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Arsenic Contamination in Groundwater and its Bioremediation: A Study in Meerut, Western Uttar Pradesh, India

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Abstract: Arsenic, a metalloid that is present in the water, soil, rock, living organisms and atmosphere. Arsenic becomes free in the environment through the natural (erosion, biological, weathering and volcanic activity) and anthropogenic (mining, combustion of fossil fuels, agriculture and wood preservation) activities. Although anthropogenic activities cause local problems and the most severe environmental problems are caused by the mobilization of arsenic under ordinary conditions. High arsenic concentrations occur naturally in groundwater at certain places and result in serious health issues when this groundwater is supplied as drinking water. The present work describes a comprehensive analysis of the contamination of arsenic in groundwater within the Meerut district of Western Uttar Pradesh, India.

Keywords: Arsenic contamination; Bioremediation; Bioaccumulation; Biotransformation; Heavy metal; Arsenicosis; Ground water; Ganga Canal.

1. Introduction:

In today's context, water pollution is one of the serious attention problems that are being faced today. One of the major causes of environmental pollution is metal contamination in different resources. Arsenic exposure to human is mostly carried out through drinking water. Heavy metal contamination of drinking water is a serious problem that will probably become more outward in coming future, as scientists and engineers measure the true extent of the problem. Arsenic has been broadly spread in soil, minerals, water and biota [1]. Natural processes, as well as anthropogenic activities, are responsible for the discharge of arsenic into the environment. For instance, arsenic in soil comes from human inputs, such as sewage, insecticides, fertilizers, atmospheric outcome of smelters and fossil fuel combustion. Arsenic is a toxic element which comes into the environment moreover by natural phenomena (weathering, volcanic activity) or by anthropogenic activities [2]. The major inorganic arsenic present in aqueous, aerobic environments is in the form arsenate, although arsenite form is more dominant in oxygen deficient environment. Arsenic accumulation is a severe problem in many parts of the India, markedly Bangladesh, Uttar Pradesh and Bihar where arsenic contaminated groundwater is consumed by many people [3].The Groundwater pollution is reported in three district Varanasi, Gazipur and Ballia which are situated in western part of Uttar Pradesh, the upper-middle Ganga plain, India. This has been a rising global distress of arsenic contamination in drinking water (WHO). The research reports of recent years revealed that arsenic contaminated drinking water is a drastic disaster in the Gangetic alluvial zones of West Bengal and western parts of Uttar Pradesh [4, 5]. In the environment due to the presence of large quantity of arsenic, various bacterial strains have been developed having resistance capacity towards the arsenic compounds. In addition to natural geochemical contamination, groundwater pollution can occur in various ways such as by infiltration of polluted surface water, through leakage of pipelines, from landfill leachates etc. Groundwater contamination can be caused by three main categories (i.e., contamination by organic compounds, by microorganisms, and by inorganic pollutants, such as toxic heavy metals). The uncleanness of groundwater with metals embraces severe environmental problems because these are not biodegradable so cause severe adverse effects on human health.

The way by which heavy metals can be removed from contaminated water is bioremediation. In this process, a microorganism helps to remediate the waste like heavy metals. For example, the organic waste disposed into the environment in the form of household waste is acted upon by microorganisms which use them as food thereby degrading them. Similarly, some microorganisms can be targeted towards the hazardous pollutants in the environment, thus serving to restore the quality of the environment. This method requires knowledge of the strain of microorganisms that should be directed in the polluted sites. The use of microorganisms has been found advantageous for the treatment of wastes in controlled eco-friendly conditions. Either bacteria that already exist at the place of pollution or new strains of bacteria that have been proven effective for removal of the pollutants may be used for bioremediation [6].

2. Chemistry of Arsenic

As a metalloid, third-row, group V element, arsenic is sat down by nitrogen and phosphorus in the periodic table and thus has a surplus of electrons and unfilled orbitals that steady formal oxidation states from V, III, 0, -III under the normal pH conditions in aquatic systems. The predominant forms are inorganic oxyanions of trivalent arsenite (As(III)) or pentavalent arsenate (As(V)). The harmfulness of different arsenic species diverges in decreasing order as arsenite, arsenate, monomethylarsonate, dimethylarsinate. The electron configuration for neutral arsenic is [Ar] $3d^{10}4s^{2}4px^{1}\ 4py^{1}\ 4pz^{1}$, a state that supplies up to five valence electrons for participation in chemical bonding and empty *p*-orbitals for electron occupation. In context to electronegativity of arsenic, it is greater than nitrogen and resembles to phosphorous in most actions [7], as arsenic has a greater oxidation potential. Arsenic is having the mysterious ability to change color, behavior, reactivity, and toxicity which makes it elusive. For example, two arsenic sulfide minerals, red-colored realgar (As_4S_4 mineral) and bright yellow orpiment orange-yellow arsenic sulphide (As_2S_3), were described by the ancient Greeks, but they considered to be two entirely different substances [8].

In comparison to pentavalent state of arsenic, trivalent arsenic is more toxic by about 60 times. On the other hand, inorganic arsenic compounds are about 100 times more toxic than organic arsenic compounds. In aqueous solutions, arsenic forms the oxo-anions arsenite $H_3As^{3+}O_3$or commonly written $As^{3+}(OH)_3$] and arsenate,$H_3As^{5+}O_4$. Aqueous arsenic species in most natural waters (pH ~4–10) are the neutral species H_3AsO_3 for As^{3+}, and $H_2AsO_4^{-}$ and $HAsO_4^{2-}$ for As^{5+} [9, 10]. It is usually present in natural water resources at minimal concentrations (micromolar to nanomolar), its adsorption and desorption work on mineral surfaces play an important part in regulating its concentration in groundwater and surface water. Surface coverings of iron oxides on detrital minerals are supposed to be important sorbents of arsenic in groundwater aquifers and have been exploited as remediation technologies for arsenic elimination [11]. Arsine gas (AsH_3), which is formally the most reduced form of arsenic (analogous to ammonia, NH_3), is notable for its extreme toxicity. It is a volatile gas only which is slightly soluble in water. Because of its volatility, its importance in the environment as a microbiological end product may be overlooked [12, 13].

3. Sources of Arsenic in Groundwater

In many regions of India, the main source of drinking water is deep tube-well which can be contaminated by arsenic compounds (Fig 1) and causes serious issues due to arsenic poisoning to the large number of people mainly villagers. The groundwater nearer to the surface classically has spent a shorter time in the ground, so likely absorbs a lower concentration of arsenic; water deeper than 100m is unprotected to much older sediments which have even now been depleted of arsenic [14].

Pyrites, realger and orpiment are most important ores of arsenic. Arsenic is introduced in water and soil during the weathering of rocks and minerals followed by subsequent leaching and runoff. Based on arsenic geochemistry three probable mechanisms have been proposed for arsenic mobility in groundwater [15].

I. Mobilization of arsenic due to the oxidation of arsenic bearing pyrite minerals. Insoluble arsenic bearing minerals such as arsenopyrite (FeAsS) are rapidly oxidized when exposed to the atmosphere, releasing soluble arsenite, sulfate and ferrous iron [16].

II. Dissolution of arsenic rich iron oxyhydroxides FeO(OH) due to the onset of reducing conditions in the subsurface. Under oxidizing conditions, and in the presence of iron, inorganic species of arsenic are predominantly retained in the solid phase through interaction with FeO(OH) coatings on soil particles. The onset of reducing conditions in such environments can lead to the dissolution of FeO(OH) coatings [17,18].

III. Release of arsenic absorbed to aquifer minerals by competitive exchange with phosphate ions that migrate into aquifers from the application of fertilizers to surface soil [19].

The source of arsenic in groundwater of the lower Gangetic delta is accordingly considered to be the arsenic-rich sediments, transported from the Chotonagpur-Rajmahal highlands and deposited in sluggish streams under reducing conditions [20]. Continued extensive pumping triggered the reduction process by inducing the movement of groundwater having highly reduced degraded organic products.

Fig 1: Arsenic species commonly detected in the environment

4. Stretch of Arsenic Pollution in Meerut

The concentration of these minor constituents (arsenic compounds), including iron and nitrate is of concern as a large amount of groundwater is extracted by drilling, wells both in rustic and urban areas for drinking and irrigation persistence. There are sixteen state in India, i.e. Andhra Pradesh, Bihar, Delhi, Gujarat, Haryana, Jammu and Kashmir, Karnataka, Kerala, Madhya Pradesh, Maharashtra, Manipur, Orrisa, Punjab, Rajasthan, Tamilnadu, and Uttar Pradesh have already been acknowledged endemic to fluorosis [21]. Arsenic contamination occurs naturally in groundwater. However, indiscriminate pumping of water in Uttar Pradesh has increased the arsenic concentration beyond safe or permissible levels. The Upper Ganges Canal is the original Ganges Canal, which starts from the Bhimgoda Barrage near Haridwar navigates Meerut and Bulandshahar and continues to Nanu Canal in the Aligarh district, where it diverges into the Kanpur and Etawah branches (Fig 2).

The study of Meerut, Uttar Pradesh

Topography: The metropolitan city of Meerut is one of the important industrial towns of western Uttar Pradesh. It is situated about 85 km from Delhi. The metropolitan city occupies an area of about 142 km^2 and lies between 28°57’ to 29°02’ N latitude and 77°40’ to 77°45’ E longitude. The metropolitan city of Meerut is a part of the Indo-Gangetic plains.

Population: As per the 2021 census, the population of Meerut is 1,728,000 and it's increasing day by day.

Climate: Meerut city has a moderate type of climate. From October to March a cool, dry winter season, from April to June a hot, dry summer season and from July to September a warm rainy season is observed. During June to September, in the monsoon season average annual rainfall is about 1000 mm, and significant diurnal variations in hydro-meteorological parameters like precipitation, temperature and relative humidity also exist.

Fig 2: Arsenic Affected Areas in Uttar Pradesh

Geology and soil type: The area is lacking of any significant relief topographies and is a collection of unconsolidated alluvial deposits. The advancement of land surface lies at 220 m from mean sea level. Meerut City is a measure of Indo-Gangetic plains, which is mostly composed of Pleistocene and sub-recent alluvial sediments elated and dumped by river action from the Himalayan region. Lithologically, sediments consist of clay, silt and fine to uneven sand. The deposits of sandy horizons of varying thickness are the main source of groundwater in the area. For growing wheat, sugar cane and vegetables this type of soil is very fertile. Conversely, along the sandy river course, fruit orchards are also common. Despite enormous surface and groundwater resources, western Uttar Pradesh faces a problem scarcity of water for irrigation in dry months, when groundwater is extensively used for irrigation. The varying lithology of the geological formations considerably influences the groundwater conditions in all alluvial parts. The main source of water, which sustains the groundwater body in fine to bristly grained sands, is rainfall; the other sources of groundwater replenishment are intrusion from the river, return discharge from irrigation and inflow from neighboring areas.

Water Resources: The two important rivers Yamuna and Hindon lie from north to south of this area. The stream Ganga and the river Yamuna flow from the eastern and the western boundaries of the area. However, the other two rivers flowing in the area are Kali and Krishna. At a distance from these rivers, the Upper Ganga Canal also drains the zone. In certain of the above mentioned recharge sources, rainfall is probably the most important. The rate and amount of infiltration of rainwater depend on several factors. In this area, shallow and deep tube wells are the common source of groundwater. Burrowed wells have become almost rare in the region; the surface water in the region is fully utilized. Regarding the occurrence of groundwater, the principal groundwater reservoir in the region is unconsolidated alluvial deposits. Recent studies in the region indicated that the top aquifers are unconfined in nature and deeper aquifers are semi-confined in nature.

5. Arsenic Biotransformation and Metabolism

In the environment and biological systems, a large number of arsenic compounds have been discovered. During the biogeochemical cycle, arsenic is converted and transported to two different chemical forms in the environment. The toxicity of arsenic is dependent on its total amount or its chemical species [20]. For oral administration, the moderate lethal doses (LD_{50})to mice are as follows: 3mg/kg for AsH_3,14 mg/kg for arsenite [As(III)], 20 mg/kg for arsenate [As(V)], 700-1800mg/kg for monomethylarsonic acid (MMA), 700-2600 mg/kg for dimethylarsinic acid (DMA) and 10000 mg/kg for arsenobetaine (AsB) and arsenocholine (AsC). As(III) and As(V) inorganic species are thus more toxic as compared to their methylated forms, MMA, and DMA [22, 23].

Studies revealed that arsenic is absorbed by cells in the form of arsenite is transported by aquaglycoporins [24] and arsenate form may be transported by the phosphate transporter [25]. The consumed arsenic can also be broken down into various composite species in the human body. Arsenic compounds are absorbed after methylation followed by secretion in the urine. Methylation of arsenic compounds happens to some extent to lower the toxicity of ingested inorganic arsenic species. This excretion pathway can therefore protect the body from being damaged upon exposure to arsenic compounds. Methylation primarily occurs in the liver, although it can also take place in a smaller amount in some other organs such as the kidneys and lungs. During the process, inorganic arsenic is methylated by alternating reduction of As(V) to As(III) and the addition of a methyl group from S-adenosylmethionine. Glutathione and possibly other thiols, serve as reducing agents [26]. Methylated arsenic species are excreted much faster than arsenical inorganic species [27].

The major metabolites of inorganic arsenic species in the human body is MMA and DMA (Fig 1). Most of the arsenic in the body are excreted about 10-30% as inorganic forms, 10-20% as MMA (III) & (V), and 60-80% as DMA(III) & (V), although their ratios may differ in different populations [28]. The intermediate reduced forms of the methylated metabolites, i.e., MMA(III) and DMA(III) have been detected in human urine. Methylated trivalent metabolites are highly reactive and more potent inhibitors of GSH reductase and thioredoxin reductase enzymes [29,30] in comparison to arsenite or arsenic pentavalent metabolites. Methylated trivalent arsenic species are the only arsenic compounds that were observed to damage naked DNA and thus considered to be genotoxic [31]. This also means that the biotransformation process of arsenic in the human body should not exclusively be regarded as a simple detoxification process.

6. Arsenic Bioaccumulation

Arsenic poisoning or arsenicosis is a condition caused by the ingestion, absorption or inhalation of higher levels of arsenic. Arsenic is a natural semi-metallic chemical that is abundant all over the world mainly in groundwater. Arsenic poisoning can cause major health complications even death if not treated timely. Because of the risks involved, precautions are required for the protection of populations and workers at risk of arsenic poisoning. Arsenic can be accumulated in different forms in plants, animals and human beings [32].

6.1. Bioaccumulation in plant

As(V) is taken up and metabolized by the plant through phosphate transporter channels [33]. Understanding how arsenic is taken up by plants and subsequently accumulated in different plant parts is essential to estimate the risks posed by arsenic contaminated soils to humans and wildlife populations. Arsenic is found in the environment in both organic and inorganic forms. Arsenate [As(V)] and arsenite [As(III)] are the inorganic and more predominant phytoavailable forms of arsenic in soil solution, as well as the most common arsenic species in crop plants. Organic species such as MMA and DMA are present in the environment to a lesser degree [34, 35]. Arsenate and phosphate are chemically similar and arsenate can act as a phosphate analog, thereby being transported into the cell via the phosphate transporters [36]. Through a series of As(V) and Inorganic phosphate (Pi) transport studies, it was concluded that suppression of the high affinity Pi uptake system decreases by As(V) uptake [37-40]. In contrast, As(III) and undissociated methylated arsenic species are transported through the nodulin 26-like intrinsic (NIP) aquaporin channels [41]. Both inorganic forms of arsenic are highly toxic as As(V) interfering with phosphate metabolism (such as phosphorylation and ATP synthesis) and As(III) binds to vicinal sulfhydryl groups of proteins affecting their structures or catalytic functions. Exposure to As(V) generates reactive oxygen species (ROS) in plant tissues and induces oxidative stresses such as lipid peroxidation [42, 43]. Exposure to As(III) also enhances the activity of several enzymes involved in the antioxidant responses [44, 45]. Some genes or enzymes involved in glutathione synthesis and metabolism as sequestration are upregulated in rice seedlings exposed to As(V) [46]. This probably reflects a higher demand for GSH under arsenic stress. Arsenate is readily reduced to As(III) in plants, which is detoxified by complex formation with either thiol-rich peptides for example reduced glutathione (GSH) or phytochelatins (PCs) or vacuolar sequestration or a combination of both. To reduce the arsenic uptake through the consumption of contaminated plant foods, the mechanisms of arsenic uptake and detoxification is prerequisite to understanding. PCs are GSH derived peptides that chelate arsenic and participate in the first step of arsenic detoxification.

In higher concentrations, arsenate in plants inhibits photosynthesis through the interference of the pentose-phosphate pathway [47]. Arsenite(III) penetrates the plant cuticle to a greater degree than arsenate(V) and generally results in the loss of turgor pressure [48].

6.2. Bioaccumulation in animals

Chronic exposure to arsenic involves a biotransformation process leading to the excretion of methylated metabolites, such as mono-methyl arsenic and dimethyl arsenic as well as the parental inorganic species (arsenite and arsenate). Carcinogenic mechanisms could include blocking of DNA repair, stimulating angiogenesis, altering DNA methylation patterns, dysregulation of cell cycle control, and introducing of aneuploidy and blocking apoptosis. The generation of reactive oxygen species (ROS) is one of the possible mechanisms suggested for arsenic toxicity [49]. Arsenic metabolism is characterized by two main types of reactions: (i) reduction reactions of arsenate to arsenite and (ii) oxidative methylation reactions in which arsenite is sequentially methylated to form mono-, di- and trimethylated products using S-adenosyl methionine (SAM) as the methyl donor and glutathione (GSH) as an essential co-factor [50].

6.3. Bioaccumulation in the human body

Several studies in humans indicated both soluble arsenic compounds also known as the organic arsenic species are well absorbed across the gastrointestinal tract [51]. The As(III) may undergo enzymatic methylation primarily in the liver to form MMA and DMA. However, ingested elemental arsenic is poorly absorbed and largely eliminated in unchanged form. Soluble arsenic compounds are rapidly absorbed from the gastrointestinal tract. Arsenic(V) and organic arsenic are rapidly and almost eliminated via the kidneys [52-54]. Inorganic arsenic may accumulate in skin, bone, liver, kidney and muscles [55]. Its half-life in humans is between 2 and 40 days [56]. Inorganic arsenic is eliminated from the body by the rapid urinary excretion in an unchanged state both in the trivalent and pentavalent forms of arsenic or by sequential methylation to MMA and DMA in both III and V valenced states [57, 58]. Limited short-term studies on humans indicated that the capacity to modulate inorganic arsenic is progressive, but not completely saturated when daily intake exceeds 0.5 mg [59]. Arsenic concentrations in the hair, nail or urine are established biomarkers of arsenic contamination.

7. Bioremediation of Arsenic by Microorganisms

The microbial activity could reduce the arsenic through absorption, biomethylation, complex formation, and oxidation-reduction processes. Microorganisms have developed the biochemical mechanisms to utilize arsenic, either as an electron acceptor or electron donor for anaerobic respiration or to perform chemoautotrophic fixation of CO_2 into cell carbon, respectively. The microbes belong from both prokaryotes bacterial and archaeal domain which reduce dissimilatory arsenate. It uses arsenate (As(V)) as an electron acceptor and reduces it to arsenite (As(III)). ATP synthesis is inhibited by disruption of phosphorylation reaction by the As(V). CO_2 is used as a carbon source and arsenite as an electron source by chemoautotrophic arsenite oxidizers and microorganisms. They oxidize arsenite to arsenate for energy in the presence of arsenate reductase enzymes. Although, heterotrophic arsenite oxidizers utilize oxygen in place of CO_2 as an electron acceptor and oxidize arsenite to arsenate. Arsenate resistant microbes reduce arsenate into arsenite, which allows the elimination of toxic form from the cell [60-62].

Advantages of arsenic bioremediation

The various advantages of bioremediation are listed below-

- In the *in situ* treatment, the pollutants are removed on the site itself by the microbes thereby decreasing the need for excavation and transportation of the contaminated medium.
- In the case of physical and chemical processes of remediation, the disposal of the removed pollutants is a problem as there is a chance that they might again cause pollution. Whereas, in the case of bioremediation, the pollutants are consumed and degraded into harmless gases and water. Hence there is no release of harmful secondary byproducts in the environment.
- Once the pollutants are cleaned up by the microbes, the microbes die due to the lack of the pollutants which has been serving as their food. Thus, any harmful effects of the microbes are also ruled out.
- Aesthetically acceptable.

8.0 Conclusion

The atomic absorption spectroscopy is used to examine and measure heavy metals. It is a very sensitive method for the determination of metals, as it can detect metals even in a concentration of less than one part per million in a sample solution. It is essential to address the pollution issues that have been caused in the past as soon as possible to safeguard natural resources. One solution for remediation is the use of microorganisms which is an environmental friendly process and is well suited to remediate pollutants without causing secondary pollution. It is a sustainable approach to be expanded in future for the both aspects of remediation of the contaminated sites and safe agricultural creation in Arsenic contaminated sites.

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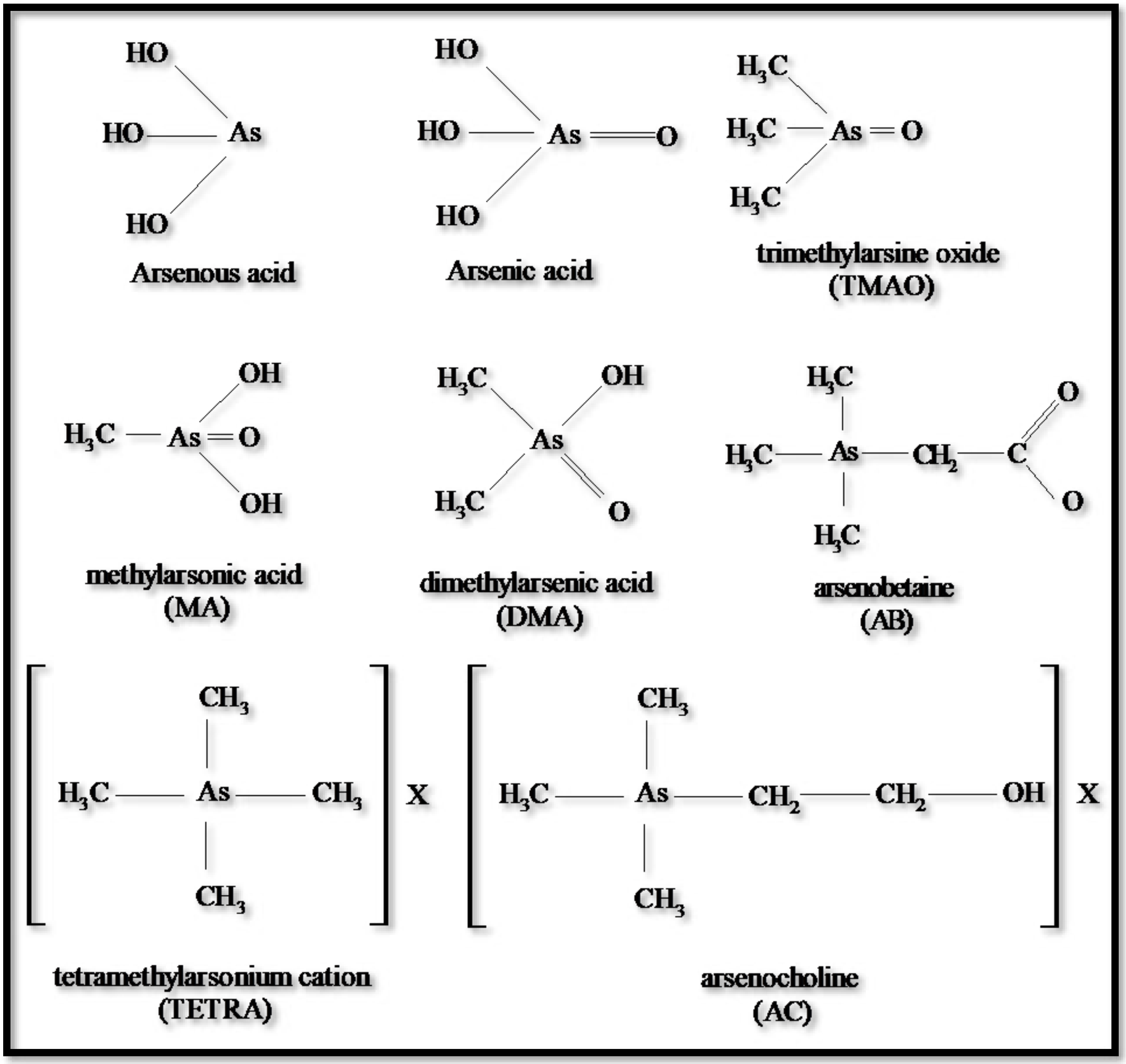

Fig 1: Arsenic species commonly detected in the environment

Fig 2: **Arsenic Affected Areas in Uttar Pradesh**