Microbial Influence on Iodine Speciation at the 200 West Hanford Site-16549

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ABSTRACT

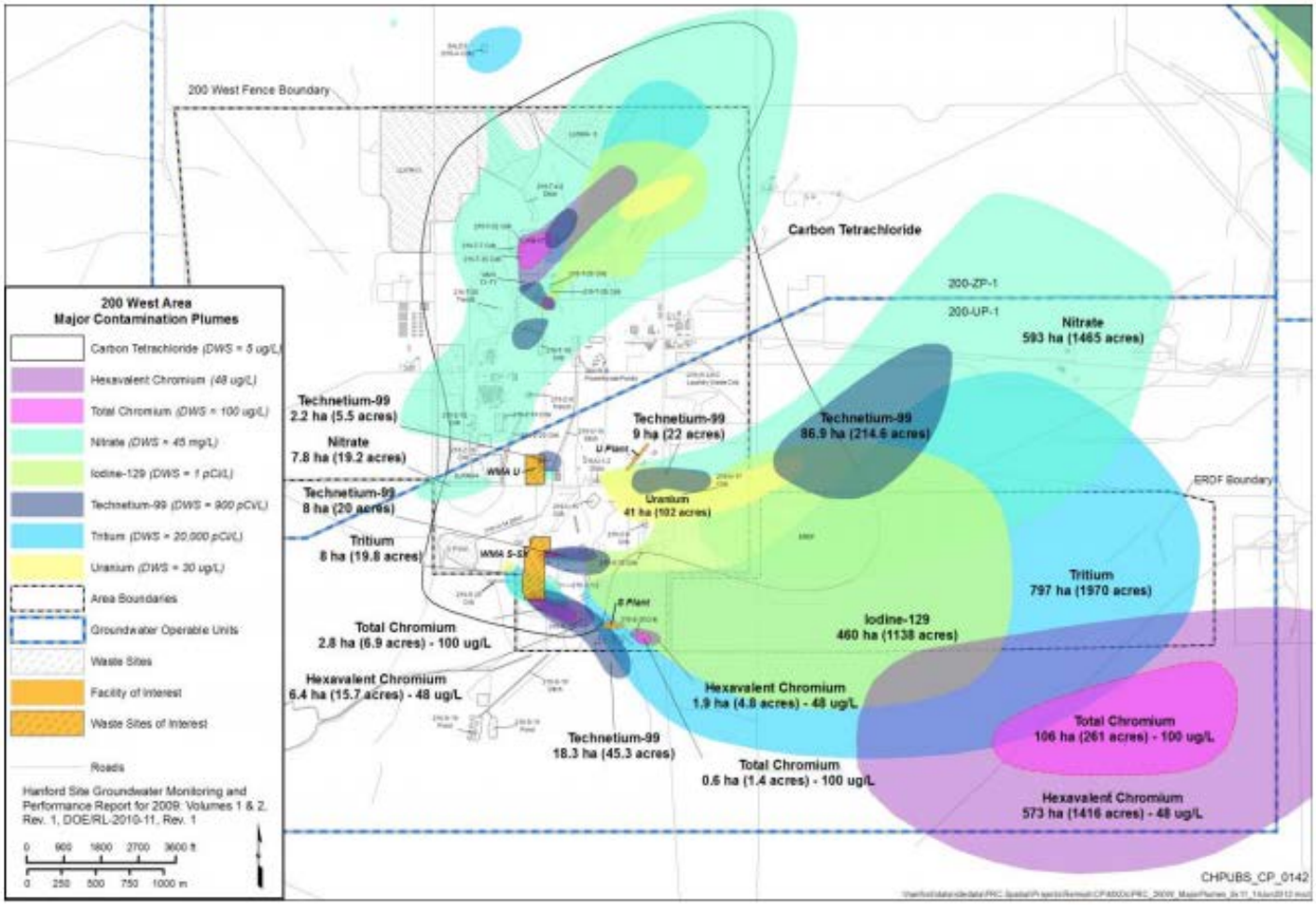
A waste product from nuclear fission, the radioisotope iodine-129 (^{129}I) is of environmental concern due to its long half-life (~16 million years), mobility, and hazardous potential to humans through bioaccumulation in the thyroid gland. The Hanford Site in Richland, Washington, contains two separate ^{129}I contaminant plumes over 1,500 acres with concentrations of ~3.5 pCi/L in groundwater samples, exceeding the federal drinking water standard of <1pCi/L. Consequently, understanding the mechanisms and contributors to iodine speciation is important in order to develop appropriate remediation strategies for iodine in non-aqueous environments such as the vadose zone, the subsurface environment located directly above groundwater. A lack of current remedial techniques is due to the complexity of microbial influence and presence of dissolved organics that impact the transport and speciation of ^{129}I. Although, iodide (I^-) is thermodynamically favored in the geological support material, based on current pH and Eh ranges at the Hanford Site, groundwater sampling of monitoring wells within the 200 West Area have found the dominant species to be iodate (IO_3^-) (70.6%) while organo-iodine (25.8%) and iodide (3.6%) were found in far lower quantities. Enrichments of Lower Ringgold sediment from the 200 West Hanford site allowed the isolation of microbial species metabolically involved in the speciation of iodine. Through a series of batch studies and spectrophotometric assays, isolates were found to couple nitrate (NO_3) reduction with iodate (IO_3), where iodate reduction was not observed in the absence of nitrate. Additionally, isolates able to oxidize iodide were also identified. Currently, analytical techniques are being developed to further understand the kinetics and enzymatic activity for both of these redox reactions. Ongoing research involves these isolates and their influence on iodine speciation in the presence of organics such as humic acid or lignocellulose.

BACKGROUND

At the Hanford Site in Richland, WA, numerous overlapping contaminant plumes of varying radioisotopes exist, a remnant of the mass plutonium production during World War II and throughout the Cold War. The majority of this waste produced was stored in underground tank systems at both the Hanford and Savannah River Sites (Aiken, SC) [1]. At least 200-square miles of groundwater beneath the site is contaminated, 80 of which are above federal drinking water standards. Radioactive iodine (^{129}I), discharged from former disposal cribs, in the 200 West Area of the Hanford Site is of particular environmental interest as the two existing ^{129}I plumes contain concentrations of ^{129}I at roughly 3.5 pCi/L and span an area of over two square miles (Fig 1). The federal drinking water standard is <1 pCi/L identifying

^{129}I. While iodine (I^{127}) is a necessary micronutrient for thyroid hormone production in humans, the radioisotope, iodine-129 (I^{129}), is thought to be potentially toxic through bioaccumulation in the food chain and in the thyroid gland of humans, leading to thyroid cancers. Currently, the high levels of this radioisotope in multiple DOE sites has spurred a desire to improve the efficiency of current remediation approaches for mitigating the risk associated with ^{129}I above regulation levels in groundwater.

Unfortunately, little is known about the biogeochemical cycling and mobilization of trace element iodine due to the complexity of microbial influences and interactions with dissolved organics in subsurface environments. This is especially prevalent in areas such as the vadose zone that are large, anhydrous, and difficult to sample. Due to the longevity of iodine-129, with a half-life of approximately 16 million years, and its mobility in subsurface environments, it is important to consider alternative and sustainable remediation techniques by first understanding the



biogeochemical cycling and speciation events of this radioisotope.

Fig 1. 200 West (200-UP-1 OU and 200-ZP-1 OU) Groundwater Plume Map

Iodine speciation at the Hanford Site

In the environment, iodine predominantly exists in five different forms: iodide (I^-), iodine (I_2), hypoiodous acid (HIO), iodate (IO_3^-), and organo-iodine species that are often associated with humic material. At the 200 Area of the Hanford Site, iodide (I^-) is expected to be the dominant iodine species based on the current ranges of pH and Eh in the complex subsurface geologic environments found on the Central Plateau [2]. However, through the groundwater sampling of monitoring wells within the 200 West Area and further speciation of these samples, iodate (IO_3^-) was found to be the dominant species (70.6%), while organo-iodine (25.8%) and iodide (I^-) (3.6%) were found in far lower quantities [2].

Figure 2 shows a schematic of the current understanding of environmental iodine cycling. When considering thermodynamics, the iodine species observed at the Hanford site are surprising and lead to interesting questions about the driving forces and mechanisms of these speciation events and the biogeochemical cycling of iodine. In this paper we will evaluate how the presence of organic material and microbial communities in subsurface environments influence the speciation of iodine.

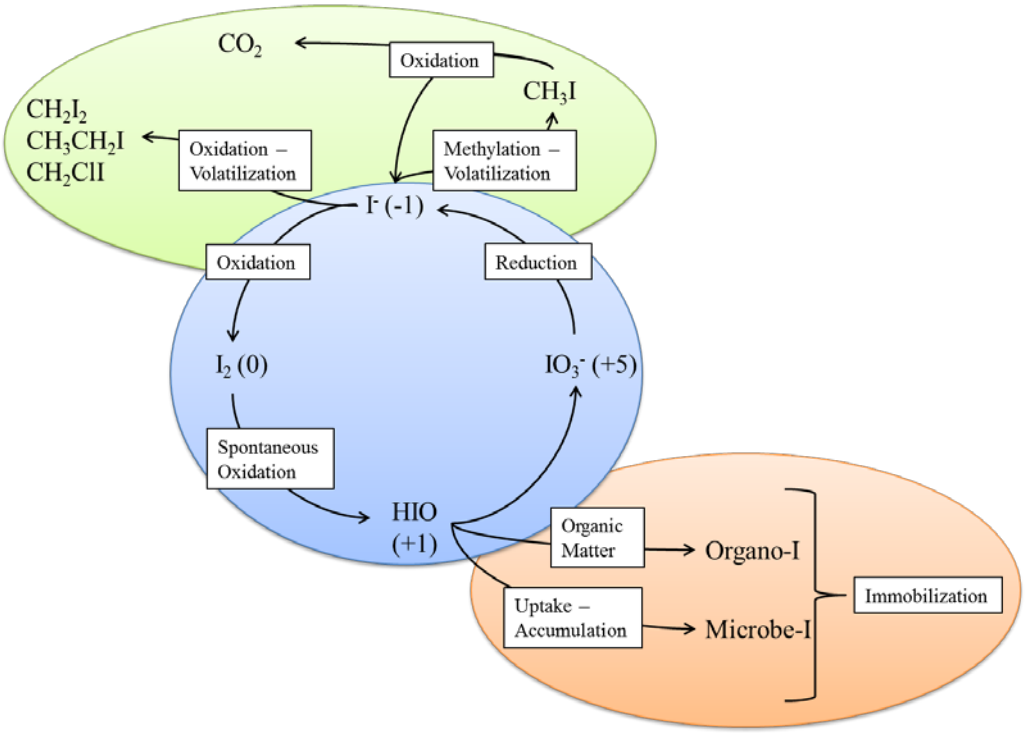


FIG 2. A current understanding of iodine cycling in the environment in gaseous (green), aqueous (blue), and subsurface (orange), environments.

Microbial influence on iodine speciation

In order to further understand the unexpected iodine speciation events observed at

the Hanford Site it is necessary to understand more specific and plausible interactions that may be occurring in this environment with the microbial community. Two reactions especially interesting are 1. The reduction of iodate in the presence of nitrate and 2. The oxidation of iodide by the laccase enzyme and the effect of naturally occurring organic materials on these oxidation events.

Currently, literature exists which describes the ability of microbial species to reduce iodate in the presence of nitrate, most often, in marine bacteria [3,4]. In these scenarios, it is suggested that this coupled reduction of iodate in the presence of nitrate is likely catalyzed by enzymes normally involved in innate nitrate reducing capabilities of these microbial species. Alternatively, other literature describes an iodate-reductase in the periplasm of *Pseudomonas stutzeri* SCT and suggested that induction of this reductase depended on only presence of iodate independent from nitrate [3]. Due to the overlapping iodate and nitrate plumes at the Hanford Site and literature describing the ability of microbial species to reduce iodate in the presence of nitrate, it is plausible that microbial species present in subsurface environments at the Hanford Site may also be capable of this metabolic activity or that this activity is relevant when considering future bioremediation mechanisms.

For decades, bacterial oxidation of iodide has been recognized as an important part of the iodine cycle in marine environments around the world by an extracellular peroxidase enzyme that, in the presence of polysaccharides, was shown to catalyze the oxidative reaction [4]. The enzyme laccase and other laccase-like multicopper oxidase (LMCO) have been shown to work in similar mechanisms especially in the presence of organic matter, most commonly by *Roseobacter* or *Proteobacteria* [5,6,7]. One microbe, *Roseovarius tolerans* is an iodide-oxidizing bacterium that was isolated from natural gas brines and found to oxidize iodide to iodine while many similar oxidation reactions of iodide to iodine have also been observed [8,9,10,11]. Additionally, a number of bacteria from the *Firmicute* family, isolated from the F Area at the Savannah River Site (SRS), have been shown to oxidize iodide to iodine enhanced by the addition of organic acids such as peroxy carboxylic acids [11]. If iodide oxidizing bacteria exist in ^{129}I contaminant plumes at the SRS similar to those of the Hanford site, it is useful to further explore these iodide oxidizing mechanisms tied t**o** laccase multicopper oxidases, especially when considering the unexpected speciation events present there.

Organic material's effect on iodine speciation and sorption

While microbial interactions with iodine species involve volatilization, oxidation and reduction of inorganic iodine species, accumulation of iodine species by microorganisms, and microbial mediated sorption of iodine in soil organic matter these processes, which are often considered individually, are connected in many ways (e.g., iodide oxidation leading to volatilization and formation of organic iodine compounds) [3,12-15,]. The aforementioned oxidase enzymes have been noted for both its participation in the oxidation of inorganic iodide and involved in the generation and catalysis of organic iodine compounds. One enzyme in particular, belonging to a group of lignocellulolytic enzymes that are involved in the breakdown of lignin and other aromatic compounds, has been found to catalyze iodination of

soil organic matter. Multi-copper oxidases, such as laccase, is involved in a process called “humification” in which polyaromatic humic acid structures become linked through phenoxy-free radical catalyzed polymerization [16]. When iodide is transformed to more reactive species such as I_2, HIO, or I_3, the reaction with laccase is more likely to result in the formation of organo-iodine [17]. In a study of soils in Chiba, Japan, soils with high laccase activity displayed a positive correlation with enhanced sorption of iodine [18]. In terrestrial ecosystems, such as peatlands, this process is thought to be key in storage of iodine in these environments. Iodine bound to the humic fraction of peat material can be stable for thousands of years, a process of immobilization important to understanding the overall fate and transport of iodine at Hanford [16,19]. Transformation studies of radioiodine and its speciation in organic-rich soils at the Savannah River Site indicated that the humic acid fraction of the soil organic matter accounted for up to 56% of the iodine in the soil [20]. The Hanford Site in Richland, WA has far lower levels of organics but demonstrates high levels of iodide oxidation and organo-iodine still accounts for up to 25% of iodine present. Because of the relationship shown between iodination of organic material and iodide oxidation, exploring how humics could attribute to the speciation of iodine at Hanford was explored.

METHODS

Isolation of microbial species at the 200 West Hanford Site

Bio traps filled with autoclaved sediment were placed in monitoring wells in the 200 West Hanford site and incubated for 50 and 150 days in high. medium, and low ^{129}I concentration plumes. Microbial species isolated directly from bio traps after these time periods were subsequently screened and chosen for their ability to oxidize iodide and reduce iodate in a series of batch microcosms prior to further experiments of bioremediation strategies for iodine-129.

Iodate reduction by *Agrobacterium* DVZ35

One isolate from bio trap material was chosen for it’s ability to reduce iodate. This microbe was dubbed DVZ35 (99% related to *Agrobacterium tumefaciens* based on partial 16s rRNA gene sequencing) and was subcultured in the presence of iodate for initial batch experiments [5]. In secondary experiments further studying the microbes ability to reduce iodate, DVZ35 was grown in the presence of iodate and nitrate. Some batches also received spiked nitrate concentrations or supplemented carbon sources to explore any possible changes this might have in iodate reduction rate or quantity. Using the same test conditions as DVZ35, two other species of *Agrobacterium* (currently designated as *Rhizobium*), *Agrobacterium radiobacter* NCIB 9042 (isolated from saprobic soil) and *Rhizobium radiobacter* TT3 (isolated from crown gall of apple seedling), were investigated to determine if other species similar to DVZ35 exhibit iodate reduction capabilities. Strains were obtained from ATCC.

Final iodine species concentrations were estimated by colorimetric assays used to estimate reduced iodate. Additionally, iodate, iodide, nitrate, and nitrite levels.

were determined by a Dionex DX-600 ion chromatograph equipped with an AS17 column with a gradient elution of 1 to 35 mM sodium hydroxide and measured using a conductivity detector. This method generated results that were 99% similar to results from colorimetric assays.

Oxidation of iodide by laccase multicopper oxidase

Four isolates from bio trap material chosen for their ability to oxidize iodide, dubbed DVZ2, DVZ19, DVZ21, and DVZ24, were used in a series of batch experiments evaluating growth and metabolic activity using optical density in four different carbon sources (glucose, xylose, acetate, and lactate). The levels of iodate, iodide, and iodine were explored using spectrophotometric colorimetric assays while laccase enzyme levels were observed using a spectrophotometric assay based on oxidation of ABTS, a common assay used to monitor multicopper oxidase activity.

Organics' influence on microbial adsorption of iodine

Utilizing soil and microbial communities from the same material incubated for 50 and 150 days in the high, medium, and low ^{129}I concentration plumes, the effect of humic acid on iodine speciation or iodate and iodide was explored in a series of batch studies. Samples of Humax® (JHBiotech, Inc., Ventura, CA) were obtained from personnel at Savannah River National Laboratory and were tested to compare humic material from different sources. Soil samples were used directly from Hanford biotrap material or supplemented with microbial nutrients to stimulate microbial communities in the sediments.

RESULTS

Iodate reduction by *Agrobacterium* DVZ35

For initial experiments with DVZ35, 200 µM iodate was reduced by 36.3% in cultures transitioning from aerobic to anaerobic growth and 47.8% in anaerobic cultures with 10 mM nitrate present, which were reduced 81.4% and 80.9%, respectively. Iodate was shown to be reduced by 84.0% and 69.2% in transition and anaerobic growth conditions, respectively, when nitrate was spiked into the growth media. DVZ35 reduced more than twice the amount of iodate than *Agrobacterium radiobacter* NCIB 9042 and *Rhizobium radiobacter* TT3. Without the presence of nitrate, DVZ35 did not reduce iodate.

Oxidation of iodide by laccase multicopper oxidase

Nine of the 14 bacterial isolates enriched from these Hanford sediments were capable of oxidizing iodide, four of which were chosen based on the highest iodide oxidation from initial batch experiments. The highest cell densities and relatedly, concentrations of oxidase enzyme were observed in glucose. In effect, higher levels of laccase enzyme resulted in increased oxidation of iodide. These findings indicate the first evidence of microbial iodide oxidation by bacteria isolated from the Hanford subsurface and support the hypothesis that microbial species, especially when

stimulated by a carbon source, are involved in iodine redox cycling.

Organics' influence on microbial adsorption of iodine

When the experiment was run with sediments that had been incubated in the ^{129}I plume for 50 days and the Sigma-Aldrich humic acid, on average, less than 9 µg/L of the 50 µg/L iodide that was added was adsorbed. Iodate adsorption was greater than 20 µg/L for all of the different treatments compared during the experiment. Adsorption results from these experiments demonstrated that adsorption of iodate was greater than adsorption of iodide under all conditions tested. Additionally, results demonstrated that addition of humic acid with microbial communities did not appear to improve iodide or iodate sorption from humic acid alone. However, in all scenarios, increased adsorption was observed when microbial growth was stimulated with a carbon source.

DISCUSSION

Iodate reduction by *Agrobacterium* DVZ35

The bacterium chosen from Hanford bio-trap material for its ability to reduce iodate in the presence of nitrate was designated as, *Agrobacterium* strain DVZ35 and was found to be closely related to (99% similar) *Agrobacterium tumefaciens* based on partial 16s rRNA gene sequencing. [5]**.** Gene expression by *Agrobacterium tumefaciens* supporting the denitrification of nitrate to nitrous oxide is linked to nitrate reductase (*nap*), nitrite reductase (*nir*) and nitric oxide reductase (*nor*). In lieu of *Agrobacterium tumefaciens'* ability to reduce nitrate it has never been described for its ability to reduce iodate [21,22]. However, it has been found that strain DVZ35 reduces iodate in the presence of nitrate. Additionally, both other *Agrobacterium* species, *Agrobacterium radiobacter* NCIB 9042 and *Rhizobium radiobacter* TT3 displayed the ability to reduce iodate, but they were not nearly as efficient as *Agrobacterium* strain DVZ35 which reduced more than twice the amount of iodate than these alternative species. In batch studies with this microbe, speciation of iodate to iodide and nitrate past nitrite was observed and confirmed using both colorimetric assays and ion chromatography only when nitrate was added to batch experiments. This suggests the potential mechanism of an iodate-reductase induced solely the presence of iodate is an unlikely mechanism for *Agrobacterium* strain DVZ35. Alternatively, this bacterium's iodate reducing capabilities are likely connected to a nitrate reductase based on DVZ3's ability to reduce iodate solely in the presence of nitrate.

These findings indicate that iodate reducing microbes are present in the groundwater at the 200 West Area at Hanford and are consistent with the overlapping iodine plume. It does not explain the high levels of iodate present in these subsurface environments if bacteria such as this are present. This leads us to ask what other influences may be in effect than those of bacterial species such as DVZ35. The significance of a groundwater bacterium (*A. tumefaciens* strain DVZ35) ability to reduce iodate under transition (i.e., slightly oxic to anoxic) and anoxic conditions in the presence of nitrate can help in understanding the speciation

mechanisms of iodine at the 200 West Area of the Hanford Site but also present very early possibilities when considering avenues to explore for bioremediation purposes.

Oxidation of iodide by laccase multicopper oxidase

The bio trap materials incubated at various ^{129}I concentrations in Hanford groundwater showed bacterial communities dominated by *Proteobacteria*. Nine of the 14 bacterial isolates enriched from these Hanford sediments were capable of oxidizing iodide, four of which chosen based on the highest iodide oxidation, dubbed DVZ2, DV19, DV21, and DV24. These four isolates were evaluated for growth and metabolic activity using optical density in four different carbon sources while enzymatic activity of oxidase enzymes was observed based on oxidation of ABTS, a common assay used to monitor multicopper oxidase activity. Evaluation of cellular optical density was the highest when microbes were grown in the presence of glucose as expected. Relatedly, the highest concentrations of oxidase enzyme present was observed in batches grown in glucose due to higher number of cells. In effect, estimated laccase enzyme presence correlated with increased oxidation of iodide. These findings indicate the first evidence of microbial iodide oxidation by bacteria isolated from the Hanford subsurface and support the hypothesis that microbial species, especially when stimulated by a carbon source, are involved in iodine redox cycling.

Organics' influence on microbial adsorption of iodine

Sediments exposed to ^{129}I plume for 50 and 150 days were compared for iodine transformation with and without the addition of humic acid from two different sources. In most cases that addition of humic acid, whether Humax® or Sigma-Aldrich, did not appear to change total iodine levels or increase sorption of iodine. While addition of humic acid did not improve iodine adsorption to the sediments overall, there was a significant difference in experiments where iodate was added as the iodine source as opposed to iodide. These results support previous research demonstrating that K_d values for iodate were higher than K_d values for iodide when adsorption isotherms were run with three different Hanford soils [23]. Additionally, stimulation of microbial activity appeared to increase adsorption of iodine species in all scenarios but further analysis of iodine speciation with addition of microbial growth supplements is required to determine the potential of remediation mechanisms utilizing natural organic matter to immobilize iodine in low organic carbon Hanford sediments.

CONCLUSION

In both avenues, microbial species isolated directly from the 200 West Area of the Hanford Site demonstrated redox capabilities in speciation of iodine namely, the oxidation of iodide and the reduction of iodate in the presence of nitrate. Whether or not organic material influences the speciation of iodine is inconclusive and requires further exploration. In effect, both aspects of iodine speciation in various subsurface environments must be well understood so appropriate and

productive remediation strategies may be developed in the future.

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