Bulletin A2.9432.4

PUREFLOW © FILTRATION SYSTEMS

THE REMOVAL

OF

ARSENIC

FROM

POTABLE WATER

FOREWORD

Arsenic's evil reputation is manifested in its old alchemists' symbol: a coiled serpent ready to strike. Considering arsenic's eminence as the preferred homicidal agent of detective novels, it is not surprising there is a great deal of anxiety over its possible presence in water, among other substances, we use on a daily basis. With this bulletin, Pureflow© hopes to provide some facts on arsenic; its occurrence, normal routes of exposure, its health effects, and finally, the various unit operations available for its removal from water.

I am an evil, poisonous smoke...
But when from poison I am freed
Through art and sleight of hand,
Then can I cure both man and beast
From dire disease ofttimes direct them;
But prepare me correctly, and take great care
That you faithfully keep watch over me;
For else I am poison, and poison remain
That pierces the heart of many a one

(Valentini 1694)

OCCURRENCE AND USES OF ARSENIC

In the earth's crust, arsenic ranks as the twentieth most common element. In the human body it ranks as number twelve (National Academy of Sciences). So, obviously, it is quite ubiquitous. Arsenic is present in all soils, ranging from 0.1 to 40 milligrams per kilogram (parts per million), and averaging between 1.5 - 2 milligrams per kilogram. The arsenic content of a soil is a function of its geologic history. Sandstones, shales and coals associated with uranium mineralization in Utah, Colorado, Wyoming and South Dakota show higher than average arsenic content. This is also the case of phosphorites found in Florida, Montana and Idaho, as well as in British Columbia, Canada, southern Australia, the Far East, notably Taiwan, Africa, and parts of Europe.

Arsenic is found in at least 245 mineral species. In base-metal mining and refining operations, arsenic, occurring along with lead, silver, zinc, copper and tin in their sulfide ores, is considered an expensive nuisance. Arsenic toxicity is also problematical near base-metal smelters. Regions of volcanic or geothermal activity usually show elevated arsenic concentrations.

Arsenic has had, and still has, many uses. In metallurgy the inclusion of 0.5 - 2% arsenic improves the sphericity of lead shot (McKee and Wolf). Inclusion of up to 5% arsenic in lead-based bearing alloys improves their high-temperature mechanical properties. Arsenic is added to other alloys to increase their hardness and heat resistance. High purity (99.999%) arsenic is used in semiconductor technology. Arsenic is used in the manufacture of glass and ceramics. Arsenic's major use has been, and remains, in farm chemicals. Approximately 97% of arsenic produced enters end-product manufacture in the form of "white arsenic," the remaining 3% as the metalloid for metallurgical purposes.

Agricultural uses accounted for about 81% of total arsenic consumption in the U.S. in 1973. Arsenic oxide is the raw material for arsenical pesticides. Compounds like Paris Green (copper acetoarsenite) used to be popular insecticides for orchard use, but are of lesser importance today. Likewise, lead and calcium arsenate have been used extensively in the U.S. for insect control on fruits, tobacco, cotton, and even some vegetables, but current use is slight.

Sodium arsenite came into use as an insecticide between 1920-30, mainly as a bait and as livestock dip. Organic arsenicals are used extensively as selective herbicides for the postemergence control of crabgrass, Dalliograss, and other weedy grasses in turf. Disodiummethanearsonate (DMSA) has been used extensively for cotton weed control. For example, in 1963 71,000 acres in Mississippi were treated with DMSA as a directed spray, and more than 329,000 acres were treated in 1964.

Arsenic acid is used extensively as a cotton desiccant in Texas and Oklahoma to prepare cotton plants for mechanical stripping. It depletes the leaves and other plant tissues of moisture, thus facilitating reaping. This is a more efficient cotton harvesting method than mechanical picking. It is estimated that in Texas alone, over two million acres of cotton were treated with arsenic acid. Arsenic, along with copper, is used as a wood preservative in such things as telephone poles and pilings. Additionally, there are uses for arsenic in human and veterinary pharmaceuticals.

EXPOSURE

The most significant and common route of exposure to arsenic is through food (Pontius, et al.). The average daily per capita intake of arsenic from food has been estimated at 50 micrograms, depending on geographical location and dietary habits. The daily per capita intake of arsenic from institutional food containing no seafood has been estimated at 400 µg (µg = microgram = 1/1000 mg) or 0.4 mg/day (Schroeder and Balassa). Saltwater fishes average about 10 milligrams of arsenic per kilogram of tissue or 10 parts per million (ppm), with mollusks and crustaceans at even higher levels (National Academy of Sciences). Four ounces (approximately 100 grams) of shrimp may contribute between 127 and 4,160 µg of arsenic, while the consumption of the same amount of tuna may add from 71 to 460 µg. Fresh water fishes, by comparison, average from 1-3 ppm.

The ingestion of arsenic from drinking water can also represent an important route of exposure. The United States Environmental Protection Agency (EPA) bases most of its Maximum Contaminant Levels (MCL) on the ingestion of two liters of water per day per person. Water at the previous MCL for arsenic would thus have contributed 100 µg. (0.1 mg) of arsenic to the average individual each day. The newly adopted MCL reduces this daily intake to 20 µg (0.02 mg) per day.

Arsenic occurs in waters worldwide. About 3% of water analyses show arsenic to be present at concentrations greater than 50 µg/l (50 parts per billion). Thermal waters in Wyoming, California, Nevada, Alaska, and Iceland are reported to range from 20 - 3,800 µg/l, and water from the Waitopu Valley of New Zealand is said to contain up to 276,000 µg/l. As might be expected, arsenic levels are higher in well waters than in surface waters (National Academy of Sciences). Well waters have been reported as ranging from "none detected" (which until the quite recent development of more sensitive methods and instrumentation could mean less than 5 or 10 parts per billion) to 1,700 micrograms per liter (parts per billion, or 1.7 parts per million) and higher. Some well waters on Taiwan are reported to contain up to 1,800 µg/l, and have been implicated in "Blackfoot Disease" and in skin cancers occurring in the southwest part of the island. In the Aomori Prefecture of Japan, groundwater is reported to contain arsenic up to 3,950 µg/l, while spring waters from the Kamchatka (former USSR) Peninsula are reported to contain approximately 1,000 µg/l (National Academy of Sciences). In one national survey, arsenic has been found in 39% of drinking water samples. For greater statistical occurrence detail, and estimated treatment costs for compliance with the variously suggested MCLs, please consult the September 1994 issue of the *Journal of the American Water Works Association*. It has been calculated, based on these national occurrence figures, that the average daily per capita contribution of arsenic from drinking water in the United States of America is about twenty micrograms.

Low levels of arsenic can be found in ambient air, probably as the result of the burning of fossil fuels, the smelting of ores, and other industrial uses (National Academy of Sciences). The Occupational Safety and Health Administration (OSHA) has recognized this, and has set a Threshold Limit Value for arsenic exposure from air at 500 micrograms per cubic meter, 200 micrograms per cubic meter for the (more toxic) arsine gas.

HEALTH EFFECTS

At low doses arsenic may be essential, or at least beneficial, for good health in humans, though no conclusive evidence to the element's essentiality has been documented.

Acute, high-dose, oral exposure to arsenic typically leads to gastrointestinal irritation, extreme thirst, hypotension (low blood pressure), and convulsions (National Academy of Sciences; Pontius, et al.). Death, when it occurs, is usually from cardiovascular collapse. The estimated lethal dose in adult humans is 70 - 180 milligrams, or between 1,000 to 2,600 micrograms of arsenic per kilogram of body weight.

While high-dose exposure is very unlikely, long-term exposure to 10 - 100 micrograms per kilogram per day commonly leads to neurological (nerve), dermatological (skin), or hepatic (liver) toxicity. Long-term exposures below 1 microgram/kg/day have not been observed to cause any detectable effects (National Academy of Sciences).

Evidence exists, however, that long-term exposure to arsenic increases cancer risk. When the exposure is by inhalation (as in some industrial exposures) the effect is an increased risk of lung cancer. When the exposure is by ingestion, the clearest effect is a greater risk of skin cancer, but there is evidence the risk of internal cancer is also increased.

REGULATORY BACKGROUND

First regulated at 0.05 milligrams per liter (mg/l) or (50 ppb) by the U.S. Public Health Service in 1942, the National Interim Primary Drinking Water Regulations (NIPDWR) affirmed this level in 1975 as the Maximum Contaminant Level (MCL) (McKee and Wolf). Arsenic has been classified as a carcinogen (cancer causing agent), thus the Maximum Contaminant Level Goal (MCLG) must, by law, be set at zero. The Environmental Protection Agency (EPA) published the final Arsenic Rule in January 2001 lowering the maximum contaminant level (MCL) from 50 µg/l to 10 µg/l (0.050 mg/L to 0.010 mg/L). States must adopt the Federal MCL within two years of promulgation. The effective compliance date is February 2006. Community Water Systems exceeding 5.0 ppb (one half of the arsenic MCL) must notify their customers in their annual Consumer Confidence Reports published after March 23, 2002.

CHEMISTRY AND TOXICITY

Because of its appearance, arsenic is considered a metalloid, and is not a true metal. Arsenic exists in four valence (oxidation) states. Valence is defined as the combining power of an element with other elements or compounds. These four states are:

1. Arsenate (As^{+5})
2. Arsenite (As^{+3})
3. Elemental arsenic (As^{0})
4. Arsine gas ($As^{-3)}$

In water, if arsenic is present, it is predominantly in the As^{+5} (arsenate) form. Arsenite (As^{+3}), when present, is readily oxidized to arsenate in aerobic waters at pH values above 7.0. Conversely, arsenate (As^{+5}) can be reduced to arsenite at low pH values.

Of the two predominant species, the trivalent form, arsenite (As^{+3}) is considerably more toxic than the pentavalent (As^{+5}) form, although it has been demonstrated that As^{+5} is better absorbed by the human body because it tends to react less with the membranes in the gastrointestinal tract (National Academy of Sciences).

Arsenic metabolism is a two-step process: arsenate (As^{+5}), entering a cell, is reduced to arsenite (As^{+3}), which is then methylated to arsenate. The methylation is a detoxification step, since neither MMA or DMA are as toxic to the system as is inorganic arsenic, although the chronic effects of MMA or DMA are not known (McKee and Wolf).

ARSENIC REMOVAL PROCESSES

The EPA will identify a number of treatment options for the removal of arsenic as Best Available Technology (BAT). Listed and discussed below, in no particular order of applicability, preference, efficiency or treatment cost, are some of the processes under consideration.

REVERSE OSMOSIS

GRANULAR FERRIC HYDROXIDE

ION EXCHANGE

NANO-FILTRATION

ELECTRODIALYSIS

ACTIVATED ALUMINA

LIME SOFTENING

COAGULATION/FILTRATION

COAGULATION ASSISTED MEMBRANE FILTRATION

REVERSE OSMOSIS

This membrane process is primarily designed for the desalting of saline or brackish waters by the application of hydrostatic pressure (Montgomery). This overcomes osmotic pressure and drives the water to be treated through a semi-permeable membrane designed to allow passage of water, but not of dissolved contaminants. The process requires expensive and fragile membrane stacks, either cellulose-acetate or thin film composite. Cellulose-acetate membranes can be operated at up to 400 psi, but are subject to biological attack and hydrolysis. They also allow the salt passage to double after a service life of about 3 years. The more expensive thin film composite membranes are capable of the same or greater flux rate, but at half the applied pressure. These allow only a less than 30% increase in salt passage after 3 years. Both require considerable pre-treatment to prevent scaling, plugging, and colloidal or biological fouling of the membranes.

Since the recovery of product water, as a percentage of feed water, is a function of applied hydrostatic pressure (up to 400 psi or more), the process tends to be quite energy intensive. Most reverse osmosis plants are designed for 75-80% recovery, i.e., up to 25% of the flow must be disposed of as a concentrated, possibly hazardous, waste (Montgomery). Reverse osmosis is quite capable of the removal of arsenic to very low levels. Process operation and maintenance costs, as well as labor intensity, will tend to rule out its application for all but small volume treatment systems.

GRANULAR FERRIC HYDROXIDE (GFH)

Granular Ferric Hydroxide (GFH) is an absorptive medium designed for the removal of arsenic, phosphates, chromium and other heavy metals. Raw water pH and contaminant concentration (e.g. iron, manganese, chromium, organics, silica, phosphates, etc.) determine the life of the media. Preoxidation of raw water is not required and both arsenic valence states are removed. Periodic backwashing of the media is required depending on raw water quality. GFH is presently classified as a non-regenerative media that must be removed from the filter vessel when exhausted, and replaced with new media. Research is being conducted to determine the feasibility of regenerating GFH. Spent GFH media is disposed of in a landfill. TCLP testing is a federal test commonly used to determine if the spent media is classified as hazardous or non-hazardous. State regulations should be consulted to verify acceptable testing methods and media disposal requirements.

ION EXCHANGE

Capable of complete removal of all dissolved matter, including arsenic, from water, this process is widely used for the production of deionized water. One great advantage of Ion Exchange is that no pH adjustment is necessary. Recent advances in resin technology have replaced the weak-base anion resins with strong-base ones. Pentavalent arsenic (As^{+5}), being present as the divalent anion $HAsO_4^{-2}$, appears to have a greater affinity for this type of resin. Strong base resins permit the use of ordinary sodium chloride brine for regeneration, and eliminate the need for the use of strong acids. Regeneration is a slow and water-intensive process. Typically, columns are rinsed with 1-2 bed volumes to displace the regenerant. This is followed by a fast rinse for about 10 minutes at design flow. The used regeneration brine, containing arsenic, is a hazardous waste and must be disposed of accordingly.

ACTIVATED ALUMINA

"Activated alumina has a long history of use as an adsorptive treatment technology for arsenic removal. The media is a byproduct of aluminum production. It is primarily an aluminum oxide that has been activated by exposure to high temperature and caustic soda. The material is extremely porous and has a high average surface area per unit weight (350 m^2/g). The capacity for arsenic removal by activated alumina is pH-dependent, with the maximum removal capacity achieved at pH 5.5. Adjusting the pH of the source water, therefore, provides removal capacity advantages. As the pH deviates from the 5.0 – 6.0 range, the adsorption capacity for arsenic decreases at an increasing rate. Process demonstrations have shown that arsenic removal capacity has been reduced by more than 15% at pH 6.0 compared to that of pH 5.5 (Rubel, 1984).

Fluoride, selenium, and other inorganic ions and organic molecules also are removed by the same pH adjustment activated alumina process. The process, however is preferential for arsenic at the optimum pH level of 5.5. Other ions that compete with arsenic for the same adsorptive sites at other pH levels are not adsorbed in the pH range of 5.0 – 6.0. Included are silica and hardness ions that are adsorbed in the pH range of 7 – 10.

Activated alumina either can be regenerated or can be replaced with new media when the selected breakthrough point is reached. At the optimum pH for arsenic removal, fluoride, selenium, some organic molecules, and some trace heavy metal ions are adsorbed; however, these are also completely regenerated along with arsenic. Because these ions compete for the same adsorptive sties with arsenic, their presence might deplete the alumina capacity for arsenic. When excess fluoride and arsenic are present in the water supply, a special treatment technique is required (Rubel and Williams, 1980).

The adsorptive capacity of many adsorptive media, particularly activated alumina, is pH sensitive; removal capacity increases with decreasing pH. Employing pH adjustment, therefore, generally provides cost advantages regardless of whether the media is regenerated or replaced. Because the pH adjustment chemicals are usually the same chemicals that are used for regeneration, it is generally advantageous to couple regeneration with pH adjustment systems when the media can be regenerated." Rubel (2003)

LIME SOFTENING

Excess Lime Softening is the addition of a sufficiently high lime dosage, at times in excess of 1 gram per liter, to obtain a pH greater than 11.5. It has long been used for the removal of calcium and magnesium carbonate hardness, and is also capable of the removal of approximately 90% of any arsenic that may be present (Dutta and Chauderi).

The removal of trivalent arsenic appears to be dependent upon the precipitation of magnesium hydroxide $\{Mg(OH)_2\}$. The addition of powdered activated carbon, though apparently not absolutely necessary, appears to enhance removal efficiency. While this is an old tried and true process, and while apparently quite capable of arsenic removal, the process remains plant and chemical intensive, requires the recarbonation of the water, and produces large volumes of sludge. For these reasons, unless there is also a demonstrated need for softening, the process may not be deemed economically viable.

COAGULATION-FILTRATION

Conventional coagulation/flocculation/filtration, using iron salts, is effective in the removal of up to 90% of arsenate (As^{+5}) at pH levels of 7 or less (Edwards). Above a pH of 7 flocs from iron salts effectively remove arsenic. Iron coagulants will remove about 50% of trivalent arsenic (As^{+3}) (Montgomery). Thus, it is very important to fully oxidize As^{+3} to As^{+5} with chlorine or another strong oxidant prior to coagulation.

High arsenic concentrations are frequently found in anaerobic waters. These same waters are generally high in ferrous iron (Fe^{+2}), and manganous manganese (Mn^{+2}), necessitating their removal. Iron and manganese removal processes relying on the oxidation, and subsequent precipitation of the metals as hydroxides, will also effectively remove soluble arsenic by co-precipitation and/or adsorption reactions. It is almost as though the presence of the iron and manganese to be removed is analogous to a natural coagulant addition, as it facilitates the removal of arsenic. Removals of soluble arsenate (As^{+5}) during ferrous iron oxidation and precipitation processes are very significant. This is likely not the case during soluble manganese (Mn^{+2}) oxidation alone. In such cases, the addition of iron salts is indicated.

Of the arsenic removal processes available and discussed in this bulletin, iron coagulation, whether practiced primarily for the removal of arsenic, or also of iron and manganese, appears to be the most promising and proven process available. This process is capable of the removal of 90% or more of any arsenic present. Furthermore, many such plants are already in existence, and operating efficiently, albeit they were initially designed for the removal of iron and manganese only.

Some iron and manganese removal processes (Pureflow© PM-100 is one) rely on a proprietary adsorptive media for their removal efficiencies. In these systems, the iron/manganese (along with any arsenic present) are oxidized, then the iron and manganese are precipitated as hydroxides, adsorbing arsenic. A second adsorptive reaction occurs at the water/media interface where localized zones of high pH assure not only the continued formation, but the maintenance of an active, adsorbent, hydroxide floc.

The mechanism is not unlike the reactions occurring in an Activated Alumina system, with the exception that the adsorptive media process requires no regeneration and can be backwashed like a conventional sand filter. The backwash water can be decanted and reclaimed allowing approximately 99% recovery and water recycle. Since the backwash water is non-hazardous, it can also be drained into a sanitary sewer.

ELECTRODIALYSIS (ED) AND ELECTRODIALYSIS REVERSAL (EDR)

ED is an electrochemical membrane process initially developed for the treatment of saline or brackish waters (Montgomery). Instead of hydrostatic pressure, the process uses an applied direct current (DC) voltage to move dissolved anions and cations from alternate cells through semi-permeable membranes. This purifies a portion of the feed water, while concentrating another. While capable of removing arsenic to low levels, the process is equipment, energy and labor intensive. It also creates a concentrate which must be disposed of, and is quite wasteful of water.

EDR is an ED process which reverses the polarity of the electrodes on a controlled time cycle, which reverses the direction of ion movement in a membrane stack. Reversing polarity provides automatic flushing of scale forming minerals from the surface of the membrane. EDR typically requires little or no pretreatment to minimize fouling of the membrane. ED/EDR systems are not considered to be economically viable for any but very small installations.

NANO-FILTRATION

This process, also known as “membrane softening” uses an ultra-low-pressure membrane designed to allow only passage of particles less than 1 nanometer (10 Angstroms) in size. It is, thus, very efficient (more so than Reverse Osmosis) in the removal of dissolved matter, but is, of course, not selective for arsenic only. Like all other membrane processes, extensive pretreatment is necessary to prevent fouling of the delicate and expensive membranes caused by particulate matter, scaling, or biofouling.

COAGULATION ASSISTED MEMBRANE PROCESS (CAMP)

Coagulation assisted membrane process (CAMP) is considered to be a promising technology for arsenic removal because it can be applied over a wide range of water quality that contains high turbidity, iron, manganese, sulfate and nitrate. Low pressure membranes (e.g. microfiltration and ultrafiltration) are very effective in removing particulate arsenic, but without a pre-coagulation step, low pressure membranes are ineffective at removing soluble arsenic.

Metal based coagulants, such as ferric chloride, can be used to bind the arsenic which is removed with the ferric floc on the membrane. The use of low pressure membranes eliminates the breakthrough of arsenic-laden coagulant flocs (a typical occurrence with conventional granular media filters) by taking advantage of the membranes’ particle barrier. Factors affecting the CAM process include ferric chloride dosage, pH, mixing and floc formation (contact time). As with all membrane processes, provision for adequate pretreatment to control feed water quality should be taken to protect the membrane from fouling caused by particulate matter, scaling and biofouling to optimize membrane performance and life. Disposal of the reject coagulant (which is not considered to be a hazardous waste) can be to a sanitary sewer.

PILOT TESTING

Due to the fact that varying ground water quality can significantly affect arsenic removal processes, pilot testing at each well site is recommended. Raw water quality analyses should be made prior to pilot testing to determine all of the constituents in the water that can affect arsenic removal processes. The pilot filter system should be designed to treat all of the constituents in the raw water that will affect the efficiency of the treatment process. On-site testing should be verified by raw and treated water samples that are tested by an independent certified laboratory. The pilot process should include pretreatment equipment as dictated by the raw water quality analysis to assure continued treatment to below EPA standards and to maximize process runs and optimize media/membrane life. The pilot system must verify removal of arsenic throughout the process run cycle as well as determine pretreatment chemical requirements, and the following costs: operations, labor, media disposal and replacement, membrane disposal and replacement, and/or regenerant brine disposal.

CONCLUSION

Arsenic, long known as the poison of choice because of its legendary toxicity at high doses, has been implicated in skin and internal carcinogenesis, thus the requirement to be regulated at the current EPA level of 0.010 mg/l or 10.0 µg/l.

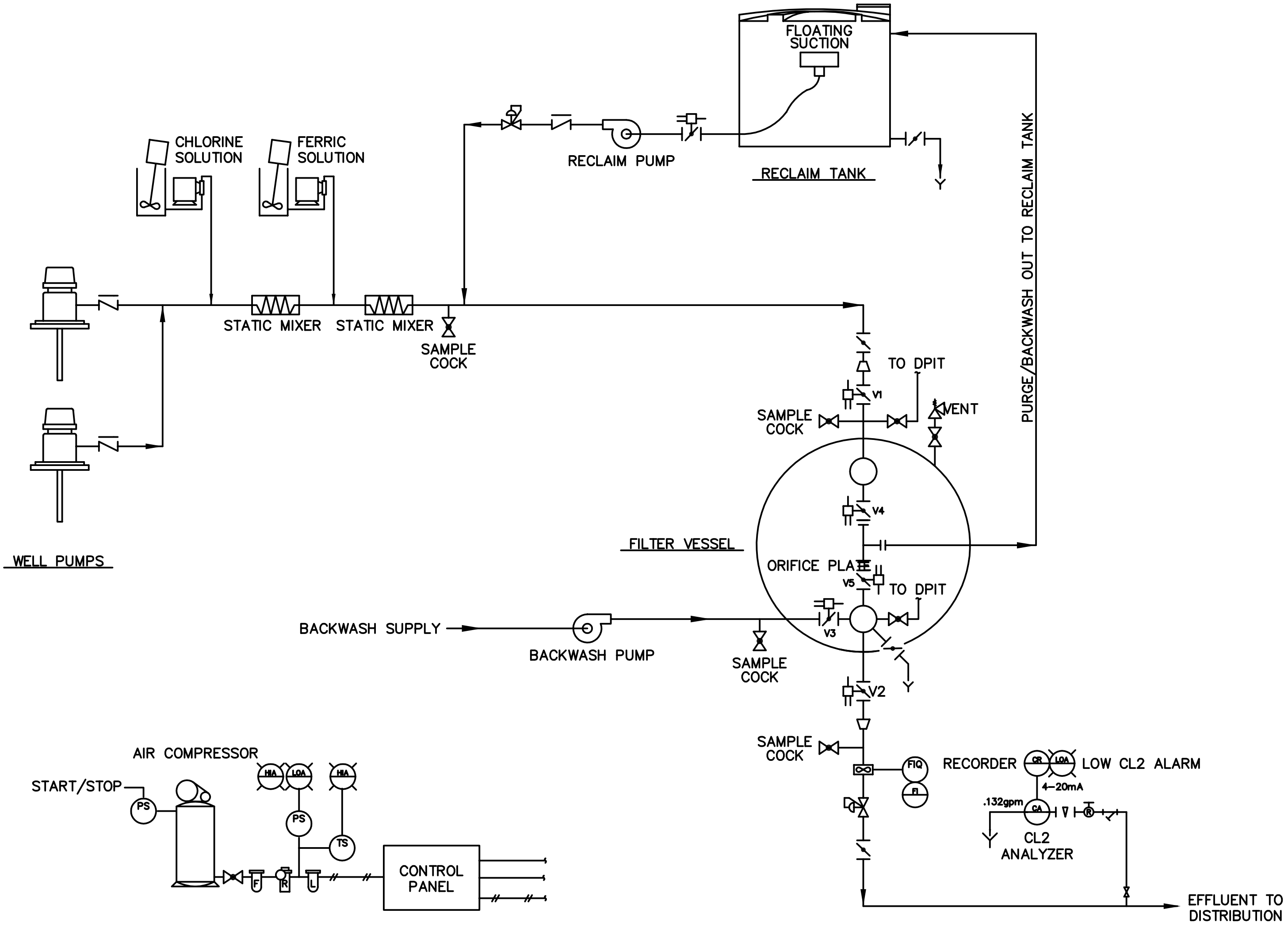
The new MCL will largely drive and dictate the installation of arsenic removal processes. Of the removal processes available, iron and arsenic oxidation, followed by coagulation/ filtration, appears to be the most promising. Meanwhile many researchers are exploring modifications to conventional treatment, with a particular emphasis on enhancing existing coagulation/flocculation operations.

Pilot testing should be conducted at each well site to verify arsenic removal rates, process design and operational costs.

APPENDIX

This bulletin was prepared for Pureflow© by Frank Baumann, PE., formerly Chief of the Sanitation and Radiation Laboratories Branch of the Department of Health Services, State of California. It provides a perspective on arsenic, an important, somewhat mysterious, and somewhat controversial contaminant of water. This bulletin explores the sources, chemistry, occurrence and toxicity of arsenic, as well as possible methods for its removal.

Opinions contained herein are those of the author, and do not necessarily reflect policy of the State of California Department of Health Services.



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DWG. NO. 0474
JOB NO.
SCALE
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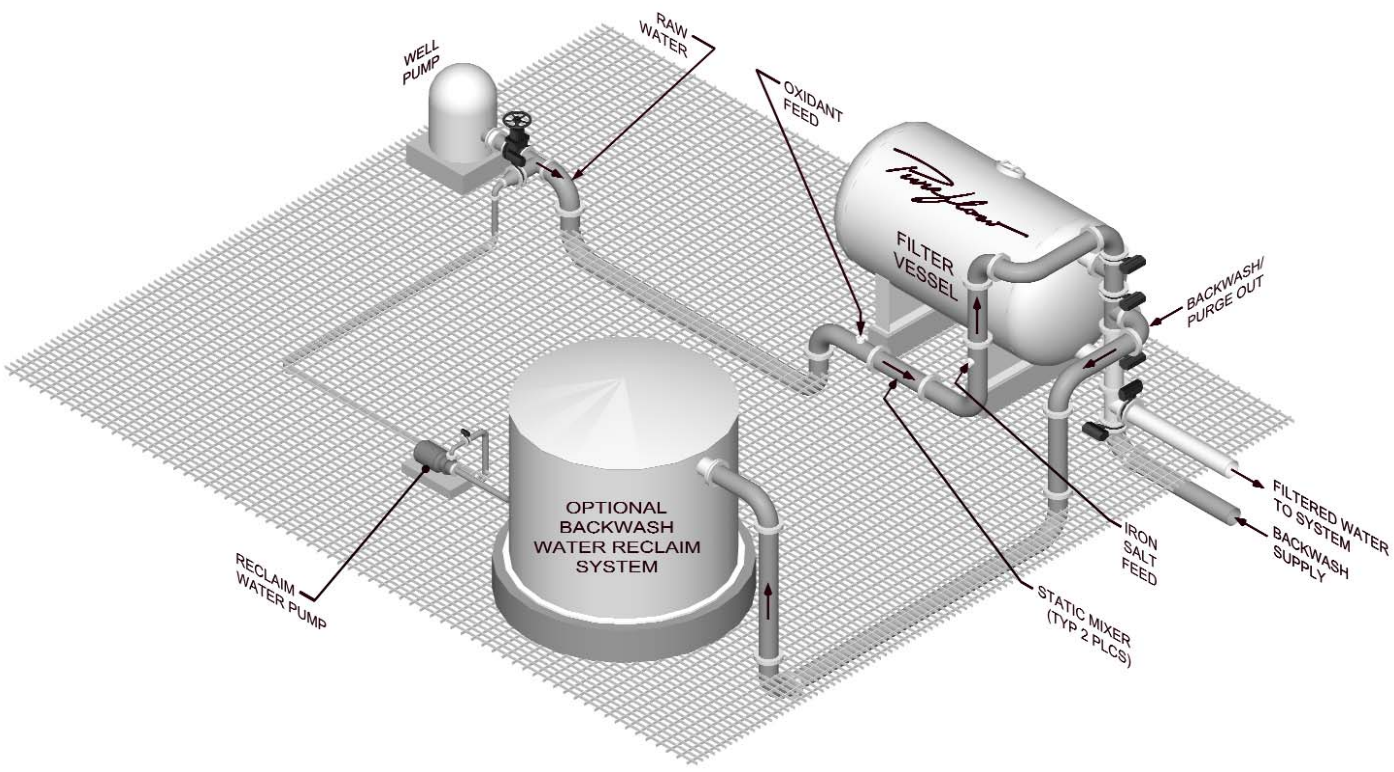
REV. 3

*COAGULATION / FILTRATION PROCESS FOR ARSENIC REMOVAL



TYPICAL INSTALLATION

*Refer to pages 7 and 8 for a description of the coagulation/filtration process for arsenic removal

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NOTES

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