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RESEARCH ARTICLE

Rare earth elements and heavy metals in coastal springs of southern Kerala: a hydrogeochemical analysis

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Abstract

The present study deals with the hydro-geochemical and trace metal analysis of coastal springs of Varkala cliff, Kerala, South India. Varkala beach (Papanasam Beach) is one of the most popular tourism destinations in Kerala. The cliff section at Varkala consisting of clay, lignite band, sandy-clay, sandstone of Mio-Pliocene age. The coastal springs emerge from the contact between sandstone and underlying carbonaceous clay stone of the Warkalli formation of Mio-Pliocene age exposed on the uplifted block south of the Achan kovil shear zone. Spring water samples collected from five different regions of the study area during pre and post monsoon season. The pH is considerably low in all samples (4 - 4.64). The sediments and the percolating spring water have some sort of pungent odor of sulphur and these reason leads to spring water shows the sulphide content but not exceed the standard value. The rare earth metals show different concentrations in water samples under study. According to heavy metals analysis, all samples comparatively low heavy metal concentrations. The exceptions were Al, Fe, Ba and Mn were present in high concentrations in some of the samples than other analyzed heavy metals.

Keywords

Metal analysis

Spring

Sediments

Sulphide

Heavy metals

Introduction

Springs are defined as places where groundwater flows naturally from a rock sediment or soil onto the land surface or into a body of surface water. Springs are dynamic and evolve ebb and flow in response to changes in climatological, topographical, geological, and geomorphological conditions.A spring is a component of the hydrosphere; it is the surface water flows from underground.

Geological and hydro-chemical properties are the major parameters to explore classifications of springs. According to various geological origins, springs can be divided into three categories - springs emerging from igneous, metamorphic and sedimentary rocks [1]. Hydro geological properties of rocks, genesis, water discharge, rock structures and temperature are some of the key aspects used for classification of springs [2]. Springs can also be classified into

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different water types based on compositions of their major ions [3]. Major ions in spring water include HCO^{-}, Cl^{-}, SO^{2-}, SiO_2, Na^{+}, K^{+}, Ca^{2+} and Mg^{2+}, which mainly originates from dissolution and mineralization of rock [4-7].

Springs that contain significant amounts of minerals are sometimes called 'mineral springs'. Mineral springs are naturally occurring, which produce water containing minerals or other dissolved substances that alter its taste or give it a purported therapeutic value. Water from springs is usually clear. Water from some springs, however may be 'tea-coloured'. Its red iron colouring and metals enrichment are caused by groundwater coming in contact with naturally occurring minerals present as a result of ancient volcanic activity in the area. Salts, sulphur compounds and gases are among the substances that can be dissolved in the spring water during its passage underground [8]. Minerals become dissolved in the water as it moves through the underground rocks. This may give the water flavour and even carbon dioxide bubbles depending on the nature of the geology through which it passes. Springs waters are believed to have many therapeutic properties and used to cure off diseases like arthritis, many skin diseases etc [9].

Chemistry of spring water

The chemistry of spring water reflects the interaction of groundwater with the aquifer host rock as well as any chemical constituents that may be introduced from surface sources. Spring water chemistry is therefore, not intrinsically different from groundwater chemistry and the same principles apply. Aqueous chemistry in general and spring water chemistry in particular can be approached from two points of view. One can begin with the minerals that make up the host rock and their thermodynamic properties and calculate mineral solubilities and interactions.

From this in principle, the water chemistry can be predicted. The common cations in spring waters are Ca^{2+}, Mg^{2+}, Na^{+}, and K^{+}; while the most common anions are HCO^{3-}, SO_4^{2-}, and Cl^{-}. Together, these form the basis for the classification of spring water. The dominant chemistry usually depends on the host rock of the aquifer from which the spring emerges [10]. The amount of water that flows from springs depends on many factors, including the size of the caverns within the rocks, the water pressure in the aquifer, the size of the spring basin, and the amount of rainfall.

Water analysis plays a decisive role in the regulations that determine water quality for specific uses, especially for drinking water quality. Due to the importance of drinking water for human life, lack of water sources and the increased consumption of mineral waters, there is a need to assess the quality of water from various potential water resources. Besides this, as a part of water resources management, water protection measures need to be prioritized and implemented in order to ensure sustainability for future generations. The particular problem in the case of water quality monitoring is the complexity associated with analyzing the large number of measured variables.

Heavy metals are metallic elements including the transition series, which include many element required for plants and animals nutrition in trace concentrations but which become toxic at higher concentrations. These are essential elements because they are necessary for biological functions i.e. without them life does not exist or exists at unhealthy level. Excessive levels of trace elements occur naturally or due to human release of metals into the environment. Heavy metals exist partly in solution and partly in suspension adsorbed to organic or inorganic particulate matter and they are generally considered to be toxic in soluble ionic forms but toxicity is known to be reduced by water hardness [11]. Heavy metal contamination of water is an important environmental issue because most heavy metals are bio-accumulative in aquatic food webs. They can not only have effect on ecosystem, but also increase the potential health risk for wildlife and humans [12]. The background concentrations of heavy metals in natural aquatic environment are trace levels, but they have increased since the increases in industrial wastes, mining activities and landfill leachate. Cu and Zn are essential elements for various enzymes and are required by many biological processes in all life, however large amount of Cu and Zn would cause health problems. Unlike Cu and Zn, Cd and Pb are nonessential elements and toxic elements which could exhibit extreme toxicity at trace levels.

The occurrence of heavy metals in ground and surface waters can be due to natural sources, such as dissolution of naturally occurring minerals containing trace elements in the soil zone. Human activities and other anthropogenic sources such as mining, fuels, smelting of ores and improper disposal of industrial wastes, significantly contribute to elevated levels of heavy metals in waters. Surface water bodies are particularly vulnerable to contamination from industrial and municipal wastewater, leaching or runoff of agrochemicals and dissolution of air-borne pollutants. Exposure to heavy metals has been linked to developmental retardation, various cancers, kidney damage and even death in instances of very high exposure. The quality of the water in the local ground-water system will generally determine the quality of spring water. The quality of water discharged by springs can vary greatly because of factors such as the quality of the water that recharges the aquifer and the type of rocks with which the groundwater is in contact. The rate of flow and the length of the flow path through the aquifer affect the amount of time the water is in contact with the rock and thus the amount of minerals that the water can dissolve. Objectives of the study include seasonal hydro geochemical analysis of water samples from major springs of Varkala cliff area, analysis of rare earth elements and trace metals in spring water of Varkala cliff area and statistical analysis of obtained data.

Study Area

Varkala is a small coastal town located 51 km northwest of Thiruvananthapuram and 37 km southwest of Kollam in Kerala that is also an important religious place for the Hindus. Varkala beach (Papanasam beach) is one of the most popular tourism destinations in Kerala. The area is located on the sea coast of Papanasam, approximately 3.5 km from Varkala town. The long sandy beach, towering red laterite cliffs and soothing mineral springs have turned Varkala into a beautiful destination. The cliff section at Varkala, Kerala consisting of clay, lignite band, clay, sandy-clay, sandstone of Mio-Pliocene age. This sedimentary sequence is overlain by a thick laterite. The cliff section of nearly 27.69 m thick and consists of clay, lignite band, thick horizon of clay and sandstone. This thick sedimentary sequence is overlain by laterite.

The Cliff contains mineral spring that is considered to be holy water with medicinal properties [13]. The coastal belt of Thiruvananthapuram district of Kerala in Varkala has plenty of coastal springs in the cliff area. The rapid growth of coastal tourism that has been seen in the last 10 years is frequently described as one of the major reasons for the development of these areas and also is responsible for many current coastal problems (Rajan *et al*., 2007) and direct and indirect human influence and interference such as population explosion, climate change and erratic monsoon and overall environmental degradation are the major causative factors for the cliff erosion. Due to cliff erosion now coastal spring only existing Chilakoor and Papanasam helipad regions.

Methodology

Analysis were carried out for various water quality parameters such as temperature, pH, TDS, Conductivity, Chloride, Phosphate, Nitrite, Sulphide, Na, K, DO, BOD and MPN. In addition to the normal hydro geochemical parameters heavy metals and rare earth minerals were also analysed using sophisticated technologies. Temperature, pH, TDS and Conductivity were measured by multiparameter. Multiparameter is manufactured by Eutech Bench meter PC 2700. Chloride concentration was measured by $AgNO_3$ titrimetric method. Sodium and potassium was analyzed using flame photometer. Flame photometer is manufactured by Systronics128. Phosphate and nitrite was analyzed using Atomic Absorbtion spectrophotometer. It is manufactured by Systronics119. Sulphide was measured by iodometric titration method. DO and BOD by the titrimetric method. The microbiological quality of samples was analyzed in terms of most probable number (MPN) of faecal coli forms using MacConkey broth. The inductively coupled mass spectrometry system Thermo Scientific ICAP Qc was used for the trace metal analysis. The reagents used for the analysis were AR grade for preparation of solutions.

Rare earth metals and heavy metals analysis

The inductively coupled mass spectrometry system Thermo Scientific ICAP Qc was used for analysis of rare earth metals and heavy metals. The external calibration solutions were prepared from standard certified multi element solution. Thermo scientific BRANSTEAD Smart to pure water containing 1% suprapur grade Nitric acid was used to get a range of concentrations 25, 50, 100, ppb for all elements. Samples containing higher concentration of elements than this calibration range are diluted and analyzed applying dilution factors. The ion optics was tuned using Thermo scientific Tune-B ICAP-Q solution in standard mode and KED mode. Mass and detector calibration was conducted using Thermo scientific Setup solution ICAPQ. In this analysis rare earth metals like Yttrium, Lanthanum, Cerium, Praseodymium, Neodymium, Samarium, Europium, Gadolinium, Terbium, Dysprosium, Holmium, Erbium, Thulium, Ytterbium and Lutetium are analyzed. Heavy metals include Aluminum, Chromium, Manganese, Iron, Cobalt, Nickel, Copper, Zinc, Arsenic, Strontium, Cadmium, Barium and Lead analyzed by this technique.

Statistical analysis of the present data was interpreted using Standard deviation and Pearson correlation coefficient. It was analyzed to identify the association between pairs of variables for sampling status. Standard deviation is the square root of the mean of the squares of the deviation of all values of a series from their arithmetic mean. Correlation coefficient is a numerical value; it shows the degree or the extent of correlation between two variables. Statistical analysis is carried out using SPSS 11.5 software platform.

Results and discussion

The chemistry of spring water reflects the interaction of groundwater with the aquifer host rock as well as any chemical constituents that may be introduced from surface sources. Spring water chemistry is therefore, not intrinsically different from groundwater chemistry and the same principles apply. The present study evaluated the water quality of spring water and its seasonal changes (pre-monsoon and post-monsoon) in hydro geochemical and trace elements analysis of coastal springs of varkala cliff. Parameters such as Temperature, pH, Total Dissolved Solids, Conductivity, Chloride, Phosphate, Nitrite, Sulphide, Sodium, Potassium, BOD, DO and the MPN were analyzed. In addition to the normal hydro geochemical parameters heavy metals and rare earth metals were also analyzed using sophisticated technologies and statistical analysis.

Yttrium is a highly crystalline iron-gray, rare-earth metal. Yttrium is fairly stable in air, because it is protected by the formation by the formation of a stable oxide film on its surface, but oxidizes readily when heated. It reacts with water decomposing it to release hydrogen gas and it reacts with mineral acids. Yttrium never occurs in nature as a free element. It is found in almost all rare earth minerals and in uranium ores. Yttrium is mostly dangerous in the working environment, due to the fact that damps and gaseous can be inhaled with air. This can cause lung embolisms, especially during long-term exposure. Yttrium can also cause cancer with humans, as it enlarges the chances of lung cancer when it is inhaled. Finally, it can be a threat to the liver when it accumulates in the human body. Yttrium will gradually accumulate in soils and water soils and this will eventually lead to increasing concentrations in humans, animals and soil particles. Lanthanum is a soft, malleable, ductile, silver-white metal. It is chemically active; it is one of the most reactive of the rare-earth metals. Cerium is a malleable, soft, ductile, iron-grey metal, slightly harder than lead. It is very reactive: it tarnishes readily in the air; it oxidizes slowly in cold water and rapidly in hot water. Cerium is the most abundant of the rare earth elements. It makes up about 0.0046 % of the earth's crust by weight.

Praseodymium is a soft malleable, silvery-yellow metal. It is a member of the lanthanide group of the periodic table of elements. It reacts slowly with oxygen: when exposed to air it forms a green oxide that does not protect it from further oxidation. It is more resistant to corrosion in air the other rare metals. It reacts rapidly with water. Like all rare metals praseodymium is of low to moderate toxicity. Neodimium is a lustrous silvery-yellow metal. It is very reactive in air and the coated formed does not protect the metal from further oxidation, so it must be stored away from contact with air. It reacts slowly with cold water and rapidly with hot. Neodymium is the second most abundant of the rare-earth elements (after cerium) is almost as abundant as copper. It is found in minerals that include all lanthanide minerals, such as monazite and bastnasite. Samarium is a silvery-white metal belonging to the lanthanide group of the periodic table. Samarium is the fifth most abundant of the rare elements and is almost four times as common as tin. It is never found free in nature, but in contained in many minerals, including monazite, bastnasite and samarskite. Europium is a soft silvery metal both are and expensive. It is the most reactive of the lanthanide group. Europium is one of the less abundant rare-earth elements: it is almost as abundant as tin. It is never found in nature as the free element, but there are many elements containing eruropium**.** Gadolinium is being used as an environmental tracer to prove that drinking water has been contaminated by inadequately treated waste water**.** If gadolinium continues to be found in drinking water, there's danger the risk may spread to even those who do not get injected directly with the substance. Terbium is a soft, malleable, ductile, silver-gray metal member of the lanthanide group of the periodic table. It is reasonably stable in air, but it is slowly oxidized and it reacts with cold water**.**

Dysprosium is a lustrous, very soft, silvery metal. It is stable in air at room temperature even if it is slowly oxidized by oxygen. It reacts with cold water. Holmium is a malleable, soft, lustrous metal with a silvery colour, belonging to the lantanides series of the periodic chart of elements. It is slowly attacked by oxygen and water and dissolves in acids. It is stable in dry air at room temperature. Holmium has no biological role and it is considered one of the least abundant elements present in human body. It has been noted that holmium stimulates metabolism, even if it appears to have a low acute toxic rating. Erbium is a soft, malleable, lustrous, silvery metal. It is very stable in air; it reacts very slowly with oxygen and water and dissolves in acids. Its salts are rose coloured and it has a sharp adsorption spectra in visible, ultraviolet and infrared light. Thulium is a lanthanide element; it has a bright silvery-gray luster. It is the least abundant of the rare earths and its metal is easy to work. It slowly tarnishes in air, but is more resistant to oxidation than most rare-earth elements. It also has some corrosion resistance in dry

air and good ductility. Naturally occurring thulium is made entirely of the stable isotope Tm-169. The element is never found in nature in pure form but it is found in small quantities in minerals with other rare earths. It is principally extracted from monazite, which contains about 0.007% of thulium and bastnasite (about 0.0008%). Soluble thulium salts are regarded as slightly toxic in taken in large amounts, but the soluble salts are completely not toxic.
Lutetium is a rare earth metal and perhaps the most expensive of all rare elements. It is found in small amounts with all rare earth metals and is very difficult to separate from other rare elements. This is largely because of the way it is found in nature. Ytterbium is a soft, malleable and rather ductile element that exhibits a bright silvery luster. Compounds of ytterbium are rare. Strontium is an alkaline earth metal that is found naturally in the minerals celestine and strontianite. In the present study analyzed these rare earth metals concentration is much lower than prescribed level in all the samples. The rare earth metals show different concentrations in water sample under study. These metals are highly correlated with each other (+0.77 to +0.99).

Aluminium is the most abundant metallic element and constitutes about 8% of the earth's crust. Aluminium exposure is a risk factor for the development or acceleration of onset of Alzheimer disease (AD) in humans. The concentration of aluminium in natural waters can vary significantly depending on various physicochemical and mineralogical factors. Dissolved aluminium concentrations in waters with near-neutral pH values usually range from 1 ppb to 50 ppb but rise to 500 – 1000 ppb in more acidic waters or water rich in organic matter. At the extreme acidity of waters affected by acid mine drainage, dissolved aluminium concentrations of up to 90000 ppb have been measured. Chromium is widely distributed in the Earth's crust. Chromium is present in small quantities in the environment. The toxicity of chromium depends on its physicochemical form; hexavalent salts are considered the most dangerous. Desirable limit of chromium is 50 ppb (BIS). Concentration of chromium is much lower than prescribed level in all the samples. Prolonged consumption of water containing elevated concentrations of chromium, can damage liver, kidney and nervous tissue.

Manganese is an essential element for humans and other animals. Adverse effects can result from both deficiency and overexposure. Adverse neurological effects following extended exposure to very high levels in drinking-water. In surface waters, manganese occurs in both dissolved and suspended forms, depending on such factors as pH, anions present and oxidation–reduction potential. Anaerobic groundwater often contains elevated levels of dissolved manganese. The divalent form (Mn^{2+}) predominates in most water at pH 4–7, but more highly oxidized forms may occur at higher pH values. It bioaccumulates in lower organisms. At higher concentration levels manganese causes an undesirable taste in drinking-water. The presence of manganese in drinking-water may also lead to the accumulation of deposits in the distribution systems. The desirable limit of manganese is 100 ppb. Among the two samples are higher than the desirable limit prescribed by BIS (S I – 120 ppb, SII – 200 ppb). Iron can affect the flavour and colour of food and water. Iron is biologically an important element which is essential to all organisms and present in haemoglobin system. The concentration of dissolved iron in water is dependent on the pH, redox potential, turbidity, suspended matter and the concentration of aluminium and occurrence of several heavy metals, notably manganese. Iron is an essential element in human nutrition. Iron is an essential micronutrient element required by both plants and wildlife at significant concentrations, as it is a vital part of metabolic enzyme formation and the oxygen transport mechanism in the blood of all vertebrate and some invertebrate animals. Iron is considered as an essential nutrient but in the concentration above could change the taste of water and produces adverse effects. Iron causes indigestion and constipation in human beings. Aeration of iron-containing layers in the soil can affect the quality of both groundwater and surface water if the groundwater table is lowered or nitrate leaching takes place. Dissolution of iron can occur as a result of oxidation and decrease in pH. Desirable limit of iron is 300 ppb (BIS). Among the two samples are higher than the desirable limit prescribed by BIS (S II – 660 ppb, SIII – 2800 ppb).

Cobalt is a natural element found throughout the environment. Cobalt has both beneficial and harmful effects on human health. It is beneficial for humans as a part of vitamin B12, which is essential to human health. At elevated levels cobalt provokes harmful health effects. It can cause pulmonary syndrome, skin syndrome, allergy, gastrointestinal irritations, nausea, cardiomyopathy, haematological disorders. Nickel is a lustrous white, hard, ferromagnetic metal. Nickel may also be present in some ground waters as a consequence of dissolution from nickel ore-bearing rocks. Nickel concentrations in ground water depend on the soil use, pH, and depth of sampling. Long-term exposure to elevated concentrations of nickel causes weight loss, heart and liver damage and dermatitis. Copper is found in surface water, ground water, seawater and drinking-water, but it is primarily present in complexes or as particulate matter. Copper concentrations in drinking-water vary widely as a result of variations in water characteristics, such as pH, hardness and copper availability in the distribution system. The symptoms of severe copper poisoning in the human body are extensive hemolysis, hepatic necrosis, nephropathy and coma and if not treated may lead to death. Wilson's disease is well known. It is characterized by inability to digest copper in the body resulting in degenerative changes in the brain and cirrhosis of the liver. Desirable limit of Copper is 50 ppb (BIS). Copper contents were within allowed limits.

Zinc is an essential trace mineral that is found in water and food in the form of salts or organic complexes. Zinc occurs in small amounts in almost all igneous rocks. The principal zinc ores are sulfides, such as sphalerite and wurzite. In natural surface waters, the concentration of zinc is usually below 10000 ppb and in ground waters, 10000– 40000 ppb. The desirable limit of zinc is 5000 ppb (BIS). Zinc concentrations within allowed limits. Arsenic is introduced into water through the dissolution of rocks, minerals and ores from industrial effluents, including mining wastes and via atmospheric deposition. The clinical symptoms of zinc deficiency are anorexia, pica, impaired taste acuity, menstrual lethargy and disturbances, via; rough, dry skin, impaired wound healing and increased susceptibility to infection, chronic deficiency in paediatric and adolescent age group causes growth retardation and delay of sexual maturation.

Acute zinc toxicity results in hemodialysis, characterized by nausea, vomiting, fever and severe anemia. However, concentrations above 5 mg/l can cause a stringent taste and opalescence in alkaline water. Long-term exposure to arsenic in drinking-water is causally related to increased risks of cancer in the skin, lungs, bladder and kidney, as well as other skin changes such as hyperkeratosis and pigmentation changes. The desirable limit of arsenic is 50 ppb (BIS). Arsenic concentrations fall within allowed limits. Strontium shares many physical and chemical properties with calcium and barium and is highly susceptible to chemical changes. Strontium that occurs naturally in the earth has four stable isotopes Sr-84, -86, -87 and -88. Twelve other strontium isotopes are unstable, meaning they are radioactive. Strontium-90 is the most prevalent radioactive isotope in the environment. Strontium occurs naturally in the environment. Air, dust, soil, foods and drinking water all contain small amounts of strontium. Ingestion of small amounts of strontium is not harmful. However high levels of strontium can occur in water drawn from bedrock aquifers that are rich in strontium minerals. The risk posed by strontium depends on the concentration ingested and on the exposure conditions and EPA current reference concentration indicates that ongoing exposure to strontium at levels of more than 4000 parts per billion per day may lead to negative health effects. There is no evidence that drinking water with trace amounts of naturally-occurring strontium is harmful. Strontium is comparatively low concentrations in all the samples. Cadmium has carcinogenic properties as well as long biological half-life, leading to chronic effects as a result of accumulation in the liver. It can also cause kidney damage, as well as produce acute health effects resulting from overexposure to high concentrations. The desirable limit of cadmium is 10 ppb (BIS). Cadmium concentration fall within allowed limits.

Barium is present as a trace element in both igneous and sedimentary rocks. Barium in water comes primarily from natural sources. The acetate, nitrate and halides are soluble in water, but the carbonate, chromate, fluoride, oxalate, phosphate and sulfate are quite insoluble. The solubility of barium compounds increases as the pH level decreases. The highest levels to be found in drinking-water are likely to be associated with groundwater of low pH from granite-like igneous rocks, alkaline igneous and volcanic rocks and manganese-rich sedimentary rocks. Organic barium compounds are ionic and are hydrolyzed in water. The concentration of barium ions in natural aquatic systems is limited by the presence of naturally occurring anions and possibly also by the adsorption of these ions onto metal oxides and hydroxides. At high concentrations, barium causes vasoconstriction by its direct stimulation of arterial muscle, peristalsis as a result of the violent stimulation of smooth muscles and convulsions and paralysis. The desirable limit of Barium is 10 ppb (BIS). Barium was present in high concentrations in all the samples. Lead is the commonest of the heavy elements. Lead occurs naturally in the environment. However, the most significant concentrations of this element found in the environment are the result of the human activities. Lead is toxic and it accumulates in kidneys and skeleton. Over centuries lead became recognized as a cumulative general metabolic poison. Lead contamination of the ground water may result from industrial effluents, old plumbing household sewages, agricultural run-off containing phosphatic fertilizers and human and animal excreta. The desirable limit of lead is 50 ppb (BIS). Lead concentrations within allowed limits.

The present study focusing on heavy metal determinations in spring water samples in Cliff regions showed the presence of aluminium, manganese, barium and iron. Aluminium was present in high concentrations in all the samples than manganese, barium and iron. Analyzed other heavy metals concentration is much lower than prescribed level in all the samples. These metals include Al, Cr, Fe, Co, Ni, Zn, As, Sr, Cd, Ba and Pb are highly correlated with each other (+0.50 to +0.99). Mn is negatively correlated with other heavy metals (except Ba +0.62). Exposure to heavy metals has been linked to developmental retardation, various cancers, kidney damage and even death in instances of very high exposure. Excess of too much copper causes narcotic hepatitis [14]. Arsenic, barium, cadmium, chromium and nickel are toxic metals affecting the internal organs of the human body. Arsenic is widely distributed in waters at low concentrations, with associated instances of higher concentrations in well waters. Presence of metals in potable waters is a matter of serious concern because of the toxic nature of these materials. Iron is vital in the formation of haemoglobin in human being.

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