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Title: Perspective Review: Bromide, Ways of Exposure and Environmental Effects.

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Abstract. Nowadays, individuals come to contact with many health hazards without awareness of the ways of exposure, one of these is bromide exposure. Humans directly ingest many forms of bromide in food, water and medicine. Other indirect routs to contact with this element are swimming pools, electronic products, fire retardants materials, dyes and sanitizers. People are exposed to bromine every day through their jobs as individuals working in ports, cloth shops, farmers or even military. Exposure to bromine in both organic and inorganic forms does not only have effect on human health but also have environmental impacts. Some of the routes of exposure and how it affects the human health through water and food cycle will be highlighted.

Key words: Methyl bromide, Fumigation. Brominated vegetable oil, water disinfection.

Introduction

Bromine (Br) is located in the element periodical table intermediate between chloride and iodine. To achieve the full electron configuration as krypton inert gas, bromine need to gain an electron and may exist as (Br_2) or more preferable is converted to bromide salts (Br^-). Thus, bromine which is more chemically reactive than bromide does not occur free in nature, but exist in colorless soluble crystalline mineral halide salts[1].

Halide salts naturally occur in water as a dissolved form. It exists with sodium bromide (NaBr), potassium bromide (KBr) and other cations such as calcium[2]. Bromate is another type of ion, in which oxygen atoms occur as well (BrO^{3-}). Both inorganic and organic bromide such as methyl bromide CH_3Br (MB) and Ethylene dibromide (BrCH2CHsBr) may be present in various environmental media and commodities[3]. The two forms require different analytical methods to identify.

Routs of Bromine Exposure

Calcium bromide brine (CBB)

People who work in the oil industrial field can come to contact to CBB which is a mixture of calcium bromide and calcium chloride. It is used as a completion fluid to control wellbore pressure in oil and gas operation[4].

The exposure to the dry constituents of CBB as well as the aqueous solution may cause tissue loss, particularly if in prolonged contact with the skin. There is considered to be no significant hazard of systemic toxicity through contact with intact skin[4].

Methyl bromide (MB)

Methyl bromide (MB) is a natural substance manufactured for many uses. Although toxic, it is used as fumigant to control pests in the

soil[5, 6], food and cereals. MB is used to control insects and to fumigate areas in flour and feed mills[7]. Due to globalization of trade, the fear of spreading alien species leads to increased use of fumigation with MB. It is therefore used on nonfood commodities including cloth, and furniture before shipment [8, 9] as well as warehouses, buildings and cargo ships. [10, 11, 12] It is also used on small barges transporting goods in small rivers and water canals. Recently, regulations require fumigation with MB of wooden packaging, flooring and wooden goods in imported cargo containers[13].

The imported containers and the fumigated products are shipped deep inside the destination country before being opened, unloaded, distributed and used by workers to reach the final public consumers who are also in the danger of fumes exposure. Exposure due to off gassing is likely since MB persists on clothes, leather, and rubber brought home as well as storage facilities where highly fumigated products are stored [11, 14, 15].

There are two more factors which increase the risk of using MB, First is the wind factor and second is the leakage from the facility[16]. As much as 51–95 % of MB escapes into the air[17] and contribute to ozone depilation which affect human health. As a result, there will be an increase in the concentration of MB used in the facility to overcome leakage and protect grains without much loss. It was suggested that a higher concentration than 5 part per million (ppm) can affect the lung and nervous system[18, 19]. Unfortunately, odor detection is not sufficient to provide adequate warning against potentially toxic levels. Also, MB is three to four times heavier than air, and can spread efficiently near ground level[20].

The absorption of MB usually takes place by the lungs; therefore, expected toxicity in humans and animals is through inhalation of MB gas [21, 15]. It may be also absorbed from the digestive tract. The liver may show degeneration, and the spleen, kidneys and stomach may be hyperemic. In contrast, it is hardly absorbed through the skin, although it may cause severe skin damage[22]. As a result of exposure to MB, malaise, headache, visual disturbances, nausea and vomiting may appear[23]. If the exposure was severe whether by duration or intensity, tremors develop, passing to convulsions, epileptiform, this may lead to coma and death[24, 25]. Small exposure as 0.9 ppm bromine (Br_2) for 5 min. results in coughing, headache, and irritation of the eyes, nose and the upper respiratory tract[26].

MB is also used in quarantine pre-shipment (QPS) of goods, plants seeds and other structures and it's still in use in almost all countries. According to the United Nations Environment Programme (UNEP), Saudi Arabia's use of MB in QPS is considered low. Table (1) shows the Saudi consumption of MB in QPS from 2000-2016 in metric tons which declined from 42 to 15 tons. Before 2000 no Data has been found.

Table 1: The consumption of methyl bromide in Saudi Arabia QPS from 2000-2016 in metric tons.

Year	Quality intone	Year	Quality intone
2000	No data	2009	10
2001	No data	2010	15
2002	13	2011	18
2003	35	2012	15
2004	42	2013	12
2005	42	2014	15
2006	6	2015	15
2007	6	2016	15
2008	No data		

http://ozone.unep.org/ar/data-reporting/data-centre 13/2/ 2018[27]

In terms of production Saudi Arabia is not a producer of MB. In contrast, United States, China, Japan, India and Israel are big producers (Table 2) in which Saudi is still importing food and goods from. Japan consumption and production of MB is exactly the same.

Table 2: Major countries in Production and Consumption of methyl bromide in metric tons in 2016.

Country name	Production	Country name	Consumption
China	1410.528	China	1141.6262
India	2510.654	India	1180.873
United States	3041.6	United States	739.6
Israel	1316.9	Israel	10.4
Japan	434.068	Japan	434.068

http://ozone.unep.org/ar/data-reporting/data-centre 13/2/ 2018[27]

The several hundred tons of MB used annually in major ports around the world for the fumigation represents a significant environmental and human health risk[28, 29] Fumigation of delivery containers with MB is a standard procedure, particularly in Asia[30]. Fumigation is a major source of both MB and bromides which can be produced from the former[31].

The effect on health and environment is greater in under developed countries where there is no strict monitor to bromide levels and not all workers are certified to deal with MB. The claim that it has a short half-life of 7 month or it decomposes in the soil to other products might be not enough to overcome the steady increase of bromine levels in the environment this increase will lead to a further increase in ingested bromide by humans and animals.

Methyl bromide in fire retarding and distinguishers

More than half the bromine produced worldwide each year is used in the fire retardant industry. Methyl bromide is a volatile liquid incapable of ignition or of propagation of a flame. At high temperatures, MB dissociates to form free bromine atoms, in a process that stops free radical propagation chain, hence stops fire. On the other hand, at high temperatures it splits up to form hydrogen bromide (HBr), which is a toxic gas. Unfortunately, the same property causes sunlight to yield free bromine atoms in the atmosphere, causing ozone depletion. Applying bromide fire retardants have not been stopped on furniture and wooden materials. Brominated compounds are also widely used as flame retardants for printed circuit boards which exist nearly in every mechanical and electronical machine. The thermal disposal of these materials adds dangerous hydrogen bromide, and high molecular weight brominated compounds causing additional hazard to the environment[32].

A possible link between MB and serious health problems are mentioned in many references. This include prostate cancer risk[33] from occupational and community exposure. MB is also a genetic[33] and neurotoxic agent[34]. Despite it is genotoxic, MB has not been classified as a human carcinogen. Instead it's only considered as a health and environmental hazard[35]. Ozone depletion, insect resistance, and residues on grain surface are the problems with the use of these chemical fumigants[25].

Potassium Bromate ($KBrO_3$)

In the old days, bakers depend on oxygen in the air to act as an oxidizing element to form molecular bridges. They expose flour after milling to open air for weeks. Alternatively, potassium bromate chemically age's flour in a short period with a beautiful white fluffy soft end product. The amount allowed for this procedure is 15-30 ppm. Under perfect conditions this amount should be converted to potassium bromide during baking. However, if bromated flour isn't baked long enough or baked at a high temperature, or if high amounts of potassium bromate is added in the first place, the final product will contain greater quantities of this harmful additive. Potassium bromate appears as E number E924 on food labels[36, 37]. Foods that contain potassium bromide include breads, tortillas, crackers and other baked goods. Since 1990 many countries such as China the European Union (1990), Canada (1994) and Brazil has banned the use of Potassium bromate as an oxidizing agent in the flour industry. However the United States is still using it. Instead of banning BM, Food and drugs Administration (FDA) reduced the amount of potassium bromate to 30 ppm or just put a carcinogenic warning as in California State[38]. Saudi Arabia is still importing flour and wheat products and potato chips from the USA on which MB is still used.

Brominated Vegetable Oil (BVO)

Brominated vegetable oil (BVO) is made by adding bromine across the double bonds of fatty acids in vegetable oil usually soybean. Like plain vegetable oil BVO dose a good job of dissolving water insoluble food flavor, fragrance and coloring agents which serve as a carrier for these agents in soft drinks[39]. Although BVO is prohibited in many countries 8ppm was detected in North American's soft drinks.

Bromide in vegetables and fruits

Methyl Bromide is the most widely used chemical fumigants for fresh fruits, vegetables and flower cuts[40]. Despite MB use as a pesticide is declined under the Montreal Protocol (2005) mainly because its role in ozone depletion[41], many countries are still using it such as Australia, USA and Japan in less quantity. Some crops, like strawberry and potato, are exempt and MB is still used on them as a pesticide. Most countries continue the use of MB under the critical use exemptions (CUE) clause. The CUE allows continued use of MB where no adequate alternative is available[42]. In Saudi Arabia, it seems that date fumigation with MB was stopped in accordance with the Montreal Protocol on the first of January 2015[43]. No clear information about other crops is available.

In 2003, MB was the most commonly used pesticide among California growers[44, 45]. In addition, since 2001 grapes are required to be fumigated in the US with MB[46, 47]. Consequently many agricultural products contain bromide predominantly strawberries grown in California. United States is the largest strawberry producers and fumigates strawberries exported to Asia with MB[48].

Bromide is detected in vegetables. The concentration of bromide ranged from 3.65 to 14.42 mg kg–1 in capsicum, 4.50 to 9.30 mg kg–1 in potatoes, and 3.63 to 19.02 mg/ kg in fungi. All content of bromide in these vegetables were below the concentration of 20mg/Kg which is established by Spanish legislation[49]. In contrast, in areas still using bromide in agriculture as in Japan[50], it was found very high bromine content in different vegetables which may reach 1000-16200 ppm.

Bromine and water

Bromate formation during oxidative treatment of bromide containing drinking water has been a source of concern ever since bromate

was classified as potentially carcinogenic by the International Agency for the Research on Cancer (IARC)[51]. One of the coast effective procedure to disinfect drinking water and treatment of wastewater is ozone[52]. Unfortunately, harmful bromate ion is formed during the process.

Bromate may be found in some drinking-water samples as a by-product of ozone disinfection. Elevated bromide levels in rural areas may be attributed to bromine leaching from organic matter in soil and biomass [53, 54]. The amount of produced bromate also increase with many factors such as bromide ion concentration, pH of the source water, the amount of ozone and the reaction time used to disinfect the water. Ozone applications on water is not restricted on disinfection only it is also used in reducing taste and odor[52].

Bromine is a microelement present in waters, both in inorganic and organic bromide but at lower concentrations[55]. The concentration of naturally occurring bromide in water summarized in Table (3). Oceans are a net sink for atmospheric MB, where it is slowly degraded by chemical and biological processes[56].

The main byproduct formation in the presence of bromide during the ozonation of drinking water is bromate. And the level of bromide depends on the amount of bromide found in the source of water. Saudi Arabia's ground water is naturally high in bromide content and the process of water sterilization with ozone will result in more bromide production. This is shown in a study of bottled water in Saudi which detected two times more bromide than the average labeled on the bottle and some exceeded the permissible limits by KSA and WHO which is $\leq 10\mu g/l$[57]. On the other hand, the a study for the surface water quality in the eastern province of Saudi Arabia was affected due to agricultural activities[58].

Fumigation with MB as pesticide and irrigated with regenerated waste water will contribute to the increase of bromine recycle in food and environment[59]. Freshwater contamination leading to increased salinity probably occurring with a parallel increase in bromide. In addition, the runoff bromide following winter salting of roads also affect water and soil in the area[60].

Table (3): The concentration of naturally occurring bromide in water (mg/ml).

Water	Br^-(mg/ml)	Ref
Sea water	65-90	[60]
Dead sea	5000	[61]
Underground water	0.1	[55]
Fresh water	0.5	[62]
Desalinated water	1	[62]
Bottled water	0.11-0.039	[55]
Tab water	0.1-1.0	[55]

Swimming pools and spas

Since the early 1900's, the use of seawater in swimming pools and water parks has been widely practiced in Europe or on cruise ships and ferries[63]. The normal bromide level in the sea water is reported to be between 65-90 mg/L Table (3).

The disinfection of the water of these leisure places weather by chlorination or bromination lead to the formation of brominated disinfection by products (DBPs) in seawater swimming pools treated with chlorine. The high levels of brominated trihalomethanes (THM-Br) and brominated acetic acids

(BAA) found as carcinogenic and mutagenic brominated DBP such as brominated trihalomethanes and brominated haloacetic acids were found in swimming pools and presented a real problem of health and safety. The levels of these by products were measured at levels up to 18 folds greater than the maximum contaminant levels of 60 and 80 mg/l in drinking waters. Swimming pools contained a high amount of human body fluids such as urine, sweat, hair and skin [64, 65], perfumes, care products cosmetics and sunscreens [66] and some stools. This resulted in the formation of chlorination byproducts. The process of pools and leisure disinfection involves input of high concentrations of chlorine or bromine products. Both bromination and chlorination of bromide-containing water, including the natural sea water, is known to produce brominated by products[67, 68, 69] These brominated compounds tend to show higher toxicity than their similar chlorinated compounds[70, 71, 72, 73]. Elevated bromide levels together with the continuous inputs of chlorine-based disinfectant in addition to the organic compounds brought by bathers explain the highest concentrations of THM-Br compounds in these pools[74].

Because of the long plasma half-life of bromide in

Bromide in medicine

Bromine exists as a base in over-the-counter pharmaceuticals such as brompheniramine maleate used for colds and allergies, and hyoscine hydrobromide used for travel sickness[75]. Elevated levels can result from the use of common bromine based over-the-counter pharmaceuticals such as those used for colds and allergies[75].

Bromide exists in many pharmaceutical preparations as hydrobromide such as FHBr or as bromide salt forms such as Calcium Bromide (CaBr) which is used in neuroses medication. Formulas like $C_{18}H_{28}BrNO_2$ or $C_{20}H_{22}BrFN_2O$ may exist under different names with no indicator to inexperienced people that the formula may contain bromide. The percentage of these compounds in preparations ranges from 2-5%[76]. Bromide is also used in the military as a medicine to protect solders against possible nerve gas attacks[77].

How much of bromide is safe

The daily intake of bromide from normal diets is an average of 8 mg/day for adults[75]. The joint FAO/WHO acceptable daily intake (ADI) for humans is 0-1mg/Kg body weight[78]. Blood levels of inorganic bromide in the general population have been quoted as 1-5.5 mg/l[79, 80].

A random study in Australia on male and females aged from 18-71 years indicated that bromide level in human blood was 5.3 mg/l with female slightly higher bromide than males (5.5 mg/ml). The same study showed no associations between bromide levels and variables including age, gender, weight, height, and postcode address[81]. According to the U.S. Department of Health and Human Service (1978) the threshold for bromide inhalation is 20 ppm which is equal to 80 mg/m^3. The half-life of bromide in plasma of human is considered to be long (10-12) days[23].

Bromide is also used in a wide range of products for example, potassium bromide in hair styling products. Other forms of bromide such as lithium bromide are used in explosives, wool, hair industry and in cooling systems[82, 83]. Photography, optics, spectrophotometry, paper industries, anti-convulsion and antiepileptic medication for pets[84] dyes and sanitizers all contain bromide products[85]. Bromide presents in coal at trace levels (<0.001 %). Burning coal causes its bromine content to be oxidized and enter the atmosphere as a toxic air pollutant with a consequence hazard on health and the environment[86, 87]. Finally, continuous wars

with the use of bromide in the military have its effect on fresh and underground water. A study in Iraq concluded that both water from Euphrates River and the ground water in some regions contains very high levels of bromine and this is due to the gulf war[88].

Conclusion

It is clear that there is a high daily exposure of inorganic and organic bromide through a wide range of routes Significant quantities of bromide are being discharged into the environment which affect human health and increase bromine levels in both water and soil. Being a country in conflict and war regions, we are exposed to more bromine levels. As the 2030 vision approaches with huge changes, Saudi Mills will be sold to foreign companies which may still use and produce MB and should be monitored closely for the health of our citizens. Precaution and research is recommended to investigate the impact of bromide on workers exposed to this element through jobs like farming, fumigation and quarantine sectors since symptoms can be delayed from hours to days after exposure[35]. When ozonation is considered as a treatment of drinking or waste water strict limits and continuous monitoring on the acceptable levels of bromate levels must be applied.

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مراجعة منظور: البروميد، طرق التعرض والآثار البيئية

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مستخلص. في الوقت الحاضر، يتم التعرض الي العديد من المخاطر الصحية دون وعي بطرق مختلفه واحدي هذه المخاطر هي: التعرض للبروميد. ويكون ذلك بطريقه مباشرة كتناول العديد من أشكال البروميد في الغذاء والماء والدواء. وهناك طرق أخرى غير مباشرة للاتصال بهذا العنصر وهي حمامات السباحة والمنتجات الالكترونية ومواد مقاومة الحريق والأصباغ والمطهرات. وقد يتعرض الافراد للبروم يوميا من خلال وظائفهم مثل من يعملون في الموانئ ومحلات القماش والمزارعين أو حتى في المجال العسكري. إن التعرض للبروم في الأشكال العضوية وغير العضوية لا يؤثر فقط على صحة الإنسان ولكن له أيضا آثارمضره علي البيئة. وسيتم تسليط الضوء على بعض طرق التعرض وكيفية تأثيره على صحة الإنسان من خلال دورة المياه والغذاء.