According to the World Health Organization (WHO), more than 80% of human illnesses are related to the drinking water quality; according to this indicator, Ukraine ranks 95th among 122 countries. According to the official data from the National Security and Defense Council of Ukraine, up to 80% of the country’s surface water bodies are unsuitable as sources of drinking water, and about half of groundwater (GW) is supplied to consumers with large deviations from standard requirements. Therefore, continuous monitoring of surface and groundwater composition is a mandatory requirement (Odnorih et al., 2020), in addition to the introduction of wastewater treatment technologies to prevent hydrosphere pollution. For these purposes, biological (Malovanyy et al., 2014), reagent (Tulaydan et al., 2017) or adsorption (Sakalova et al., 2019) technologies are most often used. The study of regional features of drinking water supply of Ukraine and elucidation of the role of water factor in the formation of non-communicable diseases is still relevant. In particular, it is necessary to establish the role of different combinations of mineral components in

Fluorides as an Indicator of Physiological Completeness of Mineral Composition of Drinking Waters of Separate Regions of Ukraine

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ABSTRACT

More than 80% of human diseases are related to the drinking water quality. In Ukraine, up to 80% of surface water bodies, according to official data, are unsuitable as sources of drinking water. A significant part of groundwater is supplied to consumers with significant deviations from quality standards. The urgency of the work is due to the need to assess the physiological value of the mineral composition of drinking water in some regions of Ukraine, in particular the content of fluoride in surface and groundwater sources. The paper evaluated the level of fluoride balance as a possible factor influencing the health of the population of certain regions of Ukraine. In Ukraine, there are four main geochemical regions in terms of fluoride content in drinking water (from very low concentrations of fluoride to high content). On the example of Odessa and Poltava regions, it was noted that the foci of endemic pathology are usually concentrated in the area of development of adverse natural or man-made processes. Both deficiency and excess of fluorides are important factors in shaping the health of the population, which determines the rationale for fluoridation or fluoridation of drinking water. Depending on the components of the environmental risk of high or low fluoride content, comprehensive caries and fluorosis prevention schemes were also recommended.

Keywords: fluorides, sources of drinking water supply, mineral composition, surface waters, groundwater.
shaping the health of drinking water consumers. The role of the water factor in the supply of the body with essential minerals needs to be further specified. Finally, it is advisable to determine the degree of adaptation of the population to different concentrations of mineral compounds in drinking water, which would determine the ranges acceptable to justify the requirements for the drinking water quality in some regions of Ukraine. In this regard, the physiological balance of the mineral composition of drinking water is not only an indicator of drinking water quality, but also an important factor in shaping the health of the population, because both deficiency and excess of physiologically important chemical elements – trace elements – provoke specific human diseases, i.e. diseases (symptoms) caused by deficiency, excess or imbalance of trace elements in the body (Avitsyn & Zhavoronkov, 1991). For example, fluoride – depending on its amount of in drinking water – it can be both beneficial and harmful.

It will be recalled that “fluorine” in drinking water does not mean fluoride itself (fluorine) – one of the chemical elements (pale yellow gas with a pungent odor), and fluorides (fluoride) – chemical compounds of fluorine with other elements. Fluoride in natural waters is in the form of simple and complex fluoride ions: \( F^- \), \( [AlF_3]^- \), \( [FeF_3]^- \), \( [FeF_6]^{3-} \), \( [CrF_5]^3- \), \( [TiF_6]^{3-} \) etc. The migration capacity of fluoride in natural waters depends on the content of \( Ca^{2+} \) which form sparingly soluble compounds with fluoride.

According to the WHO recommendations, drinking water should contain no more than 1.5 mg/dm\(^3\) of fluorides, and 0.5–1.0 mg/dm\(^3\) in the case of artificial fluoridation of water. According to sanitary norms 7525–2014 “Requirements and methods of quality control of drinking water” (2015) the maximum permissible concentration (MPC) of fluorides in drinking water is 0.7–1.5 mg/dm\(^3\). According 2.2.4-171-10 (State sanitary norms, 2010) depending on climatic zones, the content of fluorides in tap water and water from bottling points and pumping stations is in the range of 0.7–1.5 mg/dm\(^3\); in the water from wells and catchments ≤ 1.5 mg/dm\(^3\); in packaged water ≤ 1.5 mg/dm\(^3\). One of the indicators of the physiological value of the mineral composition (PVMC) of drinking water is the content of fluorides, the mass concentration of which is determined by using the photometric and potentiometric methods. In contrast to other indicators of drinking water PVMC (total hardness, total alkalinity, dry residue, iodine, potassium, calcium, magnesium, sodium, dry residue), the optimal concentration of fluorides varies within very narrow limits (0.7–1.2 mg/dm\(^3\)) (State sanitary norms, 2010). It is this range of optimal fluoride content that is taken into account when estimating PVMC of drinking water in certain regions of Ukraine.

The urgency of the work is due to the need to assess the PVMC of drinking water in certain regions of Ukraine, in particular the content of fluoride in surface and groundwater sources.

**MATERIALS AND METHODS**

The purpose of the study was to assess the level of balance (optimality) of one of the indicators of the mineral composition of drinking water – fluoride, as a possible factor influencing the health of the population of certain regions of Ukraine.

Estimation of the content of fluorides in the composition of drinking water from surface and groundwater sources of water supply in some regions of Ukraine is given by the results of research of regional chemical and bacteriological laboratories. The research is based on critical analysis, synthesis, analogy and generalization of information on the content of fluorides in the drinking water of certain regions of Ukraine. The published data, as well as materials of own studies were used while performing the work.

**RESULTS AND DISCUSSION**

It is known that the amount of fluoride in the human body depends on its content in the skeleton (about 0.007%), exceeding the content of iron, iodine, copper, zinc and other trace elements (Trygub, 2013), so fluoride is one of the most important chemical elements on human life in general. In minimal amounts, fluoride is necessary for metabolic processes in the body and constitutes a vital trace element after copper, zinc, iron, manganese, iodine and cobalt (Neiko et al., 2001). The reason for this statement is the ability of fluoride to prevent the development of dental caries and its therapeutic effect in some bone diseases. Therefore, both insufficient and high concentrations of fluoride entering the body have a negative effect, primarily on mineral metabolism and contribute to various abnormalities in human organs. In addition, fluorides affect the
reproductive system. Damage to chromosomes by fluoride compounds is due to the defeat of enzymes that provide DNA synthesis and repair (Trygub, 2013, Boyko, 2012).

The mineral composition of drinking water, in particular the content of fluorides, is the most important factor in determining dental health, especially the development of endemic caries, which is confirmed by numerous publications of R.D. Gabovich and his followers.

Fluorides are a recognized anti-caries agent, but the use of drinking water and foods with their high content, as well as excessive intake of fluorides in the atmosphere for a long time lead to oversaturation of the body with fluoride, the external manifestation of which is dental fluorosis (Dental fluorosis, 2021). It should be noted that the content of fluorides in drinking water is not the only one in determining the risk of carious lesions. It is known that the main source of both essential and non-essential minerals for the human body are foods, but 99% of fluorides enter the human body mainly with drinking water.


If excessive fluoride intake is not prevented, it can provoke not only damage to teeth, but also disorders of the musculoskeletal system and joints, which significantly affects human life. Fluorosis most often occurs in children because their body, unlike adults, is more susceptible to the harmful effects of environmental factors and easily adsorbs fluoride (Rybalov & Skikevich, 2006).

The main source of fluoride in the human body is drinking water. In Ukraine, there are four main geochemical regions in terms of the fluoride content in drinking water (Fig. 1): 1) regions where fluorides are absent in drinking waters or their number is very low (Zakarpattia, Ivano-Frankivsk, Chernivtsi, Lviv, Volyn, Ternopil and Rivne regions); 2) regions where the content of fluorides in drinking waters is reduced (Kyiv, Zhytomyr, Khmelnytsky, Vinnytsia, Odessa, Mykolaiv, Kherson, Zaporizhia regions and the Autonomous Republic of Crimea); 3) regions with normal fluoride content in drinking water (Chernigiv, Cherkasy, Luhansk, Sumy and Kharkiv regions); 4) regions where the content of fluorides in drinking water is high (Poltava, Kirovograd, Dnipropetrovsk and Donetsk regions).

![Figure 1. Zoning of the territory of Ukraine in terms of the fluoride content](image-url)
These geochemical regions are selected rather conditionally, because within these regions there are local differences.

Most of the surface waters of Ukraine contain a small amount of fluorides, the waters of some rivers and reservoirs may contain 1.3–1.8 mg/dm³ of fluorides; their concentration in the waters of artesian wells, as a rule, does not exceed the maximum allowable (0.7–1.5 mg/dm³), but the underground sources of some regions are characterized by an increased content of fluorides. This is confirmed by the data on the content of fluorides in drinking water of centralized water supply systems of some industrial and urban agglomerations (IUA) of Ukraine.

The average annual fluoride content in the waters of the Dniester River (source of centralized water supply) is 0.249 ± 0.031 mg/dm³, in the tap water of the Odessa IUA – 0.157 ± 0.012 mg/dm³, i.e. below the value of the minimum physiological norm (0.7 mg/dm³). An alternative source of water supply is the interstratal groundwater of the Upper Sarmatian aquifer of the Miocene, which is operated by artesian wells drilled in different parts of the Odessa IUA. The concentration of fluorides after purification in pumping complexes is 0.03–0.64 mg/dm³, which is also below the minimum standard value (Boyko, 2012). The main source of centralized water supply of the Mykolayiv IUA involves the surface waters of the Dnieper river. The average annual content of fluorides in the waters of the Dnieper River is 0.35 ± 0.041 mg/dm³, in the tap water of the Mykolayiv IUA – 0.24 ± 0.011 mg/dm³, which is below the value of the minimum physiological norm. Kherson IUA is located on the right high bank of the Dnieper, but, despite the availability and accessibility to surface water sources, the centralized water supply system is based on groundwater. The main part of fresh groundwater is concentrated mainly in the Neogene aquifer complex. This complex is widely branched across the Kherson region and provides almost 100% of groundwater production. The content of fluorides in groundwater does not exceed the optimal range (0.7–1.2 mg/dm³). The centralized water supply of the Dnieper IUA is provided by the waters of the Dnieper. At Kaidatsky and Lomovsky water intakes, the concentration of fluorides in river water varies between 0.10–0.21 mg/dm³, and in their clean water reservoirs – 0.08–0.12 mg/dm³, i.e. much lower than the minimum normative value. For the centralized water supply of Kharkiv IUA use water from the Pecheneg reservoir, which is filled with surface waters of the Seversky Donets River (74.1% of the total supply), water from the Chervonopavlovskiy reservoir, which is filled from the Dnieper-Donbass canal, and artesian water (2.4%). The average annual concentration of fluorides in the water of the Seversky Donets is 0.34 mg/dm³, and in the water of the Krasnopavlovsk Reservoir – 0.27 mg/dm³, i.e. below the value of the minimum physiological norm. The water supply of Lviv IUA is carried out from underground sources. One of the main is the aquifer of Upper Cretaceous sediments. The fluoride content varies in the range of 0.15–0.40 mg/dm³, which is below the minimum standard value (Safranov et al., 2016; Safranov & Husieva, 2016).

In Ukraine, studies have been conducted on the problem of fluoride supply to people living under different environmental conditions. The areas characterized by different concentrations of fluoride in drinking water were identified, including those that are at risk for the symptoms of hypofluorism and fluorosis.

On the example of the Odessa and Poltava regions, it was noted that the foci of endemic pathology are usually concentrated in the area of development of adverse natural or man-made processes.

According to the data (Svitlychna, 2013) the following average concentrations of fluorides are found in the drinking waters of the districts of the Odessa region (Fig. 2) (mg/dm³): Ananivskiy – 0.55 ± 0.05 (↓); Arcizkiy – 1.48 ± 0.15 (↑); Baltskiy – 0.44 ± 0.03 (↓); Berezivskiy – 0.73 ± 0.11 (↓); Bilgorod-Dnistrovskiy – 0.73 ± 0.11 (↓); Bilyaevskiy – 0.48 ± 0.08 (↓); Bolgradskiy – 0.35 ± 0.09 (↓); Velykomikhailivskiy – 0.24 ± 0.06 (↓); Ivanivskiy – 0.65 ± 0.07 (↓); Izmailivskiy – 0.50 ± 0.10 (↓); Kiliyskiy – 0.28 ± 0.03 (↓); Kodymskiy – 0.47 ± 0.09 (↓); Limanskiy – 0.12 ± 0.02 (↓); Podilskiy – 0.39 ± 0.05 (↓); Oknyanskiy – 0.65 ± 0.07 (↓); Lubashivskiy – 0.45 ± 0.09 (↓); Mukolaivskiy – 0.38 ± 0.07 (↓); Ovidipol’skiy – 0.55 ± 0.05 (↓); Rozdilynskiy – 0.32 ± 0.03(↓); Renisskiy – 0.52 ± 0.03 (↓); Savranskiy – 0.40 ± 0.05 (↓); Saratskiy – 1.25 ± 0.11 (↑); Tarutinskiy – 1.80 ± 0.23 (↑); Tatarbunskiy – 1.48 ± 0.15 (↑); Zakharivskiy – 0.53 ± 0.07 (↓); Shiryavskiy – 0.63 ± 0.11 (↓).

Concentrations of fluorides, the values of which are higher (↑) or lower (↓) than the standard (0.7–1.2 mg/dm³) (State sanitary norms, 2010) are indicated in bold.
As it can be seen from the data above, excessive concentrations of fluorides are characteristic only for some districts of the Odessa region, namely for interlayer PV of Sarmatian Neogene deposits of Artsysk, Tarutynsk, Saratsk, Tarabunsk and Belgorod-Dnestrov regions, which can be attributed to fluorosis of dangerous areas. For example, in the Artsysk region the main indicators of carious process in children 7, 12 and 15 years were much lower than in other districts of the Odessa region. In many other areas and in the Odessa IUA the content of fluorides in drinking water does not exceed 0.7 mg/dm³. The level of prevalence and intensity of major dental diseases in children of different ages in the Odessa region exceeds the national average by 25–30%. The highest is the prevalence and intensity of caries in Bilyaivka city, vill. Velyka-Mykhailivka, Kodyma city, vill. Mykolaivka, vill. Savran & Shiryaevo. Depending on the components of environmental risk of high or low fluoride content, as well as the hardness of drinking water, prevention schemes are recommended, involving the integrated use of bio-flavonoids, natural trace elements, oral hygiene products. It is recommended to repeat the prevention schemes every 6 months.

Almost the entire territory of Poltava region can be attributed to the fluorine-bearing hydrogeogeochemical province, where in many settlements drinking water is characterized by a fluoride content of 2.5 to 8.8 mg/dm³. Usually, the increase in fluoride concentration is due to the contact of groundwater with the Buchach-Kaniv aquifer complex of Paleogene sediments. The dominant form of fluoride migration is $F^-$, MgF$^+$, NaF$^+$. In biogeochemical terms, the most active is hydrocarbonate-chloride-magnesium calcium-free groundwater, which contains fluoride in the form of an anion $F^-$ (98.21%). When assessing the degree of environmental risk, it is advisable to use the coefficient of biogeochemical activity, which is determined by the ratio $F^-/MgF^+/CaF^+$ or $F^-/MenP^{-n-m}$ (Zhovinsky & Kravchenko, 2021).

According to Poltavastandartmetrologiya, the highest content of fluorides is characteristic of the drinking water from the Reshetylivskiy, Shishatskiy, Chutivskiy, Dykanskiy, Myrgorodskiy, Novosangarskiy, Mashivskiy and Karlivskiy district, especially in the Buchack aquifer (Fig. 3). In some settlements, the fluorine content in drinking water ranges from 2.0 to 7.8 mg/dm³. As for the water from the Poltava water supply system, for many years in a row during the state supervision it was found that the fluoride content is 1.0–1.3 mg/dm³, which is below the sanitary-toxicological norm, but slightly above the upper limit of the range of optimal values (State sanitary norms, 2010).

According to the research of I.A Boyko (2011, 2012), E.Ya. Zhovinsky and others (2006, 2021) it is shown that in some districts and cities
of Poltava region the content of fluorides in waters meets the requirements of 2.2.4-171-10 State sanitary norms, 2010, but the fluoride content in the groundwater in the Buchatsk-Kaniv aquifer complex reaches 4.5 mg/dm³ and more. The influx of fluorides into the groundwater is due to the imbalance during the dissolution of phosphorite-containing rocks and tectonic disturbances. In the area of oil and gas deposits, which are located west of Myrhorod, the waters contain up to 4 mg/dm³ of fluorides, and near the location of salt deposits – up to 8.8 mg/dm³. The areas with fluoride content in water in the range of 3.6–5.0 mg/dm³ were found near the Khorol city. This means that the increase in the fluoride content in groundwater is due not only to the inflow of fluoride-containing waters due to tectonic disturbances, but also the development of oil and gas deposits, i.e. the action of both natural and man-made factors. Intensive inflow of fluorides into aquifers (complexes) of Poltava region creates risks for the share of the region’s population (Boyko, 2012), as centralized water supply is based on these aquifers. The risks arising from the consumption of water with high fluoride content can be divided into environmental and social. Soluble fluorine compounds move easily along the soil profile and enter groundwater, and from there into surface water bodies. The migration of fluorides helps to improve soil composition, it does not bind to soil absorption complexes, so it becomes available for root nutrition of plants.

Since Poltava region is one of the regions with the highest fluoride content in groundwater (Nazarenko et al., 2014), there is a high probability of fluorosis, so the studies of the effects of fluoride on human life to develop the measures to prevent this disease are relevant.

Fluorosis is a type of enamel hypoplasia caused by an excess of fluoride ions that inhibit ameloblasts during intramaxillary tooth formation and mineralization. Observations have shown that the degree of fluorosis of teeth depends on many factors, which include: the concentration of fluoride in drinking water; duration of water consumption with high fluoride content; age and nature of feeding the child at 1–2 years of age; the nature of nutrition; socio-hygienic living conditions; transferred diseases; general condition of the body; landscape-climatic and ecological factors, etc. (Kaskova & Amosova, 2015). If the concentration of fluorides in drinking water is higher than permissible (1.5–2 mg/dm³), then up to 30–40% of the population is affected by dental fluorosis, mainly I and II degree. The use of water with such a concentration of fluorides may be temporarily permitted in a local water supply. In the case of centralized water supply, it is necessary to carry out defluoridation or dilution of water. At high concentrations of fluorides in drinking water (2–6 mg/dm³) the incidence of fluorosis in the population is 30–90%, and in 10–50% of them is fluorosis III–IV degree. Among children, there are often cases of developmental delay and bone mineralization. These disorders when drinking...
water containing 2–3 dm³ of fluorides are temporary. In some people who drink water with a fluoride content of 4–6 dm³, there is an increase in bone density and impaired conditioned reflex activity. In this case, mandatory defluoridation or dilution of water is required (Kotlyar et al., 2008). At a very high concentration of fluoride in drinking water (6–15 mg/dm³ and more) 90–100% of the population is affected by dental fluorosis with a predominance of severe forms, significantly increased abrasion, brittle teeth. In children, disorders of bone development and mineralization are often found, in adults – changes in the bones by type of osteosclerosis. There is suppression of thyroid function, changes in the activity of certain enzyme systems of the blood, changes in the myocardium, inhibition of bioelectrical activity of the brain, as well as disorders of other internal organs (eg, liver), which are detected during functional examination. Water defluoridation is also a mandatory measure.

On the basis of clinical observations, it was found that at a concentration of fluoride in drinking water of 0.8–1.2 mg/dm³ fluorosis is practically not observed. Changes in the concentration of fluoride in drinking water significantly affect the metabolic processes in the body, which leads to impaired mineralization, the formation of the protein matrix and dentin of the teeth. Children are most sensitive to fluoride intoxication. This is due to the fact that in the bones of the growing organism, fluoride is deposited faster deposited and in greater amounts. Fluorosis usually affects the permanent teeth of the children who are in an area endemic for fluorosis, from birth or from 3 to 4 years of age. The initial forms of fluorosis do not heal over time, and after a year begin to transform into more severe, which leads to brown pigmentation and destruction of the enamel. Therefore, it is necessary to implement appropriate medical measures for the children living in regions with different fluoride content in drinking water (Kosenko, 2011).

Since children are the most sensitive to fluoride intoxication, in order to study the level of their incidence of fluorosis, surveys of students of secondary and high school Takhtaulovsky educational complex of the Poltava region were conducted. According to a survey of children examined by a dentist, it was determined what percentage of patients with fluorosis use artesian pressure water, and what – well water. The study showed that of the 112 students examined, 33 (29.5%) had signs of fluorosis, of which 24 (21.4% of the total number of examined and 72.7% of the number of patients) confirmed that they constantly used interstitial (artesian) water with a fluoride concentration of 2.8–3.5 mg/dm³, and the other 9 students (8.1% and 27.3%, respectively) – well (ground) water with a fluoride concentration of 0.64–0.79 mg/dm³ (except for the village of Zhubki – 1.29 mg/dm³). The students who have used well (ground) water and are ill with fluorosis live in different parts of Takhtauli village council. It is known that fluorosis can be formed at a concentration of fluoride in water in the range of 1 mg/dm³ in children from 3 months to 8 years. The children with signs of fluorosis have an excess of fluoride, which is manifested by chronic intoxication. Elevated concentrations of fluoride disrupt collagen synthesis and affect the degree of bone mineralization. Although the content of fluorides in well (ground) water is lower than in interstitial (artesian) water, the lifestyle and nutrition of children also influenced the development of fluorosis.

In order to implement the measures to prevent fluorosis, practical recommendations were developed for school students and their parents in the form of booklets and distributed to all interested parties at student conferences and school parent meetings.

On the example of the Odessa and Poltava regions, which are referred to different geochemical regions in terms of fluoride content in drinking water, it can be seen that there is a problem of deficit and excess of these indicators PVMC. It was obtained that for some areas, the level of fluorides in drinking water may differ, which determines the justification of defluoridation, and for others fluoridation of drinking water.

If the fluoride content in natural waters is too high, it must be reduced to an acceptable level. Water defluoridation is carried out in the cases of high (more than 1.5 mg/dm³) fluoride content in drinking water and a large number of patients with fluorosis, when it is impossible to change the source of water supply or dilute it with water with low fluoride concentration. In order to reduce the concentration of fluorides in water, two methods of defluoridation are used: sorption or precipitation of fluorides by precipitation of aluminum or magnesium hydroxide or calcium phosphate, and filtration (ion exchange method) of water through fluorselective materials (Water defluoridation, 2021). In order to determine the severity of fluorosis, use the classification of I. Mueller, which
contains five degrees of the disease: doubtful, very weak, weak, moderate and severe fluorosis with the corresponding signs of the disease.

In the areas with a deficiency of fluorides in drinking water, it is advisable to justify the possibility of artificial fluoridation. Since fluorine is a trace element, which is characterized by a relatively sharp transition from physiologically useful concentrations to the concentrations that cause toxicosis, in the domestic and foreign literature there are convincing arguments, both supporters and opponents of fluoridation of drinking water. Many countries around the world have adopted a regional principle of normalization of fluorides in drinking water, when its optimal concentration is determined by the maximum daily air temperature, due to the fact that the amount of water consumed by humans depends on it. In the countries where artificial fluoridation of tap water is carried out (USA, Canada, Great Britain, Ireland, etc.), its safety and effectiveness for caries prevention are noted. The principles of enrichment of fluoride drinking water are considered in State Sanitary Norms 2.2.4.-005-98 “Fluoridation of water in the pipelines of centralized water supply” (1998). In some countries, there is no technical possibility for water fluoridation, and other countries add fluorides to table salt, bottled drinking water or milk instead of water fluoridation. It is also possible to ingest fluorides when using toothpaste, mouthwash, tablets, gels and the like. The benefits of fluoride for caries prevention have now been confirmed.

Both deficiency and excess of fluorides are important factors in shaping the health of the population, in particular the dental health of children and adults, which justifies defluoridation (Levinsky et al., 2006) or fluoridation (Fluoridation of water, 1998).

CONCLUSIONS

The optimal content of fluorides in drinking water of certain regions of Ukraine is an important indicator of the physiological value of their mineral composition. Most of the surface sources of centralized water supply in the regions of Ukraine contain a small amount of fluorides. Groundwater sources are also usually characterized by fluoride concentrations within the range of optimal values (0.7–0.2 mg/dm³), except for certain areas and aquifers in some regions of Ukraine (for example, in Poltava and Odessa regions). In the case of deviation of the fluoride content in drinking water to a greater or lesser extent, the fluorination or defluorination stage must be integrated into the drinking water treatment technology. Depending on the components of the environmental risk of high or low fluoride content, comprehensive caries and fluorosis prevention schemes are also recommended.

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