Chlorates:
A Literature Review of Aspects
Relevant to the Aquatic Environment

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Introduction

Chlorates are powerful oxidizing agents used for several industrial, agricultural and pharmaceutical purposes (Table 1). They can form explosive mixtures with common materials and must be handled with caution. Chlorates also occur as inorganic by-products of processes that use chlorine dioxide; they are present in bleached kraft pulp mill effluents and water disinfected with chlorine compounds. Their occurrence is expected to expand with increasing chlorine dioxide substitution at bleached kraft pulp mills (Bonsor et al. 1988). A few studies have focused on the effects of chlorates as water pollutants, including field and laboratory experiments. Chlorates can be toxic to humans and animals, and can be harmful to the environment. The purpose of this document is to summarize the aquatic information available on chlorates, and to provide a list of pertinent literature.

Methods

A comprehensive literature search was conducted through the Alberta Environment library. Initially, not many references were found specific to chlorates, so the literature search was expanded to include sodium, potassium, calcium and magnesium chlorate. General information was obtained from chemistry, hazardous chemicals and pesticide reference books. Also, six computer databases were searched. The most useful was the Hazardous Substances Data Bank (HSDB). This source, and the Register of Toxic Effects of Chemical Substances (RTECS), are on-line databases available from the United States' National Library of Medicine that provide specific chemical data and references. The other four databases searched were Enviroline, Biosis, Water Resources Abstracts and Chemical Abstracts. These are on-line indexes to published literature.

Chemistry

Chlorates are oxyhalogen compounds that are a combination of a metal or hydrogen cation and the ClO_3^- monovalent anion. They are manufactured by the electrolysis of chloride solutions. For example, to produce sodium chlorate, the reaction is:
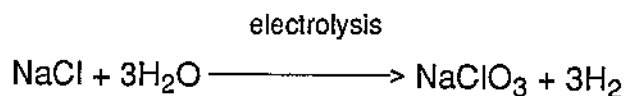
$$NaCl + 3H_2O \xrightarrow{\text{electrolysis}} NaClO_3 + 3H_2$$

Table 1. Various uses of chlorates.

| Compound	Type of Use		Percentage Use for each compound
sodium chlorate	industrial	bleaching agent for paper pulps	85
		perchlorate formulation	6
		uranium ore processing	4
		explosives and matches; pharmaceutical aid;	
		dyeing and printing fabrics; tanning and finishing leather;	2
		rocket fuel oxidant	
	agricultural	soil sterilant; cotton defoliant; dessicant; harvest aid	3
potassium chlorate	industrial	match tips	80–90
		pyrotechnics; explosives and propellants;dyeing of furs;	
		cotton and wool; pulp and paper manufacturing	10–20
	pharmaceutical	gargles; mouth wash; dentifrices and throat lozenges	
calcium chlorate	industrial	processing waste gases and boiler waste; pyrotechnics;	
		photography; potassium chlorate production	nd
	agricultural	herbicide; insecticide; seed disinfectant	
magnesium chlorate	agricultural	formerly used for cotton defoliation	nd

Note: nd = no data

Sources: Farm Chem. Hdbk. 1988; HSDB on-line

Chlorate is commercially produced for the pulp and paper industry by the reaction of hypochlorous acid and hypochlorite ion (Aieta and Berg 1986). The reaction is:

$$2HOCl + OCl^- = ClO_3^- + 2H^+ + Cl^-$$
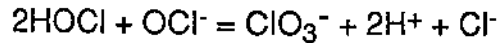
Manufactured chlorate products are colourless, white or pale yellow crystals, and are very soluble in water. There is no indication in the literature that the chlorate ion occurs naturally, except as a by-product of the manufacture, and breakdown in acidic solutions, of chlorine dioxide (ClO_2). In fact, only one oxyhalogen compound, sodium iodate, does occur in nature in appreciable quantities. It is found in sodium nitrate beds in Chile (Sisler *et al.* 1961).

Chlorates are strong oxidizing agents and are stable at normal temperatures when kept isolated from other materials (HSDB on-line). Chlorates themselves are not explosive but they form flammable or explosive mixtures with organic matter, sulfur, sulfides, powdered metals and ammonium compounds. Cloth, leather, wood and paper are extremely flammable when impregnated by chlorates. Even water solutions react in this way if chlorate concentrations are greater than 30%, especially if the solution is warm (Weiss 1985). There have been warehouse explosions and severe fires caused by chlorates (Envir. Can. 1985). When on fire, chlorates melt and decompose to yield oxygen gas that increases the intensity of fire; the breakdown reaction is:

$$2NaClO_3 \longrightarrow 2NaCl + 3O_2$$
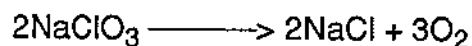
Chlorates also yield toxic chlorine fumes when on fire. Water is the best agent for fighting fires involving chlorate, but runoff from fire control or dilution may result in pollution (HSDB on-line).

Sources

Chlorates occur in solution as a by-product of chlorine dioxide when used for certain industrial processes, particularly pulp bleaching and water disinfection. Chlorine dioxide is an unstable chemical that cannot be transported effectively, so all bleached kraft pulp mills prepare it on-site from sodium chlorate and one or more reducing agents such as sulphur dioxide or methanol (Bonsor *et al.* 1988).

Chlorine dioxide is an attractive alternative for chlorine in drinking water

disinfection because it effectively inactivates bacteria and viruses over a broad pH range and does not produce carcinogenic trihalomethanes. All chlorine dioxide for drinking water treatment is generated from sodium chlorite (Aieta and Berg 1986). The reaction is:

$$2NaClO_2 + Cl_2 = 2ClO_2 + 2NaCl$$
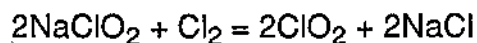
In addition, chlorate ion may be formed in the generation system as an undesired by-product by the competing reaction:

$$NaClO_2 + Cl_2 + OH^- = NaClO_3 + HCl + Cl^-$$
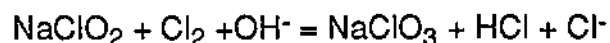
In alkaline solutions, chlorine dioxide disassociates to form chlorite and chlorate and in acidic solutions with exposure to sunlight, chlorine dioxide decomposes to yield chlorate (Condie 1986). Also, chlorate may be present as a feedstock contaminant from water intake supplies (Aieta et al. 1984). However, very little chlorate is formed in drinking water treated with chlorine dioxide, assuming an efficient generation system is used (Aieta and Berg 1986).

Human Toxicity

The available information on the toxic effects of chlorates on humans consists mainly of summaries of toxicological data and reports/observations of accidental or suicidal poisonings (HSDB on-line; RTECS on-line).

The principal toxic effects of chlorates are the production of methemoglobin in the blood and destruction of red blood cells (Sax and Lewis 1989). The latter may lead to irritation of the kidneys. Damage to heart muscle has been reported. The probable oral lethal dose of sodium chlorate for humans is 50 to 500 mg of chlorate per kg of body weight (mg/kg) (HSDB on-line). This amount is between one teasponful and one ounce for a 70-kg person. A common source of poisoning by potassium chlorate is ingestion of match heads; a lethal dose of this chemical for adults is estimated to be 5 to 30 g. Lethal doses of calcium and magnesium chlorate were not reported in the literature. The toxic symptoms and effects of chlorates on humans are listed in Table 2.

Numerous studies have been conducted with chlorate, as a by-product of chlorination processes in water disinfection. A short-term study that involved administration to human volunteers of chlorate in drinking water (up to 2.4 mg/L)

Table 2. Toxicity of chlorates to humans.

Route of Exposure	Symptoms and Effects
skin contact	not readily absorbed but does cause irritation
eye contact	irritation; conjuctivitis
inhalation of dust	local irritation of skin and mucous membranes; respiratory difficulties; may cause respiratory failure
ingestion	absorbed in alimentary tract[a]: gastroentric pain; nausea; vomiting; diarrhea; dyspnea; cyanosis; hemolysis; blood urea increase; acute nephritis; anuria; kidney injury; liver injury; convulsions; coma; death

Note: [a]effects listed in approximate order of increasing severity
Sources: Envir. Can. 1985; RTECS on-line

did not result in any detectable alterations in blood parameters, serum or urine chemistry values, or adverse physical symptoms (Lubbers *et al.* 1982). Similarly, no effects were detected after 84–day exposures of volunteers to 5 mg/L of chlorate in drinking water. Another study found that when humans drank water containing chlorate concentrations of less than 5 mg/L, there were no adverse thyroid effects (Bercz *et al.* 1982). In the same study, there were harmful blood formation effects to monkeys that drank chlorate up to 400 mg/L (as ionic equivalents to chlorine dioxide). Rats, mice and chickens treated with chlorate in drinking water, at concentrations of up to 100 mg/L, also demonstrated alterations in formation of blood components (Couri *et al.* 1982).

Environmental Effects

Terrestrial Aspects

Chlorates are formulated as herbicides and defoliants. They act as nonselective contact poisons, are translocated and may be absorbed from soil to kill both plant roots and tops (Farm Chem. Hdbk. 1988). Soil applications are best for a sterilant effect. Chlorate compounds, however, can be sprayed or applied dry to soil or plants, for control of both grasses and broadleaf weeds, and to kill trees and stumps. Chlorates kill all plant growth, except moss. When applied in less than lethal doses, chlorates cause chlorosis of leaves and a starch depletion in stems and roots. Sodium chlorate is a non-selective herbicide used against established vegetation at 200–600 kg/ha and against annual weeds at 100–200 kg/ha (HSDB on-line). The Agricultural Ministry of Japan permits the use of sodium chlorate as a weed killer because it is entirely decomposed to sodium chloride in the fields before it is absorbed by rice plants (Ito 1982).

Animal poisonings have occurred when using sodium chlorate as a herbicide (HSDB on-line). Following application of it (amount unknown) to a field, six of fifteen cattle died. In another case, two cattle died and others showed symptoms of poisoning after approximately 0.4 kg of sodium chlorate had been scattered on a pasture to kill thistles. The toxic effects of chlorates on animals are not well known. In lethal doses, however, death occurs by severe methoglobinema. Table 3 shows toxic concentrations and effects of chlorates on some animals.

Table 3. Toxic effects of chlorates on animals. Doses administered orally.

Compound	Animal	Concentration/Amount	Effect
sodium chlorate	mouse	596 mg/kg	LD_{50}
	dog	700 mg/kg	LD_{LO}
	rat	1 200 mg/kg	LD_{50}
	cat	1 350 mg/kg	LD_{LO}
	rabbit	7 200 mg/kg	LD_{50}
	chicken	5 g/kg	lethal
	horse	250 g	lethal
	cow	500 g	lethal
potassium chlorate	dog	1 200 mg/kg	LD_{LO}
	rat	1 870 mg/kg	LD_{50}
	rabbit	2 000 mg/kg	LD_{LO}
calcium chlorate	rat	4 500 mg/kg	LD_{LO}
magnesium chlorate	rat	508 mg/kg	effects on fertility
	guinea pig	1 500 mg/kg	LD_{LO}
	mouse	5 235 mg/kg	LD_{50}
	rat	6 348 mg/kg	LD_{50}
	rabbit	8 660 mg/kg	LD_{50}

Notes: LD_{50} = lethal dose fifty, a calculated dose of a substance that is expected to cause the death of 50% of an entire defined experimental animal population when exposed by any route other than inhalation;

LD_{LO} = lethal dose low, the lowest doses of a substance introduced by any route, other than inhalation, over any given period of time in one or more divided portions and reported to have caused death in humans or animals

Sources: HSDB on-line; RTECS on-line

The Baltic Sea Situation

Chorine dioxide is used in the pulp and paper industry as a bleaching agent. Chlorate occurs in pulp mill effluents in concentrations from 1 to 70 mg/L, depending on the amount of chlorine dioxide used in the bleaching process and on the type and efficiency of the effluent treatment (Lehtinen *et al.* 1988). The amount of chlorate removed in effluent treatment does not seem to be well documented. Pulp mills that use low chlorine dioxide substitution in the bleach plant are able to recover the by-products for make-up in the pulping chemical cycle; the chlorate content of the effluent is insignificant. High chlorine dioxide substitution, however, is being favoured as a method of lowering the amount of chlorinated organic matter discharged in wastewater and when this process is used, it may be impossible to use all of the by-products. Up to 3 kg of chlorate per tonne of pulp may be found in <u>untreated</u> wastewaters (Bonsor *et al.* 1988). For a 1000 t/d bleached kraft mill this would correspond to a maximum of 3000 kg/d. If the mill produced 1 m^3/s of wastewater, this would result in 35 mg/L of chlorate in the <u>untreated</u> wastewater.

The environmental effects of chlorates in pulp mill effluents are little documented. In one area of the brackish Baltic Sea, originally dense stands of a brown algae called bladder-wrack (*Fucus vesiculosus* L.) were eliminated from a 12 km^2 area in the vicinity of a bleached kraft pulp mill outfall after the mill introduced chlorine dioxide substitution in the bleaching process (Lehtinen *et al.* 1988). In 1983, a field study was conducted to determine whether chlorate was the causal agent. Well-developed specimens of bladder-wrack were transplanted from an unpolluted area into the area from which the algae previously had vanished. As a control, specimens were also transplanted within the unpolluted area. The transplanted *Fucus* plants were killed within 3 to 5 months as far as 1.5 km from the effluent source and reduced growth occurred at 4 km from the source whereas the plants within the control area continued to grow (Rosemarin *et al.* 1986).

To confirm the results of the field study, laboratory and model ecosystem experiments were conducted that show chlorates cause harm to bladder-wrack and its associated fauna. In 1983, excised apical fronds were exposed to chlorate and effluent containing chlorate for periods from a few hours to three days (Rosemarin *et al.* 1986). Although the concentrations of chlorate used were relatively high, 0.4 and 4.0 mg/L, similar effects were observed when comparing similar levels of chlorate in mixed effluent and pure chlorate alone.

Polyphenol efflux rates from *Fucus* were enhanced 2–fold in 0.4 mg/L solution and 5–fold in 4 mg/L solution. After 70 hours exposure to chlorate, the nitrate uptake rate was reduced by about one-third in 4 mg/L solution with no effect at 0.4 mg/L solution and the net photosynthetic rate was reduced about two-thirds in 4 mg/L solution and about one-sixth in 0.4 mg/L solution. In another study, the radioisotope chlorate–36 was used as a tracer to show that chlorate in mill effluent was taken up by *Fucus* (Rosemarin *et al.* 1986). Chlorate uptake was determined in 1 hr experiments using excised apical fronds of fresh *Fucus* in 10 mL volumes of seawater. Uptake was linear in chlorate concentrations ranging from 16 µg/L to 16.6 mg/L. Uptake was also linear over time up to 1 hr in these experiments. Pure chlorate and chlorate in mixed effluent were taken up at similar rates. It was also discovered that nitrate stimulated chlorate uptake, contrary to previous findings. This may contribute to seasonally-enhanced uptake of chlorate in receiving waters, in response to variations in nitrate level.

Model ecosystem experiments were conducted in 8000 L circular pools containing 15 L of transplanted *Fucus* (Rosemarin *et al.* 1986; Lehtinen *et al.* 1988). In three 6-month growth seasons (May to November) from 1982 to 1984, a continuous flow of seawater and effluent (48 hr renewal period), including different concentrations of mixed effluent containing chlorate and pure chlorate alone, were tested. The first signs of toxic effects were that primary production was reduced, followed by an impairment of growth of apical fronds and decreased biomass. The whole model ecosystem, from initially being autotrophic, switched into a heterotrophic state at chlorate concentrations of 85 µg/L. In the tests conducted with pure solutions of chlorate, the lowest effective concentration of chlorate at five to six months continuous exposure that damaged bladder-wrack was 10 to 20 µg/L. The effective concentration whereby 50% inhibition occured was observed at about 80 µg/L chlorate. Several other species of brown algae were found to be equally sensitive to chlorate, whereas blue-green, green and red algae showed much lower sensitivities.

The action of chlorate is reported to be due to its reduction to chlorite via nitrate reductase (Rosemarin *et al.* 1986). For this reason, the ambient nitrate level plays an important role in determining the degree of chlorate uptake and toxicity. Low nitrate levels contribute to higher chlorate uptake while high nitrate levels inhibit chlorate uptake. Ammonium can also inhibit chlorate uptake. Rosemarin *et al.* (1986) concluded that chlorate toxicity should therefore occur most often in nitrogen-limited systems.

Secondary effects due to the damage and disappearance of bladder-wrack may include changes to invertebrate macrofauna production and pelagic fish reproduction (Lehtinen *et al.* 1988). The short-term consequence of the damage to bladder-wrack is an increase in total food supply (the decaying bladder-wrack) to the faunal components and hence, invertebrate production will remain normal for a short period of time. The increased edibility of the alga will soon result in a reduction of the available surface needed for invertebrate shelter. Consequently, before the bladder-wrack disintegrates and disappears, it becomes over-crowded by invertebrates. In the long run, the habitat destruction, the lack of space and shelter and the decrease in primary production following the elimination of bladder-wrack results in a shift from herbivore and omnivore species to detritivores and filterfeeders. Although green and red filamentous algae replace the bladder-wrack, only a small part of those animals previously dependent on bladder-wrack can be supported on the replacement species. The shift from perennial to annual algal populations will result in an increased transport of organic material to deep soft bottoms, favoring filter feeders and the fish species that feed on them. Possible long-term secondary effects of the disappearance of bladder-wrack may also include decreased reproduction in fish populations; the effects on herring have been discussed by Lehtinen *et al.* (1988). A decrease in bladder-wrack biomass in the Finnish archipelago has been correlated with higher abundance and biomass of filamentous algae. Unfortunately, exudates from decaying filamentous algae are toxic to herring eggs. This impairment of the quality of spawning substrate may be detrimental to the reproduction of herring; the survival of eggs and larvae may decrease to about a third of the normal value. Thus, the secondary effects of the disappearance of bladder-wrack may result in decreased recruitment of pelagic fish populations.

Freshwater Studies

A few other studies have addressed the environmental effects of chlorates as water pollutants. Experiments were conducted to determine the effects of temperature on the toxicity of three materials, one of which was sodium chlorate, to fish (Shifrer *et al.* 1974). Sodium chlorate was present in the treated sewage at a concentration of 13,690 mg/L (equivalent $ClO_3^- = $ 10,730 mg/L) and accounted for 18.62% of the total contribution required to produce 50% mortality in the bioassays, assuming that the toxicities of all toxicants in the waste were additive. Schifrer *et al.* (1974) also cite work others indicating threshold toxicity of sodium chlorate to 3 species of fish in static

bioassays at 8000–13,000 mg/L.

In another study, the effects of sodium chlorate on natural and simulated stream communities were examined by Matida *et al.* (1975). A herbicide containing 50% sodium chlorate as the active ingredient was aerially spread over a 70–ha area of mountain forest in Japan. The chemical was applied at a rate of 200 kg/ha. Sodium chlorate concentrations in the water increased to 57 mg/L (equivalent ClO_3^- = 45 mg/L) within 30 minutes after the beginning of application, and decreased to 1.9 mg/L (equivalent ClO_3^- = 1.5 mg/L) after 4 days. Concentrations at the confluence of the contaminated and control streams followed a similar pattern; values were high at the beginning of the experiment, 38 mg/L (equivalent ClO_3^- = 30 mg/L) and decreased to 1.1 mg/L (equivalent ClO_3^- = 0.86 mg/L) after 4 days. Sodium chlorate was not detected in the control stream. The herbicide did not significantly affect numbers of species and individuals, or total weight of aquatic invertebrates and trout showed no mortality, histopatholigical or external changes. Also, trout did not descend the stream, which indicates that environmental conditions were not unfavorable. In a subsequent study (Matida *et al.* 1975), the effects of sodium chlorate on an artifical stream community were studied. Sodium chlorate concentrations were regulated so that values simulated those observed when the chemical was aerially applied. Bottom dwelling and drifting invertebrates and fish were not adversely affected by the herbicide.

In other work, calcium chlorate was classified as a low toxicity substance with weak cumulative properties (Avezbakiev and Demidenko 1979). A maximum permissible concentration of 5 mg/L, calcium chlorate and chloride combined, in drinking water was recommended based on taste as the limiting factor; toxic levels to rats and mice were also studied. A freshwater static bioassay by Environment Canada on rainbow trout resulted in an LC_{50} estimate of 1750 mg/L as sodium chlorate (equivalent ClO_3^- = 1372 mg/L) (Beech 1983).

Water Quality Guidelines

There are no air or water quality guidelines specific to chlorate in Canada or the United States (Envir. Can. 1985). Because of the impact of chlorates in the Baltic Sea, Sweden is proposing to regulate chlorate discharge (Kringstad 1988).

Environmental Fate of Chlorates

In general, there is little information on the fate of chlorates in the environment and degradation data are not available. They will not persist in a high-organic-contact environment because they are strong oxidants, and react with organic materials in the presence of sunlight. The longevity of chlorates when applied to soil appears to be site-dependent. They can persist for three to six months (Farm Chem. Hdbk. 1988). When applied at rates of over 300 kg/ha, chlorates give persistent control for about six months but are leached from soil by high rainfall (HSDB on-line). However, it has been reported that chlorates are most persistent in areas of low rainfall where they may remain toxic as long as five years.

Compared to some other chlorinated compounds, chlorates can not be considered persistent (Lehtinen *et al.* 1988). To quote from Bonsor *et al.* (1988, p. 4–83):

> "Chlorate is readily converted to chloride in biological treatment systems. Ekstrom (1987) reported 90% removal at the Nymolla mill in Sweden, so it seems probable that it is reduced naturally in receiving waters in the case of mills which do not have biological treatment systems.
>
> We do not consider chlorate discharges to be a serious environmental problem, although they will almost certainly increase since the current trend toward adopting high chlorine dioxide substitution to reduce total organically bound chlorine discharges will probably accelerate in the near term."

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