 to 1476.5 m·s⁻¹, and the mean value of the values obtained was 1476.1 ±2.1 m·s⁻¹. A difference appeared already in the first sample with 20 kg·m⁻³ phosphate content, for which the calculated velocity ranged from 1502.2 to 1502.3 m·s⁻¹ and the mean value was 1502.3 ±2.1 m·s⁻¹.

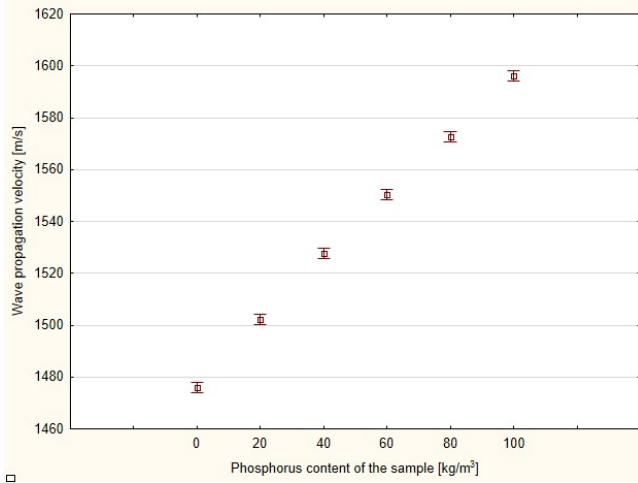


Fig. 2. Ultrasonic velocity calculated from the laboratory samples, phosphorus content

The average wave propagation velocity for the other samples was 1527.7 ±2.2 m·s⁻¹ for the 40 kg·m⁻³ sample, 1550.4 ±2.2 m·s⁻¹ for the 60 kg·m⁻³ sample, 1572.7 ±2.2 m·s⁻¹ for the 80 kg·m⁻³ sample and 1596.3 ±2.3 m·s⁻¹ for the 100 kg·m⁻³ sample.

Based on the results obtained, a linear approximating function (Fig. 3) (4) was determined using CurveExpert 1.4 software, whose correlation coefficient was r = 0.9995.

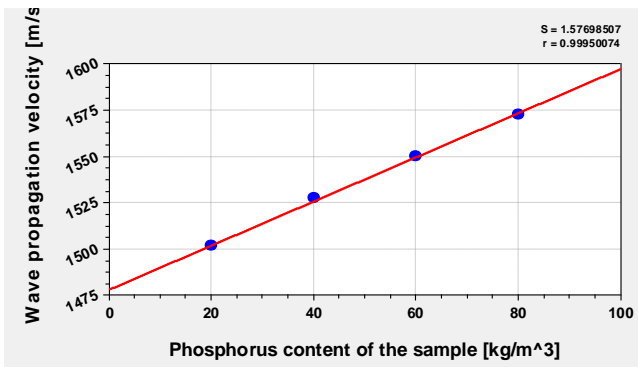


Fig. 3. Linear approximating function, phosphorus content

$$v = 1477.97 + 1.19 \cdot y_1 \quad (4)$$

where: v – ultrasonic wave propagation speed [m·s⁻¹], y₁ – phosphate content of the test sample [kg·m⁻³].

The second part of the study, testing the sample for changes in the characteristic parameter due to temperature change. Using the same sample continuously, the results were 1469.4 ±2.1 m·s⁻¹ for a temperature of 12°C to 1518.5 ±2.1 m·s⁻¹ for a temperature of 30°C. Changes in the ultrasonic wave propagation velocity values were observed with each temperature change (Fig. 4).

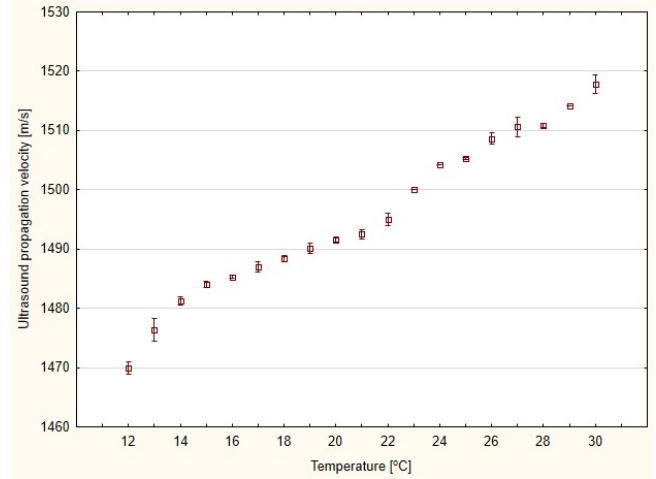


Fig. 4. Ultrasonic wave propagation velocity, temperature

Analogous to the first part, a linear approximating function (Fig. 5) (5) was determined using CurveExpert 1.4 software, whose correlation coefficient was r = 0.9906.

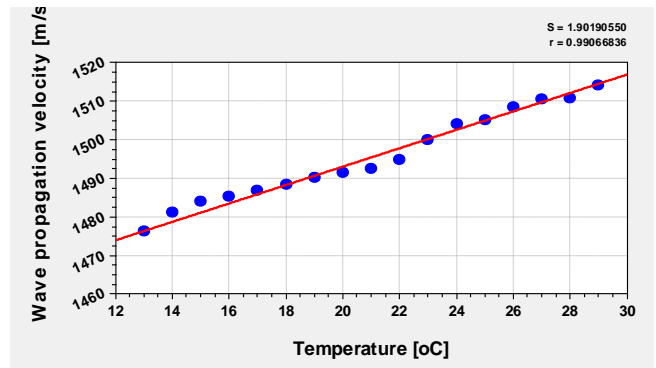


Fig. 5. Linear approximating function, temperature

$$v = 1444.21 + 2.44 \cdot y_2 \quad (5)$$

where: v – ultrasonic wave propagation speed [m·s⁻¹], y₂ – temperature of the tested sample [°C].

In conclusion, the first part of the study shows that the ultrasonic wave propagation velocity not only increases with increasing phosphate content in the test sample but also this increase is close to linear, as the propagation velocity changed by 25 ±3 m·s⁻¹ for each 20 kg·m⁻³ of phosphate content in the samples. This suggests that each 1 kg/m³ increase in phosphate concentration should increase the wave propagation velocity by approximately 0.25 m·s⁻¹.

In the second stage, the significant influence of the test sample temperature on the test results was confirmed. Each change in temperature caused significant differences in the average propagation of the ultrasonic wave in the sample. This shows that in ultrasonic testing, it is necessary to monitor and maintain the temperature of the samples being tested constantly.

6. Conclusions

As a result of the first part of the study, it can be concluded that the proposed method is useful for the measurement of phosphate content in laboratory samples, as the

value of the propagation velocity of the ultrasonic wave is dependent on the content of phosphate in the test sample.

The measurements obtained indicate that the propagation speed of the ultrasonic wave not only increases with increasing phosphate concentration, but that this increase is also close to linear, as the propagation speed changes by $25 \pm 3 \text{ m}\cdot\text{s}^{-1}$ for every $20 \text{ kg}\cdot\text{m}^{-3}$ of phosphate concentration in the sample. This shows that each $1 \text{ kg}\cdot\text{m}^{-3}$ increase in phosphate concentration should increase the wave propagation velocity by approximately $0.125 \text{ m}\cdot\text{s}^{-1}$.

As a result of the study, it can be concluded that the proposed method is useful for measuring phosphate content in water with an accuracy of $\pm 2.5 \text{ kg}\cdot\text{m}^{-3}$.

The second part of the study shows how much influence the sample temperature has on the measurement results. A temperature deviation of as little as 0.1 degree causes

a significant change in the wave propagation speed. Therefore, it should be kept constant during future tests with the proposed method so that its value can be ignored.

The aim of publishing the paper in an automotive journal is to present the developed method for measuring contaminant content. As is well known, contaminants are also found in engine oils. They contribute to the degradation of the lubricating properties of oils [39]. According to the work [23], soot contamination of the lubricant can damage the power unit, so the rapid detection of its accumulation may be desirable to prevent damage. Adaptation of the presented method in vehicle diagnostics could extend the available methodology to a non-destructive and quick way of determining the properties of engine oils. A pilot study on contaminated engine oils using ultrasound is planned for the near future.

Nomenclature

c	speed of sound in water	S	water temperature
c°	speed of sound in water under standard conditions	t	time of ultrasound wave transition through the test medium
CTD	conductivity, temperature, depth (pressure) data analysis	T	water salinity
K	modulus of volumetric elasticity	v	ultrasonic wave propagation speed
P	water pressure	y1	phosphate content of the test sample
PAM	passive acoustic monitoring	y2	temperature of tested sample
r	correlation coefficient	ρ	liquid density
s	distance between measuring heads		

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Paweł Wesołowski, MEng. – Faculty of Technical Sciences, University of Warmia and Mazury in Olsztyn, Poland.
e-mail: pawel.wesolowski@student.uwm.edu.pl



Maciej Neugebauer, DSc., DEng. – Faculty of Technical Sciences, University of Warmia and Mazury in Olsztyn, Poland.
e-mail: mak@uwm.edu.pl

